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Radon Concentrations in Mineral Waters of the Sikhote-Alin, Primorsky Krai

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Abstract—The geological and hydrogeological conditions of 19 occurrences of thermal waters in the southern Russian Far East are considered in this paper. Radon concentrations were measured in waters, and uranium and thorium concentrations were determined in host rocks and secondary minerals. The results were compared with the chemical composition of waters and gases in the mineral springs. It is established that water-bearing rocks are classified into rocks of low and normal radioactivity. The contents of uranium (<0.56 μg/L) and thorium (<0.1 μg/L) in the waters are low. Measurements of radon concentrations in mineral water springs in Primorye indicate that most of them are weakly radioactive (0.2–1.5 kBq/L), whereas two are moderately radioactive (1.5–7.5 kBq/L). From a medical-balneological point of view, waters in 7 of 19 springs were defined as radon waters, with 4 of them classified as nitrogen thermal water and 3 as CO_2-rich mineral waters. The CO_2-rich mineral waters of the Shetukha Group exhibit the highest level of radioactivity. Such natural objects reflect the state of the geological environment and can be utilized both for medical purposes and for monitoring a wide range of hydrogeochemical parameters. Based on the analysis of geological conditions and geochemical features of thermal water occurrences, the factors that determine the radioactivity level were identified and general schemes for the radon transport to the surface in different geological and hydrogeological conditions were proposed.

Keywords: mineral waters, radon, uranium, thorium, thermal waters, CO_2-rich mineral waters, Sikhote-Alin, Russian Far East

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INTRODUCTION

Discovery of the spontaneous decay of uranium, thorium, and other elements at the boundary of the 19th–20th centuries attracted geologists and geochemists to the problem of natural radioactivity of the Earth's crust. Numerous studies established that radon accounts for the most part of total radiation dose in the radiation background of the Earth. The studies of radon in natural objects have several applied aspects: ecological (radon hazard), medical (balneological), and geological (prediction of earthquakes and volcanic eruptions; marker of groundwater recharge area, and so on). Radon studies are widely carried out to determine the radon abundance in subsoil air, since ^{222}Rn in building from ground air significantly contributes to the radiation dose of inhabitants. Depending on the geological conditions, radon concentrations in groundwaters differ. The high therapeutic effect of radioactive waters was proved by numerous experimental and clinical studies and is widely discussed in literature [3, 4, 7, 18]. Although Far East hydromineral resources have been studied for over 100 years and, from the very beginning, have traditionally been considered from the therapeutic point of view, the radioactivity of waters from most natural springs of Primorye has been unknown yet. Few data on the radioactivity of CO_2-rich mineral waters of the Shmakovka spa are reported in [19], and data on the Shetukha Group are reported in [25].

The aim of this work was to determine the radon contents in mineral waters of the Sikhote Alin and to elucidate the reasons and factors that define the radioactivity level of different types of mineral waters.

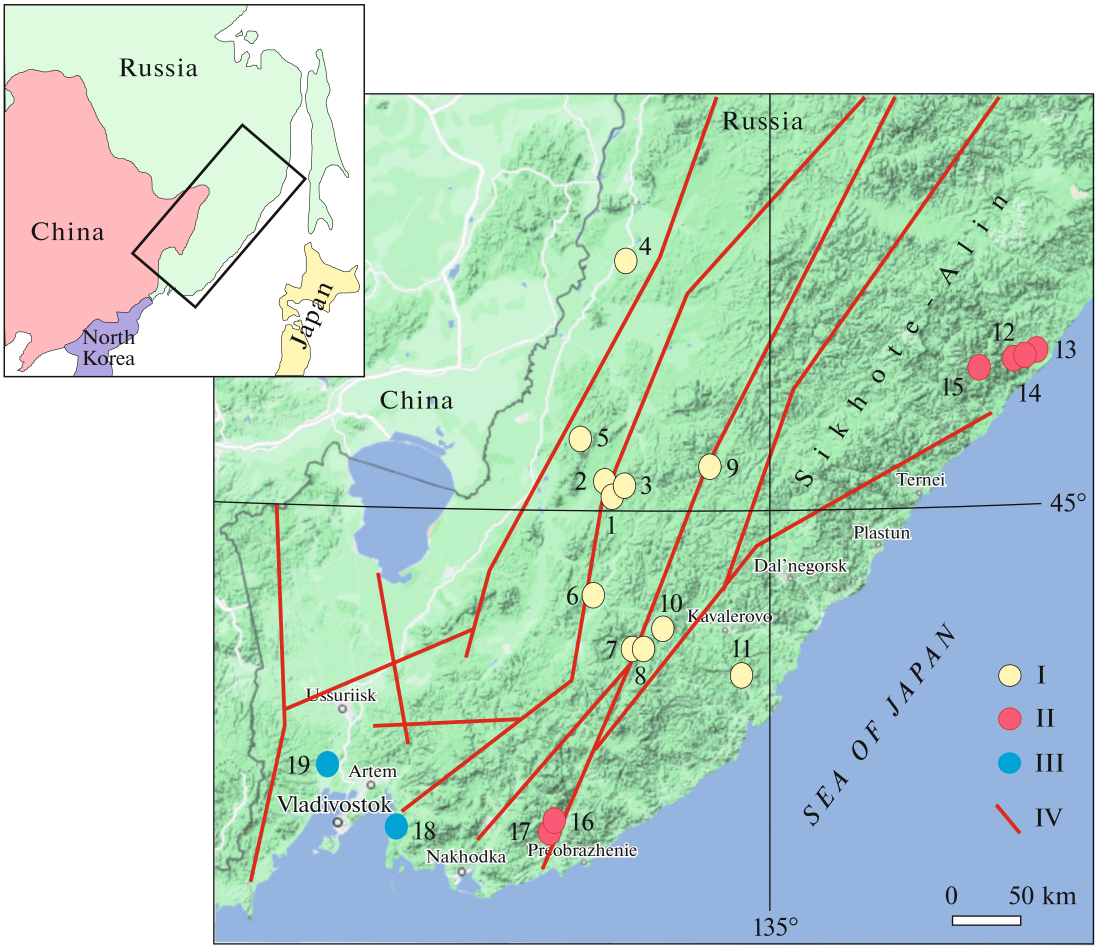


Fig. 1. An overview map of sampled occurrences of mineral waters of the Sikhote-Alin.
(I) CO_2-rich mineral waters: (1) Nerobisnky spring; (2) Bol'shoi Klyuch spring; (3) Fabrichny spring; (4) Lastochka deposit; (5) Eastern Ussuri site of the Shmakovka deposit; (6) Pokrovsky spring (well); (7) Ivanov Klyuch spring; (8) Kurortnyi spring; (9) Ariadnensky (upper) spring; (10) Pukhovsky spring; (11) Gornovodnoe. (II) Nitrogen thermal waters: (12) Teplyi Klyuch (spa, well); (13) Saion spring; (14) Balanov Klyuch spring; (15) Svyataya Elena spring; (16) Goryachii Klyuch deposit; (17) Chistovodnoe deposit. (III) Nitrogen–methane waters of elevated salinity: (18) Rechitsa well; (19) Razdolnoe deposit. (IV) Large faults and structural sutures.

OBJECTS AND METHODS

The study objects are CO_2-rich, nitrogen–methane, and nitrogen waters of Primorye. All occurrences of the thermal waters are confined to the large tectonically active structure of the region, the Sikhote-Alin orogenic area (Fig. 1), which is extended along the Sea of Japan for 1200 km. The Sikhote-Alin hydrogeological massif occupies ~70% of the Primorsky krai.

