ENHANCED NATURAL RADIOACTIVITY IN POLISH HARD COAL MINING INDUSTRY

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Abstract

The radiation risk due to the exposure to natural radionuclides, especially to short-lived radon progeny, is a component of the radiation hazard, common in the natural environment and working environment of people. The effective dose, caused by the exposure to radon (radon progeny), exceeds 50% of the average dose for a man from all sources of the ionising radiation. Under specific circumstances, for example as a result of working in confined space with low ventilation rate (cellars, underground galleries, tunnels, mines), the risk caused by radon and its progeny can be enhanced and can reach significant levels.

In Polish mining industry the radiation hazard, caused by natural radionuclides is one among many other natural hazards. It is worth to be point out that besides radon another source of radiation hazard in coal mines is radium, present in underground brines. Sediments, precipitated out from such waters, have enhanced radium content and may cause the increase of external gamma radiation dose rate as well as internal contamination due to accidental ingestion or inhalation.

Investigation of that specific problem has been started in hard coal underground mines in Poland in early 1970’s. At the end of 1980’s the first regulations were issued: the national standard have been prepared, in which dose limits and requirements of the radiation monitoring have been established. In following years in some branches of underground mining (mainly in coal mining industry) internal regulations for monitoring and mitigation measures were developed on the basis of the Polish standard.

Due to being in force regulatory acts - Geological and Mining Law, Decree of the President of State Mining Authority and Decree of the Ministry of Economy – the monitoring and prevention against natural radiation is obligatory in all Polish underground mines since 1989. This duty is strictly supervised by local offices of State Mining Authority in co-operation with other governmental agencies. Monitoring and mitigation measures are obligatory not only for active mines but also for mines, excluded from the exploitation, and used for other purposes as museums, balneotherapy spas etc. Such solution is unique in non-uranium mining.

Far less attention has been paid to environmental burden caused waste with natural radioactivity enhanced due to mining activity. Such waste, mainly sediments differ significantly from “classical” nuclear materials and the derived radiation risk is usually associated with risk caused by other pollutants and can not be controlled applying rules designed for pure radioactive waste. Existing data have pointed out strong needs to take into account mining industry as a special case of radiation risk and enclose them in frame of the formal control. But up to now there are not reasonable and clear regulations in this matter. As a result, the coal mining industry is not aware of problems connected with natural radioactivity or they would expect negative consequences in case of implementing radiation protection measures. The modification of widely comprehended environmental legislation with requirements taken from radiation protection seems to be the first step to solve this problem and make awareness about enhanced natural radioactivity for all stakeholders of concern.

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1. Natural sources of the ionising radiation in underground mines

The radiation hazard is one of the natural hazards in underground mines, like methane hazard, tremor hazard etc. The investigations of the natural radioactivity in underground mines in Poland have been started in early 1970’s [Tomza & Lebecka, 1981]. Firstly, efforts were focused on solving the problem of occurrence of radium-bearing waters and sediments with enhanced radioactivity in underground galleries [Lebecka et al., 1985]. Later the importance of the exposure to radon progeny have been noticed [Chalupnik et al., 1986].

During our investigation have been found, that main sources of natural radiation in Polish coal mines were as follows [Lebecka et al., 1985] short-lived radon progeny and deposits with enhanced radioactivity, precipitated out of radium-bearing mine waters.

The phenomenon of the radioactivity of saline waters from coal mines in Poland was discovered in the 1960’s [Sałdan, 1965]. Later, investigations showed, that radium concentration in water was correlated with the salinity [Tomza & Lebecka, 1981]. In some cases, the total dissolved solids concentration (TDS) exceeds 200 kg/m³, whilst the radium concentration may reach 400 kBq/m³. The analysis of the radium isotopes in inflows showed, that the input of $^{226}$Ra was of about 725 MBq per day, while the corresponding value for $^{228}$Ra was roughly 700 MBq per day [Wysocka et al., 1996]. Only 40% of radium remained in the underground galleries and gauntons, whilst 60% were transported in pumped waters to the settling ponds on the surface and later to rivers [Lebecka et al., 1994]. It was an important source of contamination of the natural environment.

Two types of brine were distinguished. In coal mines In type A waters ions of barium and radium are present, while in type B waters only radium ions and sulphate occur, but no barium [Lebecka et al., 1986]. From type A waters radium is very easily precipitated out with barium carrier as sulphates after mixing with waters rich in sulphate ions. This is the source of sediments with enhanced radium content. Such process often occurred in underground galleries, leading to the radioactive contamination and enhancement of gamma-dose rates. The highest levels of contamination were always connected with release of A type waters and precipitation of insoluble deposits with enhanced radium content.

