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ARTICLE

Screening for risk assessment around closed uranium mining sites

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Abstract – The aim of this study is to present the environmental impact and health risk 20 years after the closing of the uranium mining industry. The areas under observation are 31 former classical underground uranium mining and exploratory sites in Bulgaria. The geometric mean value of the gamma dose rate (0.33 μSv h^{-1}), measured 1 m above ground in the mining sites, was three times higher than the mean of the gamma dose in the nearby settlements (0.11 μSv h^{-1}). The geometric mean of specific activities for ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{210}Pb in mining site soil was found to be: 437 Bq kg^{-1}, 291 Bq kg^{-1}, 65 Bq kg^{-1}, 835 Bq kg^{-1} and 318 Bq kg^{-1}, respectively. Analysis of variance and correlation were applied. The external dose and offsite external hazard index, as health risk indicators, were estimated based on the results. After twenty years there is still environmental contamination in some locations that may cause a health risk, so remediation and monitoring should continue and be maintained.

Keywords: soil sampling / gamma radiation / outdoor radon / risk assessment

1 Introduction

Apart from the impacts of the uranium mining industry arising from chemical pollution and physical changes to the environment, radioactivity is a particular human health risk. Radiation exposure to humans and the ecosystem can occur due to radioactive materials migrating from or near the site, as well as the use of such materials for construction purposes. The presence of populations, industry and agriculture close to or actually in such areas can result in radiation protection problems.

Uranium mining residues contain naturally occurring radioactive materials. The human radiation exposure arising from mining and milling waste is influenced by the physical, chemical and radioactive properties of the waste materials. The radionuclides in undisturbed and unprocessed ores exist in conditions close to secular equilibrium. Mining and processing of ore alters the state of secular equilibrium as well as the physical and chemical state in which the radionuclides exist. This changed state may enhance their ability to migrate into the environment and give rise to exposure pathways to humans. Several studies worldwide have investigated the environmental contamination around mining and milling sites and the efficacy of remediation measures to reduce radiological exposure (IAEA, 1997, 2004).

The uranium mining and milling industry in Bulgaria was established after World War II, in 1945. In the whole country, 48 ore deposits were found and developed. According to Vapirev *et al.* (1996), there are 298 waste heaps covering an area of 85 ha and 8×10^6 m^3 (13×10^6 t). The waste heaps from the underground mines have an average activity concentration of 6 kBq kg^{-1}.

A lot of countries in Europe closed their uranium mining and milling industry and started remediation programs after the crash of the market in the 1990s (IAEA, 1997). The extraction and processing of uranium ores in Bulgaria was closed in 1992 and liquidation of the classical uranium mining and in situ leaching sites has been completed. During the mining and milling of uranium ores many other sites were only explored for the opportunity of mining. These sites were not included in the remediation program until 2007 and have not been rehabilitated so far. Environmental pollution surveys were carried out the for the purpose of remediation as well as epidemiological study (Chobanova *et al.*, 2003). After 2005, the remediation activities were limited to technical and biological remediation of the same sites, monitoring, and purification of the mining waters leaking to the surface, regeneration of ion exchange resins, and maintenance of and guarding the tailing ponds (EC, 2009). The polluted mine waters are purified only in three sites in Bulgaria, although other mining sites have contaminated water outflow (Ivanova *et al.*, 2015). Those mining sites are under assessment for appropriate water treatment techniques.

The objective of this survey is to investigate radionuclide variability and potential radioactive environmental contamination in the areas of the classical underground uranium mining

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Figure 1. Locations of the former uranium mining sites observed.

and exploratory sites 20 years after their closing. The indicators for screening for assessment of possible health risks are presented in this work. The data generated in the study will provide the values of natural radioactivity in or around the sites contaminated due to past activities and may be useful for the authorities concerned with implementation of radiation protection standards and monitoring programs. This work also contributes to the development of a procedure for screening for risk assessment of contamination sites and for grading them for control and remediation purposes.

