

Soil-to-dwelling radon isotope ratio in the Baikal region

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Abstract

Relations between indoor and soil gas radon were experimentally studied in villages of the Baikal region. On the basis of the obtained data, the soil-to-indoor radon ratio was calculated, which can be used for prediction of radon pollution in buildings of the same kind.

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Introduction

Natural gas radon is an inevitable source of radiation impact on people as it is present both indoors and outdoors. For the Baikal region the problem of radon hazard is especially important because of climatic and geological features of the territory.

The inhabited areas are required to be zoned according to the degree of radon hazard to distinguish the territories where measures for reducing the radon impact are urgent and to carry out geographically correlated medical and epidemiological studies on the estimation of radon effect on the population health. This qualitative zoning should be based on the qualitative estimate and comparison of various parameters controlling radon pollution.

The International Commission on Radiological Protection (Protection., 1995) recommends to measure the radon concentration in a representative sample of dwellings as this is the safest procedure for outlining radon-hazardous zones. However, the today's financial support of regional programs is not sufficient for carrying out large-scale studies of radon pollution in inhabited areas. Therefore, to estimate the situation, at least at the first stages, we have to use radiogeochemical data on potential radon hazard of a certain area rather than direct measurements of indoor radon.

The main source of indoor radon is the ground on which the house is built (Nazaroff and Nero, 1988). The simplest way to estimate the radon pollution is to measure concentrations of radon in soil gas. However, any attempt to make a

forecast from single measurements of soil radon usually fails. The territory of an inhabited area can be characterized by strongly differentiated Rn contents in soil. Where primary rocks and soils have high concentrations of parental radionuclides of U and Th (granites, carbonaceous-siliceous schists, phosphorites), a high density of Rn flow is recorded. Fractures in the soil cover and active tectonic faults in underlying rocks also promote a higher emanation of radon from soil (Chernyago et al., 1996). The soil permeability can vary by several orders of magnitude within even a small site (Reimer and Gundersen, 1989; Varley and Flowers, 1994). The rate of Rn emanation from soil and its accumulation in dwellings as well as the degree of radiation hazard depends on many (not only geochemical) factors, which considerably hinders modeling and quantitative forecast of radon pollution for a particular locality or a building. The point is that a representative base of data on soil gas is difficult to obtain. The problem might be solved by estimation not from solitary measurements but from a large data base.

On the basis of summarized results of many years of radioecological study in the Baikal region, we propose here an approach to estimate the radon hazard of the territory hosting a given inhabited area from the content of soil gas radon.

Methodological

We have analyzed the data on radon pollution in inhabited areas of the southern Baikal region obtained in the period 1995–2000. The objects of study were the most typical villages in the southern Irkutsk Region, situated, chiefly, in radon

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hazard zones differing in radiogeochemistry and geology: Bol'shaya Zima, Bol'shaya Rechka, Maloe Goloustnoe, Listvyanka, Podkamennaya, Bol'shie Koty, Karluk, Oek, and Odinsk. The measurements performed in these villages were the most representative in kind and volume and, therefore, the summarized results should be sufficient for the reliable estimation of the radon pollution in the villages of the same type within the study area.

One-story wooden houses predominant in villages are the most vulnerable to radon. Most likely, the radon pollution should depend on radiogeochemical properties of radon occurrences.

Preliminary zoning, or classification of the territory of a particular village according to radon hazard, was based on analysis of geochemical information on the rocks and soils with a higher content of natural radionuclides and on the related tectonic faults, and on a few direct measurements of volume activities of indoor radon and thoron on the study area. Oek and Odinsk are situated in relatively safe sites, while the radon pollution was supposed to be considerable in the other chosen villages, especially Belaya Zima.

Methods of study

The complex study of radon pollution implied measurements of ^{222}Rn and ^{220}Rn activities in soil gas evenly over the area within each village and measurements of radon volume activities within dwellings and public buildings. All outdoor studies were carried out in the warm season, whereas indoor measurements were made the year round and were not only single but also integral in time. Both within and beyond the village the radon flow density was measured at the soil-atmosphere boundary and soil samples were taken for gamma-spectrometric analysis for natural radionuclides.

