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Evaluation of Existing Exposure around the Sborishte Former Uranium Mining Site (Case Study)

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Abstract. Uranium mining industry in Bulgaria started operation in 1945, followed by in situ leaching technology in the beginning of 1970-ties. Two decades later (1990) all uranium mining sites in the country are closed. However the radioactive discharges from these sites continue cause various environmental problems and after the closing phase.

Sborishte (Southern Bulgaria, Sliven region) is a one of uranium mining site, where two methods: underground mines and in situ leaching (ISL) were applied. The aim of the study is to assess the radiological impact on living environment, related to former uranium mining activities in the vicinity of Sborishte village. The assessment is performed on the base of systematic control on radiological parameters. Systematic control on radiological parameters involves: measuring gamma dose rate and analyzing uranium contamination in surface water and specific activity of natural radionuclides in soil and sediment samples. For evaluation of radiological contamination in environment, the concentration of uranium in water and sediment were tracked in time. The range of gamma dose rate around the site is typical for the natural background. A high value up to 2.56 μSv/h was measured on waste piles. The range of uranium concentration in water flowing from the mining sites is from 0.287 to 1.0 mgU.l^{-1}. The range of uranium concentration in sediment was from 1.2 to 19 mgU.kg^{-1}. The sediment was sampled from the same sites where the water were. In order to evaluate the existing exposure of people, conservative dose assessments were done using IAEA recommended models.

Keywords: uranium mining; radiological impact; environmental, natural uranium.

1 Introduction

The uranium mining in Bulgaria has been started in the aftermath of World War II. According to their geographical location the uranium deposits correspond to four major zones - Balkan, Srednogorska, Rhodope and Thracian. Extraction activities have taken place on 48 uranium sites and additional 30 deposits were studied as potential mining sites [1]. Sborishte area has been studied in 1954 with aero-gamma measurement. Uranium mining was carried out by two methods [2]: classic extraction – by open pit mining and underground mines in grade of ore between 0.5% and 15% U3O8, and geotechnological method (in situ leaching – ISL) under very low grade ore. The choice of extraction method is determined by the depth, shape and type of uranium mineralization. It depends on composition, hydrogeology and geotechnical aspects of the surrounding rock formations. Classic extraction method was generated output and embankments (heaps), related to mine ventilation. Fine particles of radioactive dust polluted the road system during ore transportation process. One of the sources of pollution in geotechnological method was spills on the technological chain. In Sborishte the technology of deposit exploitation combined classic extraction (underground mining) and geotechnological method. The site has eight adits, one vertical shaft and multiple exploratory and geotechnological drillings. Investigation activities in Sborishte started in 1960. Sixteen years later (1976) began a commercial yield from the mining area. In 1992 the uranium extraction site is closed. Experimental uranium mining is conducted by underground methods of leaching with solutions of sulfuric acid and soda from 1983 to 1985. According to the records, the production from the site amounted to 710 kg uranium, 128 085 ore tons and 87 512 kg metal.

The project on technical and biological recultivation of Sborishte site was outlined in 1996. The vertical shaft 3 (VS 3) and adit 4 (M4) were liquidated, according to this project.

The company, which is responsible for the remediation of mining and uranium mining is “Ecoengineering-RM” Ltd., according to Bulgarian Council of Ministers Decree No. 74/1998 [3].

2 Area under Investigation

The former uranium mining site Sborishte is situated at the southern slopes of the Elena-Tvarditsa Mountain, Central Stara Planina Mountain. The deposit is located 2 km. northeast of Sborishte village, Tvarditsa municipality and 3 km southwest of Shivachevo town. Geological structure is dominated by metamorphic sedimentary rocks, diorite formations and poly-metallic ores. River Blyagovitsa, Dolap dere and Domuz dere rivers, as well as their tributaries (Hambarchukuru and Kaziolu) form a river network and provide water supply to the agglomerations in the vicinity (Sborishte and Shivachevo). The climate in the area is moderate continental with the influence of the mountain climate, typical of places on high altitude. The temperature range is 41°C to 20°C.