2 Material and methods

2.1 Mining sites observed

The uranium mining sites with classical mining methods are characterized by a complex geological structure. They are situated in the mountainous regions (Stara Planina, Rhodopes massif, East Sredna Gora) at an altitude above 1000 m, with a mountainous climate characterized by relatively low temperatures, heavy rainfall and continuous snow retention. In this work, the environmental contamination and the risk of potential public exposure were evaluated at a total of 31 former classical underground uranium mining and exploratory sites. Any uranium mining site consists of one or several mines with adits and waste rock heaps. At each mining site the facilities (adit, waste rock heaps, etc.) were monitored. With the purpose of assessing the influence of the mining site, measurements were made in the nearby settlements too. Usually, the nearest village is located approximately 500 m^{-3} km from the mining site. Characterization of environmental contamination around the mining site was done through the following activities: a site visit; field measurements; and soil sampling, sample preparation and analysis. The locations of the sites observed and their codes are presented in Figure 1. The U code is used for mining sites with the classical underground method of uranium extraction, and the E code is used for exploratory sites.

2.2 Measurements

2.2.1 Outdoor gamma dose rate

The gamma dose rate was measured in accordance with the accredited procedure. All mining facilities, such as adits and waste rock heaps, were scanned at one meter above the ground. The average value of measurements for the mining site was then defined. The gamma dose rate was measured at the center of settlements, with the assumption that the value is representative of the whole village (Larssen *et al.*, 1999). For these purposes, the Geiger-Muller-type RADOS RDS-110 dosimeter, calibrated in the National Metrological Laboratory, was used. The dosimeter's measuring range is 0.05 μSv h^{-1} to 100 mSv h^{-1}. At the position of the measurement, an average value of 12–15 dosimeter records registered at 5-min intervals gave the gamma dose rate of a location. The relative uncertainty of the method was assessed to be 12%.

2.2.2 Outdoor radon concentration

In situ outdoor radon activity concentration measurements were carried out with an AlphaGuard PQ2000 radon monitor (Genitron, Germany) in the center of settlements where the gamma dose rate was measured. The AlphaGuard monitor is suitable for radon concentration measurements in the range from 2 to 2 000 000 Bq m^{-3}. The monitor was calibrated in the Bundesamt für Strahlenschutz, Germany. During the measuring procedure, the monitor was situated on the ground. Following the laboratory procedure, the values of outdoor radon concentrations were registered at 10-min intervals over approximately a 24-h period. At each location, the outdoor radon concentration was presented as a mean value of all records obtained.

Table 1. Descriptive statistics of measured quantities in air and soil from mining sites (MS) and settlements (S).

Sample	N	AM	SD	Min	Max	Med	KS test (p value)	Skew.	Kurt.	GM	GSD
D (MS) (μSv h^{-1})	31	0.37	0.20	0.17	0.91	0.29	0.316	1.346	0.815	0.33	1.61
D (S) (μSv h^{-1})	31	0.11	0.03	0.08	0.17	0.11	0.551	0.649	−0.762	0.11	1.25
^{222}Rn (S) (Bq m^{-3})	18	34	13	15	55	34	0.809	0.064	−1.236	32	1.49
^{40}K (MS) (Bq kg^{-1})	28	869	226	393	1169	961	0.131	−0.571	−0.837	835	1.35
^{210}Pb (MS) (Bq kg^{-1})	19	485	416	46	1414	395	0.824	0.919	−0.237	318	2.77
^{226}Ra (MS) (Bq kg^{-1})	28	577	614	32	2350	316	0.644	1.381	1.369	291	3.77
^{232}Th (MS) (Bq kg^{-1})	28	78	53	10	264	68	0.537	1.949	4.009	65	1.87
^{238}U (MS) (Bq kg^{-1})	27	1211	2883	42	15 183	460	0.873	4.446	19.041	437	3.78

The results from measurements in mining sites are noted with (MS) and measurements in settlements with (S). Table includes the minimum (min), maximum (max), median (med), arithmetic mean (AM), standard deviation (SD), skewness (skew.), kurtosis (kurt.), geometric mean (GM) and geometric standard deviation.