Volume activity of soil gas radon was measured by RGA-01 radiometers of alpha-active gases. To determine the amounts and ratio of ^{222}Rn and ^{220}Rn isotopes, a known technique of time-separate measurement was used (Kuznetsova and Polyachenko, 1986) based on a considerable difference between half-lives of these isotopes (3.82 days and 54 s, respectively). We measured twice the soil gas sample taken from a blast hole 0.6 m deep, with an interval of 10 min. The volume activities of radon and thoron in soil gas were calculated by means of programs that take into account the time of each measurement and interval between two measurements. The measurements were made either near the houses in which the contents of indoor radon were determined or at a uniform grid on the territory of the village at a step of 50 to 100 m. The run of measurements within a certain village was performed within three days at a relatively dry soil (in the absence of rain) either in summer or in early fall. The measurements were considered satisfactory if at control points (about 10% of total volume) the radon activities differed from each other by no more than 30%.

Direct measurements of radon content in dwelling and public buildings of the villages were carried out, at least, in

40 houses, which was usually 5 to 12% of the dwellings in each village with a population of up to five hundreds. The houses to be examined were chosen in a random way and as evenly as possible over the area of the village so that the sample be representative. The main type of housing system in the near-Baikal villages is quite the same — one-story wooden houses with a cellar or basement used for storing provisions the entrance to which is, as a rule, within the house.

The volume activity (VA) of indoor radon was determined by rapid and integral methods. To determine the seasonal VA of indoor radon, we used passive nitrocellulose-film track detectors. The time of exposure of radon track detectors varied from 50 to 90 days. Rapid measurements of radon were carried out with RRA-01 radiometers and additionally, if the radon activity was very high, with RGA-01. They were used mostly to control integral measurements and to estimate variation in indoor radon contents. The main run of measurements was made in winter after the ventilation regime typical of heating period had reached its steady state. In spring, summer, and fall the measurements were only rapid and took about 30% of the winter measurements. Nevertheless, they were used for correcting the estimate of annual contents of indoor radon.

Results and discussion

According to the available radiogeochemical data, the villages were conventionally divided into three groups (or categories) of radon hazard. The categorization involved all parameters of radon hazard: Ra and Th contents in soil, volume activity of Rn in soil gas, density of Rn flow from soil to atmosphere and its indoor activity, with safe standards [MU 2.61.715-98] on radon hazard zoning taken into account. In particular, radon hazard zoning can be made according to the levels of radon activity in soil (kBq/m^3): less than 10 (category I), from 10 to 50 (category II), and more than 50 (category III).

Radon and thoron in soil gas and at the soil-atmosphere boundary. Data on the measurement of volume activities of radon isotopes in soil gas on the territory of near-Baikal villages were rather representative as compared with other characteristics of radon pollution. Figures 1 and 2 show distribution patterns of experimental data on volume activity of radon isotopes in soils within separate villages. The distribution of frequencies of volume activities of radon isotopes in soils is lognormal. Therefore, to characterize the territory of a village in frequency distribution, we can use geometric means of volume activity of radon in soil.

The distribution patterns for the activities of ^{222}Rn and ^{220}Rn (thoron) in soils on the same territory have the same view but displaced peaks. In the Baikalian soils, the thoron activities are about half an order of magnitude higher than the radon activity (Chernyago et al., 1996), which is typical of the territories of villages.

Volume activities of radon in soil and radon flows from soil to atmosphere averaged over several studied areas are

