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Rehabilitation Technology of Radioactive Sterile Dumps

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Abstract

One of the most aggressive pollution comes from mining. Large areas are negatively affected either by daily mining or by deposition of residues from the underground mining. The tailings from uranium mining which is disposed in the dumps from Baita, Baita Plai, Fintinele etc. have a significant negative impact on the environment. The disposal area of uranium tailings at Baiata Plai covers 60 ha. The measures of remediation process are: characterization of the contaminated areas, dump designing, the dump covering, dump fertilization and acid reaction correction, crop structures. By fertilization organic and mineral fertilizers are applied. By organic fertilization 100 t/ha of well decomposed animal manure or 70 t/ha of compost prepared from animal manure are applied. The mineral fertilization consists of applying 200 kg/ha of nitrogen (applied in 2-3 fractions), 150 kg/ha of phosphorus and 200 kg/ha of potassium. The quantity of potassium applied has to increase at 250 kg/ha, if animal manure is not applied. The different tested plants (*Lotus corniculatus, Dactylis glomerata, Lolium perenne, Bromus inermis, Festuca pratensis*) had a very good behavior on the covered dump, giving high yields, a high degree of land covering and a good protection against erosion. Best results would be obtained if the plants would be cultivated in a mixture (each of them with a weight of 20 %).

Keywords: *uranium mining, dump, isotope, remediation, fertilization, plants*.

1. Soil specific features

One of the most aggressive pollution comes from mining. Large areas are negatively affected either by daily mining or by deposition of residues from the underground mining.

The tailings from uranium mining which is disposed in the dumps from Baita, Baita Plai, Fintinele etc. have a significant negative impact on the environment. The disposal area of uranium tailings at Baiata Plai covers 60 ha.

The radioactive isotopes are spread everywhere in the environment.

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In addition the nuclear industry, the industry of gas and ores extraction, the ores processing, the fertilizers and abrasives production, the military activities as well as the coal energy production have a high contribution to the radiologic doses received annually by the population [27].

Large areas in Europe have been affected by in a certain degree by the emissions from Cernobil. An area of over 200,000 km^2 from Europe were contaminated with radioactive cesium (over 0.04 MBq of $^{137}Cs/m^2$), from which 71 % were located in the most affected countries (Belarus, Russia and Ukraine). The deposits were very heterogeneous and strongly influenced by the rain at the moment of the contaminated air passing [1].

Since now, there are no any criteria or international standards established for defining a system for radiological protection of the environment.

The ecological risk evaluation (ERE) of the chemical products is based on methodologies well defined, but radionuclides have not received the same attention. The ERE of radionuclides will be based on the following scheme: i) the analyze of source duration and of the scenario of its releasing in the environment; ii) the identification of the exposure ways; iii) the potential induced to the biological effects at different levels (from sub-cell to individual level, with an additional level of populations and of environmental aspects) and estimations of the non-effect and final values; iv) the evaluation of risk associated to the presence of the contaminants. The risk is evaluated by the ratio between the predictable concentrations in the studied source at i) and the non-concentrations as they were defined at iii) [3, 18].

The uranium is extracted by mining, being used as fuel for the nuclear power plants which produce electric energy (for example, fuels for transport). In addition the electricity is produced by the nuclear energy without high emissions of greenhouse gases and thus the future severe consequences on the environment may be avoided [4]. From 1987, the content of plants and animals in radionuclides were determined mostly by the interaction between radionuclides and different soil components, because the soil is the main reservoir of the radionuclides stored on long term within the terrestrial ecosystems. These processes control the availability of the radionuclides plants and animals uptakes and also influence the radionuclides migration within the soil column [1, 24, 25, 26].

The most important factors that affect the rate of dissolving the fuel particles in soil are the acidity of soil solution and the physical and chemical properties of the particles (mainly the degree of oxidation). At values of pH lower than 4,0, the time of dissolving 50 % of particles were about one year, while at values of pH higher than 7, 0, 14 years were needed [1, 29].

For the program feasibility evaluation, uranium was chosen as a model of radionuclide, because it meets the whole criteria described above as lack of knowledge. In addition, being an emitter of α particles and having a heterogeneous distribution within the compartments of organisms, the uranium was the adequate element for studying the biological effects induced by the internal contamination. It also may have chemical and radio toxicity, which may lead to a better opportunity for identifying the adverse effects at reduced doses. The environmental experimental strategy is divided in three main areas:

i) evaluation of the radionuclides losses and their behavior within the exposed compartments (water, soil solution, sediments). The aim is to determine the radionuclides availability in soils and at the surface of ores sediments and of the organisms. The mobility, species and the residence time of the radionuclides in soils and sediments depend on the geochemical cycle of the entire elements, combined with the influence of abiotic processes (thermodynamic and sorption/desorption) as well as with the biological processes (reaction of microorganisms, the influence of plant roots and macro-fauna).

ii) study of the processes which lead to organisms contamination (for ex. bioavailability). These are interactions between radionuclides and different organisms which result from complex combinations of the biological and chemical processes, both of them being governed by kinetics and thermodynamics. These processes are essential for highlighting the best exposition which predicts the biological responses at different organizational levels.

iii) establishment of the dose – effect relationships by evaluating systematically a large number of functions of the organisms at individual level (mortality, growth, reproduction, behavior). The aim is to cover a variety of effects and actions, which may take place within an ecosystem. Thus, the primary mechanisms involved at sub-cell level are investigated (biochemical responses, genetic toxicity) and the potential consequences on the population dynamics by modeling the uptake of liquid and solid nutrients.