2.2.3 Sample collection and analytical methods

In order to characterize the mining sites, the soil samples were taken where the measured gamma dose was the highest. According to the sampling procedure, the sampling area is approximately 100 m^2. Five sub-samples are taken from every angle and center of the square as a form of envelope of 15 cm of the topsoil. The sub-samples are combined and homogenized into one sample with a weight of 1.5 kg. The activity of radionuclides in soil samples was analyzed from 28 mining sites. The natural radionuclides were analyzed as typical contaminants in the region. Out of 28 mining sites, ^{210}Pb was only measured in 19 of them where the ^{226}Ra content was enhanced. Due to the long period and natural reclamation process, there was usually a complex of material from waste heaps and forest soils. Organic materials and pebbles were eliminated during sampling and sieving. The samples were packed in PVC bags, labeled and transported to the laboratory for analysis. In the laboratory, they were dried, homogenized and sieved through a 2-mm sieve. Then, the samples were placed in a 1-l Marinelli beaker, weighed, sealed and stored for at least three weeks to allow secular equilibrium between ^{226}Ra and its decay products. Then, each sample was measured using gamma spectrometry and the weighted mean values of radionuclide specific activities were reported in Bq kg^{-1} air dry weight. The gamma spectrometry measurements were carried out by a laboratory validated method based on the International Standard IEC 61452 (1995-09), with a HPGe detector (45% relative efficiency, resolution of 1.95 keV at 1.33 MeV). A certified mixed calibration source with an energy range of 47−1836 keV and the same 1-l Marinelli beaker geometry was used for detector efficiency calibration. Under standard measurement conditions, the minimum detectable activities (MDA) of ^{40} K, ^{210}Pb, ^{226}Ra, ^{238}U and ^{232}Th in soil samples were in the order of magnitude of a few Bq kg^{-1}.

2.3 Statistical methods

The measured data was approximated as log-normal distribution; an alternative hypothesis was tested by applying the Kolmogorov-Smirnov (KS) test. In order to reduce the influence of extreme values in the further analysis the data were normalized due to the ln transformation. The significance of the type of the mining site for the measured quantities was tested by analysis of variance. Parametric ANOVA was performed only where the variances within the group were homogeneous; otherwise, in cases of heterogeneous variances, the non-parametric Kruskal-Wallis (KW) test was applied. The homogeneity of variance was tested by the Bartlett test. Person's correlation coefficients were used to examine linear relationships between the gamma dose rate and radionuclide specific activity in the soil of mining sites as well as its relation to the gamma dose rate and outdoor radon concentrations in settlements.

3 Results and discussion

The descriptive statistics for the gamma dose rate (D) in mining sites, and the gamma dose rate and outdoor radon concentration (^{222}Rn) measurements in settlements as well as the specific activity of ^{40}K, ^{210}Pb, ^{226}Ra, ^{232}Th and ^{238}U in soil sampled from the mining sites are presented in Table 1. The analysis was performed for long-lived natural radionuclides for investigation of the influence of past mining on their activities and correlation. The null hypothesis that a data set can be approximated with a lognormal function was not rejected due to the Kolmogorov-Smirnov test, at an error probability $p > 0.05$. Therefore, the average values of measured quantities are presented with the geometric mean (GM) and geometric standard deviation. The values of skewness and kurtosis differing from 0 indicate that the data do not follow normal distribution. The relatively higher kurtosis indicates peaked distribution, while the higher values of skewness are in relation to tailed distribution on the left (negative skewness) or on the right (positive skewness).

The results from many worldwide studies summarized in UNSCEAR reports show that the outdoor terrestrial gamma dose rate ranges from 0.01 to 0.20 μSv h^{-1}, with an average value of 0.06 μSv h^{-1} (UNSCEAR, 2000). In this survey, the variation of the gamma dose rate measured in settlements, which were situated approximately 500 m to 3 km from uranium mining sites, was in the range of 0.08 μSv h^{-1} to 0.17 μSv h^{-1}. The measurement values were in diapason of the average background radiation reported by UNSCEAR, which confirmed the representativeness of the measurement in the center of the villages. On the other hand, the geometric mean of the gamma dose rate of 0.11 μSv h^{-1} in the settlements

was three times lower than the geometric mean value of 0.33 μSv h^{-1} obtained from the measurements in the mining sites. As expected, the variation of the gamma dose rate in the mining sites was in a wider range: from 0.17 to 0.91 μSv h^{-1}, registering elevated radioactivity in the surrounding environment in many locations. Compared with the gamma dose rate measured in the vicinity of the uranium mining and milling sites of Mailuu Suu in Kyrgyzstan, the results are quite similar. In that survey, the authors reported mean values of the gamma dose rate for some households of Kara Agach (0.10 μSv h^{-1}), Bedresay (0.20 μSv h^{-1}) and a value of 1.10 μSv h^{-1} measured at the mine entrance (Corcho Alvarado *et al.*, 2014).