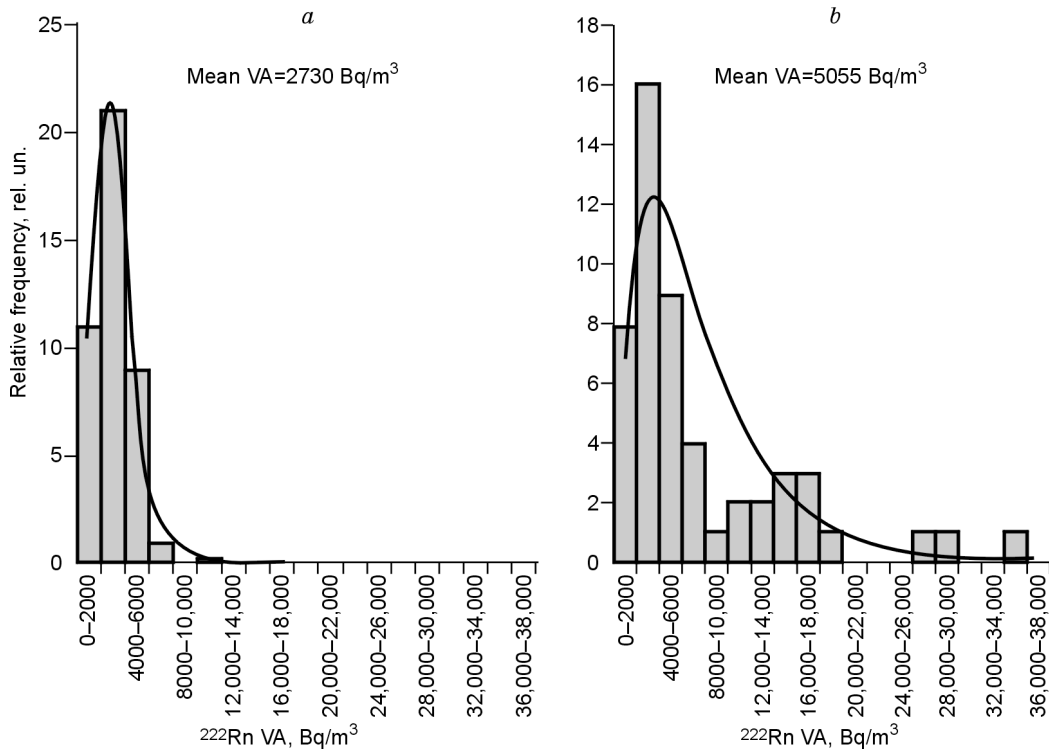


Fig. 1. Typical histograms and functions of distribution of volume activities of ²²²Rn in soils: *a* — Odinsk (safe zone); *b* — Podkamennaya (potentially radon-hazardous zone).