In B type waters there is no convenient carrier for radium, therefore precipitation of radium scales doesn’t occur. Radium from B-type waters is slowly adsorbed on suspended matter, therefore the level of contamination of underground work places and environment is much smaller in comparison with A type waters.

Taking into account mine crew the main source of the radiation hazard is the exposure to short-lived radon progeny, present in the ventilation air [Skowronek et al., 1997]. In underground galleries radon and radon progeny concentrations are higher than outdoor, because that are places with limited ventilation, confined in the rock body with many cracks and fissures. In coal mines in Upper Silesia region the radon concentration up to 15 000 Bq/m³ was measured [Skowronek, 1991]. Moreover, in mines of Lower Silesian Coal Basin galleries with radon concentration up to 150 000 Bq/m³ were found [Lebecka et al., 1985].

In monitoring of radon progeny in mines hardly ever concentrations of particular short-lived decay product of radon are measured. Usually the term of potential alpha
energy concentration (PAEC) is used. The PAEC values are expressed in joules per meter cubed and this term can be explained as the total energy, released during decay of radon progeny with alpha particles until their complete disintegration. Till now, the highest value of PAEC, measured in Polish coal mines, was equal 63 µJ/m³. But one should mention that this measurement was made close to old uranium workings. In coal mines of Upper Silesian Coal Basin (USCB) the highest PAEC values never exceeded 15 µJ/m³ [Skowronek, 1999].

During several years of extended investigations main pathways of radiation hazard in underground mines have been recognised. Methods of measurement have been developed, then suitable instrumentation for underground application was designed and system of radiological protection was implemented in all underground mines in Poland.

2. Legal basis of radiological protection in Polish mines

Systematic obligatory monitoring of radiation exposure to natural radionuclides at workplaces in Polish coal mines has been carried out since 1989. At first the legal basis of the monitoring was the Polish standard PN-88/Z-70071 “Radiation protection in underground mines. Limits of miners exposure to natural radionuclides and methods of monitoring” [PN, 1988]. On this basis Ministry of Mining issued „Guidelines of monitoring of radiation exposure to natural radioactive substances in hard coal mines” [Guidelines, 1988]. According to these legal acts following parameters had to be monitored in underground galleries:

- the concentration of short-lived radon progeny in mine atmosphere;
- the gamma radiation dose rates;
- the concentration of $^{226}$Ra and $^{228}$Ra in mine waters;
- the concentration of natural radionuclides in mine sediments.

The most important thing was the emphasis on workplace monitoring, followed by individual dosimetry in particular cases. Direct monitoring of potential alpha energy concentration was applied, because radon progeny caused 90% of collective dose equivalent for miners. The equilibrium factor varied in a wide range in underground galleries, making radon monitoring insufficient for dose calculations.

The most important regulatory act - The Geological and Mining Law - was issued in 1994, and amended in 2004 [Mining Law, 2005]. Accordingly to this law, criteria of the hazard assessment, also natural ones, must be establish by the President of State Mining Authority in co-operation with Ministry of Economy. Additionally, in case of radiation hazard, all regulations must be co-ordinated with President of Polish Atomic Energy Agency. The safety requirements for underground workings were issued by Ministry of Internal Affairs and Administration [Decree, 2004]. In this decree criteria of the assessment of all natural hazards occurring in underground mines were described, including radiation hazard caused by presence of enhanced concentration of natural radionuclides. The requirements concerning the assessment of radiation hazard were completed with requirements for protection against artificial and natural radionuclides, prevention measures and monitoring set forth in the Decree of the
Ministry of Economy [Decree, 2006], issued in agreement with the President of Polish Atomic Energy Agency. Also requirements of EU Basic Safety Standards were included into new regulations [BSS, 1996].

In the Decree of the Ministry of Internal Affairs and Administration underground galleries with enhanced radiation risk were splinted into two classes:
- Class A – galleries, where the annual dose equivalent for workers may exceed 6 mSv;
- Class B – galleries, where the annual dose equivalent may lay within the range 1-6 mSv.