The measured outdoor ^{222}Rn concentration in the settlements varies in the range from 15 to 55 Bq m^{-3}, with an overall geometric mean value of 32 Bq m^{-3}. The values are higher than the average outdoor ^{222}Rn of 10 Bq m^{-3} estimated by UNSCEAR (2000). On comparing the measured outdoor radon values with those of some other studies, it is observed that the range value of this work is in consistence with the measured range from 22 to 77 Bq m^{-3} reported for some Bulgarian settlements (Kunovska *et al.*, 2014) and the range from 2 to 60 Bq m^{-3} measured in Mailuu Suu, Kyrgyzstan (Corcho Alvarado *et al.*, 2014). The measurement of the indoor radon concentration in some villages near uranium mining sites showed that an average of 50% of houses have around or above the reference level of 300 Bq m^{-3} (Ivanova and Badulin, 2012). This allows assuming that these areas could be considered as radon-prone areas.

Radionuclide activity in the soil samples varied within the study area. The geometric mean of ^{238}U was 437 Bq kg^{-1}, and the highest activity of 15183 Bq kg^{-1} was registered. The geometric mean value of ^{226}Ra was 291 Bq kg^{-1}, and the highest activity was 2350 Bq kg^{-1}. The decay of radium after twenty years leads to accumulation of ^{210}Pb in the samples from mining sites. The GM of ^{210}Pb was 318 Bq kg^{-1}, and the highest activity was 1414 Bq kg^{-1}, the GM of ^{232}Th was 65 Bq kg^{-1}, and the highest activity was 264 Bq kg^{-1} and the GM of ^{40}K was 835 Bq kg^{-1}, and the highest activity was 1169 Bq kg^{-1} (Table 1). The world average soil activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K are estimated to be 33, 32, 45 and 420 Bq kg^{-1}, respectively (UNSCEAR, 2000); the corresponding average concentrations in Bulgarian soil are 40 Bq kg^{-1} (8–190 Bq kg^{-1}), 45 Bq kg^{-1} (12–210 Bq kg^{-1}), 30 Bq kg^{-1} (7–160 Bq kg^{-1}) and 400 Bq kg^{-1} (40–800 Bq kg^{-1}) (UNSCEAR, 2000). Dragovic *et al.* (2006) reported ^{40}K, ^{232}Th and ^{238}U activities in the range: 271–919 Bq kg^{-1}, 18–83 Bq kg^{-1} and 15–53 Bq kg^{-1} in soil of the neighboring country Serbia. The activities of radionuclides from the ^{238}U chain in Greek surface soil ranged from 4–480 Bq kg^{-1} for ^{238}U, 0.5–372 Bq kg^{-1} for ^{226}Ra and 19–434 Bq kg^{-1} for ^{210}Pb (Anagnostakis *et al.*, 2005). The results of the current study are generally higher than results reported for normal soil.

3.1 Statistical analysis of data

Given approximate log-normality, all further statistical tests were performed only for ln-transformed values of our data, since this reduces the influence of extreme values on the statistics.

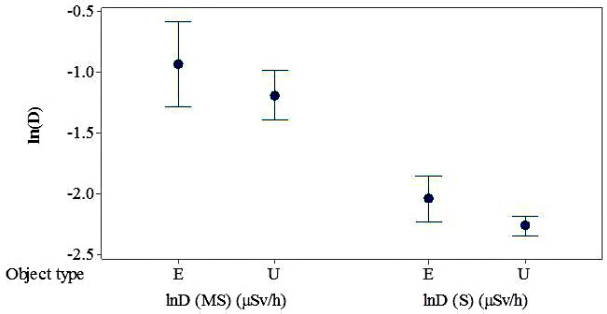

Figure 2. Interval plot of ln D in μSv h^{-1} measured in mining sites (MS) and settlements (S). Bars are at 95% CI of the mean.

In order to analyze variation of the measured quantities in function of the mining site type we classified our data from mining sites and settlements into two groups. The first group, marked U, contained the measured results from underground uranium mining sites, while the second group, marked E, refers to results from exploration sites.

