Criteria and application of remediation methods for territories polluted with heavy metals

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Remediation methods can be evaluated according to the following criteria: clean-up effectiveness, lif survival, groundwater safety (i.e. ecological criteria, they are summed up in the index of ecological effectiveness), remediation cost, duration of remediation process, economical effect (these criteria are summed up in the index of conditional effectiveness). The index of ecological effectiveness calculated according to clean-up effectiveness, life survival and groundwater safety is more important than the index of conditional effectiveness, because soil remediation from heavy metals is first of all an ecological issue. These indices were calculated by nine remediation methods: phytoextraction, electrokinetic method, chemical immobilization, gravimetric concentration, soil removal & scatter, soil removal & heaping, afforestation with HM – proof tree species, remediation with sorbents, and an intrinsic method. According to the index of ecological effectiveness, most acceptable are the following method: soil removal & scatter, phytoextraction, and gravimetric concentration. In respect of conditional economical effectiveness, the first are electrokinetic and phytoekstraction methods. Therefore, considering the necessity to give a priority to ecological criteria, phytoextraction should be regarded as most acceptable, because it stands in the beginning of the list according to both indeces.

Key words: soil remediation methods, contamination with heavy metals, criteria for evaluation

INTRODUCTION

Pollution is one of the most topical environmental problems. It is especially urgent in Lithuania, where hundreds of thousands of hectars are contaminated with oil products and heavy metals (HM). It is the heritage of Soviet military stations, polluted territories of oil depots, places of large accidental spills, overfilled and uncared-for dumps. A certain threat to the environment and population health comes from abandoned Soviet industrial sewerage filtration fields extremely contaminated with HM. The other problem is connected with the storage of HM-polluted sludge. Recommendations to use it for fertilizing the farmlands is not ecologically secure. Often plants, even growing in highly polluted land show no marks of HM accumulation, because they can eliminate HM excess by transpiration. Evaluation of plant HM accumulation capacity is complicated and requires reliable research methods, very precise analysis, therefore, hasty conclusions and recommendations can result in damage to the environment and human health.

For remediation of contaminated territories scientific grounding is essential, because each territory to be recultivated is characterized by an individual environment (geographical position, contamination nature and level, pH and Eh, certain soil features, etc.). The main criteria for choosing the methods are ecological effect, remediation cost, possible economical effect.

For the analysis of the most world-famous remediation methods, the Internet information and literature sources were used. Every method was dealt with distinguishing its advantages and faults. Besides, preliminary cost calculations for the remediation of HM-polluted territory was performed for each method (the case of Molainiai filtration fields was taken).

All the methods of soil remediation from HM can be classified into the following groups: bioremediation, electroremediation, chemical, physical, physico-chemical and intrinsic. Electromediation method, physico-chemical in its nature, is excluded because of its technological peculiarity. The following chapted gives a short description of the chosen methods.

REVIEW OF REMEDIATION METHODS

From all the above-mentioned classes of remediation methods the following methods were chosen for

the evaluation by different criteria: phytoextraction, electrokinetic, chemical immobilization, gravimetric contentration, soil removal & scatter, soil removal & heaping, afforestation with HM-proof tree species, sorbent application, and intrinsic. These methods belong to different groups, they were selected from a great variety of the worldwide used methods as most effective for Lithuanian ecological and economical conditions.

Phytoextraction is based on the features of some species of plants (called hyperaccumulators) to accumulate metals in their tissues. It is only one of numerous bioremediation methods [1, 2, 5, 7, 10, 11]. The electrokinetic method is based on the feature of metallic ions to move in electrolytic solution affected by electric field [9, 12, 16]. The method of chemical immobilization, mentioned in this presentation, is not classical chemical, it is the Geo-Remediation™ technology (called accelerated metallo-organic mineralization) offered by one US company [4]. Most chemical methods are very unfriendly to soil. The Geo-Remediation™ method is not described as one whose impact on soil life could be seen clearly, but it seems to be very effective, because metals are chemically converted into their insoluble minerals bound into the alumino-silicate portion of the soil or sediment and immobilised. The method of gravimetric concentration (part of TerraWash™ technology) is a physical method offered by another company. The principle of this method is related to the gravimetric separation of the heaviest soil fraction which contains HM. This causes a slight loss of soil volume [6]. The territory afforestation with trees that can survive under high HM concentrations is one of the most simplest and cheapest methods. Application of mineral sorbents is based on the ability of sorbents to enclose metals in their molecular grating [3, 8, 13]. The intrinsic approach rest upon the premise that nature knows better how to treat the polluted land, it is allowed to take its course with regard to the fate of contaminants and vegetation [15]. The rest two methods – soil removal & scatter [14] or soil removal & heaping (together with isolation of contaminated soil by geomembrane) – do not require detailed presentation.