According to European Directive 2013/59 [4] the former uranium mining site Sborishte could be defined as an existing exposure situation of past activities. Radiation factors on living environment in the area are monitored and controlled on a regular basis in order to collect and evaluate of radiation contamination in Sborishte.

Radioactive contamination is usually spread into the environment by migration of radionuclides. One of the potential sources of exposure from Sborishte is using the contaminated water and consuming vegetation from farm animals. Materials, taken from waste piles for construction of house are other way of explosion of the peoples.

The present paper provides analysis the contamination on the site Sborishte and trends of environmental pollution during the years. The screening assessment of the radiation dose at two objects on the sites is discussed in order to identify the main ways of exposure of the population. The results from control of water and sediment from the mining drilling facility at vertical shaft and gamma dose rate on the waste piles and contaminated area are presented.

3 Materials and Methods

The site study involves qualitative assessment by collecting information on exploitation activities and subsequent restoration of the environment, as well as results of site inspections. The circumstances with potential relation to public exposure are examined.

Qualitative analysis of the place is based on: site inspections; communication to the local people; identification of the mine workings near Sborishte; recording precise GPS coordinates by Global position system (GPS) GARMIN device and take a photo documentation of the objects. The photo archive provides information, arranged on annual basis, and serves as important tool in tracking and assessing the changes in the examined area [1]. Qualitative analysis of the former uranium mining and milling sites is made using the model for assessment of contaminated sites developed by Canadian Council of Ministers of the Environments (CCME) [5]. Following this assessment the former uranium mining sites are classified according to pollution characteristics of the sites and their potential to affect the environment. Sborishte site corresponds to Class 2 - average radiation risk sites, which should be investigated on every two years, according to the plan of inspection. The investigation of the sites encompasses exploration of the living environment and measurement (control) of the radiation parameters in order to determining the dose or the risk of the population in the regions. In this paper the following objects and parameters of the living environment are examined in Table 1.

The mine workings (adits, shafts, drillings and waste piles), identified during the living environment monitoring in Sborishte area, are the sources of pollution. The focus of the current study is the results from two investigated points: the mining drilling facility at vertical shaft and the waste pile and contaminated area.

Table 1. Objects of the environment and controlled parameters

Objects of the environment	Controlled parameters
natural gamma radiation outdoors	ambient gamma dose rate
sediments	natural radionuclides
mine waters	natural uraium, gross beta

Both of the points present a trend of spreading radiation contamination on region. The choice of investigated points is determined because of their location close to village Sborishte and their potential as source of contamination of the living environment. Point P1 represents a drilling of uranium mining by geotechnological method. Point P2 is waste pile of mining production and contamination area.

Point P1 – drilling of VS 3 is located approximately 350 meters of vertical shaft 3, in the vicinity of Sborishte village (GPS coordinates N42°41'28.6" E25°59'40.1"). From the drilling continuous water is flowing with the flow rate which varies depending on the season. During the summer the surface waters in the region as well as the drillings in Sborishte dry up.

The concrete structure of the drilling is partially collapsed and the tube is missing. The measured flow rate in August 2016 is 0.5 l/s. This water is used as drinking water from animals and their shepherds sometimes.

Point P2 – waste pile and contamination area close to adits on the site Sborishte.

Affected zone from radiation contamination is an area of approximate 60 m^2 (GPS coordinates N42°41'20.4" E25°59'40.2"). The point P2 is located about 700 m from P1. On this point reclamation activities on the area have been not carried out.

To achieve the objective of this study the gamma dose rate on the contaminated area was measured and water and sediments samples were analysed. Discharged water from drilling is traced in the period 2004–2016. For the analysis were used validated analytical methods, control procedures and standard operating procedures that follow internationally accepted practices (ISO Standards).

