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Long term changes in the concentration of radium in discharge waters of coal mines and Upper Silesian rivers

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ABSTRACT

According to the latest guidelines of the International Atomic Energy Agency (IAEA, 2016), coal mining is one of the most important contributors to occupational exposure. Coal mining contributes about 45% of the total annual collective dose obtained by workers due to the exposure at places of working. One of the sources of exposure in mining are formation brines with elevated concentrations of natural radionuclides, the most common are radium ^{226}Ra and ^{228}Ra. Radium isotopes often occur in formation waters in underground collieries in the Upper Silesian region (USCB) in Poland. Significant amounts of radium remain underground in the form of radioactive deposits created as a result of spontaneous deposition or water treatment. This phenomenon leads to the increase of radiation hazard for miners. The remaining activities of ^{226}Ra and ^{228}Ra are released into the rivers with mine effluents, causing the contamination of bottom sediments and river banks.

The results of radioactivity monitoring of effluents and river waters are presented here to illustrate a trend of long-term changes in environmental contamination, caused by mining industry in the Upper Silesian Region.

1. Introduction

Located in the Southern part of Poland, the Upper Silesian Coal Basin (USCB) is characterized by a complicated geological structure of the sedimentation basin, moreover the area is also strongly affected by mining activity. Nowadays, there are approximately 30 underground coal mines in the region. These mines extract, on average, 80 million tons of coal per year. To allow this exploitation, dewatering of mine workings must be undertaken to remove inflows of formation and technological waters. The total release of water from these mines exceeds 700,000 m^3/day. The salinity of these brines often far exceeds that of ocean waters, the amount of total dissolved solids carried with mine waters to the rivers is roughly 10,000 t/day. The range in concentrations of total dissolved solids (TDS) may vary in discharge waters from 2 to 3 g/l up to 120 g/L. The most frequently found ions in these brines, include Cl^- and Na^+. Apart from Cl^- and Na^+, abundant concentrations of Ca^{2+}, Mg^{2+} and SO_4^{2-} can be found in brines together with significant amounts of other types of ions (Tomza and Lebecka, 1981; Różkowski et al., 1993). The mine effluents are released to the tributaries of the two main Polish rivers – Vistula and Oder. Since 1980, the discharge waters from several coal mines, located in the catchment area of Oder river, were collected (gathered) and released into the Oder River via the so-called OLZA Pipeline (see Fig. 1).

The Pipeline was used to transport saline waters from coal mines in the south-western part of the Silesian Coal Basin (USCB). The aim of this system was to protect small rivers from salinity, present in waters, released from coal mines. In these mines, the process of co-precipitation of barium and radium sulphates occurs mostly in underground galleries (spontaneously or due to treatment technologies). This is why the radium activity in waters discharges into the Oder River via he OLZA Pipeline have been very low since the early 1990's (Lebecka et al., 1994).

Due to their high salinity, brines cause damage to the natural environment. Significant environmental impact is also due to high concentrations of radium isotopes, present in these type of waters. Waters with enhanced radium content of up to 390 kBq/m^3 of ^{226}Ra