The following chapter is dedicated to the criteria for choosing the remediation method most convenient in all respects.

CRITERIA FOR APPLICATION OF REMEDIATION METHODS

The main criteria for selecting the method are as follows: remediation cost, duration of remediation process, clean-up effectiveness (result), ecological effect, economical effect. All the methods described in the previous chapter were evaluated according to these criteria (Table).

In order to select the most advantageous remediation method, each of them should be evaluated in line of different criteria and then the final score should be calculated in complex indices. For that purpose, more criteria were added to the previously mentioned. Ecological criteria could be as follows: clean-up effectiveness (result), life survival, groundwater safety. They could be summed up in the index of ecological effectiveness. Cost, duration and economical effectiveness are the criteria that could be summarized in the index of conditional economical effectiveness. For ecological criteria an evaluation system of 10 scores was applied. The cost was calculated using as an example the situation of the Molainiai sewerage filtration fields. The same area was taken for calculation of remediation process duration, which is given in years. More complicated was evaluation of the economical gain of the method itself. The calculation of economical gain that the recultivated land could bring in future was not performed, because it must be the same no matter what method was applied, and, consequently, it does not help in comparing the methods. Therefore, the economical effect was evaluated just for the remediation process and was graded into three cathegories: positive (the remediation project pays dividends), negative (economical effect is formed only by expenses for the project), neutral (expenses buv off partly). Remediation methods according to these criteria and indices are characterized in Table 2.

Compared by clean-up effectiveness most methods are acceptable, even the soil removal & heaping (when the contaminated soil is stored in an artificial hill, isolated from the environment by a geomembrane, nevertheless, it keeps the former threat to the environment). Less acceptable methods are afforestation and intrinsic (the clean-up is very slow, the danger to surroundings remains). The electrokinetic method and sorbent application also got less marks, because they still lack exhaustive research on their applicability in soil remediation from HM.

A very important criterion is the method's friendliness to soil life; it was called life survival. In line of this criterion, the most sterilizing and evaluated by the least score are electrokinetics, gravimetric concentration and chemical immobilization. While applying these methods the environment becomes inimical to soil organisms. The intrinsic method got a low score as well, because HM left in the soil endangers some species of organisms. Life in an ecosystem is ensured by HM-imbiding sorbents and HM-proof tree forest, but the highest score is given to phytoextraction and soil removal & scatter.

Table 1. **Comparison of remediation methods (cost and duration calculated for Molainiai sewerage outlet filtration fields (about 100 ha)**

No.	Methods	Cost	Duration	Result	Ecological effect	Economical effect
1	*2*	*3*	*4*	*5*	*6*	*7*
1.	Phytoextraction	Depends on: cost of plant seeds. cost of sowing and harvesting, costs of biomass storage or utilisation, costs for monitoring, under 1 mln. litas (Lt)	Shortest – 5 years for different HM duration depends on: 1. different HM concentrations 2. ability of plants to absorb different HM Duration can be unequal in different parts of area because of: 1. unequal territory contamination 2. unequal thickness of polluted layer	HM concentration is reduced to acceptable extent	Soil remains viable	Expenses could be reduced if the metals in biomass were recycled
2.	Electrokinetic method	Cost of electric energy up to 3.5 mln. Lt. Some extra cost must be added for metal excavation and transportation	Not clear, could be about one warm season (6 months).	HM concentration in the soil reduced by 95%	Unknown is the long-term influence of electric field on soil organisms; probably negative	Expenses could be reduced by recycling of extracted metals
3.	Chemical immobilization	According to Geo-Remediation™ technology, 50 USD for 1 m^3 or about 120 mln. Lt for all the area	2,5 years if company works round year, 5–6 years if excluded cold season.	All the contaminants are converted and bound	Unclear influence of this technology on soil organisms	Negative (only remediation expenses)
4.	Gravimetric concentraion	Accordint to Terra Wash™ technology, 13 USD for 1 m^3 or about 22 mln. Lt for the whole area	Depending on capacity equipment and work schedule, from 0.5 to 4.5 years for all the area	HM are removed from the soil which loses 1/500 of its volume	Unclear influence of this technology on soil organisms	Negative (only remediation expenses)
5.	Soil removal & scatter	At least 3 mln. Lt	Depends on labour productivity (technology, transport, number of workers): 1 to 3 years	HM concentration reduced to acceptable level by scattering soil	Dozed land stays without soil (needs recultivation); filled area gets a dose of HM and organics	Negative (expenses for dozing and scattering)

Table 1 (continued)

