THE RADIOLOGICAL SITUATION AROUND THE FORMER URANIUM PROCESSING PLANT MAPE MYDLOVARY, CZECH REPUBLIC

by

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The uranium processing plant MAPE Mydlovary in South Bohemia, Czech Republic, was in operation for about 30 years, from 1962 until 1991. Extensive remediation and reclamation work has been done in the area. In the study presented here we measured mass and volume activities of certain radionuclides in soil and water samples from the surroundings and measured gamma equivalent dose rates at the same locations. The average activity concentrations of ^{40}K, ^{226}Ra, and ^{238}U in soil were 307.3 4.4 Bq/kg, 133.4 0.8 Bq/kg, and 113.2 3.8 Bq/kg, respectively, whereas in water they were 5.7 0.3 Bq/L, 0.30 0.03 Bq/L, and 1.8 0.16 Bq/L, respectively. The gamma equivalent dose rate at 5 cm and 1 m height above ground was 0.15 0.04 μSv/h and 0.15 0.03 μSv/h, respectively. As shown by comparison with the findings for similar sites elsewhere in the world, as well as with the results of measurements at uncontaminated locations, these values are compatible with regulation limits and there is no reason for concern regarding the radiation protection for workers involved with further remediation and reclamation, or carrying out other activities in the area.

Key words: radionuclide, activity concentration, equivalent dose, remediation

INTRODUCTION

Mining and processing of uranium ore in the Czech Republic began in the 1950s and it has left its mark on the environment in many places. One example is the pollution associated with the operation of the uranium processing plant MAPE Mydlovary (MAPE stands for "magnesium perchlorate") [1]. It is located in the South Bohemian Region, 20 km northwest of Ceske Budejovice (Budweis). The plant was in operation from 1962 to 1991 [2] and processed uranium ore from deposits all over the western part of Czechoslovakia, mainly from Okrouhla Radoun, Pribram, Dolni Rozinka and Straz pod Ralskem [3]. A total of 16.8 million tons of uranium ore were processed, producing 28.5 thousand tons of uranium in the form of yellow cake and leaving a total of 35.8 million tons of radioactive sludge deposited in tailing ponds covering an area of 285 ha. The uranium tailings have a residual uranium content of 0.014 % (2.4 thousand tons of uranium in total) [2].

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Tailing ponds are, of course, a foreign element in the landscape, posing threats to the environment due to possible contamination with radioactivity or other hazardous agents such as heavy metals [4]. One problem consists in polluted groundwater which due to its natural movement transports contaminants elsewhere. Another problem is contaminated dust, which the wind spreads in the surroundings. Health risks to humans are expected from ingestion of contaminated water, inhalation of contaminated dust, exposure to radon in the air and external exposure to radiation from radionuclides deposited on the ground [5].

Currently, the uranium processing equipment has been dismantled, the area of the plant itself has been decontaminated and part of the complex has been sold. For the most part, the tailing ponds have undergone remediation and reclamation work which consisted in drying them out and partly covering them with 2-7 m car tyres and ashes from waste incineration, 0.5-0.7 m clay and 0.3-0.5 m top soil. This should prevent the entry of rain water into the (former) pond, shield against radiation from deposited materials, prevent the spreading of contaminated dust and reduce emissions of radon. However, the tailing ponds were never insulated against the substratum. A great part of the radioactive sludge was dumped in exploited lignite deposits whose bottom was considered to have "a low permeability", but no active measures were ever taken to prevent the contamination of groundwater [1].

Due to the nature of the activities carried out in the area (the processing of uranium ores), the main agents affecting the radiation situation are radionuclides associated with natural uranium. Therefore, attention has to be focused mainly on the uranium – radium decay chain, of which the most important isotopes are ^{238}U, its daughter products of ^{226}Ra and ^{222}Rn, as well as ^{210}Pb and ^{210}Po [3]. Taking into account the properties of the processed material, which comes mainly from the mines around Pribram and other West Bohemian mines, other natural radionuclides are not expected to play a role, namely those from the thorium series and potassium (^{40}K). In the processed raw materials, the contents of thorium (^{232}Th) or related nuclides were on levels comparable with non-ores. Due to the low content of ^{235}U in natural uranium ores, contributions from the actinium series was also negligible [3].

