Radioactivity of Mine Waters in Upper Silesian Coal Basin and its Influence on Natural Environment

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ABSTRACT

Waters with high mineralization occurring in coal mines in the Upper Silesian Coal Basin often contain natural radioactive isotopes, mainly radium. In these waters elevated concentrations of ^{226}Ra, ^{228}Ra have been found. Concentration of ^{226}Ra in waters flowing out from the rock into underground mine workings may be as high as 390 kBq/m^3, and concentration of ^{228}Ra is sometimes higher than that of ^{226}Ra.

Sometimes radium-bearing waters contain also barium ions, which concentration may reach 1.5 kg/m^3. Waters containing radium and barium ions as well have been called radium waters type A. Another type of radium-bearing waters, called type B does not contain barium ions, but contains SO_4^{2-}. From waters type A which contain barium, radium is always coprecipitated out by sulfates. Concentration of ^{226}Ra in such deposits may be as high as 400 kBq/kg. Precipitation takes place in underground workings, settlement ponds, pipelines, small rivers and so on. Radioactive deposits cause pollution of the natural environment and sometimes serious technical problems as well.

On the contrary, radium waters type B do not form radioactive deposits, because no carrier such as barium for coprecipitation is present. In this case radium is transported with waters to main rivers, where dilution takes place. Enhanced radium concentrations have been observed even over hundred kilometers downstream from the source.

INTRODUCTION

One of the most serious problems met by coal extraction in Upper Silesia in Poland are waters with very high salinity occurring in carboniferous strata, described by many authors [1,2]. These waters often contain high concentrations of natural radionuclides, mainly radium isotopes. Upper Silesian radium-bearing waters were first mentioned by M. Saldan [3] and later investigated and described by I.Tomza [4] and others. As described in these papers, the dominating radioisotope in these waters was radium-226, a member of uranium series. Concentration of ^{226}Ra in Upper Silesian brines is usually between 1 and 100 kBq/m^3, with the maximum value 390 kBq/m^3. Since the amount of saline water drained by underground mine workings is as high as 91000 m^3 per day, also the total activity of ^{226}Ra is rather high reaching 650 MBq per day. Some of these waters contain not only radium, but barium ions as well. When mixing of such waters with other natural waters containing sulfate ions occurs, radium is coprecipitated with barium as sulfates. So originated radioactive deposits sometimes cause significant radiation hazard for miners and for the natural environment, where radium-bearing waters are eventually released.

Natural waters with similarly high radium concentration were also observed by other authors [5], especially in oil fields. Hot springs in Iran, described by Khademi [6], contained up to 330 kBq/m^3 of ^{226}Ra. Also in a coal mine in Germany [7] occurred radium-bearing waters with ^{226}Ra concentration up to 63 kBq/m^3. Although there are several publications on elevated ^{226}Ra concentration in water, there is much less known about radium isotopes from the thorium series - ^{228}Ra and ^{224}Ra, since these isotopes are much more difficult to measure. We have investigated ^{228}Ra and ^{226}Ra in radium-bearing mine waters and deposits from the Upper Silesian Coal Basin and their influence on the natural environment in mining areas.

INVESTIGATION SITE

The Upper Silesian Coal Basin (USCB) is located in the Southern-West part of Poland. Presently there are 66 underground coal mines there extracting approximately 150x10^6 tons of coal per year. The depth of mine workings is from 350 to 1050 m. Upper Silesia is characterized by a very complicated and differentiated geological structure with numerous faults and other tectonic dislocations [8]. Additionally, the area is very affected by mining

Two hydrological regions of the Coal Basin have been distinguished. The first region is located in southern and western Silesia with thick strata of sediments covering carboniferous formation. This overlay is built mainly by Miocene clays and silts. The thickness of this rock is up to 700 m. Such strata make almost impossible migration of water and gases. In the second region Miocene clays do not occur. Carboniferous strata are covered by Mesozoic and Quaternary sediments, slightly compacted.