The processes of nuclear fusion regarding of the atomic weapons testing and electric energy generation, have contaminated the soils with an additional number of radionuclides. However just two of these have enough long life for having significant effects in soils: strontium 90 (half-life period = 28 years) and cesium 137 (half-life period = 30 years). The medium level of ^{90}Sr in United States soils is around 14.4 kilobecquerel on m^2 (qBq/m^2) or mCi/km^2. The medium level of ^{137}Cs is around 22.9 kBq/m^2 (620 mCi/km^2) [7, 21]

2. Rehabilitation of tailing dumps from uranium industry

The mining and uranium ores grinding have led to the occurrence of a variety of residual materials at soil surface (enrichment in particles transported by air), sterile materials, non-mineralized rocks (removed in order to reach the ore layer). In addition, the residual rocks which have low rates of mineralization and thus may not be grounded during the uranium extraction have significant contents of radionuclides and other minerals which are harmful for the environment. Other important residues are the miill scrap (consisting of ores from which the uranium was extracted) and sludge (containing metals and radionuclides from the water treatment plants). There different treatments for the residues. At world level, large areas with uranium mining are abandoned and need to be rehabilitated [31]

The main objectives of the protection against radiations are to prevent the harmful effects occurrence and to limit the probabilities of occurrence of late genetic effects on the acceptable considered limits [9].

Uranium, in its natural state, is a heavy radioactive toxic metal, which is uptaken with the solid and liquid food. The mammals have a high sensibility to uranium. Once the uranium it is in organisms, the uranium is transferred by blood in other organs. The uranyl (UO_2^{2+}) is the transported soluble which forms different complexes the protein and anions. The risk determined by the exposure to uranium may be either chemical or radiological. The first type of risk is regarding of uranium binding with the biological molecules. This risk is mainly harmful for kidneys because of high uranium concentrations presence during excretion processes. The humans uptake daily from food 2 – 4 µg of uranium. However, the total weight of the consumed uranium is governed mainly by the uranium concentration in the drinking water. A case study in the north of Germany showed that, in 20 % of cases, the uptake was doubled if mineral waters were consumed [8].

The remediation criteria for the ground waters within the mining plants are generally based on mining unit (the production-injection wells from field) as a whole. The first technical restoration may consists of different methods such as natural attenuation, groundwater cleaning, clean water injection [4].

The total removal of the historical contaminants, from economic point of view is not feasible. If this is feasible, the risk control of releasing the contaminants is accomplished by: 1) excavation, relocation and settlement, 2) decreasing the rate of releasing by using a cover, 3) rapid treatment of the contaminated mining/infiltration waters. If a remediation option is selected, then a specific solution is applied according to the place and object and that solution ensure an optimum ratio between the remediation cost and environmental benefits [16].

If it feasible, the target is to maximize the number and size of the lands rehabilitated for non-restrictive uses. In terms of remediation effort, this means: a) relocation of the contaminated soil, b) a perimeter of the area in order to ensure the surface runoff, c) erosion control (usually vegetation). Sometimes, the high costs and the relative low risk associated with an contaminated area do not justify the rehabilitation for a non-restrictive use. However, in many cases, the rehabilitation is just for restricted uses such as, industry or forestry, and not agriculture for food production or building houses. In such as cases, the land contaminated at surface with a specific activity of 1 Bq/g is excavated and the subsoil contaminated with more than 0,2 Bq/g, but less than 1Bq/g is covered in order to maintain the effective doses under 1mSv/a [16].

Two tailings dumps were built near Pecs (Hungary) in order to store the uranium ground tailings. The total area of the two dumps is about 1600000 m^2 and the estimated weight of the tailing is 20000 kt. The dumps contain 1382 tU (TP1:71.3 g/tU, TP II: 55.8 g/Tu). Remediation of the tailings dumps was the most important part of remediation of the mining complex. After uranium mining stopped, remediation started immediately, mainly because of the need to protect the underground water. Remediation of the tailings dumps was of an equal importance from the standpoint of the total site remediation. The steps for implementing the complete remediation of the two tailings dumps were: 1) rebuilding of the old drainage system, 2) groundwater restoration, 3) fine tailings stabilization, 4) re-contouring and preparation of the surface for covering, 5) covering, 6) biological remediation, re-vegetation [13, 23].

The information available at present related on dose-effect relationships is more extensive for leukemia than for other malignant conditions. The natural incidence of thyroid cancer is a function of age. The United States data show an incidence of four cases per year per million for person under 25 yr of age. The incidence rises linearly with age afterwards [10, 19].

Anthropogenic U contamination of agricultural soils is closely correlated with fertilization practices. Particularly, P fertilizers may add significant U loads to soils, which exceed significantly the harvest uptake. U in soils may be accumulated or lost to adjacent ecosystems by leaching and run-off. Site -specific thresholds for admissible U contents of soils are not yet defined [17, 22].

In the context of a mining site, the practical outcomes of a remediation program are to minimize the contaminant release rates (including oxidation rates, especially of acid generating materials) and to minimize the erosion within the waste storage structures to such as rates which may be received by the environment without any adverse impacts and if the impacts occur, they are low enough and may be considered as „acceptable to the community” [14, 31].

Uranium (and also tungsten) particles determine genetic changes in the cell elements and cancer occurrence. Uranium determines anomalous inflammation in the lung, kidney, brain and other living tissue of rats and neurological effects in case of mice. Uranium determines chromosome damage at miners and Gulf War Veterans [28].

The term of remediation is used for all the activities which lead to reduction of the radiation exposure and improvement of environmental and/or economic values. The terms of rehabilitation and restoration are interchangeable. Remediation consists of: pollutants removal, encapsulation and monitoring of non-intervention [5]. A critical element is to characterize the contamination and the various environmental compartments affected by contamination, in order to be able to evaluate the applicability of the remediation techniques. The chemical or mineralogical form of the contaminant will critically influence the efficiency of the chosen remediation technique [5].

Phytoremediation comprises different perspectives such as phytoextraction, phytominig, phytostabilization, phytovolatilization and phytofiltraton. Phytoextraction Is only an economically important way to restore soils, if they are slightly contaminated, because the heavily contaminated soils require metal resistant plant species. The three basic elements for a successful phytoextraction are: first, the degree of contamination; second, the degree of plant availability and third, the capacity of plant to accumulate metal in shoots. The problem of highly contaminated soils is that only plants with low biomass grow on them, so that the cleaning procedure takes a long time, 10-20 years, so that, within one century less than 1% of the metal will be removed [12].