During field works in summer 2021, the radon contents (^{222}Rn isotope) were measured in water samples of the 19 most accessible and utilized thermal water occurrences of the region (Fig. 1). Water, spontaneous gases, water-bearing rocks, and secondary minerals were sampled for chemical and isotope analyses.

The most widespread are the CO_2-rich mineral waters form Primorsky krai, which are localized mainly on the western slope of the Sikhote-Alin [8]. In this area, the Far East largest Shmakovka CO_2-rich water deposit has been explored. On the eastern slope, only one large Gornovodnoe deposit was explored. The central part of the Sikhote-Alin is characterized by the minimum reserves of CO_2-rich mineral waters (nos. 6–10, Table 1).

The nitrogen thermal waters are ascribed to the Eastern Sikhote-Alin Province of nitrogen alkaline thermal springs and surround the Primorsky region of CO_2-rich mineral waters from the east, forming a narrow band along the Sea of Japan coast. They are united into the Chistovodnoe (nos. 16, 17, Table 1) and Amga (nos. 12–15, Table 1) groups of low-temperature mineral waters.

The nitrogen–methane waters with elevated mineralization were identified in the southern Primorye, in the junction zone of the Sikhote Alin and Khanka spurs. These springs are confined to the coastal parts

Table 1. The isotope-geochemical characteristics of mineral waters of Primorye

Ordinal no.	Spring	Host rocks	Age of rocks/ intrusions	Water type	Mineralization, g/L	U	Th	^{222}Rn
						µg/L		Bq/L
	Water							Gas
	CO$_2$-rich mineral waters							
1	Nerobinsky	Sandstone, granite	T_3/K_1	HCO_3-Ca-Na	0.3	0.19	0.02	6990
2	Bol'shoi Klyuch	Sandstone, granite	P_2/K_1	HCO_3-Ca	2.0	0.21	0.005	7365
3	Fabrichny	Tuffstone, andesite	P_2/K_2	HCO_3-Na-Ca-Mg	1.5	0.02	0.004	30
4	Lastochka	Sandstone	J_2/abs.	HCO_3-Mg-Ca-Na	3.0	0.02	0.007	60
5	Shmakovka (Eastern Ussuri)	Granite	D, C	HCO_3-Ca-Na	0.5	0.01	0.02	648
6	Pokrovsky	Tuffstone	K_2/abs.	HCO_3-Ca-Na	1.6	0.02	0.005	64
7	Ivanov Klyuch	Cherts, sandstone	J/K_1	HCO_3-Na-Ca-Mg	0.7	0.09	0.01	30
8	Kurortnyi	Sandstone, cherts	T_3/K_2	HCO_3-Ca-Na	1.6	0.08	0.007	48
9	Ariadnensky	Tuffstone, granite	P_2/K_2	HCO_3-Na-Ca	0.2	0.02	0.003	83
10	Pukhovsky	Shales	T_3/abs.	HCO_3-Ca-Na	0.1	0.17	0.11	48
11	Gornovodnoe	Tuffs, granite	K_2/K_2	HCO_3-Ca-Na	1.0	0.03	0.005	100
	Nitrogen thermal waters							
12	Teplyi Klyuch (Amgu)	Granite	K_2	$HCO_3(CO_3)$-Na	0.3	0.06	0.01	387
13	Saion (Amgu)	Tuffs, granite	$K_2/\text{₽}_1$	$HCO_3(CO_3)$-Na	0.2	0.21	0.01	185
14	Balanov Klyuch (Amgu)	Tuffs, granite	$K_2/\text{₽}_1$	$HCO_3(CO_3)$-Na	0.2	0.46	0.003	122
15	Svyataya Elena (Amgu)	Tuffs, granite	$K_2/\text{₽}_1$	$HCO_3(CO_3)$-Na	0.2	0.56	0.003	66
16	Goryachii Klyuch	Cherts, granites	P_2/K_2	$HCO_3(CO_3)$-Na	0.3	0.02	0.01	438
17	Chistovodnoe	Granite	K_2	$HCO_3(CO_3)$-Na	0.3	0.05	0.003	354
	Nitrogen–methane waters							
18	Rechitsa	Sandstone, siltstone	K_2/abs.	HCO_3-Cl-Na-Ca	26	0.02	0.007	117
19	Razdolnoe	Sandstone, tuff	K_2/abs.	HCO_3-Na	3	0.03	0.008	132

and were recovered at the end of 20th century during prospecting works for hydrocarbons or thermal waters. Wells drilled to a depth over 2.5 km made it possible to find water types that were previously unknown in Primorye. At present, only Razdolnoe spa located in the valley of the eponymous river is exploited. The large prospects are related to the unique Rechitsa occurrence of weak brines in the Sukhodol Bight (no. 18, Table 1).

Radon is an inert gas, has a high molecular weight (222) and short half-life (3.82 days), which constrains its migration. The volume activity (VA) of radon was measured by the Alpha-rad+ measurement complex (NTM-Zashchita, no. 49013-12 in the state list of measurement devices) directly on spring. Verification certificate no. MA 0177828. The volume activity is an activity per unit of volume of a source (water or air). The method is based on the circulation transfer of radon together with air from the water sample into working camera of VA measurement block of radon through bubbling. Water was sampled in 0.046-L capsule using a technique reported in [16]. The sample was loaded in the bubbler system, and air was passed through water to cause an intense ^{222}Rn release from liquid into a gas phase. Through connecting pipes, the released radon was supplied into the radiometer measuring chamber. In compliance with technique requirement, the radon was transferred from the bubbler system into measuring chamber camera for five minutes. In some cases, concentrations of radon and thoron were measured in soil air by an SRS-05 radiometer. It should be noted that the measurement error of radon VA on any device at present accounts for no more than 20%. Each sample was measured five times and the average value was calculated.

The amount of radioactive matter was measured not only in mass units (gram, milligram, and so on), but also in the activity equal to the number of nuclear disintegrations (decays) per time unit. The greater the nuclear disintegration of atoms of given matter is per second, the higher its activity is. Since the decay rates of radionuclides are different, the same weight of different radioactive isotopes has different activities. The activity unit in SI is measured in the decay per second (decay/s). The Becquerel is the measurement unit of activity of radioactive source in the International System of Units (SI). One becquerel is defined as the activity of a source in which one nucleus decays per second. In different countries, the values of the maximum permissible concentrations of radon in drinking water vary from 11 Bq/L (United States) to 300 Bq/L (Europe). According to the Radiation Safety Standards NRB-99/2009 and SP 2.6.1.1292-2003 [12], the radon content in drinking water should be no more than 60 Bq/L. Mineral waters with no less than 185 Bq/L are classified as radon waters [10]. Based on the radon concentrations, the waters are divided into waters with low radon concentrations (200–1500 Bq/L); waters with moderate radon concentrations (1500–7500 Bq/L); and waters with high radon concentrations (>7500 Bq/L).

The main source of radon is rocks with elevated contents of radioactive elements. Owing to its inert behavior, radon relatively easily leaves the crystalline lattice of radiogenic mineral. The radioactivity of rocks is caused by the concentrations of natural radionuclides that are parental for uranium and thorium. According to the decay schemes of ^{238}U and ^{232}Th, the stable isotopes in the uranium and thorium families are ^{206}Pb and ^{208}Pb, whereas ^{222}Rn and ^{220}Tn are intermediate decay products. The uranium, thorium, and lead concentrations in samples of water-bearing rocks were determined by ICP-MS at the Analytical Center of the Far East Geological Institute, Far Eastern Branch of the Russian Academy of Sciences (Vladivostok). We analyzed rock samples from the largest deposits: Lastochka, Gornovodnoe, Ivanov Klyuch, and Rechitsa. The core was sampled during hydrogeological works. At the thermal water deposits, rocks in the vicinity of natural vents were analyzed.