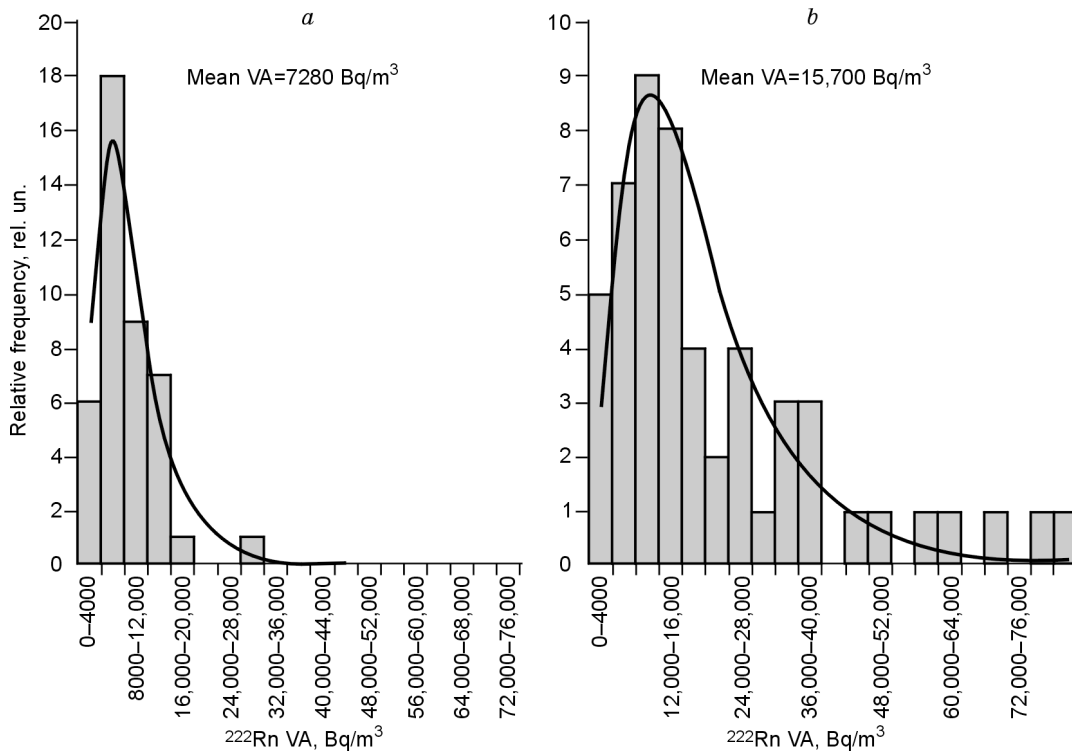


Fig. 2. Typical histograms and functions of distribution of volume activities of ²²⁰Rn (thoron) in soils: *a* — Odinsk (safe zone); *b* — Podkamennaya (potentially radon-hazardous zone).

reported in Table 1. Mean values of radon activity in soil gas as well as density of radon flow were calculated only over points where these measurements were made. These points in a village numbered three to six. Within one village the relative

scatter of volume activities and radon flows did not exceed 30–40%, which is comparable to the error of measurements of these parameters at a certain point. These parameters are proportional and seem to depend on the geology of the area.

Table 1
Averaged (over observation stations) contents of ^{222}Rn in soil and its flow from soil on the territory of some villages of the Baikal region

Village	N , kBq/m^3	J_{exp} , Bq/m^3
Listvyanka	20.6	0.119
Bol'shaya Rechka	10.6	0.059
Karluk	7.7	0.035
Podkamennaya	5.1	0.028
Odinsk	2.7	0.015

Regression equation for radon flow is

$$J \approx 6.1 \cdot N, \quad (1)$$

where J is the ^{222}Rn flow from soil to atmosphere, in $\text{mBq/m}^2\cdot\text{s}$; N is the volume activity of ^{222}Rn in soil, in kBq/m^3 .

The resulting linear dependence of ^{222}Rn flow from soil on its content in soil gas implies the same behavior of volume activity of indoor radon.

Indoor ^{222}Rn in near-Baikal villages. Despite complicate mechanisms of supply of radon (and thoron) into dwelling and public buildings, we suppose that within restricted inhabited areas characterized by a uniform geological structure and the same housing type the volume activity of indoor radon will be linearly correlated with the radon flow from soil to atmosphere and with its source, i.e., with the volume activity of soil gas radon.

It is likely that this dependence holds for averaged statistically representative data (i.e., typical of the entire inhabited area or its part) rather than for single measurements of radon activity both indoors and outdoors.

Figure 3 shows typical experimental data on radon pollution in three groups of near-Baikal villages situated in different zones of radon hazard. The first group (relatively safe zone, see Fig. 3, *a*) includes Odinsk Village in the Angarsk District. The geometrical mean of ^{222}Rn volume activity within the houses of this village was 46 Bq/m^3 . The second group (potentially hazardous zone, see Fig. 3, *b*) is exemplified by Podkamennaya Village of the Shelekhovo District, with an average VA of 111 Bq/m^3 . The third panel (Fig. 3, *c*) shows data of VA measurements in houses of Belaya Zima Village of the Tulun District, Irkutsk Region, whose territory is categorized radon hazardous (VA averages 301 Bq/m^3). Comparison of inspective measurements made in other villages with the forecast based on radiogeochemical data, including data on radon isotope contents in soil gas, confirms this estimate.

The histograms of distribution of volume activities of indoor ^{222}Rn are described by a lognormal function. The figures show not only qualitative but also quantitative difference between parameters of indoor radon VA distribution in villages classified into different hazard categories.

Relations between indoor and soil ^{222}Rn activities. A sample was composed of the data obtained on inspecting the villages, taking into account the combined measurements of soil and indoor radon volume activities.

The paired measurements of indoor and soil ^{222}Rn concentrations totalled to 147 for the inspected villages of the Baikal region.