Plants are considered hyperaccumulators if they contain more than 100 mg kg^{-1} Cd, more than 1000 mg kg^{-1} Co, Cu, Cr, Pb and Ni, and more than 10.000 mg kg^{-1} Mn and Zn. In case of plants with high biomass, the phytoextraction is not be suitable because of an imbalanced time/cost ratioo. In these cases, the chances for identifying a plant species that hyperaccumulates U is extremely low [17].

Because of the cation-exchange properties of soils, the levels of radionuclides in most soils are not high enough to be hazardous. Most of the ^{90}Sr and ^{127}Cs from soil are adsorbed by the soil colloids and exchange other cations previously adsorbed, thus, the plants take up mainly the exchanged cations rather than the adsorbed radionuclides [7, 15].

In our case, the discussion relates just on the measures for encapsulation associated with replanting the tailing dumps.

An annual effective dose of 10 mSv coming from all the sources is considered as reference for remediation measures. If this dose is less than 10 mSv, the remediation measures are not necessary. If the dose is higher than 100 mSv, remediation measures have to be applied. The decision related to application of remediation measures will take into account, besides the protection against radiations, other factors such as, people who might be affected if the remediation measures are not applied. Public acceptability can be a major factor in selecting a particular remediation technique [5].

The fundamental rules for safety management of radioactive wastes and used fuel established by the Order of the president of the National Commission for Nuclear Activities Control no. 56/2004 stipulate that the authorization holder have to manage the radioactive wastes and used fuel by following the next outlines [30, 31]:

- Human health protection;
- Environmental protection;
- Protection beyond the national borders;
- Protection of the future generations;
- Avoiding unnecessary burdens for the future generations;
- Existence of a national legislative framework;
- Control of the radioactive wastes production;
- Interdependence between wastes production and management;
- Maintenance of the equipment security;
- The polluter pays.

The management of radioactive wastes consists of the following stages: pretreatment, treatment, conditioning, intermediate storage and final storage. These stages are considered as parts of a total system, from waste production up to the final storage. In order to reduce the radionuclides release, the radioactive wastes are isolated by placing barriers around them. The barriers might be either natural or artificial. A multi barrier system provides a better isolation, thus any release of radionuclides in the environment will take place at an acceptable low level [30].

The application of a remediation technique requires detailed studies in order to evaluate the technical feasibility of the proposed measures and their impact. Thus, data from different scientific and practical areas such as health, chemistry, physics, geology, microbiology and environmental engineering, are needed for developing technical solutions. It is also necessary to include information related to the political, social and economic context. Other indicators such as costs versus availability of resources in a certain time, public perception and availability of skilled workers need to be considered [5].

2.1. Rehabilitation of the dumps with a high level of radioactivity

A dump which is out of use must have a long term itself storage capacity of the tailing. Thus the radioactivity level may be reduced to less than 1 mSv/a.

The waste rock dumps, which have a high radioactivity, are remediated in situ, in centralized sites or in an empty mine. In situ the rehabilitation of the waste rock dumps consists of the following measures that need to be applied: (a) reshaping of the dumps to a geo-mechanically stable form, and, (b) covering with a soil layer in order to reduce radon exhalation and external radiation and to limit for a long term the infiltration within the dump. After covering, (c) the surface is covered with vegetation in order to control the erosion. As much as possible the tailing dump is shaped and designed to be a part of the surrounding landscape [16].

Barnekow et al., 2005 have tested different cover concepts [2]:

- a storage layer consisting of a “clean tailing material” (mixture of tailing and granular soils).

- two layers (storage layer and sealing layer) or three layers (storage layer-drainage layer-sealing layer). The sealing layer consists of two different materials either quaternary loam or weathered Permian red-bed sediments. The storage layer consists of a mixture between tailing and a granular soil.

- a capillary barrier enclosing a storage layer of a mixture between tailing and a granular soil.

The rock wastes dumps, which have a high radioactivity, are remediated by covering in situ, by replacing at a centralized storage that provides the necessary protection condition or by reintroduction into the mines galleries.

The rehabilitation in situ of rock and rock wastes dumps requires the following remediation measures: a) to redesign the dump in a geo-mechanically stable shape and b) to cover the dump with a soil layers system designed in order to reduce the radon emissions and external radiation and to limit the long term leaching within the dump. After soil layers coverage, c) the surface is covered by vegetation in order to control the erosion. The shape of dump has to fit with the neighboring landscape elements and to resist to the impact of the precipitation with high intensity.

After the final cover, the infiltration may be limited by settling a covering and protection layer or by providing a layer of water storage that maximizes the infiltration and water storage within the soil cover, from where it will be lost through evapotranspiration. The water storage layer is more advantageous because is cheaper and has a lower risk of breaking down by the vegetation. The plastic protection layer may be penetrated by roots or may be broken by compaction, by animals or vehicles passing. The soil cover should have at least 200 years lifetime.

Because of the high radioactivity, the cover has to be very thick (2 m). The first covering layer which acts as an infiltration barrier will have 0.5 m of thickness and will be compacted as such as level at which the permeability coefficient is less or equal with 5×10^{-9} m/s. The second layer, of infiltration and storage, should have a thickness of 1.5 m and will not be compacted. This layer will be covered by vegetation in order to allow the evapotranspiration and to protect the dump against erosion. The crop rotation will be chosen according to the local pedo-climatic conditions.

The best system of protection against erosion is obtained by sowing a mixture of gramineous and perennial leguminous with 200 kg/ha of seed and planting shrubs or trees 8000 seedlings per ha, at a distance between rows of 2.5 m and a distance between shrubs or trees on row

0.5 m. 80-100 t/ha of animal manure and mineral fertilization, $N_{60}P_{50}K_{90}$, is recommended to be applied. If there is no animal manure, the mineral fertilizers doses has to be increased at $N_{120}P_{100}K_{170}$.

