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Natural Radioactivity in virgin soils and soils from some areas with closed uranium mining facilities in Bulgaria

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Abstract
It is necessary to study the natural radioactivity levels in soil to assess the dose for the population in order to know the health risks and to have a baseline for future changes in the environmental radioactivity due to human activities. The natural radionuclide (^{238}U, ^{226}Ra and ^{232}Th) contents in soil were determined for three different regions in the country using high-resolution gamma-ray spectrometric analysis. A comparison of the dynamics of their behavior throughout the years is done. Bulgaria is a country with intensive uranium mining activities in the past years. That is why radiological monitoring of closed uranium mining facilities in different regions of the country are obligatory and of great interest. This work presents results from such investigations made in regions where remediation has been done. The results have been evaluated according to the Bulgarian radionuclide environment contamination legislation. The necessity of permanent environmental monitoring is assessed.

Key words: natural radioactivity, gamma-spectrometry, soil, uranium, radium

Introduction

Most of the natural and men-made radionuclides in the environment remain mainly in the soil. The radionuclides in the earth's crust are one of the main natural sources of ionizing radiation to which human beings are exposed. An important part of the radiation monitoring is the natural radioactivity in soils. The significant contributions to the dose in humans come from the radionuclides in the ^{238}U and ^{232}Th series and ^{40}K [9]. This is a type of exposure which is "neither widely variable nor relatively constant at the surface of the globe" [8].The amount of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in soils depend on the type of rocks from which they originate and the processes of soil formation. Analysis of radionuclide content of soil, plants, water and knowledge of the behavior of the radionuclides in soil–plant system provides an important part of a data basis for dose estimation [8,9]. The assessment of activity concentration of natural radionuclides is of particular importance as the principles of long-term environmental and human protection need to take into account the natural background [6]. Human activities can cause accumulation of radioactive elements modifying in this way the natural concentrations. An example for this are areas with former uranium mining facilities The concentrations of natural radionuclides from uranium decay series in such areas are higher and the risk of higher human exposure is increased which makes them object of special interest and studies. According to the Ministry of Environment and Water in Bulgaria there are about 18 000 decare of contaminated with radionuclides grounds in the country [5]. Following the release of Resolution № 163/20.08.1992 on the cancelation of uranium mining and the related "Instruction for the termination of uranium extraction", issued November 1992, were the natural questions about the rapid and efficient recovery of the areas damaged by the uranium mining industry. In these cases regulatory target values should be evaluated against local background levels which varies within and between countries [2].

Obtaining such data is the aim of our study on the base of regular radiological monitoring of virgin soils from high mountain areas, hills and plains covering most of the territory of Bulgaria. The region around Kozloduy NPP, mountain areas and former uranium mining sites are of special interest.

Material and Methods

Sampling

Sampling of soils was done annually from referred sampling points as part of the radiation monitoring covering almost the whole area of the country. Sampling areas were specified considering the wind direction and difference in altitude. According to the altitude of the investigated areas three groups have been defined: plains – mainly North Bulgaria, (40 sampling points *[sp]*); hilly (the valleys of Struma and Mesta rivers (15 sp)) and mountains area - in South Bulgaria (25 sp). Sampling was done also in the water catchment basin on the Beli Iskar river in the Rila mountains (10 sp)) but only in one survey (in 1996) and not as a part of the regular monitoring scheme.

The soil samples were taken according to the procedure described in ISO 18589-2 for collecting samples of undisturbed soil using uniform approach, with sampling performed at depths independent of the natural variations of the soil characteristics. from the soil layer 0-5 cm.[3]The sampling sites are undisturbed, flat with minimum impact of water and wind erosion. A composite sample of at least 5 increments samples is taken from each sampling area.

Besides the areas included in the national radiation monitoring scheme sampling was done from fields with former uranium mining facilities (“Balkan”, “Buhovo”, “Sliven”, “Sborishte”).

Measurement of natural radioactivity

The soil samples were homogenized, dried at 80ºC and sieved through a 2-mm mesh before measurement with a gamma-spectrometer. The samples were stored in air-tight containers for minimum 28 days to allow ^{226}Ra to come into equilibrium with its short-lived progeny. The measurements were done following standard procedures [4]. A Canberra high-purity germanium detector with 20% efficiency an energy resolution of 1.8 keV for ^{60}Co γ-ray energy line at 1332 keV was used. The detector was calibrated with standard reference radionuclide source, type MBSS2, containing ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, ^{210}Pb, ^{203}Hg supplied by the Czech. Metrological Institute,. The measuring system included a multichannel analyzer DSA 1000(Canberra, USA). The spectrum was analyzed by GENIE-2000 software with measurement uncertainties less then 10%. Typical counting times were 19–24 h.