The geometric mean of the soil radon VA was 11.4 kBq/m^3 , with the maximum of 695 kBq/m^3 .

The geometric mean of the indoor radon VA was 187 Bq/m^3 . The average volume activity of indoor radon was five times as high as the population mean for regions with a

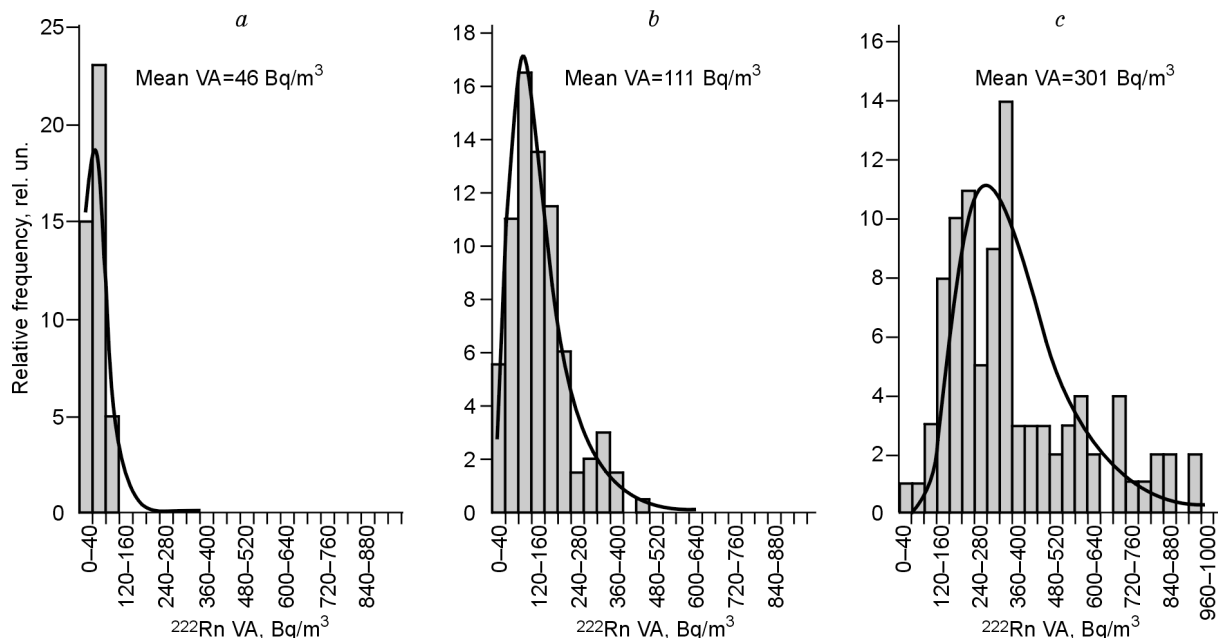


Fig. 3. Typical histograms and functions of distribution of ^{222}Rn volume activities measured by the track method in a sample of houses of three types of near-Baikal villages situated in a relatively safe zone — Odinsk (*a*), potentially hazardous zone — Podkamennaya (*b*), and radon-hazardous zone — Belaya Zima (*c*).

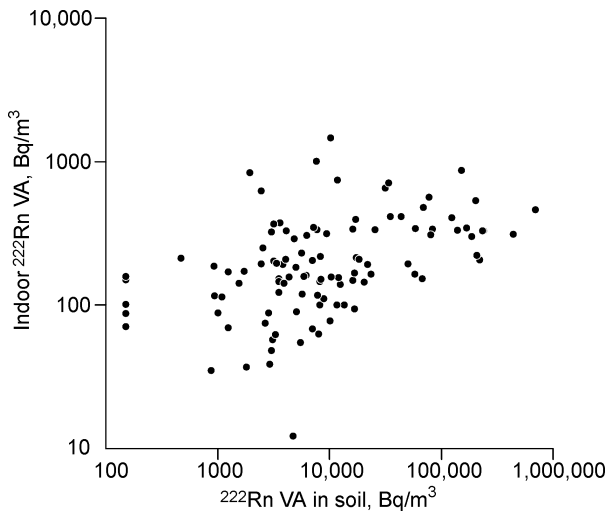


Fig. 4. Indoor radon contents against soil gas radon activity for near-Baikal inhabited areas (a sample of six villages).

moderate climate — 40 Bq/m^3 (Aleksakhin, 1994). In 43.5% of buildings the indoor concentration was more than 200, in 13.6% — more than 400, and in 4.1% — more than 800 Bq/m^3 . The maximum volume activity of indoor radon was 1470 Bq/m^3 , i.e., 3.5 times as high as the safe standard (400 Bq/m^3).