The literature data high highlight that this remediation measure costs around 29 euro/kg of extracted uranium.

The Hanford Barrier in eastern Washington is a large-scale test prototype of a maintenance-free closing system designed to store radionuclides for 1000 years. The site had been used to dispose of low-level radioactive liquid waste associated with uranium reclamation plants. However, the soil contains a high (>1,000,000 picocuries/g) contamination with strontium-90, cessium-137, plutonium-238, plutonium-239/240, and uranium between 5 and 15 m below the surface. The barrier prevents rainwater infiltration in soil and contaminants leaching to the groundwater. The covering system consisted of the following layers from the top to down: 1m of silt sediments, with a surface slope of 2 percent; 1 m of silt loam; 0.15 m of sand with geotextile; 0.3 m of gravel; 1.5 m of basalt riprap; 0.3 m of gravel; 0.15 m of asphalt.

The sand and gravel layers below the silt loam serve as a capillary break that inhibits the downward percolation and prevents the fine soil filtration downward into the riprap. The riprap and gravel layers exceed the 2:1 side slopes.

The riprap deters root penetration and animal burrowing occurrence. The asphalt is a hydraulic barrier and redundant bio-intrusion layer and lower side slopes than the riprap ones [11, 20].

2.2. The rehabilitation of the dumps with low level of radioactivity

There is a variety of technological options for disposal of low contamination levels. The approaches are broadly grouped into three categories of: 1) non-intervention; 2) isolation; 3) removal.

A decision for not site cleaning up implies depends on:

- the capacity of the natural local conditions (rocks, soils, sediments and groundwater) to mitigate the contaminant migration (i.e. natural mitigation), or on

- the physical (radioactive decay, filtration, volatilization), chemical (precipitation, co-precipitation, sorption) and biological processes (bio-mineralization, bio-sorption and microbial mediated transfer) which may lower the activity levels below those of concern.

Alternative land uses and non-agricultural measures are methods for disposal of low contamination levels. The isolation measures include: enhanced mitigation, low permeability barriers, permeable reactive barriers, immobilization, biological barriers, phyto-stabilization. Generally, any method which is based on the contaminated soil removal requires the replacement of the removed material with a clean (top) soil. There are different methods: immobilization and solidification (ex situ), surface and groundwater pumping and treatment, enhanced recovery, chemical extraction (ex situ), hydrometallurgical methods, segregation, bio-sorption, bio-leaching, phyto-extraction and rhizofiltration [5].

The specific aims of the technical measures for closing a mining waste dump are:

- Limitation of the radon exhalation;
- Limitation of contaminants leaching and erosion protection of the mininig waste dump by covering;
- Limitation of the effect of surface and groundwater on the mining waste dump (erosion, wetting of waste);
- Ensuring the geo-mechanical stability of the mining waste dump, namely the resistance to erosion, by reshaping the mining waste dump;
- Prevention of erosion and water saturation of the cover by means of a drainage system.

The remediation of a diffused contamination is still a challenge if factors such as, expected dose, cost, public perception and anxiety, and minimal disturbance of the environment, are taken into account.

Answers for solving the problem problem of contamination remediation should be found in solutions that relates on low cost, low intensity, low maintenance (passive) and often low technologies [5].

Once that the remediation objectives are established, several factors have an impact on the decision making process.

These basic evaluation criteria include engineering and non-engineering considerations:

- Effectiveness in remediating the contamination;
- Cost associated with the remediation program;
- Safety and health risks associated with the technology;

- Potential secondary environmental impacts (collateral damage);
- Prior experience with the application of the technology;
- Sustainability of any institutional control required;
- Socio-economic considerations [3]

I. The first measure that has to be considered consists of the characterization of the contaminated area in order to evaluate the applicability of the remediation measures. With this regards, soil samples are taken from the tailing dumps, at 20 cm and 40 cm depths. They are analyzed in the laboratory. The main laboratory analysis are: particle size distribution, pH, humus, total nitrogen, available phosphorus, available potassium, degree of base saturation and total heavy metal content.

The radioactivity level of the whole area is evaluated by accomplishing a large scale study. Once that the radioactivity level is established, the first choice of the remediation measures is done. It will be decided if:

a) the place will be let undisturbed and a monitoring scheme will be established in order to control the evolution of the place. This option is based on natural processes and it prevents a significant exposure. The whole process is monitorized carefully, so that alternative actions might be identified in case of need.

b) encapsulating and restricting the radioactive contaminants mobility: this action consists of contaminants immobilization within the affected area and decreasing the potential of a future contaminants migration. This may be done by covering the area with fertile soil and cultivating the soil cover.

c) removal of radioactive contaminants out of the polluted area; the contaminants are extracted, concentrated and then stored safety in another location.

The people have to accept the remediation measures. An active implication of the people in the remediation processes will increase the level of their knowledge and care regarding of this problem, will increase the acceptance of these techniques which are needed for a future development of the place and will understand the restrictions related to the use of the area.

It is established the area from which the soil will be taken and used for covering the dump. Soil samples are taken and analyzed in the laboratory.

The sources animal manure and mineral fertilizers are identified. Animal manure (100 t/ha) and mineral fertilizers ($N_{200}P_{150}K_{250}$) will be applied. A proper crop rotation is established. Theanalyze of the public perception and of the need of people for dump re-vegetation is accomplished. In order to optimize the intervention, a balance between the risk of exposure to radiations and the remediation actions cost will be done. Fig. 1 shows the decision-making phases in remediation technologies and strategies.

3. Dump designing.

The dump is designed in a way in which the risk of erosion is minimized and the soil cover is applied uniformly as much as possible. A better leveled dump, better conditions for soil cover application. If the dump has high slopes, besides leveling, it is necessary to accomplish wicker fences, placed along the slope. Thus the soil cover is better settled without any risk of its movement towards the downward of dump. The distance between the fences will be no more than 2 m.