Ambient gamma dose rate in air was determined by the standard operating procedures, using the Geiger Muller counter type RADOS RDS-110 dosimeter, calibrated in the National metrological laboratory traceable to the National Standard. The dosimeter measuring range is 0.05 μSv/h to 100 mSv/h in the energy range of 50 keV to 1.25 MeV.

The detector is suitable for environmental gamma radiation measurements. It covers majority of significant gamma radiations emitted from terrestrial sources. The calibration accuracy is $\pm 2\%$ (2σ), according the Calibration Certificate from 2015. The relative uncertainty of the method was assessed to be 12%. The ambient outdoor gamma dose rate measurements were taken in air 1 m above ground.

The gamma spectrometry measurements for concentration of natural radionuclides were carried out by laboratory-validated method based on the International Standard IEC 61452 (1995-09) [6]. It is carried out by gamma spectrometry system with an HPGe detector (45% relative efficiency, resolution of 1.95 keV at 1.33 MeV). A certified mixed source in the same Marineli beaker geometry with an energy range of $47 \div 1836$ keV was used for detector efficiency calibration. Under standard measurement conditions the minimum detectable amount or activity in the sediment samples is 0.3 mgU/kg for U_{nat} and 3 Bq/kg for ^{226}Ra. The combined standard uncertainty ($1\ \sigma$), which included the uncertainty from the calibration, the blank sample and the standard deviation of the sample measurements, was estimated at 3 to 10% and was evaluated for each radionuclide/sample.

The natural uranium concentration in the water samples was determined by the standard laboratory spectrophotometric method, based on the ability of the uranium ions, U(IV) and U(VI), to create a color complex compound with a reagent, arsenazo III. Reduction of U(VI) of U(IV) with zinc in the presence of 4N hydrochloric acid was performed. Prior to the photometric determination of uranium with arsenazo III, the spectrophotometer ZUZI4201/20 was calibrated with certified uranium nitrate solution. The plotted calibration curve gives the linear dependence of the measured absorbance at $\lambda = 670$ nm in the concentration range of 0.02–0.45 mgU/l. The combined standard uncertainty ($1\ \sigma$) of the method, which included the uncertainty from calibration and the standard deviation of the sample measurements, was estimated at 8–12% and was evaluated for each sample.

The screening evaluated of effective dose is used to determine the exposure of the population on the region. This method generally uses the simplifying assumption that leads to a very conservative estimate of the dose, such as the concentration of radionuclides at the discharge point. It is the simplest deterministic method used for determining compliance with the dose limits. When the results of the conservative (screening) assessment are under the relevant dose limit, the further detailed evaluation of the dose is not need.

In this paper for the screening evaluation on the external exposure due to gamma-radiation from the soil is used Calculation Guide Mining [7]. Radiation exposure from mining is usually calculated in terms of effective dose per year. Calculation of external exposure to gamma radiation from soil used calculation of mining-caused effective annual dose $E_{A,j}$ is based on representative measurement values of ambient dose equivalent rate outdoors at conceivable exposure sites **s** of reference person j as follows:

$$E_{A,j} = f_{\mathrm{con},j} \sum_{s} \left(\dot{H}^*(10)_s - \dot{H}^*(10)^u \right) t_{\mathrm{exp},j,s} a_s \,, \quad (1)$$

where:

- $E_{A,j}$ – effective annual dose from gamma radiation for reference person j [Sv];
- $H^*(10)_s$ – ambient dose equivalent rate outdoors at a height of 1 m at exposure site s [Sv/h];
- $H^*(10)^u$ – ambient dose equivalent rate of natural gamma radiation outdoors (background) at a height of 1 m [Sv/h];
- $f_{\mathrm{con},j} = 0.6$ is the conversion factor from ambient dose equivalent to effective dose for adult (>17 years) reference person j [7];
- $t_{\mathrm{exp},j,s} = 100$ is the annual time spent by adult reference person j at exposure site s, uncultivated areas, contaminated by mining residues [h] [7];
- $a_s = 1$ is the factor accounting for the shielding effect on gamma radiation outdoors at exposure site s, dimensionless [7].