What is worth mentioning, this decree first emphasized that galleries classified as A or B must be considered as controlled or supervised areas respectively, in the meaning of the Atomic Law [Atomic Law, 2007]. Also methods of monitoring and frequency of monitoring have been set, accordingly to the class of underground galleries.

In the Decree of Ministry of Economy requirements for radiological protection against artificial and natural radioactivity are described in detail. Very important innovation, in comparison with previous regulations, was the fact, that non-uranium mining activity was also treated as practice\(^2\). Therefore the annual dose limit for miners is exactly the same as for other groups of workers exposed to radiation as a result of their professional activity. This approach follows international recommendations. This results in that monitoring of the radioactivity in galleries must fulfil following requirements:

- in class B galleries, the monitoring of the working environment is sufficient;
- in class A galleries, not only monitoring of the environment must be performed but additionally individual dosimetry shall be carried out.

Another problem, pointed out in the regulations, is the reliability of the results of monitoring, because its have been used for the dose assessment [Mielnikow, 1996]. Due to Polish regulations, monitoring of radiation risk in underground mines must be done by certified laboratory, with an implemented system of quality assurance.

3. Methods of monitoring and measuring devices

3.1. Radon progeny measurements

The equilibrium factor between radon and progeny in mine atmosphere is very unstable and varies in space and time (we found F coefficient in range 0.05 – 0.95 [Skowronek, 1991]). Therefore it is necessary to measure in coal mine the potential alpha energy concentration of radon progeny directly, to avoid errors due to the approximation of the exposure from radon data [Page, 1988]. For this purposes a typical, gravimetric dust sampler is used. A special sampling probe, called ALFA-31, was designed and built {Lebecka et al., 1988}.

\(^2\) professional activity directly related to exposure to radiation or deliberately use of radioactive sources. Usually the term is used for activity focused on the use of radioactive sources or fissile materials.
The principle of the operation is as follows. During sampling aerosols are collected on the membrane filter. Thermoluminescent detectors (TLD), mounted in a special holder above the filter, measure the alpha radiation, emitted by radon progeny. The background measurements are performed by another set of TL detectors, placed behind aluminium spacers. The sampling can be done during the whole shift and after the exposure the readout of TL detectors is performed in a special TLD reader. The calibration of ALFA-31 probes is carried out in a radon chamber with the aid of liquid scintillation method [Chalupnik, 1996].

With the application of that device was possible to make measurements of dust content and radon progeny simultaneously, at little cost and effort. ‘Barbara 3A” gravimetric dust samplers were commonly used in all coal mines in Poland till late 90’s, because measurements of dust concentration were obligatory. Later the provision of making monitoring with application of personal dust samplers were implemented, therefore new version of the sampling probe was developed and applied. The new version is called ALFA-2000 sampling probe. The AP-2000EX device, where the probe can be installed can also simultaneously fulfill two tasks: measurement of dust concentration and potential alpha energy concentration (PAEC) of short-lived radon progeny.

According to ISO/CEN criteria concerning the sampling convention for aerosol, three fundamental classes of airborne dust can be specified. One of these is the respirable class that includes particles able to reach the nonciliary respiratory tract. The second, a so-called thoracic class is composed of particle penetrating beyond larynx. The broadest class of particles that contains the two mentioned classes is the inhalable class. All airborne particles that can be inhaled through nose and mouth belong here.
During measurement, the pump draws air through a filter with an adjusted flow rate. The device can be equipped with a cyclone to separate out the respirable fraction and measure concentration of these particles in air. Replacing of the cyclone by other type of measuring head makes it possible to measure the inhalable class as well.

The special sampling probe is placed inside the cyclone cassette above the filter and makes it possible to measure the PAEC value. The location of this additional part inside the cyclone does not disturb the intrinsic safety of the whole construction and measurement of dust concentration in air. TLDs are placed in these sampling probe and record radiation emitted by short-lived radon daughters bounded up with the respirable dust. The outcomes received as a result of readouts these TL detectors make possible to evaluate the PAEC of short-lived radon daughters averaged over quite long time. So this device can be used both for the environmental and personal dosimetry.
The device is supplied by a battery set and can continuously operate at least 8 hours. Measurement run is supervised by a microprocessor. There is a possibility to program the measurement time and flow rate that can be changed up to 2.3 dm³/min. The weight of this device is 550 g. The lower limit of detection related to measurement of the PAEC value is better than 0.01 µJ/m³ at a 5% significance level and 7 hours pumping. The same sampling probe was described more detail in Radiation Protection Dosimetry [Skubacz, Bywalec, 2003].