4. The dump covering.

The dump has to be covered with at least 30 cm of fertile soil in order to provide as much as possible optimum condition for vegetation development and to reduce the risk of heavy metals and radioactive isotopes uptake by the plant. A lot of studies show that by covering, the isotopes concentration in plants (*Lotus corniculatus, Dactylis glomerata, Festuca pratensis, Lolium perenne, Bromus inermis Leiss, Robinia pseudoacacia, Gleditsia triacanthos, Fraxinus*) are lower than detection limit. The isotope concentration in plant was lower than detection limit in case of a soil cover with a thickness of 10 and 20 cm. But in case of shallow soil cover, the risk of vegetation development is very high, because the rainfalls are not distributed uniformly. Besides the tailings have skeletal properties, with a very low water retention capacity. The heavy metals content of the grasses grown soil cover was in normal limits. So, the minimum recommended thickness of the fertile soil cover should be of 30 cm. Practically, it is almost impossible to provide this exact thickness. Because of this the soil layer applied on terrace will have a thickness of 35 cm and that one applied on slope will have a thickness of 45 cm.

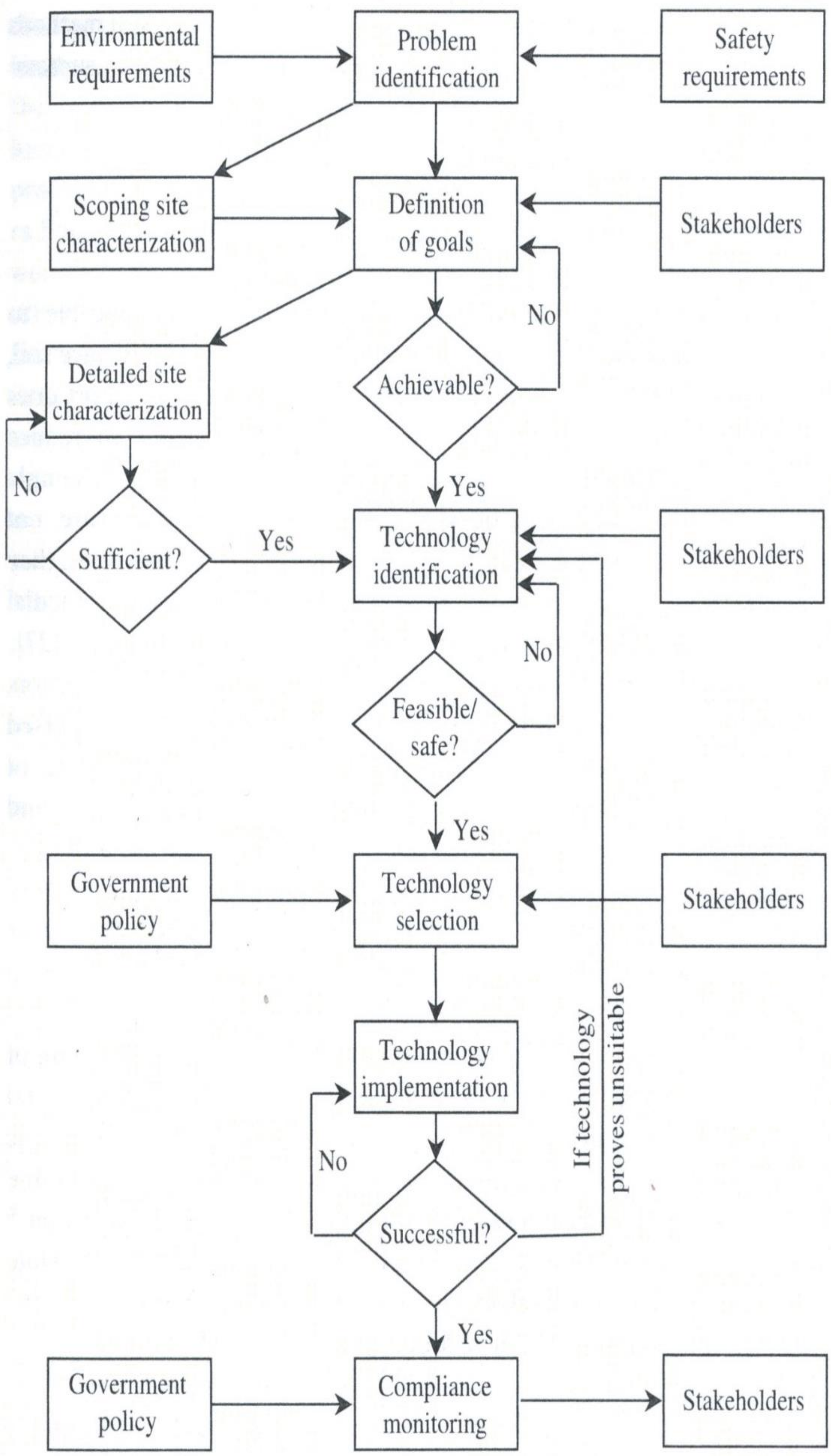

Figure 1. The phases of decision making in remediation strategies and technologies

Grabas and Koszela (2005) recommended as a remediation strategy to build up of a multi-layer bottom liner and surface cover consisting of [14]:

- Covering the bottom of the tailings dump with lime (thickness 0,6 cm);
- Covering the bottom with a geotextile (type 300 g/m^2);
- Emplacement of a layer containing dolomite aggregates (d= 10-50 mm), the thickness of the dolomite layer being 0.3 m;
- Covering the dolomite layer with geotextile (250 g/m^2);
- Material from the tailings ponds (30-50 cm);
- Bentonite (1 cm);
- Dolomite (15 cm);
- Fertile soil (with humus) 45 cm;
- Development of a vegetation cover.