For this report, soil and water samples were taken at a number of different locations in the vicinity of MAPE Mydlovary. Mass and volume activities of the radionuclides ^{40}K, ^{226}Ra, ^{238}U, and ^{235}U were determined and gamma dose equivalent rates were measured at the same locations. For comparison, sampling and measurement was carried out at control locations outside the MAPE area, at Hlincova Hora (proximity to former silver mines) and Temelin (in the area of nuclear power plants). We compare our findings with published data on the radiological situation around similar facilities in other countries.

METHODS

Sampling and sample preparation

Samples of soil were taken at 18 locations in the vicinity of MAPE Mydlovary (fig. 1). Sampling points were chosen so that all locations where radioactive materials had been deposited were represented. Thus, the sampling points included all tailing ponds and the site of the establishment itself (fig. 2).

Soil samples had a size of 20 cm 20 cm 5 cm (2000 cm^3). Sampling depth was 5-10 cm below the surface. The samples were left to air-dry at about 19 °C until their weight did not change further. After drying, each sample was carefully freed from the greater part of the soil skeleton, as well as from plant and animal remains, and was passed through a sieve to obtain fine earth. A specimen of this fine earth was used for further analysis after its exact weight had been determined.

Water samples were collected at six locations. The choice of sampling points was more limited in this

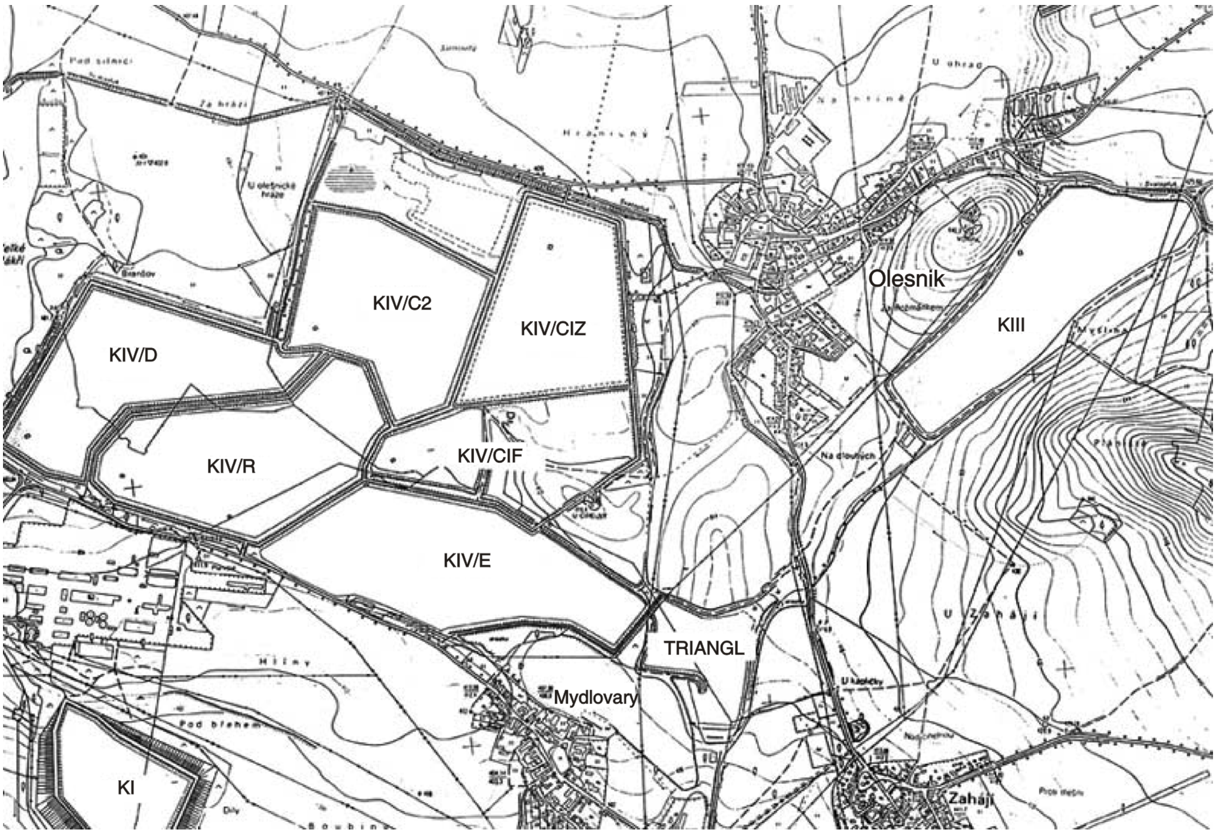

Figure 1. Map of the tailings ponds (Tomašek, 2001)