3.2. Gamma dose-rate measurements

Instead of not readily available portable gamma dose rate meters, a special dosimeter was designed (see fig.4). This device, called GAMMA-31, consists of a water- and dust-proof cassette, in which three thermoluminescent detectors are placed [Skubacz, 1986]. TL detectors of LiF:Mg,Cu,P (MCP-N type) are produced by Institute of Nuclear Physics in Cracow. The GAMMA-31 dosimeter can be used not only as a personal dosimeter but also as the workplace monitor. The read-out of dosimeters and the calibration is performed in our laboratory.

![Fig.4. GAMMA-31 personal dosimeter](image)

Technical data of the dosimeter are as follows:

- LLD = 30 nGy, for the exposure time 30 days;
- three LiF:Mg,Cu,P TL detectors;
- diameter - φ40 mm;
- weight of the device – approx. 20 g.

3.3. Measurements of radium concentration in mine waters

Radioactivity of waters from coal mines is mostly from radium isotopes - $^{226}$Ra from the uranium series and $^{228}$Ra from the thorium one. A method of chemical separation of radium, developed by Goldin [Goldin, 1961], has been modified for liquid scintillation counting [Chalupnik & Lebecka, 1993]. In this method radium is coprecipitated with barium in form of sulphates and this precipitate is mixed with liquid gelling scintillator. The prepared samples are measured by a low background
liquid scintillation spectrometer (QUANTULUS, Wallac Oy, Finland). This counter has alpha/beta separation with an anti-coincidence shield, which enables measurements of $^{226}\text{Ra}$ concentration above $3\text{ Bq/m}^3$ with simultaneous measurements of $^{228}\text{Ra}$ (LLD = $30\text{ Bq/m}^3$) and $^{224}\text{Ra}$ (LLD = $50\text{ Bq/m}^3$). In addition, the procedure enables the simultaneous preparation of $^{210}\text{Pb}$, which can separate from radium isotopes at the last stage of analysis and also can be measured in the LS spectrometer with a detection limit of $20\text{ Bq/m}^3$. Radium standards for spectrometer calibration were purchased at Amersham.

The method has been implemented in Radiometric Laboratory of the Central Mining Institute since 1993. In frame of the regular monitoring, water samples (sample volume at least 2 l) are taken into glass or plastic containers by mine services and send to the laboratory for analysis.

3.4. Measurements of natural radionuclides in mine sediments

Solid samples (deposits from settlement ponds, river beds, soil, solid waste, etc.) mainly contain radio-isotopes from the uranium and thorium decay series, $^{40}\text{K}$ and sometimes $^{137}\text{Cs}$ (from the Chernobyl disaster). For their activity concentration measurement a gamma spectrometry system was used. It comprises an HPGe detector (45%, PGT), multichannel analyser (CANBERRA) with built-in computer and the GENIE-PC software for spectrum analysis (CANBERRA). This instrumentation enables measurements of $^{226}\text{Ra}$ concentration (LLD as low as $1\text{ Bq/kg}$), $^{228}\text{Ra}$ and $^{224}\text{Ra}$, $^{40}\text{K}$ and other natural and artificial isotopes emitting gamma radiation on the similar levels [Michalik, 1995]. Calibration has been done with application of certified materials from IAEA and EPA.

In frame of obligatory monitoring mine sediments are sampled in underground galleries or on the surface by the mine services and send to the laboratory of radiometry of Central Mining Institute. After drying, sample is placed in the Marinelli beaker, stored at least two weeks and finally measured.

4. System of monitoring of radiation hazard in underground mines

The system of monitoring of radiation exposure in Polish coal mines is a part of the system of monitoring of natural and technical hazards. It is based on the following assumptions:

- a primary goal of the system is a preventive action;
- monitoring of radiation exposure should be carried out by existing mine services, preferably together with the monitoring of other hazards.
For several years of examinations of the working environment in mines different measurements have been carried out: investigations of the occurrence of radon and its progeny, determination of radium isotopes in mine waters as well as studies on precipitation processes of radioactive sediments. On this basis the requirements, concerning monitoring of individual sources of radiation have been established [Skowronek et al., 1991]. Places, where monitoring should be done, were identified in co-operation with specialists of mine ventilation and hydro-geologists and the frequency of measurements has been set (see tab.1.). All requirements were set forth in relevant regulation.