The oldest formations of this area form isolated sediments of permian or triassic limestones strongly fissured. There are numerous outcrops of coal seams. These formations enable very easy migration of water and gases.

EXPERIMENTAL

Sampling

Sampling was done by the mining staff as a routine required by Polish regulations for chemical and radiological analyses. Waters inflowing into underground workings directly from rocks or from boreholes and waters pumped out from mines were sampled as well.

Determination of radium isotopes

Determination of ^{226}Ra and ^{228}Ra was done by liquid scintillation counting after chemical separation of $RaSO_4$ with a barium carrier according to the method developed by Chalupnik and Lebecka [9]. This method enables not only determination of ^{226}Ra, but also ^{228}Ra as well. ^{228}Ra is determined by direct measurements of low energy beta particles emitted by this radioisotope, while ^{226}Ra is determined by measurement of alpha particles emitted by this radionuclide and its daughter products. Since the beta spectrum is a continuous one also beta particles emitted by ^{226}Ra daughter products are measured in the same energy range. Therefore correction of this effect is required. Necessary corrections are made by crosscalculations. The lower limit of detection (LLD) for ^{226}Ra as low as 0.002 kBq/m^3 is achieved when a low background liquid scintillation spectrometer QUANTULUS is used. Accuracy of this method was confirmed by several intercomparison runs [10].

The described methods enables also determination of ^{224}Ra but because of short half life of this radioisotope the measurement must be done shortly after sampling. Since ^{224}Ra is much less radiotoxic than ^{226}Ra and ^{228}Ra this radioisotope is not determined in a routine measurements.

RESULTS

Concentration of ^{226}Ra in waters flowing out from the rocks ranged from below 0.1 up to 390 kBq/m³. Concentrations of ^{228}Ra was usually lower, however in some waters exceeded three times or more that of ^{226}Ra. Upper Silesian radium-bearing waters are highly mineralized (dissolved matter up to 230 kg/m³) type Cl-Na or Cl-Na-Ca.

Two types of radium-bearing waters have been distinguished: type A, containing not only radium ions but also barium ions as well, and waters type B with no barium but containing sulfate ions. Concentration of barium in waters type A ranged from 20 g/m³ to 2 kg/m³, while maximum radium concentration (in terms of weight) was of several ng/m³. Concentration of sulfate ions in waters type B reached over 3 kg/m³. Presence of barium ions is essential for the behaviour of radium in the environment, because it enables coprecipitation of radium and barium sulfates. Therefore radium present in water type A is always precipitated out. As a result radioactive deposits $BaSO_4$ + $RaSO_4$ are formed. This process occurs either by spontaneous or controlled mixing of natural waters. Precipitations takes place in underground mine workings or in streams, little rivers or pipelines.

Waters with high radium concentration occur mainly in the southern and central part of the coal basin [12], where carboniferous strata are covered by a thick layer of miocenic clays. Much higher radium concentrations are observed in radium-bearing waters type A, where ^{226}Ra concentrations often exceeds 10 kBq/m³ and sometimes is higher than 100 kBq/m³. In waters type A concentrations of ^{226}Ra is usually higher than ^{228}Ra. In waters type B the dominating radium isotope is ^{228}Ra from thorium series. On the other hand, concentration of both radium isotopes in waters type B is much lower and does not exceed 12 kBq/m³. Characteristics of Upper Silesian radium-bearing waters are given in Table 1.

