Accumulation of natural radionuclides in wooden and grass vegetation from abandoned uranium mines. Opportunities for phytoremediation

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Abstract. The survey is focused on accumulation of uranium, radium and the concomitant heavy metals in forest and grass vegetation growing upon uranium waste rock dumps.

In the report are presented the final results from long-term investigations of factors on which depends the accumulation of natural radionuclides and heavy metals in vegetation.

Shown are the results of greenhouse vegetational experience with grasses and sorbents. Tracked are main growth indexes (increase in height and thickness and increase in volume) in the species of black pine-tree (Pinus nigra Arn.) and white pine-tree (Pinus sylvestris L.).

Introduction

Mining rock dumps from an abandoned uranium mines, after weathering, are sources of environmental pollution with natural radionuclides as uranium and radium, and heavy metals - iron, zinc, cadmium, lead, copper, arsenic. Frequently are observed acid rock drainage waters containing radioactive elements and sulphates as main pollutants.

Contamination of soils, groundwater, sediments, surface water and air with radionuclides and heavy metals is one of the major problems in the uranium mining areas. Different methods of remediation are known. Most of them are based on excavation of rock dumps, re-grading and stabilization the slopes, leveling the horizontal part and engineered dry covers – clay and soil masses. Traditional engineering technologies may be too expensive for remediation of most sites. Removal of metals from these soils using accumulating plants is the goal of phytoremediation (Baker et al., 1994; Brown et al., 1994; Brown et al., 1995a; Brown et al., 1995b; Blaylock et al., 1997; Carey, 1996; Chaney et al., 1997; Cunningham and Ow, 1996; Cunningham et al., 1996; Dushenkov et al., 1995; Moffat, 1995; Nanda Kumar et al., 1995; Raskin et al., 1994; Rouhi, 1997; Salt et al., 1995). Baker et al. (1991) concluded that phytoremediation, using certain species, could offer a low cost, low technology alternative to current clean up technologies.

Phytostabilization is the use of certain plant species to immobilize contaminants in the soil and groundwater through absorption and accumulation by roots, adsorption onto roots or precipitation within the root zone of plants (rhizosphere). This process reduces the mobility of the contaminant and prevents migration to the ground water or air, and thus reduces bioavailability for entrance into the food chain. This technique can be used to re-establish a vegetative cover at sites where natural vegetation is lacking due to high metals concentrations in surface soils or physical disturbances to surface materials. Metal-tolerant species can be used for restoration of vegetation in the sites, thereby decreasing the potential migration of contamination through wind erosion and transport of exposed surface soils and leaching of soil contamination to groundwater.

Though U and its daughter elements have not been shown to be essential or beneficial to either plants or animals, many plant species will absorb U and incorporate it into their biomass along with other heavy metals (Mortvedt, 1994; Sims and Kline, 1991; Sheppard et al., 1989; Singh and Narwal, 1984; Narwal et al., 1983; Moffet and Tellier, 1977). This observation suggests the possibility for remediation of U-contaminated soils through plant uptake.

The purpose of this study was the comprehensive survey of the ability of selected tree and grass vegetation to accumulate radionuclides and toxic metals from contaminated media. Assessment of growth of tree species – Pinus sylvestris L. and P. nigra Arn. in the conditions of uranium rock dumps.

The objects of this investigation were rock damps and natural vegetation of some uranium mines, situated in mountains “Stara planina” and “Rhodopa” in Bulgaria.

The Buhovo Mining Area (BMA) was the most important uranium ore region in Bulgaria. The total uranium production of BMA was 26% of the Bulgarian production. Mining activities in BMA were started in 1936 and continued until 1990. At that time mining activities developed in Goten, Chamilov kamak, Borche, Chora and in the main ore deposits – Seslavzi 1, 2 and 3, mainly as underground mines.

Buhovo ore field is located in the south slopes of “Stara planina” mountain (Kremikovci region of the Sofia Municipality). The total sum affected land from development of BMA is 2867 dca and formation of 141 dumps. Waste rock substrates of 19589.5 thousands m3 were deposited. Main rocks around the ore are clayshists, clayshists with high coal content, sandclayshistes, sandstones, granosienites, quartzsienites and pegmatites.