Table 1. The frequency of radiation hazard monitoring in Polish mines

<table>
<thead>
<tr>
<th>The source of the hazard</th>
<th>Measured quantity</th>
<th>The span of measured values</th>
<th>Required frequency of monitoring</th>
<th>The change of the frequency</th>
</tr>
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<tbody>
<tr>
<td>Short-lived radon progeny</td>
<td>$C_{\alpha}$ - potential alpha energy concentration</td>
<td>$C_{\alpha} \leq 0.5 , \mu J/m^3$</td>
<td>Once per 3 months</td>
<td>The frequency of monitoring can increase to once per month* if three consecutive measurements are lower as lower limit of the current interval</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$C_{\alpha} &gt; 0.5 , \mu J/m^3$</td>
<td>Once per month **</td>
<td>The increase of the monitoring</td>
</tr>
<tr>
<td>Gamma radiation</td>
<td>$k$ - kerma dose rate of gamma radiation</td>
<td>$k \leq 0.5 , \mu Gy/h$</td>
<td>Once a year</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>$k &gt; 0.5 , \mu Gy/h$</td>
<td>Once per 3 months **</td>
<td></td>
</tr>
<tr>
<td>Radium-bearing waters</td>
<td>$C_{Raw}$ - concentration of radium isotopes</td>
<td></td>
<td>Once a year</td>
<td>frequency is required immediately after measurement of the current interval is measured.</td>
</tr>
<tr>
<td>Mine sediments</td>
<td>$C_{Rao}$ - concentration of radium isotopes</td>
<td></td>
<td>Once a year</td>
<td></td>
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</table>

* in such case monitoring of potential alpha energy concentration in galleries, ventilated by the same air stream must be done,
** additionally monitoring of radon progeny concentration is required.

Measurements must be carried out by properly trained servicemen. In addition the person, responsible for supervising of the radiation protection from natural radionuclides must be competent having the certificate of radiation protection course. Therefore special training courses, with the programme approved by the Polish Atomic Energy Agency, are organised periodically. These courses are dedicated to the specific problems of the occurrence of natural radionuclides in underground galleries as well as in the natural environment, in the vicinity of coal mines. Within several years of about 500 miners attended such courses.

Accordingly to mentioned above law regulations, the duty of the monitoring lays upon the employer (mine management). In practice, specialised laboratories perform monitoring, because are able to provide the full service, concerning radiation hazard,
for mines. Therefore the biggest databases of radiation monitoring results are
gathered in such institutions for particular branches of mine industry.

For instance, Central Mining Institute in Katowice publishes data from coal mines
annually as a special report [Wysocka et al., 2007]. Together with reports for other
branches, like copper mines, lead and zinc mines etc., results of monitoring are
published in annual report of Polish Atomic Energy Agency, concerning general
status of nuclear safety and radiological protection in Poland.

5. Assessment of the exposure, caused by natural radionuclides in underground
mines

Results of monitoring of different parameters of radiation hazard are used for the
assessment of effective dose for particular miners as well as a collective effective
dose for all underground miners in Poland.

The annual effective dose is calculated from the following equation:

\[ E = E_\alpha + E_\gamma + E_{Ra} \text{ [mSv]} \]
\[ E_\alpha = 0.0014 \times (C_\alpha - 0.1) \times t \]
\[ E_\gamma = 0.0014 \times (K - 0.1) \times t \]

\( E_{Ra} \) is calculated as the committed dose due to inhalation and ingestion of radium-
-bearing waters and deposits [Michalik, 2005].

where:

- \( C_\alpha \) - potential alpha energy concentration [\( \mu \)J/m\(^3\)]
- \( K \) - kerma dose rate [\( \mu \)Gy/h]
- \( t \) - annual working time, usually assumed as 1800 hours

The assessment is done, taking into account the most pessimistic scenario
(maximum values of particular parameters fig. 5 and fig. 6). Results of the
consideration of annual effective dose are used for the classification of galleries and
workplaces, to ensure the proper span and frequency of monitoring and undertaking
of preventive measures, if necessary. In table 2 data concerning number of mines
with exceeded particular levels of radiation hazards, are shown. It can be clearly
seen that the most important source of radiation hazard in coal mines is radon
progeny. Nevertheless, under some specific circumstances, other sources i.e.
gamma radiation, a contamination by radium-bearing waters or sediments with
enhanced radioactivity, may increase significantly the exposure of miners (fig. 7).
Fig. 5. Maximum values of potential alpha energy concentration of radon progeny and calculated effective annual doses for last decade