Hydro- and gas-geochemical sampling was carried out using standard techniques. Water samples were filtered through 0.45-μm membrane filters. Portative pH–Eh–TDS Mettler Tolledo meters were used to measure temperature, electric conductivity, pH, and Eh. The salt composition of water was determined by ICP-MS and ICP-AES at the Analytical Center of the Institute of the Technological Problems of Microelectronics and Ultrapure Materials of the Russian Academy of Sciences (IPTM RAS), in Chernogolovka. The measurement accuracy was no worse than 15%. The concentrations of HCO_3^- and Cl^- were determined by acid and $AgNO_3$ titration at the chemical-analytical laboratory of the Geological Institute of the Russian Academy of Sciences (GIN RAS, Moscow). Stable oxygen and hydrogen isotopes were analyzed on a Picarro 2140i spectrometer (GIN RAS). The composition of spontaneous gas was studied on a Kristall-5000.2 chromatograph at the Laboratory of Heat and Mass Transfer of GIN RAS.

RESULTS AND DISCUSSION

The results of measurements of the volume activity of the region, as well as the geological-geochemical characteristics of study objects are presented in Table 1. The obtained data show that the majority of mineral waters of Primorye have radon concentrations less than 185 Bq/L and according to the radiation safety standards [12] cannot be ascribed to radon waters. Five springs are ascribed to waters with low radon content (185–1500 Bq/L) and only two springs correspond to waters with moderate radon contents (1500–7500 Bq/L). Waters with high radon content (>7.5 kBq/L) were not found. Based on the chemical composition, four radon springs are distinguished as nitrogen thermal waters and three springs belong to the CO_2-rich waters (Table 1).

Nitrogen thermal waters are widespread in crystalline massifs and usually contain radon, which is one of the main balneological factors of their use. In the Sikhote-Alin, these waters are utilized at the Chistovodnoe (no. 17) and Teplyi Klyuch (no. 12) spas and are explored at the Goryachii Klyuch deposit (no. 16, Table 1). From the hydrogeological point of view, these are sufficiently large objects with thermal water reserves estimated at 30–70 m^3/day [27–29]. The obtained values of radon volume activity for them are 350–450 Bq/L (nos. 12, 16, 17, Table 1). The lowest VA values (from 66 to 185 Bq/L) were found in the natural thermal waters, low volumes of which are supplied into alluvial horizons. The nitrogen thermal waters of Primorye are ascribed to the low-radon waters.

Thermal waters are well studied from the geological and hydrogeological points of view [6]. The deposits were studied by drilling to a depth of 250 m. Based on geological setting, the thermal occurrences of the Sikhote-Alin volcanic belt can be subdivided into two types: (1) contact zones between granite intrusions and volcanic rocks (nos. 13, 14, 15, 16, Table 1), and (2) within active parts of faults within granite massifs (nos. 12, 17, Table 1). Most of the water-bearing rocks have Late Cretaceous age, and in one case, Permian age. The youngest intrusions are ascribed to the Paleogene, while intrusions with age older than Upper Cretaceous were not found. The rocks are represented by medium-grained gray or pinkish–gray granites with uranium and thorium contents from 3.2 to 14.8 and from 11.6 to 14.8 ppm, respectively. Based on the U and Th contents, the host granites are classified as moderately radioactive rocks, although do not reach boundary values (U, 5 ppm; Th, 20 ppm).

The water temperature is 27°C in the Chistovodnoe spring group (nos. 16, 17, Table 1) and increases to 35.4°C in the north, in the Amga Group (nos. 12, 13, 14, Table 1). The characteristic feature of nitrogen thermal waters is the elevated alkalinity (pH > 8) and low mineralization (<0.3 g/L). All waters are ascribed to the hydrocarbonate–sodium type. The uranium content in the waters varies from 0.06 to 0.56 μg/L, which is much lower than the MPC (15 μg/L) for drinking waters [15]. Such concentrations are more typical of surficial waters (U = 0.04 μg/L) [2] than of waters from the supergene zone (U = 1.3 μg/L) [30].

The hydrocarbonate composition of thermal waters, low mineralization and temperature indicate that their formation likely resulted from shallow (1–2 km) circulation.

The circulation time of nitrogen thermal waters from recharge to the discharge zones was estimated using tritium measurement [28]. In particular, the value of 0.04 Bq/kg was obtained for the Goryachii Klyuch spring. The calculation of the transit time of thermal waters of the Goryachii Klyuch spring showed that these are waters of retarded water exchange: 68.5 years (by snow) and 57.3 years (by rain). This indicates that recharge and discharge zones are spatially separated, and also confirms the results of hydrogeological and hydrochemical observations, which showed that these thermal waters were not recharged with surficial and ground waters [31].

The nitrogen thermal waters show strong correlation of radon VA with mineralization ($r = 0.8$) and weak correlation with temperature ($r = 0.3$), which is likely related to the decreasing radon solubility in water with increase of these parameters (Figs. 2a, 2b). The thermal waters also show a negative correlation between radon VA and uranium concentrations (Fig. 2c). Correspondingly, the uranium content also weakly correlates with temperature ($r = 0.2$) and moderately correlates with mineralization ($r = 0.7$). Radon is ascribed to the inert gases, and from the chemical point of view, its ability to enter into chemical compounds is low. Radon usually positively correlates with uranium, and a negative correlation between them would be considered to be random. However, as is shown in Fig. 2d, low uranium contents in water are caused by redox conditions, which determine its migration (Fig. 2d). It is known that in the supergene zone, uranium shows intense migration in oxidizing environment and low migration in reduced environment [30].

Thus, the uranium contents depend on the mineral properties of water-bearing rocks (leaching areas of uranium minerals) and on the redox conditions, which could change. During interaction of water with radiogenic minerals under reducing conditions, uranium does not go into solution, but water is enriched in radon. Mineralogical studies of granites showed that sources of uranium and thorium radionuclides are thorite and uranothorite (Fig. 3). The numerous studies of uranium and thorium speciation in rocks revealed that these elements in variable contents and proportions are present in all rock-forming and accessory minerals [17].

Released radon occurs in rock in three states: in a closed pore space, in fractures, and as absorbed by the free inner surface of the entire massif [20]. The nitrogen thermal waters circulate along fractures and brecciation zones, which is consistent with a model of fractured-porous medium consisting of the main massif and chaotically arranged permeable fractures and closed pores. Under these conditions, the main mechanism of radon supply to the surface obviously is the water flow. The high solubility of radon in water allows its transfer in space and accumulation in it. Thus, the radon content in water will depend on the presence of source radon minerals, the water flow rate, and is constrained by the half-life of radon (3.8 days). When the stressed state of rocks changes (seismic event), Rn could be released from closed pores, which short-term increase of its concentration in water.

The mainly water transfer of radon is also supported by the extremely low gas factor of nitrogen thermal occurrences of the Sikhote-Alin. Nitrogen spontaneously released from water (99.6 vol %) in small amounts was established only at two objects (nos. 13, 16, Table 1). It is believed that the gas component (nitrogen) is of atmospheric origin [7, 24]. The absence of influence of juvenile gases for the thermal waters of the Chistovodnoe group was proved by the low helium isotope ratios ^{3}He/^{4}He (0.1–0.24) × 10^{-6} [5].