The screening effective doses for ^{238}U discharged into water were estimated using "no dilution" approach [8]. This method is a very simple assessment based on the conservative assumption that members of the public are exposed at the point of discharge. The dose conversion factor of 5.1×10^{-8} Sv/a per Bq/m^3 for ^{238}U represents the maximum effective dose in the 30th year of discharge that could be received by a reference person from a unit concentration of discharge into the water [9]. Screening effective doses for ^{238}U discharges were evaluated using the relationship that 1 mg of natural uranium contains 12.4 Bq ^{238}U, assuming that uranium isotopes in natural uranium in secular equilibrium [10]. For the assessment of the radiological impact on people that originates from discharges into water, three possible ways of exposure were taken into consideration. The first one by using water as drinking water, the second through the consumption of fish and the last through swimming- where each of them contributes to a total effective dose of 61%, 35% and 4%.

For the radioactive criteria is used effective dose of 0.3 mSv per year for the population from a waste disposal facility recommended by the ICRP in the application on the optimization principle [11]. The dose criteria include all pathways of exposure of the population from disposed waste, such as using the water discharges; external exposure; consume the food from site etc.

4 Results and Discussion

The radioactive discharges from former uranium mining sites in Bulgaria continue cause various environmental problems and after their closing phase. The results of the measurements of radionuclide concentrations in the water samples on the point P1, for the period 2004–2016 are presented in Figure 1 and Figure 2.

The results for natural uranium in water show that the concentration decreases over the years, while the concentrations of natural radionuclides in sediments increases. This could be explaining due to the fact that this drilling has a constant discharge of mine water and remediation measure has not taken. The values of gross beta activity in water samples increase, which could be explained by increased concentrations of natural radionuclides in sediments.

The results of the measurements of ^{238}U and ^{226}Ra in the sediment samples for the period 2006–2014 are presented in Figure 3 and Figure 4. These results indicate that deposited natural radionuclides from mine water in sediments have been increased over the time, which leads to steady trend of radioactive contamination in the area.

The calculated effective doses for ^{238}U discharged into water were estimated using the water sample analysed in

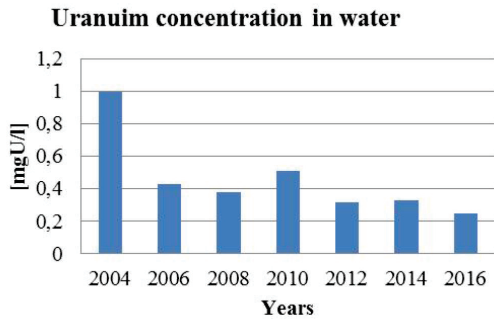

Figure 1. Analysis of uranium concentration in water samples by years.

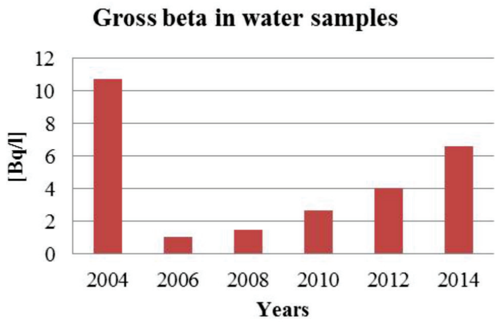

Figure 2. Analysis of gross beta activity in water samples by years.

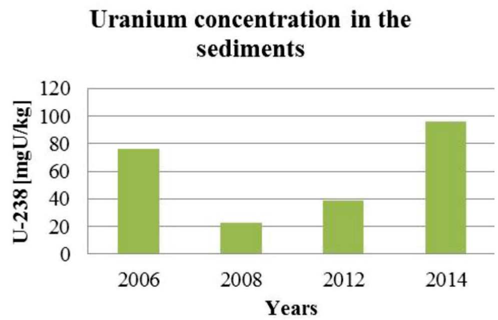

Figure 3. The ^{238}U concentration in sediment samples by years.