A typical technical cover for a tailings dump recommended by Delgado and Fernandez (2005) contains: 1) a first layer of clay used as a barrier for reducing the radon exhalation from the tailings, and as waterproofing top for preventing the infiltration of rainwater into the tailings; 2) a second layer of granular material is placed on top of this clay layer to improve the drainage of the rainwater; 3) the third layer provides protection against wind and water erosion of the lower layers, and may consist of gravel or a layer of fertile topsoil.

5. Dump fertilization and acid reaction correction

The soil within the uranium tailings dumps usually, are acid. Because of this aspect, amendments with calcium carbonate have to be applied in soil. The amendment dose is calculated by the following formula:

$$DAC, t/ha = SB\ (Vd/Vi – 1).\ K\ .\ 100/PNA$$

where:

DAC = calcareous amendment dose;

SB = exchangeable bases sum of the non-amended soil, me/100 g of soil;

Vd = degree of saturation in bases (%) which has to be attained by amending;

Vi = degree of saturation in bases (%), of non-amended soil;

K = a coefficient obtained by multiplying the thickness of amended soil layer with bulk density and with a value of 0.6. This value represents the $CaCO_3$ quantity (g) needed for neutralizing of 1 me of acidity contained in 100 g of the soil that has to be amended.

PNA = neutralizing demand of the amendment, expressed as % of $CaCO_3$.

The acid reaction correction stimulate the plants development, soil bacteria multiplication, decreasing the risk of radionuclides and heavy metals uptake in plants by reducing their mobility. After one uniform as much as possible application, the amendments are incorporated in soil. The pH of dump has to reach neutral or very close to neutral values (7.0).

By treating the soil, the uptake of radiocaesium (and radiostrontium) is reduced. The procedure may involve plowing, reseeding and/or the application of nitrogen, phosphorus, potassium fertilizers and lime. By plowing, the radioactive contamination is diluted especially in the upper soil layers, where most plant roots absorb the nutrients [1]

After the accident that took place in Chernobyl NPP, 19 regions of the Russian Federation were affected by radioactive contamination. One of the most efficient ways to reduce the radionuclide (^{137}Cs and ^{90}Sr) uptake by plants is to apply high doses of phosphorus and potassium fertilizers. The extraction of local phosphate rocks and their use as fertilizers is a promising way to solve the problem. The phosphate rocks have the potential to increase the soil fertility and productivity, to improve the quality of products by reducing their radionuclides contents in areas affected by radioactive contamination [19].

By fertilization organic and mineral fertilizers are applied. By organic fertilization 100 t/ha of well decomposed animal manure or 70 t/ha of compost prepared from animal manure are applied. The mineral fertilization consists of applyin 200 kg/ha of nitrogen (applied in 2-3 fractions), 150 kg/ha of phosphorus and 200 kg/ha of potassium. The quantity of potassium applied has to increase at 250 kg/ha, if animal manure is not applied.

Kuznetsov at all. (2002) have found that different rates of local phosphate rock application have different effect on ^{137}Cs uptake by the barley yield. By applying rates of 21.8 mg P/kg of soil, the differences of ^{137}Cs transfer in barley grain and straw were insignificant compared to the control [19].

However, at rates of 43.6 and 87.2 mg P/kg of soil the decrease of ^{137}Cs accumulation in grain and straw was about 1.2 to 1.4 times less than the control (without adding P).

The animal manure does not acts just as fertilizer, but also as physical, chemical and biological ameliorator the crop substrate. Furthermore, organic-metallic bonds formation lead to the decrease of radionuclides and heavy metals uptake by plants (especially of copper, zinc and cadmium). The phosphorus also reduces the zinc and cadmium uptake by plants and the potassium reduces the lead and radionuclides uptake by plants. It is necessary to maintain the level of available potassium in order to reduce the radionuclides transfer in plants. This is why the best manure is that produced by caws.

In case of a clayey soil, it is useful to incorporate betonite or zeolitic tuff in order to increase the cationic exchangeable capacity of the substrate and to immobilize the radionuclides. The betonite or zeolitic tuff doses have to be very high, over 30 t/ha.

Two months after seedling the field will be fertilized again with $N_{50}P_{50}K_{50}$.

The best NPK fertilizers are the organic-mineral ones, which release more slowly the nutrients and provide a longer period the nutrients needed by plants.

After an uniform application, the fertilizers are incorporated in soil.

6. Crop structures

The different tested plants (*Lotus corniculatus, Dactylis glomerata, Lolium perenne, Bromus inermis, Festuca pratensis)* had a very good behavior on the covered dump, giving high yields, a high degree of land covering and a good protection against erosion. Best results would be obtained if the plants would be cultivated in a mixture (each of them with a wight of 20 %).

The seed quantity for all the species should be 2-3 times higher than that used on normal fields. The doubled quantity is used on horizontal fields, the tripled one on slopes.

The seed quantity used on normal fields is of 30 kg/ha for *Festuca pratensis*, 25 kg/ha for *Dactylis glomerata*, 30 kg/ha for *Lolium perenne*, 35 kg / ha for *Bromus inermis* and 20 kg/ha for Lotus corniculatus.

The dump is planted with different shrubs and trees species in order to ensure a long term protection against animals and humans presence.

At the edge of the dump 3 rows of *Gleditsia triacanthos* will be planted, with a distance between rows of 1 m, 25 cm between plants in the first row and 50 cm between the plants from the second and third rows. The plants from the second row should be placed at half of the distance between the plants from the first row. The *Fraxinus* and *Robinia pseudoacacia* will be planted at a distance of 1.5 m between rows and a distance of 1 m between them on row. Before planting, the seedlings should be slushed for maintaining the roots not dried and providing a good contact between roots and soil.

Normally, if some losses occur, there are of a low level, which not affect the seedlings development. If the losses exceed 10 % then, in the next year, the gaps will re-planted.

If the tailings dump is not located in a mountain area, other crop rotation may be used, the optimum solution depending on the radiological criteria, economic, social, political aspects and scientific knowledge.