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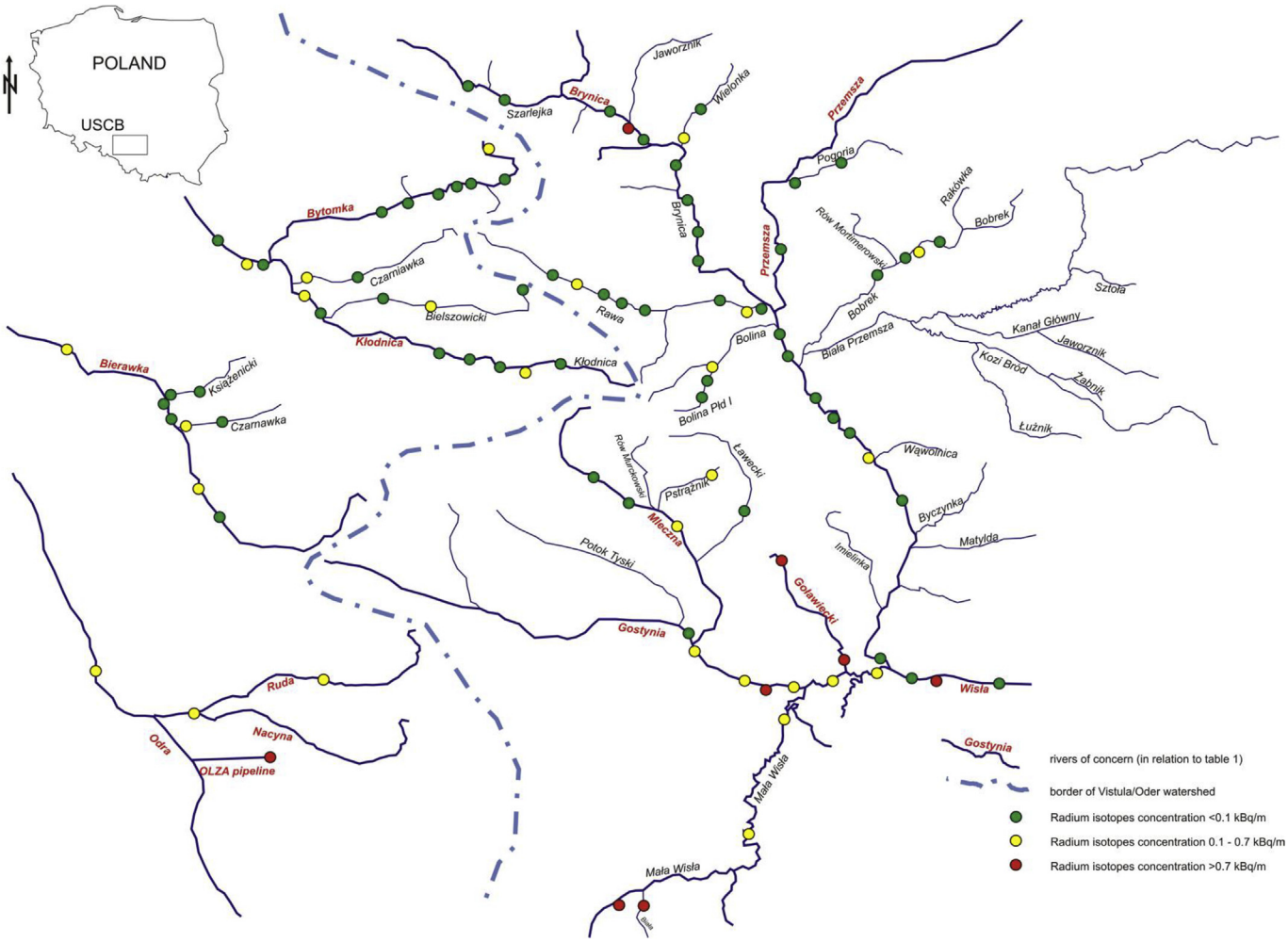

Fig. 1. Location of sampling points in rivers of USCB.

(Lebecka et al., 1994; Michalik, 2007, 2008; Michalik et al., 2013; Skubacz et al., 2009) occur in many of the coal mines located in the Upper Silesian Coal Basin. In some parts of the basin, the Carboniferous strata are overlaid by a thick layer of impermeable clays (Kotas, 1990). In such zones, there is no hydraulic connection between the surface and aquifers, because such strata restricts the migration of meteoric waters and gases. In mines, located mostly in southern and western Silesia, there is a thick cover of up to 700 m of impermeable Miocene clays and silts. Water inflows there are mostly old formation brines. Such impermeable layers inhibit the infiltration of meteoric waters from the surface, there are no water mixing zones and no dilution of salinity and radium occurs. As the radium content in mine waters is correlated with its salinity, radium-bearing waters containing additionally barium ions occur mainly in the southern and south-western part of the basin, sometimes at relatively shallow depths (300–400 m below surface). In other parts of USCB, the hydraulic conductivity of rocks covering the Carboniferous productive measures is higher and the radioactivity of mine waters at depths to 400–500 m is usually low. The hydraulic conductivity of overburden rocks is relatively high, partly due to high intergranular and fissure porosity. The permeability of overlying dolomites and limestones is often enhanced due to the historical shallow metal mining activity as well as karstic development (Różkowski et al., 1993; Różkowski, 1995). The salinity and radioactivity of mine waters increases at these locations due to exploitation of deeper coal seams. In this part of the USCB (the northern and central part of USCB), radium rich waters without barium ions are more common in mines.

The presence of barium in waters is the most important indicator of further behaviour of radium isotopes in mine galleries or on the surface (Tomza and Lebecka, 1981; de Jesus, 1984). In the USCB collieries, barium ions at concentrations of up to 2 g/l are present in radium-bearing brines. Such waters were classified in Poland as radium-bearing waters type A. Where no barium can be found in brines but radium and sulphate ions are present, radium-bearing waters were categorised as type B.

When type A waters are mixed spontaneously or intentionally with any waters, containing sulphate ions, radium and barium always co-precipitate as sulphates. As a result of the precipitation, deposits of barium sulphate are formed, with enhanced radium concentrations (Lebecka et al., 1994; Michalik et al., 2013). In the case of radium-bearing waters of type B, no precipitation occurs due to the lack of the carrier for radium.

In order to monitor the radium concentration in mine waters effluents and rivers, routine measurements of radium in discharge and surface waters started in Poland 30 years ago, in 1986. These measurements were obligatory and carried out in all coal mines located in the USCB region with a frequency of at least once per year. The measurements of radium isotopes in discharge waters and rivers were initially performed for monitoring purposes only.