Table 1. ^{238}U concentration and concentration ratios (CRs) in plants and rock substrates.

Uranium mine	Sampling location	Plant species	Number of samples	Weight basis	Plant concentration, Bq/kg	Rock substrates/soil concentration, Bq/kg	CR
„Stara planina" mountain							
BMA	Rock dumps	Robinia pseudoacacia – leaves		A	ND	160 – 27940	
		Pinus nigra – 1age needles	15	A	0.886		0.0002
		Pinus nigra – 2age needles	15	A	1.291		0.0004
		Pinus sylvestris – 1age needles	12	A	1.472		0.0004
		Pinus sylvestris – 2age needles	12	A	2.772		0.0008
		Corilus avellana – leaves	2	A	2.947		0.0008
		Populus tremula – leaves	5	A	2.089		0.0006
		Carpinus betulus – leaves	5	A	2		0.0006
		Juniperus communis	3	A	61		0.0170
		Tussilago farfara – leaves	19	A	2.425		0.0007
	Background - Hromic Luvisols/Dystric Cambisols	Pinus nigra – 1age needles	7	A	0.866	304/1955.8	0.0008
		Pinus nigra – 2age needles	7	A	4.74	304/1955.8	0.0042
		Pinus sylvestris – 1age needles	5	A	3.09	304/1955.8	0.0027
		Pinus sylvestris – 2age needles	5	A	1.894	304/1955.8	0.0017
„Rodopa" mountain							
Kalach borun	Rock dumps	Pinus sylvestris – 1age needles	15	A	1.38	958 – 1336	0.0014
		Salix caprea – leaves	5	A	3.11		0.0031
	Open pit	Pinus sylvestris – 1age needles	15	A	2.59	508 – 32938	0.0004
		Picea excelsa – 1age needles	10	A	2.18		0.0003
Kara tepe	Rock dumps	Pinus sylvestris – 1age needles	15	A	3.6	227 – 230	0.0159
		Salix caprea – leaves	5	A	2.25	228 – 230	0.0099
Sufan dere	Background – Dystric Cambisols	Pinus sylvestris – 1age needles	3	A	1.9	227	0.0084
		Picea excelsa – 1age needles	3	A	1.73		0.0076
		Salix caprea – leaves	3	A	3.03		0.0133
Sdravez	Rock dumps	Pinus sylvestris – 1age needles	5	A	2.94	50 – 1462	0.0039
		Tussilago farfara – leaves	10	A	2.59		0.0034
Narechen	Rock dumps	Mix grasses	8	A	29 – 40	254 – 39514	0.0040
		Carpinus betulus – leaves	5	A	43		0.0049
		Tussilago farfara – leaves	5	A	29		0.0033
		Pinus nigra – 1age needles	5	A	28		0.0032
		Fraxinus ornus – leaves	5	A	105		0.0120

Comments: A - ash

The uranium mines “Sdravez”, “Kalach borun”, “Kara tepe’ and “Narechen” are situated in central part of “Rhodopa” mountain. Main rocks around the ore are eruptive (granites) and metamorphic (shistses, gneisses, clayshistes, marbles).

All dumps are located on the slopes of existing gullies and often dam them. The slopes are very steep, which is a reason for transport of materials together with the surface water run-off.

Materials and methods

Samples were taken by standard ANSI-C998-83.

Experiment with 7 variants for studying of the accumulation of radionuclides and toxic metals from waste substrates with different sorbents was carried out.

The results from green mass and contents of radionuclides were worked out statistically.

Results

Many studies have been conducted on the relationships between plants and soils relevant to radionuclide accumulation by plants. In these studies it was generally observed that plant species differ in U accumulation. Uranium accumulates mainly in the roots and depth of U placement and substrate properties influence absorption by plants. There is contradictory information on the phytotoxicity of soil U to plants (Sheppard et al., 1992). Levels as low as 1 mg kg -1 in soil, well within the normal background range, have been cited as toxic.