Figure 4 compares these two kinds of measurements. Though all data are obtained from a relatively compact area, correlation between measurements of soil and indoor radon activities is not evident.

If we group the VA values of soil gas radon in intervals $0.3\text{--}1.0$; $1.1\text{--}3.0$; $3.1\text{--}10.0 \text{ kBq/m}^3$, and so on (Fig. 5), we see that as the volume activity of soil radon enhances, geometric means and maximum volume activities of indoor radon increase as well. At values of more than 30 kBq/m^3 the expected concentration of indoor radon considerably increases. According to these data, the volume activities of indoor radon exceeds the standard (200 kBq/m^3) in 82% of buildings, with soil gas concentrations more than 100 kBq/m^3 . Thus, the indoor radon content increases with soil gas radon.

Ratios of ^{222}Rn volume activities in some pairs soil-house varied widely — from 0.6 to 1500. The frequency distribution of the ratios is lognormal (Fig. 6). The geometric mean of the soil to indoor radon volume activities for the whole set of paired data is 61.4.

The volume activities of both soil and indoor radon are considerably scattered even over a restricted area (in a village). The lack of uniformity in relations between indoor and soil gas radon volume activities at separate points (observation stations) argues for the considerable effect of the above-mentioned factors on the measurements of indoor radon as well as of the character of radon measurements (single — integral). But we can mitigate the effect of these factors by using a sufficiently great deal of measurements. It seems reasonable to find a correlation between soil and indoor radon activities for averaged (over a representative and statistically significant set of measurements) values in separate villages.

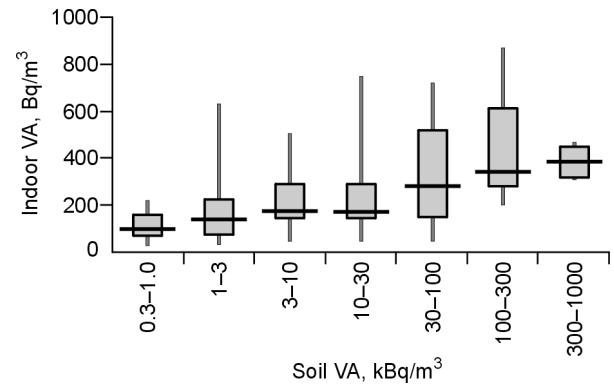


Fig. 5. Distribution of indoor radon measurements divided into groups of soil radon measurements (a sample of seven near-Baikal villages). Bottom and top of vertical lines mean 5 and 95%, bottom and top of gray rectangles — 25 and 75%, respectively, and the black horizontal line is the geometric mean for this group of soil radon measurements.

Grouped into “logarithmic” intervals, relations between geometric means of soil and indoor radon activities show their dependence on radon activity in soil. Their ratio increases from five (for soil radon concentration less than 1 kBq/m^3) to 616 (for the range from 100 to 300 kBq/m^3), i.e., the higher radon concentration in soil the lower the soil-to-indoor radon ratio. And vice versa, significant indoor radon activities are recorded where ^{222}Rn activity in soil gas is rather insignificant. This dependence is well described by a power function with a factor of 0.021 and exponent of 0.89, close to unity (Fig. 7). For the ^{222}Rn volume activities in soil of 3000 to 10000 Bq/m^3 , which are the most typical of the region, the soil-to-indoor radon ratios vary from 30 to 100.