In cases when the ln-transformed data follow normal distribution (KS, $p \geqslant 0.05$) and the variances within the group are homogeneous (Bartlett's test, $p \geqslant 0.05$), parametric analysis of variance (ANOVA) was applied in order to determine the differences between mean values of the groups. In cases where one of the conditions is not satisfied, nonparametric analysis was performed, using the Kruskall-Wallis test. The analysis showed that the gamma dose rates are not significantly different in these two types of mining sites (KW, $p = 0.107$). On the other hand, the geometric mean value of the measured gamma dose rate in settlements near E is higher than the GM near U (KW, $p = 0.027$), (Figure 2). The remediation measures carried out in the classical uranium mining sites might have reduced pollution from past activities. A significant difference between outdoor radon concentrations measured in settlements near E and U was not confirmed (ANOVA, $p = 0.139$).

Correlation analysis was applied, at the 95% confidence level, to determine relationships between the measured quantities in the mining sites and settlements. The correlations between the gamma dose rate and specific activity of radionuclides from the ^{238}U uranium chain in the soil from mining sites and correlation between the radionuclides themselves were significant. No correlation was observed between the outdoor dose rate in the mining sites and settlements. Also, no correlation was observed between concentrations of radionuclides belonging to the ^{238}U decay chain and both the outdoor dose rate and Rn concentration in settlements. This suggested that mining activities did not influence the exposure in settlements but influenced exposure in the mining sites.

3.2 Screening for risk assessment

It is generally assumed that the risk is proportional to the dose; the simplest version of this assumption is the linear dose-response relationship. Assessment of the radiological health risk can be considered in various terms. In the current study, two related quantities were deduced for screening for assessment of the risk: the offsite external index and

Table 2. Descriptive statistics of the estimated external hazard index (I_{ex}) for objects and the annual effective dose (E) in mining sites and settlements.

	I_{ex} (MS)	E (MS) (mSv y^{-1})	E (S) (mSv y^{-1})
No. of values used	28	31	31
Minimum	0.16	0.29	0.13
Maximum	4.60	1.60	0.29
Arithmetic mean	1.47	0.65	0.20
Standard deviation	1.20	0.35	0.05
Geometric mean	1.04	0.58	0.20
Geometric standard deviation	2.43	1.61	1.25

annual effective dose. More than one radionuclide contributes to the effective dose. There is a random mixture of natural radionuclides due to contamination of areas from past activities that were never subject to regulatory control. Not only is ^{226}Ra an important decay product, but also ^{210}Pb, because of its radiotoxicity and the high mobility of its decay product ^{210}Po. The offsite external index was evaluated using the idea of the index for building material (EC, 1999), as in both cases the risk of external exposure from the mixture of radionuclides is assessed. Thus, the inclusion and exclusion of radionuclides in the screening for external risk assessment is possible. The offsite external hazard index is calculated using the effective dose equivalent coefficient (Sv/Bq.s.m^{-3}) for radionuclides distributed to an infinite depth of soil assessed by Eckerman and Ryman (1993), whereas the density of air-dried soil is 1.3×10^3 kg m^{-3}. In a conservative calculation, the population outdoor occupation of 2000 h per year was used. Following the methodology of the activity concentration index for building material (EC, 1999), the offsite external hazard index for contaminated soil is derived as a criterion for health risk assessment activity:

$$I_{ex} = (C_K/5.8 \times 10^3) + (C_{Th}/370) + (C_{Ra}/540) + (C_U/3 \times 10^4) + (C_{Pb}/9.8 \times 10^5), \quad (1)$$

where C_K, C_{Th}, C_{Ra}, C_U and C_{Pb} are the specific activity of ^{40}K, ^{232}Th, ^{226}Ra, ^{238}U and ^{210}Pb in Bq kg^{-1}, respectively.

When no value is measured for some radionuclides, the index is calculated without their contribution. When the external hazard index equals 1, it will cause a radiation external effective dose of 0.3 mSv y^{-1}, which is the radiological criterion considered for a waste disposal facility, recommended by the ICRP (ICRP, 2013).

The annual effective dose equivalent received by the public due to external exposure was determined using the following formula:

$$E(\text{mSvy}^{-1}) = D(\mu\text{Sv h}^{-1}) \times 24\text{ h} \times 365.25\text{ d} \times 0.2 \quad (2)$$

where D is the gamma dose rate in μSv h^{-1} and 0.2 is the outdoor occupancy factor used by UNSCEAR (2000). The conservative suggestion was made that people spend their entire outdoor time at the mining sites. The annual effective doses were evaluated in the mining sites and in settlements in order to assess the natural level background.