Figure 2. Details of the locations at which samples where taken

case, because access to open water was required. Samples of water were drawn by immersing PET-bottles just below the water surface, taking care that there were no undesirable impurities on the water surface (leaves, grass, *etc*.). Before the measurements, the sample's precise volume was determined, then the sample was allowed to evaporate and the residue was calcined at 350 °C for 2 hours.

The sampling of soil and water was carried out under the following meteorological conditions – overcast, no precipitation and outdoor temperature around 19 °C.

Determination of mass and volume activity

For the determination of mass and volume activities of specific radionuclides in the samples, a gamma-ray spectrometer equipped with a HPGe detector (Canberra or Ortec, detection efficiency 37 % or 30 %, respectively) was used. The spectra obtained in the measurement were evaluated using the software GAMAT [6].

Soil samples were measured in Marinelli vessels and calcined water samples in a petri dish. Measuring time was 24 hours. ^{238}U was determined using the 63 keV emission of ^{234}Th which can be assumed to be in secular equilibrium with its parent in terrestrial matrices. Under the further assumption that the ^{235}U/^{238}U isotopic ratio is at the expected natural value, the 186 keV peak allowed determination of ^{226}Ra by correcting the peak for the ^{235}U contribution. ^{40}K was determined using its emission at 1460 keV [7]. The results were recorded in [Bqkg^{-1}] and [BqL^{-1}], respectively, and errors were calculated taking account also of the accuracy of weight and volume determination [8]. For each isotope, a minimal detectable activity (MDA) was calculated by the software. Only values above the MDA were recorded for the isotopes of interest in the present context [6].

Determination of gamma dose equivalent rate

Dose equivalent rates of gamma radiation were measured using a radiometer FH 40G-L10 (Eberline). Measurements were carried out at two different heights above ground, namely at heights of 5 cm and 1 m. The measurement was averaged over a 5 min period. The weather conditions during measurement were similar to those mentioned above – overcast, no precipitation and outdoor temperature around 10 °C.

Statistics

Usually, the values given below are means ± imprecision of the measuring device. In some cases, the measurements obtained in different locations were averaged; arithmetic means ± standard deviations are then stated.

RESULTS AND DISCUSSION

Radioactivity was determined in 24 samples from the area around the former uranium processing plant MAPE Mydlovary, among them 18 samples of soil and 6 of water. Measurements of gamma dose equivalent were carried out at the same locations.

Soil

Results of the radionuclide measurement of soil samples are presented in fig. 3 and tab. 1. Activity concentrations of ^{40}K ranged in the hundreds of Bq/kg (111.2 2.9 Bq/kg to 786.3 6.6 Bq/kg) with the exception of sample 7 where a value of only 25.1 2.2 Bq/kg was found. The average was 307.3 4.4 Bq/kg. For ^{226}Ra, the measured activity concentrations were between 14.8 0.4 Bq/kg and 219.6 1.1 Bq/kg, the value for sample 8 being an order of magnitude higher (1058.5 2.2 Bq/kg). Here, the average was 133.4 0.8 Bq/kg. Activity concentrations of ^{238}U lay in the range of 22.2 1.8 Bq/kg to 292.6 6.8 Bq/kg, with an average of 113.2 3.8 Bq/kg.