1	2	3	4	5	6	7
6.	Soil removal & heaping	For dozing, filling, isolating with geomembrane, 1,5 to 2 mln. Lt	Depends on labour productivity (technology, transport, number of workers): 0,5 to 2 years	Soil with HM is in a concentrated and isolated heap	Dozed land stays without soil (needs recultivation: filling with new unpolluted soil)	Negative (expenses for project realization)
7.	Afforestation with HM-proof tree species	120 to 180 thousand Lt	To reach the maturity the least of 20 to 30 years required	HM stay in the soil and accumulate in perennial plants	Risk to some species of organisms and especially ground water remains	Negative (expenses for afforestation)
8.	Remediation with sorbents	About 270 mln. Lt, using SGN sorbent	Several weeks to several months Questionable is the effect lasting	HM remain in the soil, but immobilized in sorbent	Soil becomes more viable, because toxic substances are removed	Negative (sorbent and its insertion costs)
9.	Intrinsic method (approach)	Expenses for monitoring	Depends on when a certain state of the territory is taken as suitable for a certain activity, that ignores pollution	HM stay in the soil and accumulate in perennial plants	A seeming equilibrium between contaminants and ecosystem is reached	If any economic activity is allowed, it can outweigh the monitoring expenses

The other ecological criterion is groundwater safety during the process of remediation. The safest methods in that respect are gavimetric concentration and removed soil scatter. Also rather effective methods are phytoextraction, sorbents and chemical immobilization. The situation is most complicated with the electrokinetic method and natural methods: afforestation and intrinsic (the natural water circle remains unchanged and the HM flow down to groundwater is not stopped). Conserved inside the geomembrane (in case of soil removal & heaping), contaminated soil keeps the risk to the groundwater, therefore it is evaluated meanly.

It is possible to evaluate the methods according to all the three ecological criteria by just summing up the scores. The method that has gained the highest amount of scores is most ecologically effective. Of course, it is a rather formal evaluation, but anyway it allows to compare the methods. Table 2 shows not only evaluation according to each previously mentioned criterion, but also according to a complex criterion – ecological effectiveness. Obviously, the highest scores belong to soil removal & scattering method (29), phytoextraction (27), and gravimetric concentration. Ecologically effective are methods of chemical immobilization and sorbent insertion (23 for each). Most dangerous ecologically are the intrinsic (7 scores), afforestation with HM-proof plants (10 scores) and electrokinetic (13 scores) methods.

Remediation methods must be evaluated in economic respect, which is especially important in our contemporary society. The most important indicator is the price, *i. e.* remediation cost. However, the economic effect of the very remediation process also can be considered, e. g., the recycling of extracted HM. In addition, the duration of the remediation process is also important for economy.

The lowest costs are required for realization of natural methods such as like intrinsic, afforestation and phytoextraction (Table 2). Most expensive are physicochemical methods such as sorbent insertion or chemical immobilization. The application of these methods in Lithuania is hardly possible. There are some methods of the medium cost: electrokinetic or "soil-kinetic" – soil removal & scatter or heaping.

As to remediation terms, the longest are, of course, the natural processes such as like intrinsic, or afforestation. Even phytoextraction is as long as 5 years at least (estimated for the Molainiai filtration fields). The other methods require from several months (sorbents, electrokinetic) to several years (Table 2).

Only three methods can be ("can be" does not mean "are") economically effective: phytoextraction, electrokinetic, and intrinsic. However, it is hard to

Table 2. **Evaluation of remediation methods by different criteria**

Method	No.	Result	Life survival	Groundwater safety	Ecological effect	Price	Duration	Economical effect	Conditional economical effectiveness
Phytoextraction	1	10	10	7	27	1	5	5	10.00
Electrokinetic	2	9	3	1	13	3,5	0,5	5	28.57
Chem.immobilisation	3	10	5	8	23	120	4	1	0.02
Gravimetric concentration	4	10	4	10	24	22	1	1	0.45
Soil removal and scatter	5	10	9	10	29	4	2	1	1.25
Soil removal and heaping	6	5	7	5	17	3	1	1	3.33
HM-proof afforestation	7	3	6	1	10	0,2	30	1	1.67
Sorbents	8	8	8	7	23	270	0,1	1	0.37
Intrinsic	9	1	5	1	7	0,1	1000	10	1.00
Units:		Scores (1 to 10)	Scores (1 to 10)	Scores (1 to 10)		Conditional	Years	Positive –10, Negative – 1, Neutral – 5	

speak about real profit, because it is unknown (lack of research data) whether metal output from hyperaccumulating plants will redeem the expenses for purchase of plant, sowing (or planting), handling of territory, harvesting, etc. The electrokinetic method can be even less profitable due to large expenses for electric energy. The only method that formally can bring some economical benefit is intrinsic, if a profitable function could be gained from a contaminated land (*e. g*., cultivation of quick circulation plants). But then this can be hardly called a remediation method at all. The rest methods are investment in the benefit of cleaned-up land. Indeed, there is no lack of clean land suitable for agriculture, forestry or other function in Lithuania, therefore, there is neither urgent social nor economical demand to invest in remediation of polluted spots. It will be possible if the priority is given not to economical criteria and ephemeral social needs, but to the ecological stability of the country, which in perspective would guarantee both social and economical welfare.

