Radon Emission from Coal Mines of Kuzbass Region

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Abstract. The article represents the results of a research in radionuclides concentration in coal and rocks of Kuzbass mines as well as radon concentration in operative mines and mined-out spaces. It is proved that radon concentration in mines is considerably higher than in the atmosphere and it rises drastically in the mined-out spaces. It is found out that radon is carried out from mines by ventilation flows and from open pits, generating anomalous concentrations over self-ignition areas.

1. Introduction
Coal mining causes carry-over of hazardous gases released from coal and enclosing rocks into the atmosphere. A great part of the gases released from mines is dangerous due to their toxicity and fire behavior and explosiveness at a certain concentration in the air. Most inflammable gases have been generated in the process of coal rock metamorphism. When coal beds are developed extra gases are released due to mechanical destruction of coal and its low-temperature oxidation. The rate of inflow of different gases into the mine atmosphere can vary considerably through time and space depending on geological and technological conditions.

Radon takes a special place among gases released in mines and then carried over into the atmosphere. Radon is a colorless, odorless inert gas with density 9.9 kg/m³ and boiling temperature -62 °C. Unlike conventional mine gases radon-222 belongs to a radioactive uranium-radium family and intermediate product of uranium-238 decay. Radon is produced from radium-226 with half-life as long as 1.600 years. The inert gas radon-222 which is produced at its α-decay has half-life as long as 91.68 hours. Annual average concentration of radon in the outside air is from 0.1 to 10 Bq/m³. Natural radon radiation background in the air outside premises is about 610Bq/m³ annually in average. Radon decay generates α-particles with total energy 5.482 MeV and γ-radiation with total energy 0.51 MeV [2, 3].

Concentration of radionuclides in soil and rocks depends on their initial activity as well as on mass transfer process in the earth interior caused by underground water movement and tectonic activity. That is why the amount of natural radionuclides in rocks can differ tenfold or even hundredfold. For example, specific activity of uranium -238 in coal vary from 3 to 52010 Bq/kg and its amount in soil is 10-50 Bq/kg [1, 5]. When coal is burnt its organic components turn into gases and volatilize while non-organic components and natural radionuclides remain. Thus concentration of natural radionuclides in coal ash can considerably grow compared with initial coal [2].

Radon generated by radium-226 decay in unbroken coal layers and enclosing rocks moves insignificantly in pores and produces new elements during further decay. Actual mining considerably alters the radium behavior. Due to large surface of coal and rocks in mined-out space some part of radon molecules produced from decayed radium-226 is carried out from inside coal and rocks to the
surface. Some amount of radon is accumulated on the surface of coal and rocks while the other part is released into the mine atmosphere [6]. Radon is carried out by escaping air into operating mine openings and into the atmosphere. Being inhaled decayed radon is harmful for living things. Thus it is important to determine the amount of radon in the mine air and develop ways to carry it out into the atmosphere.

2. Results and discussion
The content of radionuclides in coal and rock of some Kuzbass mines was studied by means of gamma-spectrometric analysis of collected samples. Gamma activity of samples determined the content of the four radionuclides: caesium-137, potassium-40, radium-226, thorium-232. The research showed that the content of natural radionuclides in Kuzbass coal can change 2–4 times. Average volume activity of radium in coal samples is 9.3 Bq/kg, the most radioactive coal is found in the south and north of the region. Radium content in these coals is as great as 20 Bq/kg while in coals of Kemerovo and Leninsk-Kuznetskiy it is not higher than 10 Bq/kg. Experiments showed that the total concentration of radionuclides in enclosing rocks from five to ten times higher than that in coals. Radium activity can reach 50 Bq/kg and potassium-40 is sometimes as active as 1000 Bq/kg. Measurements showed that within one coal field, mine or even coal bed there can be detected considerable changes in concentration of radioactive elements in coal and rocks.

Volume activity of radon in mine atmosphere was determined by means of adsorption method, activated carbon being used as an adsorber. This method is essential when a large number of samples is collected and when low values of radon volume activity are measured. The method has many advantages: efficiency, high sensitivity, portable equipment, availability in any atmosphere. During the research radon concentration in productive mines and mined-out space was determined. Isolated mined-out space was monitored through wells drilled down from the surface or through productive mine faces and also through wall stopings.