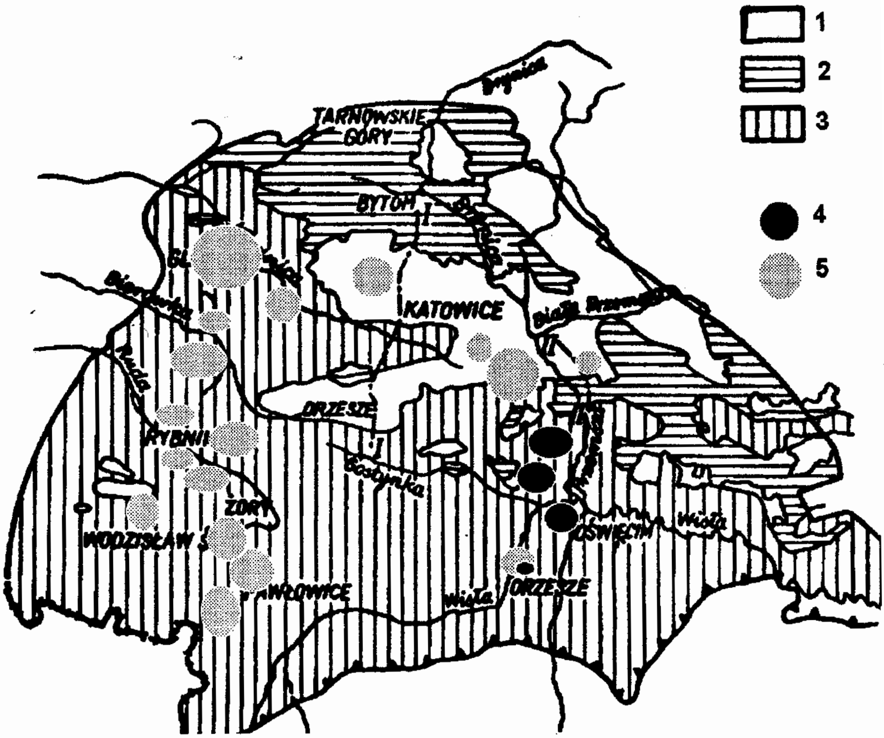
Table 1. Characteristics of typical radium-bearing waters from Upper Silesian Coal Basin

Type of water	Radium concentration kBq/m³		Chemical composition kg/m³		
	^{226}Ra	^{228}Ra	mineralization	Ba^{2+}	SO_4^{2-}
A	0.5 - 390	0.5 - 200	30 - 200	0.02 - 2	traces
B	0.1 - 12	0.1 - 20	5 - 100	not present	up to 3.5

In most cases, thanks to dilution and spontaneous or controlled precipitation of radium and barium sulfates, mine waters pumped out to the surface are free of radium. In two mines a

Figure 1.

Occurrence of radium-bearing waters in Upper Silesian Coal Basin



1. Quaternary sediments
2. Triassic sediments
3. Tertiary sediments

4. Radium-bearing waters, type B
5. Radium-bearing waters, type A

technology of underground purification of radium and barium has been implemented [13]. Nevertheless, waste waters released by 6 mines contain more radium than accepted by Polish regulations i.e. 0.7 kBq/m³ of ^{226}Ra. The total activity of ^{226}Ra released from these mines is of about 350 MBq/day, while the activity of ^{228}Ra is of 540 MBq per day. These are waters type B. Radium carried by these waters is diluted in rivers including River Vistula. present and former discharge of radium with mine waters caused enhanced natural radioactivity in the vicinity of of small streams and rivers in Upper Silesia. Comparing to the normal ^{226}Ra concentration in river water in Holland and Germany, which according to [14] is below 0.008 kBq/m³, most of Upper Silesian rivers where waters from coal mines are released have definitely higher radium concentration. Maximum value in a small river was as high as 1.3 kBq/m³ . Also bottom sediments in small and main rivers Vistula and Oder are clearly enhanced.

Although the impact of radium-bearing waters from Upper Silesian coal mines can be clearly seen, that radiation doses obtained by inhabitants are not high. Preliminary evaluation shows that annual excess doses caused by radioactive waste waters and waste rocks released by coal mines do not exceed 1 mSv per year.

CONCLUSIONS

Mine waters from underground coal mines due to high radium concentration may cause significant enhancement of natural radiation in the environment. Highly mineralized waters in Upper Silesia contain not only high concentrations of ^{226}Ra from uranium series, but ^{228}Ra from thorium series as well. $^{228}Ra/^{226}Ra$ ratio is higher than one in waters containing sulfates rather than barium ions. Elevated radium concentrations in rivers where waters from coal mines are released are observed not only in river water but also in bottom sediments many kilometers from the source.

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