Thoron (^{220}Rn) in soil and in dwellings. It is believed that the volume activity of indoor thoron, a short-lived isotope of radon (^{220}Rn), is negligible, and the researchers concerned with radon hazard often ignore the so-called “thoron” hazard. As the U-Th mineralization of the solid rocks in the Baikal region is rather specific (Chernyago et al., 1996), the thoron

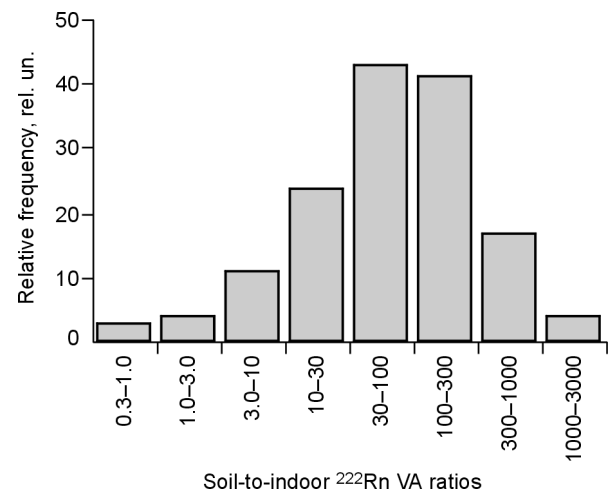


Fig. 6. Frequency distribution of soil-to-indoor ^{222}Rn activity ratios in the Baikal region.

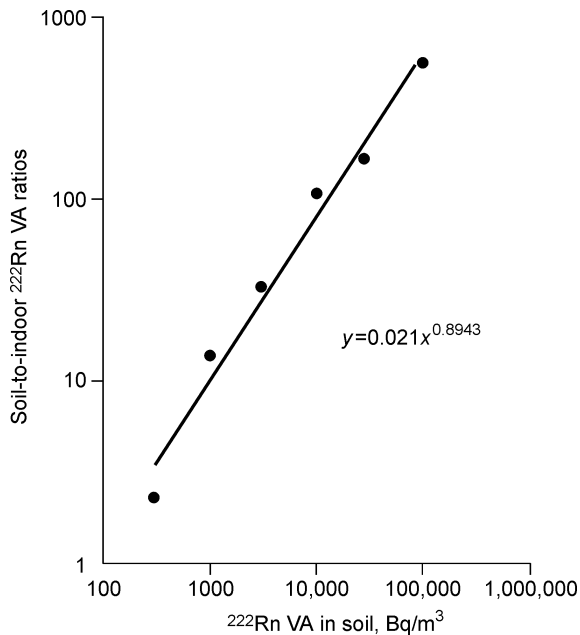


Fig. 7. Soil-to-indoor ^{222}Rn activity ratios against soil radon activity.

activity is about three times as high as the ^{222}Rn activity (see Fig. 2) and, therefore, the volume activity of indoor thoron is expected to be significant. With the dose buildup factor taken into account, the volume activities of thoron of tens of Bq/m^3 can have dose loads upon the population comparable to and even exceeding the radon impact.

We had no opportunity to make direct measurements of indoor thoron everywhere, and data of this kind are, unfortunately, very scarce. First of all, we inspected the inhabited area where high activity of indoor ^{222}Rn had already been recorded. The greatest deal of indoor thoron measurements were made in Bol'shie Koty Village, where ^{220}Rn contents were measured not only in soil but also in dwellings. With an average thoron activity in soil of $31,900 \text{ Bq}/\text{m}^3$, the volume activity of this isotope in dwelling air was $81 \text{ Bq}/\text{m}^3$. The ratio between them was about 400:1. The same order of magnitude was obtained in single measurements made in other villages of the Baikal region. Given this ratio, we expect that

village-averaged volume activity of indoor thoron will be $18 \text{ Bq}/\text{m}^3$ in Odinsk, about $40 \text{ Bq}/\text{m}^3$ in Podkamennaya, and $83 \text{ Bq}/\text{m}^3$ in Listvyanka (with average thoron activity in soil of $33\,320 \text{ Bq}/\text{m}^3$).