Fig. 6. Maximum values of kerma rate and dose equivalent, caused by external gamma radiation

Table 2.
Number of mines with galleries, where enhanced levels of natural radiation were measured in 2007

<table>
<thead>
<tr>
<th>Class of radiation hazard</th>
<th>Total number of mines</th>
<th>Radon progeny hazard</th>
<th>Gamma radiation hazard</th>
<th>Radioactive scales hazard</th>
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6. Methods of radiation risk reduction

The Geological and Mining Law and accomplished decrees contain only general regulations and requirements concerning prevention against radiation hazard caused by radon progeny. For instance, there is no guidance for the proper planning of underground workings. Therefore further efforts in the Central Mining Institute were focused on development and application of mitigation methods into practice. For the stage of exploitation of the coal seams, the requirements concerning identification of radon and radium sources, prevention measures as well as recommendation for ventilation systems were given. Additionally, needs of the prevention against radon exhalation from radium-bearing waters and deposits have been emphasised.

The insulation of goafs is of the main elements of the prevention against enhancement of radon progeny levels in the areas of longwalls. This technique is based on the principle that reduction of the ventilation rate of goaf area leads to lower exhalation of radon and radon progeny. Investigations, performed in one of coal mines, proved this thesis [Skowronek, 2000] – after the isolation of the exploited out area the PAEC dropped down from the level 6 \( \mu J/m^3 \) to values below 1 \( \mu J/m^3 \). Due to the continuous insulation of goaf’s area in this region a significant improvement has been achieved – the PAEC value never exceeded the level 1 \( \mu J/m^3 \).
Another preventive measure is the underground purification of radium-bearing waters. In several coal mines such measures have been undertaken [Lebecka et al., 1994, Chalupnik and Wysocka, 2008]. Two goals were achieved in this way – not only the diminishing of radiation hazard for miners but also the decrease of the contamination of the natural environment.

7. Environmental impact of waste with enhanced concentration of natural radionuclides

I order to determine whether the presence of sediment with enhanced concentration of radium isotopes can cause any detriment to the environment one should carry out a multi-stage evaluation process ending with real effects identification at an appropriate level. The process should identify the relationships between: an occurrence and an exposure, an exposure and a dose, a dose and effects to for non-human species representatives or the whole ecosystem. Dedicated investigations should be sequentially focused on all the stages of risk scenario creation [Michalik, 2008], such as:

- Radionuclide inventory
- Radionuclide migration and availability
- Biota exposure to external and internal radiation
- Radionuclide transfer factors into biota and committed dose evaluation
- Radiation effects on biota

7.1. Sites of investigation

In spite of the spontaneous process of precipitation and then sedimentation in underground galleries certain proportion of radium is released daily to the surface along with the other mine effluents. This results in a significant increase of radium concentration in the environment surrounding collieries [Chałupnik et al. 2005].

To control salt discharge into inland waters and allow suspended material, carried by underground brines, to settle, settling ponds were employed in coal mines. Besides artificial ones, some natural lakes or fishing ponds were adapted for this purpose. In these ponds, under advantageous circumstances radium isotopes can co-precipitate with barium and concentrate in bottom sediments. Sediments with concentration of both radium isotopes together exceeding 200 Bq/kg were found inside 25 exploited settling ponds. The total capacity of all these ponds reaches 5 millions cubic meters [Michalik 2004].

For the exemplary assessment of environmental risk two former-natural fishing ponds that had been adapted as a settling pond were chosen. Both ponds had been exploited for over 20 years. During this period more than 240 000 m$^3$ and 113000 m$^3$ of suspended material was deposited in the ponds respectively. The average radium isotopes ($^{226}$Ra+$^{228}$Ra) activity concentration in these sediments is slightly above 1000 Bq/kg in the first pond and 6500 Bq/kg in the second [Chałupnik et al. 2001]. The maximum observed of radium activity concentration in the sediment reaches 15000 Bq/kg and 67500 Bq/kg, respectively, and excides 160000 Bq/kg in scales
encrusting in the vicinity of the former discharge point of waters into the second pond [Leopold et al. 2007].