A lot of studies were targeted to the possibility of agricultural use of the tailing dumps and to the measures which should be taken in order to limit the radionuclides concentration in the food production in admissible levels. If alternative crops are studied there are some questions that need an answer:

- May be found alternative crops suitable to the existing climatic and soil conditions from the contaminated area?
- Which are the behavior of the radionuclides within the crop rotation?
- How the radionuclides will behave within the period of biomass processing and how much the radionuclides concentration are expected in the final product?
- What are the exposures during the cultivation and biomass processing?
- Will the production and alternative crops be economic feasible?
- What are the general perspectives of using the alternative crops on large contaminated areas? [5].

For understanding the behavior of the different radionuclides and their distribution in the main and secondary production and in wastes, it is necessary to know the different radionuclides fluxes. These fluxes depend on the initial level of deposition, on the factors of accumulation in plants which, in turn, depend on the soil properties and plants, accumulators of the radionuclides (for example, wood, rape, sugar-beats, etc.).

The crops used for biofuel (oils, alcohol) such as rape, wheat, sugar-beats, barley, potatoes and rye may be considered suitable alternative crops.

It is also feasible to cultivate on these areas willow for energy production. At regular time interval (3 – 5 years), the willow may be almost totally cut, because this plant grows very easy and in proper irrigation and fertilization conditions may be very often harvested. The willow is also recommended to be cultivated on heavy metals polluted areas. The Salix species is resistant to heavy metal pollution. it can absorb high heavy metals quantities, leading to the rehabilitation of the polluted soil. From this reason, the willow plant is preferred to other species, especially if in the area there is moisture excess.

Mostly, the willow is preferred on clayey soils and the other species on sandy soils. The willow may be harvested during winter time, when the field is covered by snow and the workers are protected against radiations. Furthermore, the willow cultivation does not need too intense works, which is an advantage regarding of workers protection. The willow may be a suitable instrument for the rehabilitation of highly contaminated fields, but the radionuclides concentration absorbed in wood has to be lower than the imposed levels for the wood which is used as fuel.

The following restrictions are widely applied in the USSR and partially in Scandinavia. These restrictions are related to the access in the radioactive contaminated forests and related to the use of the products from these forests. These restrictions have the aim of reducing the human exposure to the radioactive contaminants:

1. Restrictions on the large public and forest workers access as a countermeasure against external exposure.
2. Restrictions on harvesting by the public of forest food products such as game, berries and mushrooms. In many countries, the mushrooms are a common dietary component, and therefore this restriction has been particularly important.
3. Restrictions on collection of firewood by the public in order to prevent the exposures at home and in the garden when the wood is burned and the ash is disposed or used as a fertilizer.
4. Restrictions on hunting activities, with aim of avoiding the consumption of meet with high seasonal levels of radioactive cesium.
5. Fire prevention, especially in areas with large scale radionuclide deposition, in order to avoid the secondary environmental contamination [1, 6, 23].

The annual willow wood production is about 12 t/ha, but it may be higher in proper irrigation and fertilization conditions. During the conversion stage and when a wood with high contamination level (3000 Bq/kg) is burned, the radioactive concentration in the vicinity of ash collectors may exceed the of 1 mSv/a, which is harmful for humans. The contribution of other possible ways of contamination exposure is negligible (external exposure during cultivation, transport, etc.)

The plants for fibers (hemp and flax) may be also considered alternative crops for the agricultural fields with restricted uses.

The metal concentrations in the plant tissues reflect actually, the concentration of the soil in which the plant grows. However, this interrelationship differs from plant to plant, depending on plant tolerance mechanisms and other environmental factors. Mobility and phyto-availability of metals in soils depend on different soil properties such as pH, organic matter content and cation exchange capacity. The uptake also depends on the plant species. The roots of maize plants harvested from a mining area show higher U content than other plant organs (leaf, stem, grain). This may be due to a defense strategy of the plants: immobilization of metals in the roots is less dangerous for plant growth. Levels of uranium in plant ranging between 0.5 and 2 mg/kg are considered to be normal levels for plants. According to the uranium, the „tolerable daily intake" (TDI) is 0,6 µg/kg of body weight per day. The uranium content detected in maize grains (<26 µg/kg DW) allows it to be used to as feed for animals and for flours production [22].

By fencing the vegetation and limiting the access within the larger reclaimed areas the harmful effects on humans and animals are avoided until the new cultivated plants species is capable of maintaining itself under normal management practices. Criteria for establishing if the reclamation efforts are successful need to include following aspects: 1) post-mining vegetation cover and production should be equal with that's of non-contaminated area, 2) the diversity of plant species which have to be adequately supported in the planned post-mining use, and 3) a reclaimed vegetation able to sustain the environmental pressure at a rate equal with that of the surrounding areas [4].

References

[1] Alexakhin A., 2006,"Environmental consequences of the Chernobyl accident and their remediation: Twenty years of experience".Report of the Chernobyl Forum Expert Group Environment". IAEA Vienna.

[2] Barnekov U., Neudert A., Hoepfner U., 2005, „Re-contouring and final covering of Trünzig and Culmitzsch tailings ponds at Wismut". In "Recent developments in uranium exploration, production and environmental issues". IAEA- TECDOC-1463.

[3] Batandjieva B. et all., 2006, Remediation of sites with mixed contamination of radioactive and other hazardous substances". IAEA, Vienna, Technical Reports Series No. 442.

[4] Benes V., Blaise J.R., Falck W.E., Kayukov P., McMurray J.M., Paulka S., Slezak J., Stover D., Waggitt W.P., 2005, "Guidebook on environmental impact assessment for in situ leach mining projects". IAEA -TECDOC-1428.

[5] Blowes A.., 2004,"Remediation of sites with dispersed radioactive contamination". IAEA Technical Reports Series no. 424.

[6] Blum E. H. W., 1990, The challenge of soil protection in Europe". In Environmental Conservatation", 17, 72-74.