Measurements of uranium and radium concentrations in rock substrates and plants from the objects are presented in Tables 1and 2.

It was established that:

1. In the dumps of BMA - “Stara planina” mountain, that mainly consists of clayshists, granosienities and quartz, the contents of:
   - Radium in the excavated rock substrates vary from 60 to 3600 Bq/kg and in the vegetation – from 0.1 to 50.6 Bq/kg. The background contents of 226Ra in the soils of the region (Hromic Luvisols – 304.8 Bq/kg и Dystric Cambisols - 123 Bq/kg) significantly differs from the world’s characteristic - 0,03 Bq/g and from the soils in Bulgaria – 0,1 x 10-10 – 2,1 x 10-10% (Raikov, 1969; Raikov at all, 1971).
   - Uranium in the excavated rock substrates vary from 160 to 27940 Bq/kg and in the vegetation – from 0.8 to 61 Bq/kg. The background contents of 238U in the soils of the region (Hromic Luvisols – 80 Bq/kg and Dystric Cambisols – 482.5 Bq/kg) significantly differs from the world’s characteristic - 0,03 Bq/g and from the soils in Bulgaria – n x 10-5– n x 10-4% (Raikov, 1978).

2. In the dumps from objects in “Rhodopa” mountain, that mainly consists of granites crystalline shists and gneisses, the contents of:

- Radium in the excavated rock substrates vary from 39 to 29945 Bq/kg, and in the vegetation - from 1.55 to 201.33 Bq/kg. The background contents of 226Ra in the soils of the region - Dystric Eutryc Cambisols is 37 Bq/kg.
- Uranium in the excavated rock substrates vary from 50 to 32938 Bq/kg, and in the vegetation – from 1.38 to 105 Bq/kg. The background contents of 238U in the soils of the region (Hromic Luvisols – 80 Bq/kg and Dystric Cambisols – 482.5 Bq/kg) significantly differs from the world’s characteristic - 0,03 Bq/g and from the soils in Bulgaria – n x 10-5– n x 10-4% (Raikov, 1978).

Radium has slow mobility and the concentrations in the substrate are in direct connection with these in the main rock. This determines its high accumulation fron vegetation. Radium is accumulating more intensively compared to uranium. Relatively higher bioaccumulation of radium in the vegetation can be found in the objects from “Rhodopa mountain” compared to “Stara planina” mountain (CR from 0.006 to 0.12). This is result mainly from differences in: intensity of weathering of the rock substrates, soil texture, different sorption capacity of the formed during the weathering minerals, etc. For the BMA region the soil texture of fraction lower than 2 mm alters from heavy sandy clay to sandy clay, cation exchange adsorption capacity (T8.2) is represented mainly from clayish minerals of the kaolinite-illite and for the objects from “Rodopa mountain” the soil texture is from loamy sand to sandy loam and cation exchange adsorption capacity (T8.2) is represented by clay minerals muskovit and illit.

Stem analysis of model trees of Pinus sylvestris L. and P. nigra Arn.

It was established that: the white and the black pine trees can be characterized with strongly decreased values of indexes height and diameter, compared to the same from growth tables for the same species:

- Height of the modeled stem shows strongly decreased values in comparison with growth tables. The percent of such deviation has high values as it lessen with aging. This regularity can be seen in both surveyed species.
- d1,3 (DBH) – shows deviation from growth table as until 15 years of age in the white pine tree and until 20 years of age in the black pine tree has lower values and after that they increase and are greater values than the index in growth table.
- Growth in volume shows greatly decreased values compared to growth table. The percent of deviation lessens with aging which is representative for the adaptation of coniferous vegetation in aggravates growing conditions.
- Accumulation of radionuclides is in inverse proportion to the soil texture, the contents of organic substance and cation exchange adsorption capacity.

The results are presented in Tables 3 and 4.