The ^{238}U concentration was derived from the weighted mean of the photopeaks of ^{234}Th (63.5 and 92.6 keV), and the ^{226}Ra concentration was derived from ^{214}Bi (609.3 keV) and ^{214}Pb (295.2 and 352.0 keV) in the same way. In addition ^{226}Ra was evaluated at its 186.1 keV line taking into account the contribution of the overlapping line at 185.72 keV of ^{235}U calculating the specific activity of ^{235}U through the specific activity of ^{238}U (ISO 18589-3,2007). For ^{232}Th, the photopeaks of ^{212}Pb (238.6 keV), ^{208}Tl (583.1 keV) and ^{228}Ac (911.1 keV) were used. Activity concentration is expressed as Bq.kg^{-1} dry weight soil.

Results and Discussion

Natural radioactivity – local background values

The concentration of ^{238}U, ^{232}Th and ^{226}Ra as long-lived natural radionuclides of significance in the soil will be discussed in this paper. Summary of the data obtained for the period (1996-

2010) is presented on Figures 3, 4 and 5. Here again averaged values are calculated for the same groups of samples described above. The registered concentration of ^{238}U , ^{232}Th in the soils of North Bulgaria are in good agreement with the value of 40 $Bq.kg^{-1}$ estimated in UNSCEAR, 1993 report as average concentration of these radionuclides in soils of Nordic countries. Activity concentrations of ^{226}Ra are also in the range of values stated in the same report. The slightly higher concentrations in soils from South Bulgaria and the Mesta river valley are also logical as the soils in these areas are on rocks containing shale, gneiss with higher concentration of natural radioactivity [7]

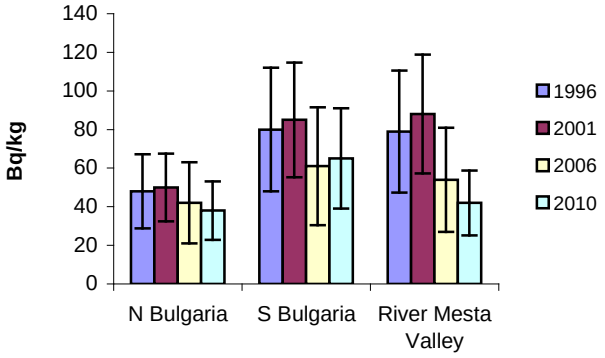


Figure 3. ^{238}U activity concentration in soil samples from different regions (1996-2010)

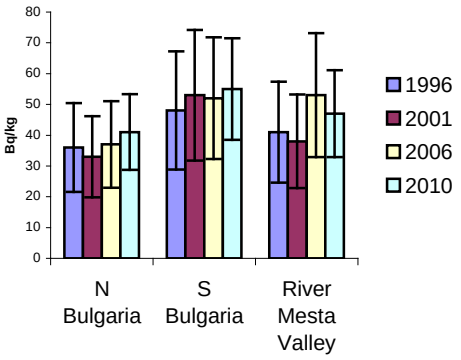


Figure 4. ^{226}Ra activity concentration in soil samples from different regions (1996-2010)

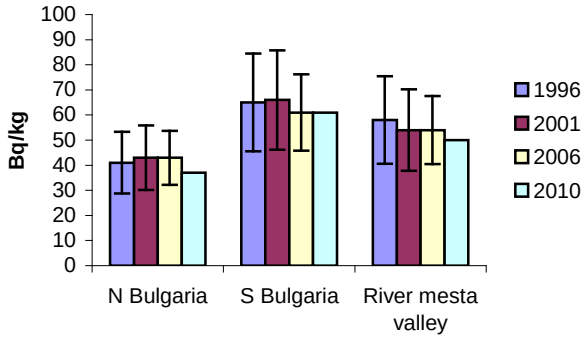


Figure 5. Content of ^{232}Th activity concentration in soil from different regions (1996-2010)

Natural radioactivity – in areas with former uranium mining facilities

All the data discussed above concern areas without former uranium mining facilities. The areas where uranium mines worked are object of special interest. In general, activities related to mining and processing of uranium ore are characterized by complex negative impact on the environmental components (soil, water and air), which is directly dependent on the extraction technology. There are several fundamental methods for extraction of uranium ore:

1. Classical methods:

а/ open-air method –via construction of quarries;

б/ classical underground method;

2. Geo-technological method – extraction of uranium concentrate by sulfuric acid.

The first of the above mentioned methods completely destroys the soil layer. Mineral masses with increased content of radioactive elements are brought to the surface of the site, the landscape of the region changes, the ecological balance is disturbed and opportunities for pollution and erosion of adjacent areas are created. These negative effects lead to serious