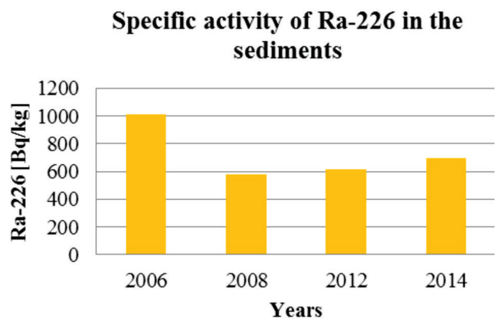

Figure 4. The ^{226} Ra concentration in sediment samples by years.

2010. The concentration of natural uranium is 6299 ± 620 Bq/m^3 and total effective doses for reference person is 0.32 ± 0.032 mSv/a. According to the used model it is distributed as follows: 0.20 ± 0.002 mSv/a by using water as drinking water; 0.11 ± 0.001 mSv/a for consumption of fish; and 0.01 mSv/a for swimming. The calculated effective dose from the use of water is higher than the dose criterion of 0.3 mSv per year for the population from a waste disposal facility recommended by the ICRP. This means that activities for more detail investigation, as well as recultivation have to be taken on the drillings of the Sborishte site in order to stop spreading of contamination trough water discharges.

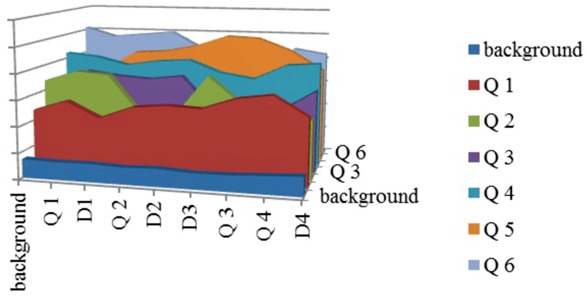

Figure 5. Measurements on gamma dose rate.

Gamma dose rate in the air measured on the investigated point P2 is used for assessment the external exposure. For this purpose, the investigated area is divided into 6 square. Ten measurements at a height of 1 m above ground of the each square were made. The measurements were done in 2016. The evaluated average value of a ambient dose rate outdoors on the contaminated area is 1.86 μSv/h with maximum value of 2.56 μSv/h.

The results of the ten measurements on the each square (Q1 to Q6) are presented in Figure 5. About 10 readings at different places on the not contaminated places were taken and the ambient mean was considered as the representative value of the gamma dose rate for the background areas. Effective annual dose from gamma radiation for the reference person, calculated using the formula (1) is 0.77 ± 0.20 mSv/a. The effective dose received of the reference person is evaluated trough the more realistic approach, as assume that person spend 100 hours of this site per year.

The calculated effective annual dose from gamma radiation of the external exposure is higher than the dose criterion of 0.3 mSv per year for the population. Although the people don't spend their all outdoor time on the sites the received dose from external exposure is high and this is the reason for taking actions more detail evaluation and for remediation.

5 Conclusions

The main factors that continue to determine the radiation contamination of the living environment are mine water discharges and waste piles from former uranium mining sites. For that reason it is necessary to perform activities for technical and biological recultivation which will improve the radio ecological status of the region and reduce the risk to public health.

Conservatively estimated effective doses from using the water and external exposure to the people on the Sborishte site are higher than the dose criterion of 0.3 mSv per year from a waste disposal facility recommended by the ICRP. Calculated doses give reason for taking activities on further investigation of contaminants on the site, migration of the radionuclides, as well as and more realistic assessment of the exposure of the population and risk of environment.

Nevertheless of the realistic risk assessment the exigent reclamation measures have to be taken for stop spreading of contamination from water flowing at the Sborishte site.

The screening assessment of health and hygiene aspects of the living environment can be applied for evaluation of the radiation contamination on other former sites of uranium industry in the country.

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