The CO_2-rich waters of the Sikhote-Alin are the best studied waters from the geological and hydrogeological points of view, but their span is very uneven due to the great number of their occurrences. Among 11 sampled CO_2-rich springs, 9 springs are characterized by low values of radon VA (from 30 to 650 Bq/L). Thus, they include both well explored large deposits (nos. 4, 5, 6, 7, 11), and weakly studied (nos. 3, 8, 9, 10) water occurrences (Table 1). The values of volume activity of radon (650 Bq/L) in a well that recovered the CO_2-rich waters in granites of the Eastern Ussuri site of the Shmakovka deposit are typical of thermal water deposits in the granite massifs (Table 1). Waters of this area also have low mineralization (0.5 g/L), which indicates a rapid water exchange and a short-term interaction with carbon dioxide. Two sources (nos. 1, 2) are characterized by moderate VA values of radon (1.5–7.5 kBq/L).

The maximum depth of geological study of the CO_2-rich waters by drilling is 300 m. Based on the conditions of formation, two main water types are distinguished: (1) fractured waters that are formed in the upper fractured zone of bedrock (hard) rocks and localized around active fault parts releasing CO_2 (nos. 1, 2, 3, 4, 5, 7, 9, 11, Table 1), and (2) formational–porous waters forming in porous reservoirs of thick (>20 m) alluvial sequences and developed above the active fault zone supplying carbon dioxide (nos. 6, 8, 10).

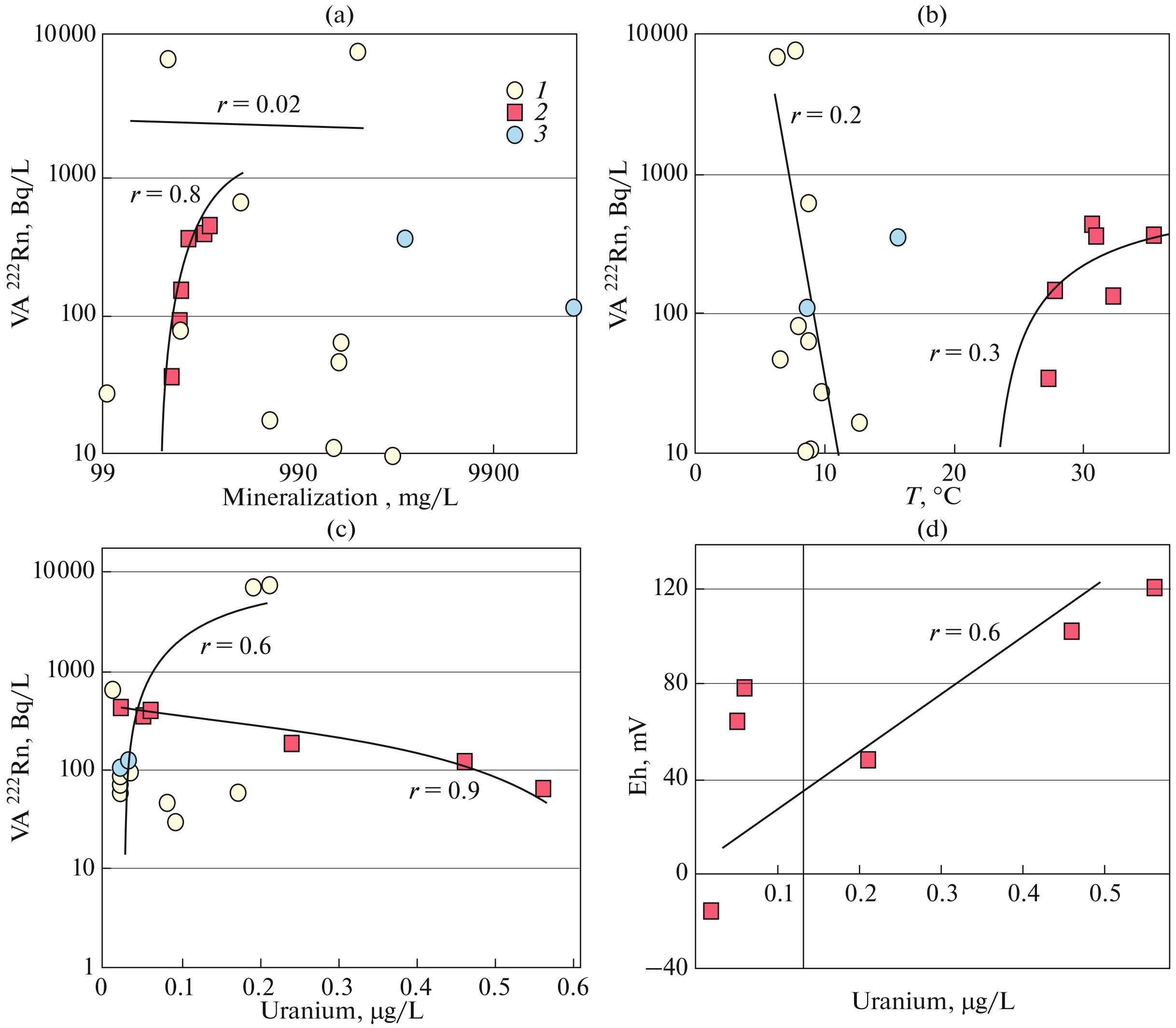


Fig. 2. Variations of radon content in water versus mineralization (a), temperature (b), uranium concentrations (c), as well as redox conditions versus uranium content in water (d). (*1*) CO_2-rich waters, (*2*) nitrogen thermal waters, (*3*) nitrogen–methane waters of elevated mineralization.

The CO_2-rich waters lie in rocks of different ages and types, which are represented by sandstones, tuffstones, siltstones, siliceous shales, as well as granites (Table 1). The age interval of the rocks varies from the Devonian to the Upper Cretaceous. Cretaceous intrusions of felsic composition are widespread. The older age was determined only for intrusions within the Eastern Ussuri site of the Shmakovka deposit. The study of core from wells drilled at the Lastochka, Ivanov Klyuch, and Gornovodnoe deposits showed that based on the U and Th contents, the host rocks are characterized as weakly radioactive (3.2 ppm U, 14 ppm Th), Th-bearing (Th/U = 4.4) rocks. Intervals of moderately radioactive rocks (5.07 ppm U, 21 ppm Th) rarely occur.

All CO_2-rich waters are ascribed to the hydrocarbonate type, with mixed cation composition and different mineralization (Table 1). The waters are cold (from 6 to 12°C), and have weakly acid or neutral pH (4.5–7.5). The concentration of hydrocarbonate ion reaches 2400 mg/L, while the contents of chlorine and sulfate ions are insignificant. The highest mineralization was determined in the hydrocarbonate sodium waters of the Lastochka deposit (3.0 g/L) and the lowest mineralization is in the hydrocarbonate–sodium–calcium waters of the Pukhovsky spring (0.1 g/L).