Conclusion. Estimation of radon hazard of an inhabited area

Radon content less than $10 \text{ kBq}/\text{m}^3$ is sufficient to create indoor concentration above the permissible limit $200 \text{ Bq}/\text{m}^3$. This suggests a considerable flow from soil to dwelling (Scott, 1992). The content of indoor ^{222}Rn increases with soil gas, which also leads to a greater number of houses out of safe standards. At a concentration of more than $100 \text{ kBq}/\text{m}^3$ in soil the permissible limit $200 \text{ Bq}/\text{m}^3$ was exceeded in more than 80% of dwellings.

To categorize the radon hazard of an inhabited area, we can level the scattered and uncertain volume activities of radon isotopes by their averaging over a considerable number of measurements performed either in a short period (under equal meteorological conditions) or in runs in different seasons. To characterize a restricted territory, e.g., a village or its part, the use of averages is permissible.

Consider characteristics of the sampled territories or separate villages. Of course, a separate area is characterized by a wider range of soil radon contents than one or two intervals of "logarithmic" scale. Table 2 shows generalized results of radiation inspection of separate villages situated in different radon-hazardous provinces of the Baikal region. The results of single and integral measurements of radon contents in representative samples of houses are compared with measurements of radon flows from soil, radon activities in soil gas and contents of natural radionuclides in soils on the territory of these villages. Volume activity of radon averaged over the sample of inspected dwellings, with upper and lower limits of variation, was obtained for each inhabited area under study.

Table 2 reports results of comparison of village-averaged radon activities in soil gas and indoor air of the inspected villages of the Irkutsk Region. Ratios are usually about 45. The ratios between averaged contents of soil and indoor radon

Table 2

Variation ranges and geometric means of volume activities (VA) of soil and indoor ^{222}Rn in villages of the Baikal region

Village	VA variation of soil ^{222}Rn , Bq/m^3	Mean VA in soil, Bq/m^3	VA variations of indoor ^{222}Rn , Bq/m^3	Mean VA in dwellings, Bq/m^3	Village-averaged soil-to-indoor VA ratio
Belaya Zima	1800–695 300	57577	37–1466	325	177.2
Bol'shie Koty	2400–38 900	12872	100–742	262	49.0
Listvyanka	3500–88 800	11540	20–960	250	46.2
Bol'shaya Rechka	2000–74 800	10600	20–1100	232	45.7
Maloe Goloustnoe	400–48 000	5197	27–434	132	39.4
Podkamennaya	600–70 500	5056	20–252	110	46.0
Karluk	900–22 000	4196	7–369	108	38.7
Oek	400–17 000	3302	17–270	68	48.6
Odinsk	550–6500	2726	17–113	66	41.3

measured in zones of different radon hazard are close (with a correction on the soil radon activity) despite varying geology of the areas under study.

A seemingly exception is Belaya Zima Village, where unexpectedly we have obtained a higher ratio against the background of a higher activity of soil radon, i.e., though the radon contents in dwellings considerably exceed the safe standards, the volume activity of indoor radon appeared to be lower than we could expect with equal soil-to-indoor radon ratios.

However, it was shown earlier (e.g., Varley and Flowers, 1994) that the soil-to-indoor radon ratio is not constant with respect to soil radon activity. This should be taken into account when summarizing the measurements and making forecasts for inhabited areas (see Table 2). Therefore, exceptions are data for Bol'shie Koty and Listvyanka Villages rather than for Belaya Zima.

The observed behavior of the soil-to-indoor ^{222}Rn ratio suggests a possibility for these ratios to be used for forecasting the radon hazard in other villages of this kind not only in the Irkutsk Region but also in the southwestern Baikal region. Though this kind of measurements is of little value for prediction of indoor radon, there is no doubt that to measure soil gas radon is a useful technique to predict an averaged level of indoor radon for this territory. However, it is necessary to note that the use of these data does not warrant a success for other territories. In various studies it appeared difficult to establish a correlation between indoor and soil gas radon. The relations between concentrations of these kinds of radon remain to be understood, but it is clear that each geographical

area will have its own features controlling the radon pollution — in geology, building constructions, and lifestyle of people.

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