In 2002 the discharge of water to the first pond was stopped. The pond was abandoned and dried. The typical ambient dose rate 1 meter above the sediments is about 0.5 µSv h⁻¹ but there are hot spots where the dose rate exceeds 3 µSv h⁻¹. Since the pond was dried wild plants have started to transgress its banks and now a large proportion of the pond is overgrown by them. The main species are: *Calamagrostis epigeios*, *Phragmites australis*, *Lepidium ruderalne*, *Cirsium vulgar e*, *Matricaria perforate*, *Atriplex hastata* [Michalik et al 2005]. The second pond was excluded from the technological process too, but still remains filled with water so that there is not noticeable dose rate increment above the water surface. But dose rate, exceeding 40 µSv h⁻¹ was measured next to the former water discharging point.

The pond sediments formed as results of radium and barium precipitation or adsorption on particles of suspended matter followed by sedimentation. Therefore these sediments have no significant uranium and thorium content. Also, the age of sediments is too short to get secular equilibrium between radium and its long-lived progeny, so the activity concentration of ²¹⁰Pb and ²¹⁰Po are negligible too. As, from the radiation protection point of view, radium is the most prominent radionuclide among the potential contaminants of concern, so that the investigation focused on the behaviour of radium isotopes ²²⁶Ra and ²²⁸Ra.

In the two sites described above a comprehensive environmental risk assessment was applied. The objective was to provide all stakeholders with well-founded and truthful information about the real environmental risk caused by the settling ponds of concern. Such information was needed in order to decide on the fate of this area.

The investigation started with the assessment of radiation effects on biota. Interactions of contaminants with plants takes place at the cellular level. Cellular response is the first manifestation of harmful effects. Therefore genetic test-systems are suitable tools for the early detection of any pollution related effects and are reliable indicators of alterations in ecosystems. Thus, genotoxicity and cytotoxicity tests were used to hazard identification. To describe the related risk the research on the behaviour of the pollutants in the environment and the derived dose assessment was applied.

### 7.2. Hazard identification

To assess the hazardous potential of the sediments (two samples of sediments from the first pond, one sample from the bank of the second pond), the sensitive and simple bioassay, *Allium* test was applied [Grant, 1982]. As a reference sample chernozem leached loamy soil was used. Sediments cytotoxicity was estimated based on root proliferation of *Allium cepa* that was quantified in parallel for all samples as mitotic index. Genotoxicity was assessed with a frequency and spectrum of chromosome aberrations and mitotic abnormalities in ana-telophase cells in root meristem of testing onion. Both the numbers of *Allium cepa* bulbs (22-23) and cells (1600-1800) scored for each tested sample were enough to fulfill the requirements to possible uncertainty in mitotic index estimations at levels of 95% confidence probability and relative probable error <20%.
Obtained values of mitotic index, \((32.84 - \text{average for three tested samples, the standard deviation equals 1.87})\) do not differ significantly from each other and from the reference one \((33.41 \pm 1.58)\). Thus, the cytotoxicity of the sediments is negligible. On the other hand, the number of cytogenetic damages to meristem cells of \textit{Allium cepa} in all samples of the tested sediments, expressed as percentage of aberrant cells, was above twice as many as the reference value, with the important contribution of such severe types of cell damage as chromosome double bridges and laggings [Geras'kin et al, 2007].

To reveal key pollutants governing biological effect observed an adaptation of the mathematical and statistical technique of multivariate analysis for reducing dimensionality of a predictor space was used [Geras'kin 2005]. The obtained variant models based on environmental concentration of pollutant and the biological endpoints measured were inconclusive. Such results support the need to know how pollutants behave in the environment, especially on the border of biotic and abiotic matter.

7.3. \textit{Radium migration and bioavailability}

The next step of the investigation focused on two objectives. The first one concerned the determination of mobility of radionuclides in waste materials. The second one was an evaluation of uptake of \(^{226}\text{Ra}\) and \(^{228}\text{Ra}\) by natural vegetation and the determination of site and plant-specific concentration factors (CF) in these areas.

Different sequential extraction procedures were tested to investigate the speciation of \(^{226}\text{Ra}\) and \(^{228}\text{Ra}\) in the matrix of waste material and in a control soil. They were: a modified by Bunzl [1995], Tessier’s method [1979], for determining the available fraction, and a method according to Community Bureau of Reference [Leopold at al., 2007]. The third method was a simple extraction steps using 1 M ammonium-acetate solution to determine exchangeable and soluble radionuclides in the samples. Finally the ratios of radium “in water”, “readily exchangeable”, “bound to carbonate” and “bound to Fe and Mn oxide” fractions were determined by Bokori [2007]. In addition, an extraction procedure according to Polish standard PN-EN 12457-4, 2006 was applied for determining the water-soluble amount of heavy metals.