Table 2. ^{226}Ra concentration and concentracion ratios (CRs) in plants and rock substrates.

Uranium mine	Sampling location	Plant species	Number of samples	Weight basis	Plant concentration, Bq/kg	Rock substrates/soil concentration, Bq/kg	CR
„Stara planina" mountain							
BOF	Rock dumps	Robinia pseudoacacia – leaves		A	22	764.6	0.0288
		Pinus nigra – 1age needles	15	A	0.211		0.0003
		Pinus nigra – 2age needles	15	A	0.166		0.0002
		Pinus sylvestris – 1age needles	12	A	0.962		0.0013
		Pinus sylvestris – 2age needles	12	A	1.026		0.0013
		Corilus avellana – leaves	2	A	7.952		0.0104
		Populus tremula – leaves	5	A	0.45		0.0006
		Carpinus betulus – leaves	5	A	1.96		0.0026
		Juniperus communis	3	A	14.2		0.0186
		Tussilago farfara – leaves	19	A	0.9		0.0012
		Лопен	3	A	50.6		0.0662
	Background – Hromic Luvisols/Dystric	Pinus nigra – 1age needles	7	A	0.516	123	0.0042
		Pinus nigra – 2age needles	7	A	0.415	123	0.0034
		Pinus sylvestris – 1age needles	5	A	0.495	123	0.0040
		Pinus sylvestris – 2age needles	5	A	4.355	123	0.0354
„Rodopa" mountain							
Kalach borun	Rock dumps	Pinus sylvestris – 1age needles	15	A	15.635	199	0.0786
		Salix caprea – leaves	5	A	10.974		0.0551
	Open pit	Pinus sylvestris – 1age needles	15	A	45.708	374-29945	0.0064
		Picea excelsa – 1age needles	10	A	201.333		0.0280
Kara tepe	Rock dumps	Pinus sylvestris – 1age needles	15	A	1.554	39	0.0398
		Salix caprea – leaves	5	A	2.385		0.0612
Sufan dere	Background – Dystric Cambisols	Pinus sylvestris – 1age needles	3	A	4	37	0.1081
		Picea excelsa – 1age needles	3	A	2.877		0.0778
		Salix caprea – leaves	3	A	2.338		0.0632
Sdravez	Rock dumps	Pinus sylvestris – 1age needles	5	A	1.287	255-2235	0.0015
		Tussilago farfara – leaves	10	A	9.872		0.0116
Narechen	Rock dumps	Mix grasses	8	A	36.8	648 - 1313	0.0376
		Carpinus betulus – leaves	5	A	45		0.0460
		Tussilago farfara – leaves	5	A	41		0.0419
		Pinus nigra – 1age needles	5	A	2		0.0020
		Fraxinus ornus – leaves	5	A	117		0.1195

Comments: A - ash

Uranium and radium uptake in grass, cultivation in greenhouse

Made is single factor disperse analysis using computer code STATGRAPHICS PLUS7.0 of the data (contents of radioactive elements in the vegetation mass) from separate treatment with corresponding repetitions.

The results for bioaccumulation of radionuclides in the comparative vegetation (Lolium perenne L.) are presented in Table 5.

The single factor disperse analysis shows good results depending on the contents of radioactive elements Ra and 236U in vegetation samples from the treatment option as the corresponding levels of significance are 0.0066 and 0.0074. With this two radioactive elements there is statistically reliable difference in control option from the other conducted options with ninety five percent “Duncan” and in it (control option) is observed the maximum accumulation. For the Th and K are not obtained statistically reliable differences in the used test between the separate options. Their contents in the overviewed experiment with Lolium perenne L. Do not directly depends on the way of treatment of the substrates.

By their effect on phytoremediation, sorbents could be divided into three groups: (i) highly reducing (a) – corresponding variants 05 for 226Ra and 06 for 238U; reducing (ab) – corresponding variants 01, 03, 04 and 06 for 226Ra and variants 01, 03 and 04 for 238U, (ii) no effect (bc, b) - corresponding variants 02 for 226Ra and variants 02 and 05 for 238U.