Analysis of proportions of main components indicates that the mineralization of CO_2-rich waters directly depends mainly on the input of CO_2 in water, the dissolution of which causes an increase of the HCO_3^- content, rather than on the influx of main cat-

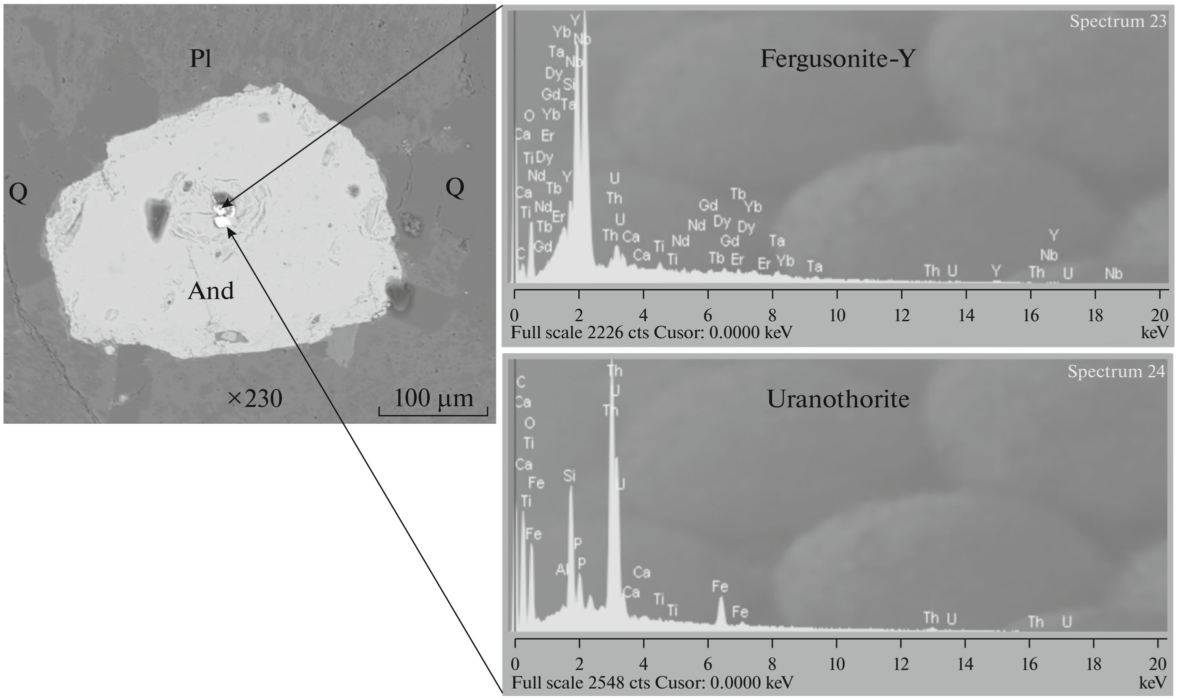


Fig. 3. The electron microscope study of granites of the Chistovodnoe thermal group (Jeol 8100, Far East Geological Institute, Far Eastern Branch, Russian Academy of Sciences). Shown are minerals, the sources of U and Th radionuclides. (Pl) Plagioclase, (Q) quartz, (And) garnet (andradite).

ions. The contents of balneological components, for instance, SiO_2 (up to 90 mg/L) and iron (up to 30 mg/L), also increase. The uranium and thorium contents in total do not exceed 0.3 μg/L, with uranium prevalence (Table 1). In general, the uranium and thorium contents are lower than those accepted for the supergene zone according to Shvartsev (U = 1.3 μg/L, Th = 0.24 μg/L) [30].

Isotope ratios of δD (from –87.8 to –103.8‰, SMOW) and $\delta^{18}O$ (from –12.2 to –14.7‰, SMOW) in CO_2-rich waters indicate their meteoric origin, while the tritium content on average accounts for 0.59 Bq/kg [28]. Calculation of water age according to the piston model shows that the minimum residence time of water in the hydrogeological system is ~21.5 years. It was established that the ^3H content in mineral CO_2-rich waters depends only on their circulation time and the degree of dilution by surficial waters [21].

The total gas saturation of CO_2-rich waters is insignificant, but is much higher than in nitrogen thermal waters. The gas factor rarely exceeds 1–3 m^3/t (the gas to water volume ratio). The main component of gas is CO_2, while subordinate components are nitrogen, argon, and helium. The CO_2 value in the spontaneously released gas mixture is within 95.0–99.9 vol %, while the contents of other components are as follows (in vol %): N_2, 0.1–5.2; O_2, 0.005–0.5; and Ar, 0.05–0.28. Other gases (CH_4, He, Ne, and others) in total account for less than 0.05 vol %.

Vents of cold CO_2-rich waters are always confined to tectonically active areas. The water discharge areas are usually localized either in zones of high seismic activity, or confined to the fault intersections. The tectonic structure of the territory (presence of permeable faults and cap rocks) provides the intense interaction in the water–rock–carbon dioxide system and formation of CO_2-rich mineral waters [11, 22]. Vents of CO_2-rich mineral waters are frequently traced as an interrupted chain along seismically active lines of significant extension or along fragments of ring structures (Gornovodnoe deposit).

In the CO_2-rich waters, the correlation of radon with the chemical type of waters, temperature, and mineralization is absent. A weak correlation was found only between radon and uranium ($r = 0.6$; Fig. 2c). This is likely related to the fact that CO_2-rich waters sharply differ from nitrogen thermal waters in host rocks and hydrogeological conditions.

The highest values of volume activity of radon (6990 and 7365 Bq/L) within the Sikhote-Alin were established for the Shetukha Group of CO_2-rich waters (nos. 1, 2, Table 1). These values are more than 15 times higher than radon concentrations measured on other occurrences of mineral waters of Primorye.

The hydrogeological position of waters of the Shetukha Group is weakly studied, and drilling was not carried out. The geological conditions of the Nerobinsky spring were studied in more detail, since it is confined to the zone of previously mined cassiterite deposit. This is the only known CO_2-rich deposit of Primorye that is located within an ore zone. It was established at the end of 20th century that the waters of this spring have the highest contents of arsenic (0.14 mg/L), as well as zinc (1.3 mg), and tungsten (0.2 μg) [29].

The mineral waters flow out in the low-density rock zone related to the fault, along which the small creek valley was initiated. The host rocks are represented by fractured Triassic sandstones and siltstones at the contact with the Early Cretaceous granites.

In terms of composition, the mineral waters of the spring are hydrocarbonate sodium–calcium, CO_2-rich, Fe-rich, and siliceous. The water temperature is +7°C. The water has low mineralization (0.3 g/L), weakly acid pH 5.1–5.2, and oxidizing conditions Eh +92...+400 mV. According to the long-term measurements, the uranium and thorium contents are 0.19–0.39 and 0.003–0.02 μg/L, respectively. The isotope characteristics show that the waters are of atmospheric origin ($\delta D = -101.4‰$, $\delta^{18}O = -14.7‰$, SMOW), while the time of their interaction with host rocks based on the tritium content is nearly 20 years. The high circulation rate is expressed in the low values of mineralization (at weakly acid water pH), and redox conditions of aqueous environment. Thus, the CO_2-rich mineral waters are the shallow-circulation waters.