Results from different extraction methods do not show significant differences in the proportion of the relevant fraction of the mobile part of radium isotopes for each tested sample [Bokori 2007]. But it became clear the mobile fraction depends on the chemical composition of the sediments. In the case where the main process of Ra incorporation into the sediment was a precipitation with the sulphate content of water or substitution in barium sulphate precipitate to create a form of \((\text{Ba (Ra)} \text{SO}_4)\) (radiobaryte), the amount of mobile fraction was less than 0.1 % of the total radium concentration. Otherwise, when radium was bound mainly to sediment grain surfaces the percentage of total mobile fraction reached 24%. In this case radium behaves like other heavy metals [Leopold et al. 2007].

The most important finding was that the relationship between the radionuclides concentration in sediment and sediment/plant CF is non-linear. This influences dose
evaluation a lot. It was shown by simple measurement and calculation that the dose caused by radionuclides incorporated to a plant (committed dose) constitutes the main proportion of the total dose. So, if the CF does not depend linearly on concentration, it is not possible to simply evaluate the total dose based just on a radionuclides' activity concentration in the environment.

7.4. Dose to biota assessment

In considering the pathway of terrestrial biota exposure, the external and committed dose was calculated for the herbaceous vegetation transgressing into the investigated sites. The external dose rates were estimated based on radioactivity concentration in the habitat of the exposed organisms, for root and above ground plant's part. Committed dose were estimated assuming that all energy of radiation emitted by radionuclides from within the plant is also absorbed by this plant [Amiro 1997]. Dose rate estimations included emissions by $^{226}$Ra, $^{228}$Ra, $^{224}$Ra and $^{40}$K and was expressed as absorbed dose in μGy d$^{-1}$.

The committed dose rates were in the range of 5 – 63 μGy d$^{-1}$. The fluctuations of external dose rates were very high, from 28 μGy d$^{-1}$ to 1659 μGy d$^{-1}$. The highest doses were noticed for root systems. Compared with the dose rates in the uncontaminated area, those in the two studied sites were significantly higher [Trabidou 2007].

The first conclusion from the obtained results is that the contribution of external exposure to the total dose was far beyond that caused by intake of radionuclides. But it is worth mentioning that both evaluated doses, resulting from internal and external irradiation are expressed as absorbed dose. If one could apply some weighting factors the committed dose would be close to the external one. The internal exposure is proportional to the concentrations of the radionuclides inside the organism, which depends on the CF. As it was already mentioned the relationship between CF and activity concentration in the abiotic environment is nonlinear. Therefore a significant part of radiation risk caused by internal exposure can not be predicted based only on the radioactivity concentration in environment.

According to the system of radiation risk to biota classification introduced by Polikarpow [1998] the dose rates to terrestrial organisms ranged in between “physiological masking zone” and “ecological masking zone”. This means that it is possible to detect minor changes in function, morphology and an increase of diseases or effects at the population level. It confirms that some proportion of the effects described above observed at the molecular level were probably caused by radiation.

8. Conclusions

The system of the monitoring of radiation hazard in Polish mining industry is a unique, complete system, first implemented in non-uranium industry. This system permits not only the assessment of miners’ exposure but due to the fact, that results can be obtained quickly, provides data necessary for preventive measures. Important part of the system is the training of miners, managers, the staff of different services.
The training makes the co-operation easier and useful for all partners (i.e. mine services and laboratory providing measurements).

Implementation of different methods of the monitoring and prevention has been started in Polish coal mines several years ago. The effect of the mitigation is the permanent decrease of the radiation hazards during 90’s.

On the other hand derived from disposal of waste with enhanced natural radioactivity environmental burden has been left out of regulatory control. There is a great need to provide mining industry operators with well-founded information about real environmental risk caused by enhanced natural radioactivity. Achieved results show that regulate the problem of water and sediments originating from coal mines would be by far better to comply existing requirements for environmental protection and waste treatment with radiation protection demands than to apply directly the rules developed for nuclear industry as “traditional” domain of radiation protection. In practice, ISO management systems, environmental impact assessments (EIA) and best available technologies (BAT) shall take into consideration enhanced natural radioactivity in cases of concern.

9. References


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