Our readings correspond with the results of other authors. Carvalho *et al*. [9] measured activity concentrations in the vicinity of former radium and uranium mining sites in Portugal and found 81 7 Bq/kg to 261 6 Bq/kg for ^{226}Ra and 123 22 Bq/kg to 337 40 Bq/kg for ^{238}U. Slightly higher values were reported by Winkelmann *et al*. [10] with ^{226}Ra activity concentrations in different waste deposits of the Wismut company in Eastern Germany in the range from 370 to 1600 Bq/kg; for ^{40}K, an average activity concentration of 860 Bq/kg was given. Again, our values agree very well with those of Tripathi *et al*. [11] who studied environmental radioactivity at the uranium processing and tailing facility at Jaduguda, India, (activity concentration of ^{226}Ra 12 to 151 Bq/kg) and with those of Mahur *et al*. [12] who carried out investigations in the surroundings of the National Thermal Power Corporation in Dadri, India, (activity concentration of ^{40}K 195.4 2.8 Bq/kg to 505.4 6.3 Bq/kg and of ^{226}Ra 32.2 5.9 Bq/kg to 120.9 4.5 Bq/kg). A wide range of values for ^{226}Ra activity concentrations around the former uranium milling facilities at the Pridnieprovsky Chemical Plant in Ukraine has been reported by Lavrova and Voitsekhovych [13] (30 to 36500 Bq/kg). Finally, we note that activity concentrations of ^{40}K and ^{238}U similar to ours (64-977 Bq/kg and 13-237 Bq/kg, respectively) were found by Tanic *et al*. around an abandoned uranium mining site at Stara Planina Mt., Serbia [14].

The control samples taken at the site of the nuclear power plant Temelín showed activity concentrations of 726 6 Bq/kg, 42 3 Bq/kg, and 39 6 Bq/kg, those at Hlincova Hora 967 8 Bq/kg, 22 4 Bq/kg, and 11 4 Bq/kg for ^{40}K, ^{226}Ra, and ^{238}U, respectively; in the case of ^{235}U, all measurements were below the limit of detectability. These measurements were in very good agreement with published values for uncontaminated places elsewhere in the world. The median values given in the UNSCEAR 2000 report [5] are 400 Bq/kg, 35 Bq/kg, and 35 Bq/kg and the population-weighted values are 420 Bq/kg, 32 Bq/kg, and 33 Bq/kg for ^{40}K, ^{226}Ra, and ^{238}U, respectively. In particular, values similar to ours have been reported for soil samples from neighbouring countries of the Czech Republic, namely ^{40}K concentrations of 520 Bq/kg, 410 Bq/kg, and 370 Bq/kg, ^{226}Ra concentrations of 32 Bq/kg, 26 Bq/kg, and 33 Bq/kg, and ^{238}U concentrations of 32 Bq/kg, 26 Bq/kg, and 29 Bq/kg for the Slovak Republic, Poland and Hungary, respectively [5].

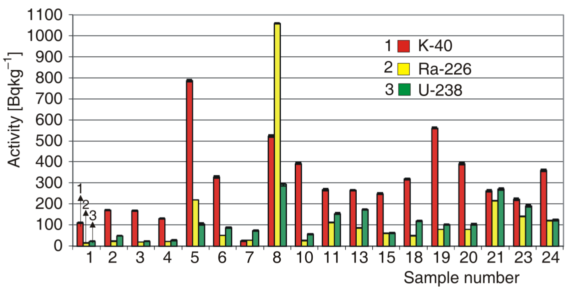

Figure 3. Activity concentrations in soil samples

Table 1. Activity concentrations in soil samples

Radionuclide	Activity concentration [Bqkg^{-1}] Minimum	Maximum	Average
^{40}K	25.1 2.2	786.3 6.6	307.3 4.4
^{226}Ra	14.8 0.4	1058.5 2.2	133.4 0.8
^{238}U	22.2 1.8	292.6 6.8	113.2 3.8

Water

Our results from the determination of activity concentrations in water samples from the area of MAPE Mylovary are summarized in fig. 4 and tab. 2. The values obtained were between 0.10 0.03 Bq/L and 0.50 0.03 Bq/L for ^{226}Ra, between 3.5 0.3 Bq/L and 9.3 0.4 Bq/L for ^{40}K, and between 0.3 0.1 Bq/L and 1.0 0.2 Bq/L for ^{238}U, in the case of sample 14 the concentration of ^{238}U was unusually high (7.4 0.2 Bq/L). As with the soil samples, the activity concentration of ^{235}U was below the limit of detectability.