Column 1 of Table 2 summarizes the I_{ex} results estimated by equation (1) and radionuclide activities in the soil samples.

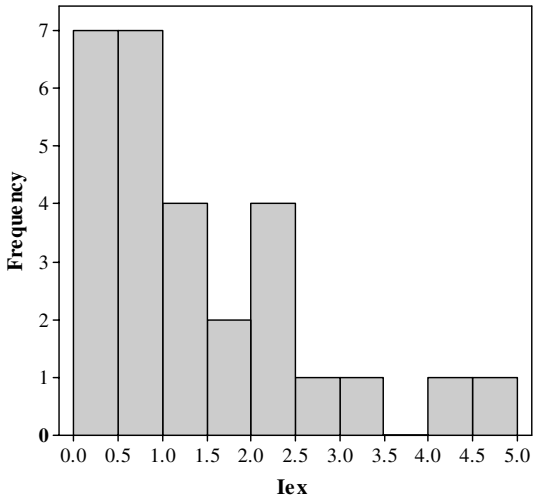

Figure 3. Frequency of values of the external hazard index.

The external hazard index due to ^{238}U, ^{232}Th, ^{226}Ra, ^{210}Pb and ^{40}K varied from 0.16 to 4.6, with an overall geometric mean value of 1.04. The results show that each site is specific and varies depending on the geology and the activities that took place at the sites. For the 14 observed mining sites the external hazard index is $\leqslant$1, and for 6 sites it ranges between 1 and 2. The I_{ex} is higher than 2 in 8 sites (Figure 3). These sites should be subject to more detailed risk assessment of exposure, taking into consideration the habits of the representative people. However, a highly conservative hypothesis was used to calculate this external hazard index and a more realistic hypothesis should be used through the use of an estimate of working time on the site. Columns 2 and 3 in Table 2 give the results of the estimated annual effective dose (using equation (2)) from external gamma radiation for the mining sites and settlements. In the present study, the values of the annual effective dose in the mining sites and settlements varied from 0.29 to 1.60 mSv y^{-1} and from 0.13 to 0.29 mSv y^{-1}, respectively. The contribution of external gamma radiation in the mining sites is quite significant, and is in the same order of magnitude as reported in the Kurday uranium mining site, Kazakhstan (0.9 mSv y^{-1}) (Salbu *et al.*, 2013). It should be mentioned that the estimated annual effective dose in mining sites might be overestimated, because it suggested that people spend their entire outdoor time at the mining sites. This is also the case for the external hazard index. Although highly conservative, this approach should be useful for screening for assessment of the residual environmental contamination and for prioritization of the sites for more detailed assessment and remediation.

4 Conclusion

The underground uranium mining and exploration sites twenty years after closing are still sources of potential environmental contamination by naturally occurring radionuclides. Most of these sites may represent a risk, having a potential radiological impact on the population and the environment.

Fourteen investigated sites have a high gamma dose rate in the mining sites and a high concentration of radionuclides from radium series in the soils. A statistically significant correlation between the gamma dose rate and specific activity of radionuclides from the ^{238}U uranium chain in the soil from mining sites was observed, as well as correlation between the radionuclides themselves. After twenty years there is still a need for reducing the potential long-term negative impact on the population and the environment by applying radiation protection principles. Taking into account the results from the survey the following conclusions and suggestions are made:

- Conservatively estimated risk indicators give evidence of exposure that cannot be disregarded from a radiation protection point of view. In the light of new recommendations (EC, 2014) some of these mining sites could be characterized as existing exposure situations.
- The program of remediation should be continued, but needs to be verified periodically on the basis of available site characterization data and the efficacy of remediation measures.
- In order to focus on available resources, sites could be graded for detailed assessment according to the screening for health risk indicators with a view to decisions on remediation activities and maintenance.
- The indicators presented for screening for risk assessment could be used for initial and periodical assessment of pollution of the sites from past activities and for implementation of a program.

In conclusion, it can be said that the main concern for the health risk is a periodic assessment of pollution and spread of contamination for consideration of further efforts, with the aim of optimizing protection and reducing any exposure that is still above the reference level.

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