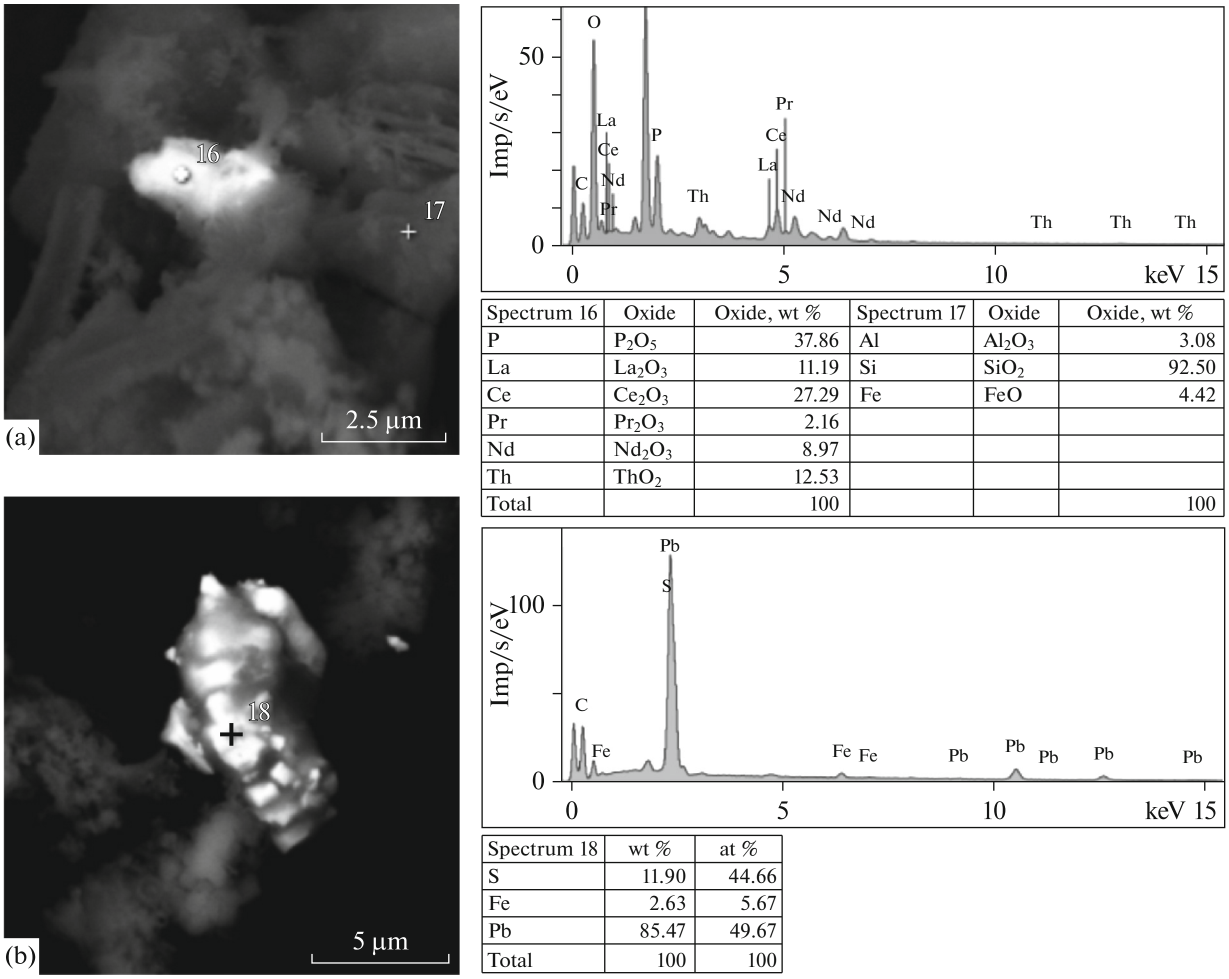
This spring is characterized by the intense spontaneous release of gas, which consists of 94.1 vol % carbon dioxide and 5.8 vol % nitrogen. The gas factor, i.e., the gas to water volume ratio is near 1 m^3/t. It was established that oxygen, nitrogen, and argon have an atmospheric genesis, whereas carbon dioxide and helium are of endogenous origin [27]. Based on the $\delta^{13}C$ and $CO_2/^3He$ ratios, the CO_2 genesis is related to mantle processes, whereas the crustal contribution is insignificant [22]. The extremely high concentrations of soil radon within the Nerobinsky mineral spring were established during previous works [25]. The results showed that VA of radon emanations vary from 20 to 360.8 kBq/m^3. The measurements of soil radon at a distance of 50 m from the spring yielded 100–136 Bq/m^3.

In addition to water and gas analysis, we studied secondary mineral deposits, which are intensely formed within the spring. Sediments are represented by iron hydroxides (Fe_2O_3 = 68 wt %) and silica (SiO_2 = 2.98 wt %), and have high contents of trace elements, Y, W (4 ppm), As (867 ppm), and Pb (372 ppm). Note that the lead content in water is as low as 0.013 mg/L. The total uranium and thorium contents in secondary deposits are 1.7 ppm, and Th, 0.12 ppm, which classify these deposits as weakly radioactive. Compared to all the studied host rocks of Primorye, secondary deposits have high levels of uranium (Th/U < 2). This is explained by the different chemical mobility of uranium and thorium in geological environments. Thorium does not form soluble minerals, and migrates in water mainly in suspended or colloid states. This is confirmed by the electron microscope study (Fig. 4a), which demonstrated that one of the sources of thorium as well as rare-earth elements in the studied deposits is monazite fragments.

Electron microscopy showed that lead occurs in galena (Fig. 4b). The issue of which part of the lead in sediments has a radiogenic nature has not been studied yet, since stable isotopes ^{206}Pb, ^{207}Pb, and ^{208}Pb are formed through the radioactive decay of ^{238}U, ^{235}U, and ^{232}Th, respectively. However, the study of the uranium and thorium distribution in the Uzon caldera (Kamchatka) showed that the lead concentrations depend on the total uranium and thorium concentrations in the hydrothermally altered rocks [1]. The lead isotope analysis of altered rocks and sediments of Uzon showed that the ^{206}Pb content, on average (final decay product of uranium–radium series), is almost 5.5% higher than the widest spread present-day lead, while the content of ^{208}Pb (final decay product of thorium series) in contrast is ~1% lower. According to the author's opinion, this is related to the decrease of thoron–radon ratio in spontaneous gases outflowing through the heated zones of hydrothermal system. The study of the lead isotope composition of host rocks of the Nerobinsky spring is an urgent task for the near future. The measured values of thoron emanation through soil in the area of the Nerobinsky spring showed extremely high values up to 930 decays. Similar significant concentrations of short-lived thoron (half-life of 55.6 s), as well as high radon contents could suggest either a shallow depth of radionuclide source or another mechanism of rapid radon supply to the surface.

In order to reach the day surface from great depths, radon (moreover, thoron) should possess high spontaneous subvertical migration, which is constrained by its high molecular weight (222) and short half-life (3.82 days) [9]. Several models were proposed in literature to explain this process. A classical model is the convective–diffusive mass transfer of radon in a homogenous porous medium [35, 38]. The "geogas" model describes the radon movement along fractures in water-saturated rocks [14]. In the model of aquifer degassing, the radon concentration in water is a function of depth of radium-saturated rocks [36]. Nevertheless, the high migration ability of radon is difficult to explain both using the classical convective–diffusive model and "geogas" theory. Considering the correlations of CO_2 and ^{222}Rn flows, some researchers suggest that the rate of endogenous CO_2 flow through the low-density zones could reach values of 0.1–1 m/s. At such rates, radon could be brought to the surface from deep horizons for several days [34, 37].