The measured activity concentrations can be tentatively compared with the limits set out in Annex 10, tab. 5, of Decree No. 307/2002 (Coll. Radiation Protection) of the State Office for Nuclear Safety. These limits, of course, apply to drinking water, whereas water from the tailing ponds would never be used as such. The measured values can also be compared with activity

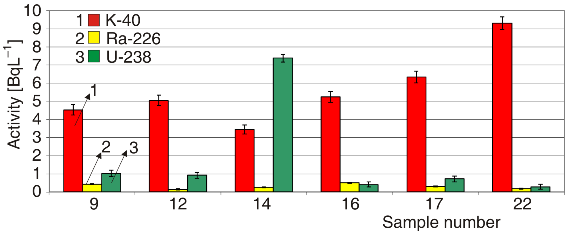

Figure 4. Activity concentrations in water samples

Table 2. Activity concentrations in water samples

Radionuclide	Activity concentration [BqL^{-1}]		
	Minimum	Maximum	Average
^{40}K	3.5 0.3	9.3 0.4	5.65 0.3
^{226}Ra	0.1 0.03	0.5 0.03	0.3 0.03
^{238}U	0.3 0.1	7.4 0.2	1.79 0.16

Table 3. Limits of activity concentrations for drinking water (according to Decree No. 307/2002 Coll. radiation protection, Annex 10, tab. 5)

Radionuclide	Limits of activity concentrations [BqL^{-1}]		
	Bottled water for infants	Drinking water for public supply, bottled table water, bottled water	Bottled natural mineral water
^{226}Ra	0.4	1.5	3
^{238}U	5	12	24

concentrations in water intended for human consumption listed in Council Directive 2013/51/EURATOM. These are 0.5 Bq/L for ^{226}Ra and 3 Bq/L for ^{238}U [15]. They were calculated under the assumption that a person drinking 730 L of this water annually would receive a dose of no more than 0.1 mSv. WHO guidance levels for radionuclides in drinking water are 1 Bq/L for ^{226}Ra and 10 Bq/L for ^{238}U [16]. Our activity concentrations of ^{226}Ra (0.3 Bq/L) were below these values – national, European and international – with the exception of one value (0.5 Bq/L) which exceeded the Czech limit for bottled water for infants (tab. 3). In the case of ^{238}U, the activity concentrations also complied with all three limits in most cases. Only one value (7.4 Bq/L) was higher than permitted by Council Directive 2013/51/ /EURATOM and also exceeded the Czech limit for bottled water for infants, but was still in line with the other Czech and WHO guidelines.

Gamma dose equivalent rate

The results of our measurements of gamma dose equivalents are shown in fig. 5. At a height above ground of 5 cm, values of 0.098 to 0.271 µSv/h were found and almost identical values of 0.109 to 0.264 µSv/h at a height of 1 m. The average values were 0.15   0.04 µSv/h for 5 cm and 0.15   0.03 µSv/h for 1 m,

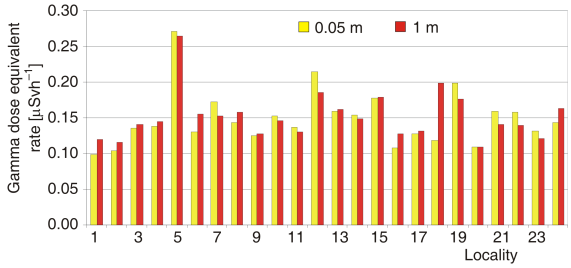

Figure 5. Gamma dose equivalent rate

respectively. This is very close to the "official" value given by the Dosimetry Department of the Office of Uranium Deposits in Pribram, which is 0.18 µSv/h (personal communication).

A person that would stay in the area for a whole year would thus be exposed to a dose of 1.3 mSv according to our measurements, or 1.6 mSv according to those of the Office of Uranium Deposits. If this estimate of annual doses was to be based on the highest measured value of the gamma dose equivalent rate, namely 0.271 µSv/h at site No. 5, we would arrive at 2.4 mSv or, with the highest dose rate of the Office of the Uranium Deposits, which is 0.286 µSv/h, we would get 2.5 mSv.

At the nuclear power plant Temelín and in Hlincova Hora we measured very similar gamma dose equivalent rates as around MAPE Mydlovary, namely 0.19 µSv/h at a height of 5 cm above ground and 0.15 µSv/h at 1 m above ground which would mean annual doses of 1.7 mSv or 1.3 mSv. It has to be kept in mind, of course, that the natural background radiation, cosmic as well as terrestrial, is included in all these measurements.