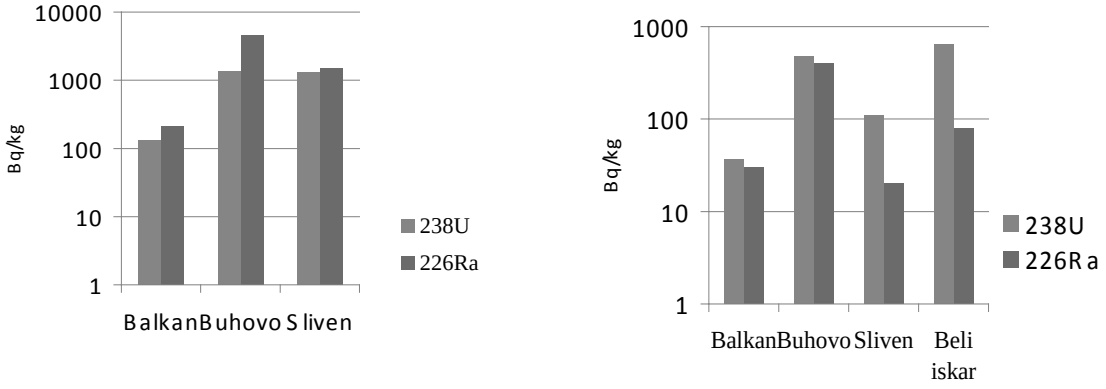
problems in dealing with the radioactive contamination and the overall landscape shaping the territory of the site.

The classical underground method does not directly disturb the integrity of the soil layer with the exception of a small area where the shafts and stulms are. The anthropogenic impact in this case is related to large amount of geological material, which is brought to the surface and takes up a significant amount of space. These materials are unsuitable substrate for growing plants and are dangerous due to the residues of uranium ore that they contain.

Especially large disturbances and changes in the landscape and soil layer at the site are caused by the geo-technological method for extraction of uranium - on one hand the integrity of the soil is mechanically disturbed during the course of drilling and blasting, the construction of the sorption system, the deployment of drilling pipes and other technological equipment and on the other hand - the soils are subject to the chemical effects of the solutions used during the mining process.

These changes and disturbances in the environmental components require precise and proper planning of the reclamation and restoration activities, combined with the recommended activities for usage of the damaged land. This is a complex and lengthy process that begins with a detailed survey of the area affected by mining works.

We have studied some of the objects with former uranium mining facilities [8]. The sampling included virgin soils and soils destroyed by the mining process activities. Information about the results obtained from 3 such areas is presented on Figure 6 (A, B).



A(disturbed soils) B (virgin soils)

Figure 6. ^{238}U and ^{226}Ra concentrations in destroyed (A) and virgin (B) soils from areas with former uranium mining facilities [Bq.kg^{-1}].

The activity concentration is presented in logarithmic scale because the variability of the results was quite high - ^{238}U (from 130 Bq.kg^{-1} up to 1360 Bq.kg^{-1}) and ^{226}Ra (from 210 Bq.kg^{-1} up to 4600 Bq.kg^{-1}). Results only for ^{238}U and ^{226}Ra are shown because they are members of one and the same decay series and the ratio between their activity concentrations may be indicative for the type of pollution recognizing at that the differences in their chemical behavior. In the soil hardly influenced by the uranium mining activities the activity of ^{226}Ra is considerably higher while in the virgin soils ^{238}U is with equal or with higher activity.

On Figure 6 (B) Beli Iskar is included as an example of undisturbed soil from a mountain region (Rila mountain) with high natural activity concentration (^{238}U – 650 Bq.kg^{-1}; ^{226}Ra 80 Bq.kg^{-1}) and ratio $^{226}Ra/^{238}U \approx 8$. This disequilibrium is most probably caused by the high amount of organic matter in this soil as uranium unlike radium makes compounds with the huminic acids in the soil [1].

Measures for decontamination and restoration of soil fertility of soils disturbed by uranium mining activities

Appropriate measures for eliminating the danger of contamination of the environment and for restoring the soil fertility on site are chosen based on the obtained results. The events can be divided into two groups according to their type:

- First group - technical activities.
  a) cleaning the site surface from large rocks, concrete foundations and other debris, leveling the surface of the piles, etc.
  b) excavation, transportation and disposal of toxic materials with high uranium concentration. Covering the less contaminated areas with substrates with specific power and appropriate physicochemical properties.
- Second group - biological events.
  a) cover disturbed areas with a layer of organic matter with a certain power;
  b) chemical reclamation of the site;
  c) planting erosion control grass and forestation of degraded areas.

Conclusions

- Caesium-137 and Strontium-90 are the main radionuclides of significance, characterizing the soil pollution with man-made radionuclides in all the areas investigated as part of this study and originate from the Chernobyl accident;
- Natural radioactivity concentrations in the investigated virgin soil are in good agreement with the values presented in the UNSCEAR reports for different soil types in the Nordic countries;
- The activity concentration ratio $^{226}Ra/^{238}U$ may be used for specifying the origin of higher natural activity concentrations in soils;
- The appropriate measures chosen for soil fertility remediation have prevented additional pollution and human exposure in the observed areas with former uranium mining facilities.

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