Table 1 shows measurements of radon volume activity in mine atmosphere of some productive mine faces of Kuzbass region.

| No | Mine, bed | Sampling site | Volume activity, Bq/kg |
|----|-----------|---------------|-----------------------|
| 1  | "Pervomayskaya", bed XXII Belt road 724 | 54 |
| 2  | "Pervomayskaya", bed XXII Air roadway | 125 |
| 3  | "Pervomayskaya", bed XXI Belt road | 62 |
| 4  | "Pervomayskaya", bed XXI Air roadway | 83 |
| 5  | "Raspadskaya", bed 6-6a Belt road 5-6-10 | 61 |
| 6  | "Raspadskaya", bed 6-6a Air roadway 3-3-11 | 82 |
| 7  | "Raspadskaya", bed 3-3a Belt road 3-3-11 | 16 |
| 8  | "Raspadskaya", bed 3-3a Air roadway 3-3-11 | 93 |
| 9  | "Raspadskaya", bed 5 Belt road 5-5-5 | 15 |
| 10 | "Raspadskaya", bed 5 Air roadway 5-5-5 | 92 |
| 11 | "Gramoteinskaya", bed Sychoevskiy 1 Belt road 526 | 23 |
| 12 | "Gramoteinskaya", bed Sychoevskiy 1 Air roadway 526 | 89 |
| 13 | "Kolmogorovskaya", bed Baikaimskiy Belt road 1304 | 175 |
| 14 | "Kolmogorovskaya", bed Baikaimskiy Air roadway 1304 | 288 |
| 15 | "Usinskaya", bed III Belt road 3-3-15 | 95 |
| 16 | "Usinskaya", bed III Air roadway 3-3-15 | 108 |
| 17 | "Yagunovskaya", bed Volkovskiy Belt road 222 | 82 |
| 18 | "Yagunovskaya", bed Volkovskiy Air roadway 222 | 91 |
| 19 | "Baidaevskaya" bed 29 a Belt road 29-31 | 48 |
Analysis of the data represented in Table 1 shows that radon volume activity in the air of productive mines does not exceed the maximum permitted limit but it is considerably higher than in the atmospheric air. But when the air from mined-out spaces is carried over into productive mine faces the maximum permitted concentration of radon in the air can be significantly exceeded.

Table 2 shows data of radon volume activity in the air of mine-out space of productive mines as well as in the air of isolated mined-out spaces.

Table 2. Volume activity of radon in the mined-out space atmosphere

| No | Mine, bed | Sampling site | Volume activity, Bq/kg |
|----|-----------|---------------|-----------------------|
| 1  | "Pervomayskaya", bed XXYII | Upper face niche 31-71 | 107 |
| 2  | "Pervomayskaya", bed XXYII | Upper face niche 724 | 134 |
| 3  | "Pervomayskaya", bed XXI | Upper face niche 12-15 | 74 |
| 4  | "Raspadskaya", bed 6-6a | Upper face niche 5-6-10 | 111 |
| 5  | "Raspadskaya", bed 6-6a | Through wall stopings | 178 |
| 6  | "Raspadskaya", bed 5 | Upper face niche 5-5-3 | 184 |
| 7  | "Raspadskaya", bed 5 | Coming out of the well | 309 |
| 8  | "Raspadskaya", bed 3-3a | Upper face niche 3-3-11 | 113 |
| 9  | "Kolmogorovskaya", bed Baikaimskiy | Upper face niche 1304 | 349 |
| 10 | "Usinskaya",bed III | Upper face niche 3-3-15 | 129 |
| 11 | "Usinskaya",bed III | Through wall stopings 778 | 273 |
| 12 | "Usinskaya",bed III | Through wall stopings 726 | 390 |
| 13 | "Yagunovskaya", bed Volkovskiy | Upper face niche 222 | 116 |
| 14 | "Baidaevskaya" bed 29 a | Well into the face 29-31 | 2594 |
| 15 | "Baidaevskaya" bed 29 a | Well into the face 29-31 | 4127 |
| 16 | "Baidaevskaya" bed 30 | Well into the face 30-35 | 1077 |
| 17 | "Baidaevskaya" bed 30 | Well into the face 30-35 | 5636 |
| 18 | "Baidaevskaya" bed 29 a | Through wall stopings | 1144 |
| 19 | "Baidaevskaya" bed 29 a | Through wall stopings | 1066 |
| 20 | Named by V.I.Lenin, bed IY-Y | Upper face niche 0-5-1-7 | 138 |
| 21 | Named by V.I.Lenin, bed IY-Y | Through wall stopings | 256 |
| 22 | "Alarda", bed 1 | Through wall stopings | 555 |
| 23 | "Yesaulskaya", bed 29a | Through wall stopings | 1580 |
| 24 | "Yesaulskaya", bed 29a | Through wall stopings | 1304 |
| 25 | "Inskaya", bed Sychevskiy 1 | Through wall stopings | 4211 |