To estimate effective doses only from external terrestrial radiation, we used the relationship given in UNSCEAR 2000 [5]

$$D = 0.462\ \mathrm{AU} + 0.604\ \mathrm{ATh} + 0.042\ \mathrm{AK}\ [\mathrm{nGyh}^{-1}]$$

where AU, ATh, and AK are the activity concentrations of the radionuclides ^{238}U, ^{232}Th, and ^{40}K in soil. For site No. 5, which had the highest gamma dose equivalent rates, we calculated an annual effective dose of 0.489 mSv, using 0.7 Sv/Gy as the conversion coefficient from the absorbed dose rate in the air to the effective dose received by adults, and 0.097 mSv with an outdoor occupancy fraction of 0.2. These calculated values of annual effective doses are well compatible with our estimates on the basis of measured gamma dose equivalent rates, which include all kinds of natural background radiation. The limit of 1 mSv per year, which is recommended by the International Commission on Radiological Protection for exposures of the general public from human activities [17], is certainly not exceeded.

The gamma dose equivalent rate was also measured by Tripathi *et al*. [11] around the uranium processing and tailing facility at Jaduguda, India. They reported markedly higher values of 0.8 μGy/h to 3.3 μGy/h. Still higher values were found by Lespukh *et al*. [18] around a former uranium mine in Tajikistan, namely 0.4 μGy/h to 22.1 μGy/h, with an average of 9.1 μGy/h. A similarly broad range of values was reported by Lavrova and Voitsekhovych [14] around a uranium processing plant in Ukraine. Most of their values were between 0.15 μGy/h and 0.30 μSv/h, but at some locations, *e. g*., right at the surface of the tailings, they were up to 30 μGy/h and 40 μSv/h. In such local "hot spots", the ^{226}Ra activity in soil samples reached 100-200 kBq/kg.

CONCLUSION

The remediation and reclamation work around the now inoperative uranium processing plant MAPE Mydlovary and its tailing ponds is still going on. Nevertheless, it can be stated that the measured values of radionuclide concentrations in soil and water samples as well as the gamma equivalent dose rates are according to expectations for a site of this kind and comparable with the results of measurements in similar locations elsewhere in the world. It is also clear that workers involved with further remediation and reclamation are not expected to be exposed to doses exceeding the limits set by radiation protection regulations.

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AUTHOR CONTRIBUTIONS

The study was planned and put in context with literature data by F. Zolzer and R. Havrankova. Measurements were carried out by J. Havranek, J. Kankovsky, and L. Repa. All authors took part in the analysis and discussion of the results. The manuscript was written by R. Havrankova, and F. Zolzer, the figures were prepared by L. Repa and R. Havrankova.

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РАДИОЛОШКА СИТУАЦИЈА У ОКОЛИНИ МАПЕ МИДЛОВАРИ, ПРЕЂАШЊЕ ФАБРИКЕ ЗА ПРОЦЕСИРАЊЕ УРАНИЈУМА У ЧЕШКОЈ

МАПЕ Мидловари, фабрика за процесирање уранијума у Јужној Бохемији, Република Чешка, радила је око тридесет година, од 1962. до 1991. године. Од тада је обављен велики посао обнављања и оживљавања околине. У истраживању које је овде приказано, мерили смо масену и запреминску специфичну активност одређених радионуклида у узорцима земљишта и воде из околине и мерили јачину еквивалентне дозе гама зрачења на истим местима. Средње специфичне активности ^{40}K, ^{226}Ra и ^{238}U у земљишту биле су 307.3 4.4 Bq/kg, 133.4 0.8 Bq/kg и 113.2 3.8 Bq/kg, док су у води биле 5.7 0.3 Bq/L, 0.30 0.03 Bq/L и 1.8 0.16 Bq/L, респективно. Јачина еквивалентне дозе гама зрачења била је 0.15 0.04 μSv/h на 5 cm изнад тла, а 0.15 0.03 μSv/h на висини од једног метра. Као што је показано поређењем са налазима на другим сличним местима у свету, као и поређењем са резултатима мерења на незагађеним просторима, ове вредности су у сагласности са законским границама и не постји повод за бригу око заштите од зрачења радника укључених у даље обнављање и регенерацију, или извођење других послова у тој области.

Кључне речи: радионуклид, специфична активност, еквивалентна доза, обнављање