The nitrogen–methane waters of elevated salinity are developed exclusively within the southern coast of Primorye (Fig. 1), which, in turn, is related to the geological and paleohydrogeological conditions. This



Spectrum 16	Oxide	Oxide, wt %	Spectrum 17	Oxide	Oxide, wt %
P	P_2O_5	37.86	Al	Al_2O_3	3.08
La	La_2O_3	11.19	Si	SiO_2	92.50
Ce	Ce_2O_3	27.29	Fe	FeO	4.42
Pr	Pr_2O_3	2.16			
Nd	Nd_2O_3	8.97			
Th	ThO_2	12.53			
Total		100			100

Spectrum 18	wt %	at %
S	11.90	44.66
Fe	2.63	5.67
Pb	85.47	49.67
Total	100	100

Fig. 4. The electron microscope study of secondary deposits of the Nerobinsky CO_2-rich spring: (a) thorium and REE contents in monazite; (b) galena. Two-beam scanning electron microscope Tescan Lyra 3 XMH + EDS AZtec X-Max 80 Standard, Far East Geological Institute, Far Eastern Branch, Russian Academy of Sciences.

area is characterized by the development of the marginal basins of subpressure waters confined to the Cenozoic superimposed basins. The basins are constrained by the discharge area. The marginal basins of subpressure waters are characterized by essentially clayey composition of their terrigenous and volcanic–terrigenous rocks, which determined the limited water capacity of the latter.

The waters of this type lie at depths of 100–500 m and are confined to fault zones. Under natural conditions, the waters are not discharged on the day surface, while well recovering them are poured out with a small flow rate. Based on mineralization and hydrochemistry, the studied waters can be subdivided into two types: (1) hydrocarbonate–sodium or hydrocarbonate sodium–calcium waters with mineralization of 3 g/L, (2) waters of chloride sodium–calcium (with calcium predominance) or mixed cation composition with mineralization more than 26 g/L (Table 1).

Waters of the first type were found in the organic-rich Mesozoic deposits. They are recovered by wells with a low flow rate (6 m^3/days) and can be exemplified by the Razdolnoe spa [32]. The characteristic feature of these waters is the high contents of hydrocarbonate ion, sodium, boron, and fluorine, at low contents of sulfate ion. In spite of the elevated salinity, the given water type is of meteoric origin ($\delta^{18}O = -12.3‰$, $\delta^2H = -85.3‰$, SMOW). Under operating conditions, the inflow of modern waters is recorded ($^3H = 2$).

The second type includes the low-salinity brines of the Rechitsa occurrence, which lie at a depth more than 250 m, have mineralization of 26–110 g/L, and chloride sodium–calcium composition [23]. The Ca^{2+} concentration in the brines is two times higher than Na^+ concentration. The bromine content is close to or higher than that of seawater, being within 65–120 mg/L. The relations of stable isotopes $\delta^{18}O$ (–9.0‰, SMOW) and δ^2H (–68.8‰, SMOW) show that such waters were formed in a warm climate. Tritium is absent, which suggests that the aquifer was closed under stationary conditions.

Radon contents in waters of these occurrences are below those typical of radon waters (VA Rn from 117 to 132 Bq/L). The nitrogen–methane springs are few, which makes statistically significant correlations impossible. The levels of radon, uranium, and thorium contents for groundwaters of the Sikhote Alin show no any anomalies (Table 1). The interval study of the core of a 100-m well drilled in the Rechitsa occurrence area (no. 18) revealed that the uranium content in Cretaceous sandstones with depth increases from 0.6 to 4.8 ppm (average content of 1.44 ppm). This dependence is not observed for thorium and its contents vary from 2.8 to 6.9 ppm (average content of 4.75 ppm). In terms of uranium and thorium contents, the sandstones are weakly radioactive (U = 11.5 ppm, Th = 3–7 ppm), thorium-bearing (Th/U = 4.6) rocks. The uranium and thorium contents in sandstones are lower than in the studied granites of other occurrences of mineral waters of Primorye.

Gas in these waters contains mainly methane (35–40 vol %) and nitrogen (35–45 vol %), while CO_2 accounts for less than 10%. The gas factor is very low, while radon transport with water to the surface is complicated under these conditions due to the low flow rate of the wells. Based on the gas geochemical studies, a gas-bearing area promising for methane was distinguished in the Sukhodol Bight [13]. It is most probable under these conditions that radon sources are host rocks, while its transport is provided by gases that are formed owing to transformation of organic matter in rocks. These results are consistent with data obtained on mineral sources of Sakhalin Island, where the volume activity of radon on the CO_2-rich water occurrences is higher than in methane-dominated waters [26, 33].

Factors Determining the Radioactivity Level of Different Water Types

Based on the considered geological conditions and geochemical features of thermal mineral waters of the Sikhote-Alin, we determined factors that determine the radioactivity level and proposed general schemes of radon transportation to the surface under different geological–hydrogeological conditions (Fig. 5).

The radon content in groundwaters is mainly determined by the following factors: (1) radon diffusion from the deeper seated geological horizons; (2) radon transport from deep-seated to subsurface horizons with other gases; and (3) filtration of groundwaters through radioactive rocks. The radon content in water is also controlled by a change of flow rate of springs (important for exploited aquifers), water temperature (on the one hand, a temperature increase causes a decrease of radon solubility in water, and, on the other hand, increases the emanating ability of rocks), conditions of water pouring out on the surface (if water flows out turbulently or falls from a depth, it loses most of its radon).

The main mechanism of radon influx in *the nitrogen thermal waters* is the circulation of atmospheric water in granite massifs and removal of radioactive emanations with water into subsurface zone. Thus, the concentrations of radon VA reflect a natural background of granite massifs spanned by the water circulation (Fig. 5a). The low values of radon VA (up to 650 Bq/L) in this area are mainly caused by the low gas factor, low discharge flow rates of thermal waters (0.5 L/s), and alkaline pH of water, which excludes intense interaction of water with rock in the subsurface zone (limited solubility of minerals).

The radon concentration in *cold CO_2-rich mineral waters* is mainly controlled by the coincidence of radon generation areas with deep-seated gas flows (CO_2). In these structures, radon and thoron are transported with gas (CO_2) flows ascending along faults from deep-seated horizons into subsurface aquifers (Figs. 5b). Radon sources in the CO_2-rich mineral waters are related to the near-contact zones of the granite intrusions with uranium- and thorium-bearing host rocks of different genesis. Such mechanism could provide the high radon concentrations in subsurface aquifers, since radon entrained by carbon dioxide flow could be supplied from deep-seated horizons for several days.

In *the nitrogen–methane waters* of sedimentary basins with retarded water exchange and low rock permeability, the radon influx in water is related to the diffusion from sedimentary water-bearing rocks, as well as from deeper geological horizons (during exploitation of the deposits) (Fig. 5c). It is more probable that the radon migration is related to gases that are formed through transformation of organic matter of rocks.