Comparing the data represented in Tables 1 and 2 we can see that in mined-out spaces radon volume activity is much higher than in operative mines. In poorly aired areas (upper or lower face niche) radon volume activity is at the maximum permitted level and sometimes higher. In isolated mined-out spaces radon concentration can tenfold exceed the maximum permitted level.
Thus, the experimental research carried out in mines proved that radon volume activity in mine atmosphere can change considerably and often exceed sanitary standards. The most important factors influencing the radon concentration in mine atmosphere are ventilation (an amount of incoming air), natural radioactivity of mined rocks, a dispersion degree of broken-down rock. Examining of the mine atmosphere showed that volume activity of radon daughters in the mine atmosphere can reach 4000–6000 Bq/m$^3$. The highest concentration of radon is found in mined-out areas which have been isolated for a long time.

Coal mining works promote carrying-over the air from the mined-out spaces onto the earth surface. The mine air containing a great amount of radon is carried out into the atmosphere mostly with ventilation flows. Gas from the mined-out spaces comes out into the atmosphere from wells and cracks in the rock. The analysis of causes stimulating carrying-over of mine gases into the atmosphere represented in [7] proved that the most important factors influencing the intensity of gas transfer are changes in gas pressure between the mined-out spaces and the atmosphere, rock permeability and a stratification depth of gas releasing rocks.

Emission of radon from the earth surface over mined-out spaces was studied at a number of Kuzbass mines. The distance between the mined-out spaces and the earth surface was from 100 to 400 m. Measurements were taken with the help of diffusion collectors with activated carbon installed on the surface. After exposure sorbent activity was determined and radon flows were calculated. Experiments showed that in Kuzbass, a natural level of radon flows from the earth surface over mine fields is about 10–70 Bq/(m$^2$ s). These values prove that mined-out spaces are properly isolated and radon does not reach the surface. Over steeply pitching coal, rock permeability increases greatly which leads to anomalous zones of radon emission. Radon flows in such zones are as great as 100–500 Bq/(m$^2$ s).

The research also showed that apart from rock permeability radon emission is effected by coal self-ignition in the mined-out spaces. High temperature in coal self-ignition places causes heat depression which intensifies carrying over of gases from mined-out spaces to the earth surface. Thus on the earth surface over places of coal self-ignition an anomalous emission of radon occurs. It can be used to find and allocate coal self-ignition areas.

Though a rise of temperature does not affect the rate of radon decay, heating can still cause an increase in carrying-over of radon into the atmosphere due to evaporation of inherent moisture from coal and rocks and growth of molecular diffusion coefficient caused be temperature rise. Decrease in radon solubility factor in water at elevated temperatures can also stimulate radon emission into the air. For example, if water is heated from 10 to 70 ºС radon concentration goes down three-fold.

Measurements of radon flows at the earth surface over underground self-ignition areas repeatedly revealed anomalous zones with high radon concentration. Areas with radon concentration of 100–400 mBq/(m$^2$ c) were detected and in some places radon concentration was as high as 1000–1500 mBq/(m$^2$ c). Radon anomalies at the earth surface coincided with vertical projection of self-ignition places and in few cases they were slightly moved apart from the projection.

3. Conclusions
1. Concentration of radium in the examined coals of Kuzbass region varies from 6.9 to 24.7 Bq/kg and in rocks it can measure from 21 to 48.8 Bq/kg which leads to background volume activity of radon in the mine atmosphere.

2. Background volume activity of radon in the mine atmosphere changes widely and often exceeds the maximum permitted level. The concentration of radon in the mine atmosphere is mostly affected by natural radioactivity of driven rocks, a degree of rock fragmentation, a speed of air flow. The highest radon concentration is found in the atmosphere of continuously isolated mined-out areas, being tenfold higher than the maximum permitted level.

3. Radon is carried out into the atmosphere mainly with ventilation flows. Some part of radon penetrates into the atmosphere through pores and cracks in the rocks. Underground self-ignition considerably intensifies radon emission generating convection gas flows to the earth surface.
4. Surface monitoring allows detecting locations of underground self-ignition areas judging by the amount of radon flows from the surface.

4. References

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