Conclusion

1. Bioaccumulation of radionuclides in tree and grass vegetation from dumps of uranium mining ores depends mainly on contents of radionuclides in the substrates, intensity of volatization and soil texture. Accumulation of 226Ra in leaf mass is more intensive than the 238U.
2. In the first years the growth of species of white and black pine trees is significantly oppressed. Later the tree vegetation is adapting to the harder conditions and shows sufficient growth.
3. Treating of the substrates with sorbents, containing organic substance and clay leads to strong reduction of bioaccumulation of 226Ra и 238U.

Table 3. Comparison of values of model stem with indexes for height (H), diameter (d1,3) and volume (V) with growth table.

Type	A(r)	H		d1,3 (DBH)		V	
		Growth table	Model stem	Growt h table	Model stem	Growth table	Model stem
White pine tree	**5**	**1.3**	**0.4**	**2.2**	**-**	**0.00304**	**0.00005**
(Pinus sylvestris L.)	**10**	**3.3**	**1.4**	**4.3**	**1.9**	**0.00897**	**0.00098**
	15	**5.6**	**3.0**	**5.7**	**5.8**	**0.01760**	**0.00444**
	18	**6.8**	**5.3**	**6.8**	**7.6**	**0.02509**	**0.00807**
Black pine tree	**5**	**1.1**	**0.3**	**1.4**	**-**	**0.00038**	**0.00007**
	10	**2.2**	**1.3**	**3.4**	**0.6**	**0.00421**	**0.00054**
	15	**3.7**	**1.5**	**5.2**	**4.0**	**0.01208**	**0.00130**
(Pinus nigra Am.)	**20**	**5.2**	**2.8**	**7.0**	**8.7**	**0.02407**	**0.00680**
	25	**6.8**	**5.2**	**9.0**	**12.0**	**0.04327**	**0.02046**
	28	**7.8**	**7.3**	**10.0**	**14.4**	**0.05570**	**0.04850**

Table 4. Percent of deviation of indexes H, d1,3 and V from growth table.

Type	A(r)	H	d1,3	V
White pine tree	**5**	**-69.2**	**-**	**-98.4**
	10	**-57.6**	**-55.8**	**-89.1**
(Pinus sylvestris L.)	**15**	**-46.4**	**1.75**	**-74.8**
	18	**-22.1**	**11.8**	**-67.8**
Black pine tree	**5**	**-72.7**	**-**	**-81.6**
	10	**-40.9**	**-82.4**	**-87.2**
(Pinus nigra Am.)	**15**	**-59.5**	**-23.1**	**-89.2**
	20	**-46.1**	**24.3**	**-71.7**
	25	**-23.5**	**33.3**	**-52.7**
	28	**-6.4**	**44.0**	**-12.9**

Table 5. Average values of radioactive elements in the leaf mass of the species Lolium perenne by options of treating of substrate and homogenic groups.

Treating option	^{226}Ra	^{238}U	^{232}Th	^{40}K
01	**76.67 ab**	**676.77 ab**	**53.33 a**	**2069.33 a**
02	**90.00 bc**	**715.00 b**	**62.50 a**	**2484 a**
03	**67.67 ab**	**543.33 ab**	**60.50 a**	**2201.67 a**
04	**78.00 ab**	**656.67 ab**	**68.33 a**	**1320.33 a**
05	**54.00 a**	**703,89 b**	**65.00 a**	**4252 a**
06	**62.00 ab**	**515.00 a**	**43.00 a**	**3104 a**
07	**115.67 c**	**916.67 c**	**75.67 a**	**3041.33 a**

Comments: The variants tick off with Latin letter formed one homogeneous group for given indicator. 01 – substrate + zeolit sorbent; 02 – substrate + modified zeolit (rhodopin); 03 – substrate + zeolit:rhodopin – (2:3); 04 – substrate + zeolite: coal ash – (5:1); 05 – substrate + vermiculite; 06 – substrate + hydrolyzed lignin + CaO + NPK fertilizer.

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