Medical and Geoecological Aspects of the Use of Radon Waters

Nitrogen thermal waters over the entire world very frequently contain radon, which is one of the main balneological factors of their use. Numerous spas operate at large deposits of these waters: Khodzha-Obi-Garm (Tajikistan), Belokurikha (Altai krai), Molokovka (Chita), Kuldur (Jewish Autonomous Region), Tumnin and Annenskie (Khabarovsk krai), Teplyi Klyuch, Chistovodnoe, and others. Patients take radon as water and air baths, shower bath, bathing in therapeutic pools, inhalations, microclyzms, and others. In the radon water, short-lived decay products cause radiation, which simulates connective tissue, epithelial, and parenchymatous cells of organism; affects the function of hypothalamic–pituitary–adrenal and sympathetic–adrenal systems; stimulates hemodynamics and exchange of biologically active

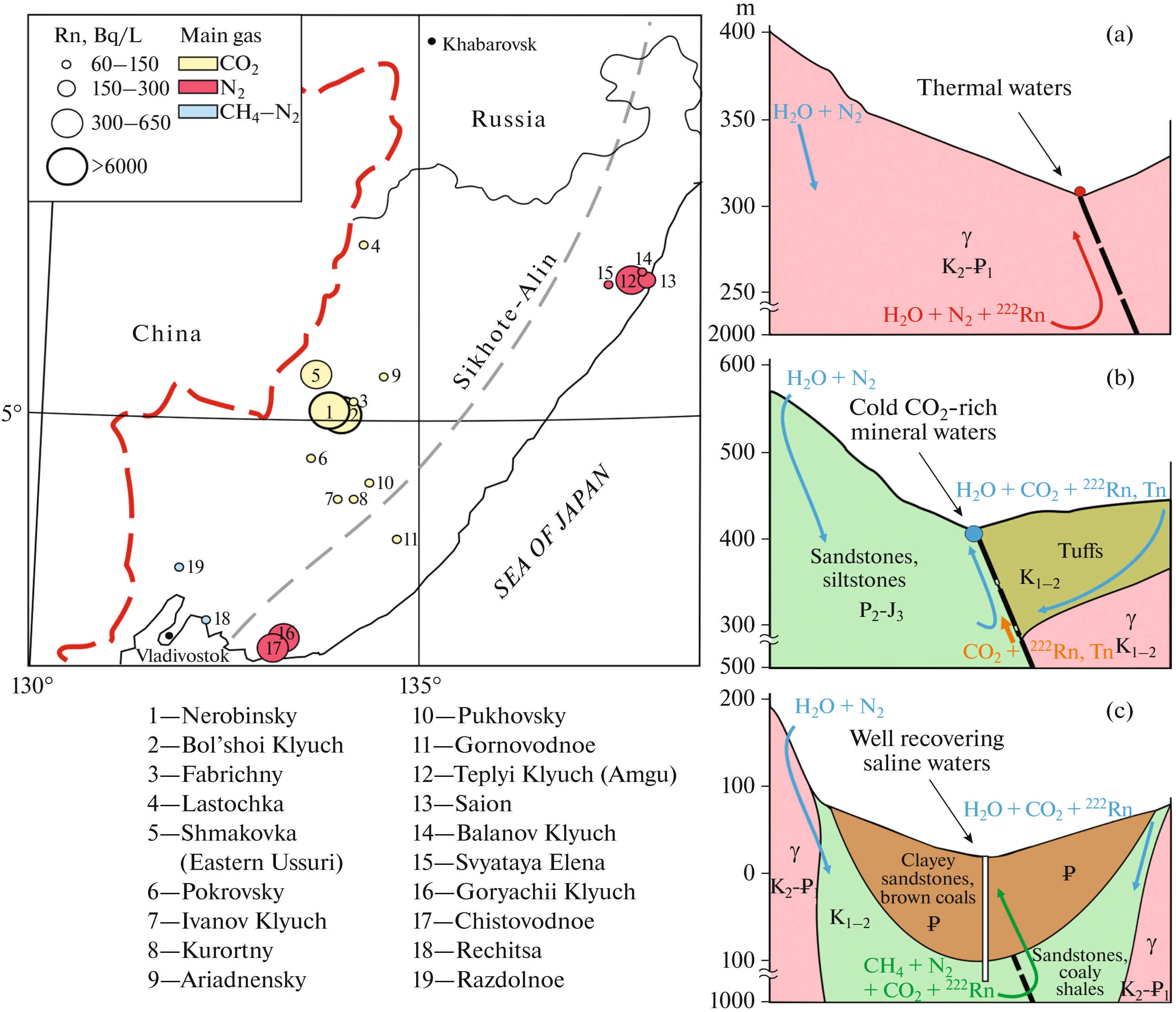


Fig. 5. A schematic map of the radon distribution in waters of mineral springs of the Sikhote-Alin. The main schemes of radon transport to the surface: (a) with water and nitrogen in granite massifs; (b) with carbon dioxide from contacts with granite intrusions; (c) with methane and nitrogen from sedimentary rocks.

matters (serotonin, histamine, catecholamines, and others) and many others [3]. The minimum effective therapeutical radon concentrations are 185 Bq/L for water baths, and 4000 Bq/L for drinking. Thus, the therapeutic effect of radon was confirmed at thermal spa resorts of Primorye. It is possible that “the wild thermal baths” (nos. 13, 14, 15) with a correct approach and development could show high radon concentrations. For this purpose, drilling and appraisal works are required.

Some cold CO_2-rich mineral springs (nos. 3, 7, 9) are independently used by the population for bathing. The data showed that such radon therapy is useless due to the low radon content. The CO_2-rich mineral waters of Primorye are traditionally used for drinking. The health risk from the low-radon drinking water is insignificant. The uranium content in water is much lower than MPC (15 μg/L) for drinking waters [15]. The main impact of radon from water on humans is related to its inhalation intake. However, unlike bathing, it is difficult to estimate the level of internal radiation in this case. It was indicated [18] that radon and its daughter isotopes could ionize tissue molecules when they enter the body and decay, in the vicinity of which the decomposition or radiolysis of water occur. The decay products could be toxic, while lead isotopes, upon entering the blood, are not removed. It is recommended to keep radon water for 10–15 hours before drinking, thus controlling its activity. For this reason, positive results of radonotherapy, as any type of therapy, could be provided only by constant medical control. The maximum permissible radiation of patient (for therapeutic course) is 34 kBq/L for water baths, 9.4 kBq/L for air baths, up to 0.7 Bq/L for inhalations of radon and its products, and 2.7 MBq at drinking radon water [3].

It should be noted from the geoecological point of view that the radon concentrations in the ground air within the Nerobinsky spring are more than 100 times higher than permissible standards for living spaces (200 Bq/m^3). Therefore, it is not recommended to set up tents and sleep on the ground, while the time spent near the spring should be reduced. The energy of alpha-particles varies from 5.48 to 7.68 MeV, which causes their strong impact on the biological tissues of internal organs. At a VA radon of 8000 Bq/m^3, it is forbidden for a person to be in the room. In this case, it is a positive thing that buildings are absent on the territory of the Nerobinsky spring and the radioactive gas is blown by wind. The measurements of soil radon at a distance of 50 m from the spring showed permissible values of 100–136 Bq/m^3.

CONCLUSIONS

Our studies made it possible, for the first time, to estimate the volume activity of radon in three types of thermal mineral waters of the Sikhote-Alin. It was established that the highest radon contents are typical of CO_2-rich mineral waters of the Shetukha Group. In addition to the known and used thermal radon waters of Primorye, three CO_2-rich mineral springs with therapeutical radon concentrations were revealed for the first time.

Based on the considered geological conditions and geochemical features of occurrences of thermal mineral waters of the Sikhote-Alin, the factors responsible for the radioactivity level were distinguished:

(1) For nitrogen thermal waters, where radon is mainly supplied through diffusion from crystalline massifs, the main factors responsible for radon concentrations are fracturing (permeability) and porosity.

(2) For cold CO_2-rich mineral waters, the main factor responsible for the radon concentration is the coincidence of radon generation areas with endogenous gas flow.

(3) In nitrogen–methane waters dominated by diffusion transfer of radon, tectonic (porosity and permeability) and gas factors likely have equal effect.

The studies showed that, on the one hand, the southern Russian Far East is a promising area for widening the possibilities of the use of mineral waters, and on the other hand, the territories with a high radon concentration could be a strong source of additional radiation.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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