[7] Brady C. Nyle and Weil R. Ray, 2008,"The nature and properties of soils". Pearson Education International, Fourteenth Edition, New Jersey 07458.

[8] Busby Chris and Schnug Ewald, 2008, "Advanced biochemical and biophysical aspects of uranium contamination". In "Loads and fate of fertilizer-derived of uranium". Backhuys Publishers, Leiden, Edited by Luit J. De Kok and Ewald Schnug.

[9] Daw H.T. and Dobson R.L. and all., 1967, " Risk evaluation for protection of the public in radiation accidents". IAEA, Vienna, Safety Series no.21.

[10] Daw H.T., Farges L., Pettersson B.G., 1978, "Principles for establishing limits for the release of radioactive materials into the environment". IAEA Vienna, Safety Series No. 45.

[11] Delgado M. Sanchez and Fernandez T. Lopez, 2005,"Uranium mill tailings piles remediation: remedial cleanup actions versus long term stewardship". Proceedings of an International Workshop Lisbon, 11-13 February 2014, „Environmental contamination from uranium production facilities and their remediation". IAEA Vienna.

[12] Ernst W.H.O., 1996, "Bioavailability of heavy metals and decontamination of soils by plants" Appl. Geochem, 11:163-167.

[13] Folding G. and Nemeth G., 2005, „Remediation of tailigs ponds I and II". In "Recent developments in uranium exploration, production and environmental issues". IAEA- TECDOC-1463.

[14] Grabas K. and Koszela J., 2005, „Remediation of the low level radioactive waste tailings pond in Kowary (Poland)". Proceedings of an International Workshop Lisbon, 11-13 February 2014, „Environmental contamination from uranium production facilities and their remediation". IAEA Vienna.

[15] Haan F.A.M., 1993, Soil quality evaluation„. International Postgraduate Course Soil Pollution and Soil Protection", International Training Centre (PHLO) Wageningen, Agricultural University.

[16] Hagen M., Jakubick A.T., Paul M., 2005, Status and results of the wismut environmental remediation project". In Recent developments in uranium exploration, production and environmental issues". IAEA- TECDOC-1463.

[17] Hanecklaus Silvia and Schnug Eswald, 2008, "A critical evaluation of phytoextraction on uranium-contaminated agricultural soils". In "Loads and fate of fertilizer-derived uranium". Backhuys Publishers, Leiden, Edited by Luit J. De Kok and Ewald Schnug.

[18] Henner Pascale, 2008, "Bioaccumulation of radionuclides and induced biological effects in situations of chronic exposure oh ecosystems – an uranium case study". In "Loads and fate of fertilizer-derived uranium". Backhuys Publishers, Leiden, Edited by Luit J. De Kok and Ewald Schnug.

[19] Kuznetsov V. K., Sanzharova N. I., Alexakhin R. M., 2002,"Evaluation of the agronomic effectiveness of rock phosphates from the Polpino deposit in the Russian Federation and their potential to reduce ^{137}Cs accumulation in plants". In "Assessment of the soil phosphorus status and management of phosphatic fertilisers to optimise crop production". IAEA Vienna, February, TECDOC-1272.

[20] Lehr Jay, Hyman Marve, Gass E.Tyler, Seevers William, 2002, „Handbook of complex environmental remediation problems". McGraw-Hill New York.

[21] Logar Z. and Pozun M., 2005, „Remediation of the Zirovski VRH uranium mine". Proceedings of an International Workshop Lisbon, 11-13 February 2014, „Environmental contamination from uranium production facilities and their remediation". IAEA Vienna.

[22] Neves M.O., Matias M.J., Abreu M.M., Magalhaes M.C.F., Basto M.J., 2005, „Abandoned mine site characterization for remediation: the case of the Cunha Baixa Uranium Mine (Viseu, Portugal)". Proceedings of an International Workshop Lisbon, 11-13 February 2014, „Environmental contamination from uranium production facilities and their remediation". IAEA Vienna.

[23] Răuță Cornel și Cârstea Stelian, 1979, "Poluarea și protecția mediului înconjurător". Editura Științifică și Enciclopedică București.

[24] Răuță C., Nastea St., Dumitru M., Bugeag Elena, 1980, "Aspecte privind influența unor emisii industriale asupra acumulării de Zn, Pb, Cu în sol și plante". Analele ICPA, Vol. XLIV.

[25] Răuţă C. şi Cârstea St., 1983, ''Prevenirea şi combaterea poluării solului''. Editura CERES, Bucureşti.

[26] Răuţă C., Dumitru M., Ciobanu C., Toti M., Dulvara M., Gamenţ E., Oşlobeanu A., Bud V., 1994, ''Efecte ale poluării produse de industria metalurgică neferoasă asupra solurilor şi vegetaţiei şi măsuri de refacere a arealelor afectate''. Lucrările CNSS, Tulcea, Bucureşti, nr.28 F, 1-34.

[27] Read D., 2005, „Characterization and risk assessment of sites contaminated by natural series radionuclides". Proceedings of an International Workshop Lisbon, 11-13 February 2014, „Environmental contamination from uranium production facilities and their remediation". IAEA Vienna.

[28] Schroeder H., Heimers A., Frentzel Beyme R., Schott A. and Hoffmann W., 2003,"Chromosome aberration analysis in peripheral lymphocytes of Gulf War and Balcans War veterans". Rad.Prot. Dosim., 103:211-219.

[29] Vegter J., 1993, "Soil protection policy in the EC and ither countries". International Postgraduate Course " "Soil Pollution and Soil Protection", International Training Centre (PHLO) Wageningen, Agricultural University.

[30] ***, Ordinul președintelui CNCAN NR. 56/2004 privind aprobarea Normelor fundamentale pentru gospodărirea în siguranță a deșeurilor radioactive și a combustibilului nuclear uzat. Monitorul Oficial al României, Partea I, nr. 223/28.III.2014.

[31] *** 2014, Lessons learned from environmental remediation programmes, IAEA Nuclear Energy Series No. NW-T-3.6