In this article an attemp was made to evaluate and compare the remediation methods suppositionally according to the complex index – conditional economical effectiveness, which was calculated using the following formula:

$$E_C = E \cdot 10 / (P \cdot D),$$

where E_C is the conditional economical effectiveness, E is a given suppositional value of economical effectiveness, P is price, D is duration; "10" is a coefficient to magnify the value for a more covenient expression.

As shown in Table 2, the highest values of this index belong to the electrokinetic and phytoextraction methods. Worth noting is the soil removing and heaping method. The other methods in terms of price, duration, and economy are less acceptable.

To summarise, from the whole variety of remediation methods, two are most suitale in Lithuania: phytoextraction (ecologically reliable) and electrokinetic (economically effective). Depending on the priorities, the method should be chosen: if ecological principles prevail against economical, then it is better to apply the phytoextraction method and *vice versa*. Of course, it cannot be a cathegoric proposal, because the calculations that were taken as a basis for the evaluation are theoretic and in reality these remediation methods can show themselves differently, however, the main features are as described above.

CONCLUSIONS

1. Remediation methods are unequal in respect of different criteria. Some of them are ecologically ineffective, but cheap. On the other hand, being cheap, they can take unequal time for realisation. Other methods can be ecologically friendly, but expensive, etc. This variety can be analysed evaluating remediation methods according to the following criteria: clean-up effectiveness, life survival, groundwater safety (ecological criteria, they are generalised by the index of ecological effectiveness), price, duration, economical effectiveness (these are criteria generalised by the index of conditional economical effectiveness).

2. From all the criteria, most important, besides the clean-up effectiveness (remediation result), are ecological criteria, according to which the possibility for life to survive and the safety of groundwater from pollution is evaluated. The index of ecological effectiveness calculated regarding the mentioned criteria is more weighty and important than the index of conditional economical effectiveness, because terri-

tory remediation from HM is first of all an environmental, ecological problem.

3. According to the index of ecological effectiveness, most acceptable are the following methods: soil removal & scatter, phytoextraction, and gravimetric concentration. In respect of conditional economical effectiveness, the first place belongs to electrokinetic and phytoekstraction methods. Therefore, considering the necessity to give a priority to ecological criteria, phytoextraction should be regarded as most acceptable, even ignoring its economical efficiency which is not the highest.

Received
15 April 2002

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SUNKIAISIAIS METALAIS UŽTERŠTŲ TERITORIJŲ REMEDIACIJOS METODŲ KRITERIJAI IR TAIKYMAS

S a n t r a u k a

Remediacijos metodus galima įvertinti pagal šiuos kriterijus: išvalymo efektyvumą, gyvybės išlikimą, gruntinių vandenų saugumą (tai ekologiniai kriterijai, kurie apibendrinami ekologinio efektyvumo rodikliu), kainą, valymo trukmę, ekonominį efektyvumą (tai santykinio ekonominio efektyvumo rodikliu apibūdinami kriterijai). Ekologinio efektyvumo rodiklis, apskaičiuotas pagal išvalymo efektyvumą, gyvybės išlikimo galimybes ir gruntinių vandenų saugumą, yra svarbesnis už santykinio ekonominio efektyvumo rodiklį, nes dirvožemių valymas nuo sunkiųjų metalų – visų pirma ekologinė problema. Šie rodikliai buvo ekspertiškai apskaičiuoti devyniems remediacijos metodams: fitoekstrakcijai, elektrokinetiniam metodui, cheminiam surišimui, gravimetriniam koncentravimui, dirvožemio nukasimui ir išbarstymui, dirvožemio nukasimui ir supylimui į izoliuotą krūvą, apsodinimui sunkiesiems metalams atspariomis medžių rūšimis, remediacijai sorbentais bei intrinsiniam metodui. Pagal ekologinio efektyvumo kriterijų patys parankiausi yra šie metodai: dirvožemio nukasimas ir išbarstymas, fitoekstrakcija ir gravimetrinis koncentravimas. Pagal santykinį ekonominį efektyvumą svarbesni elektrokinetinis ir fitoekstrakcinis metodai. Todėl, atsižvelgiant į būtinybę suteikti pirmumą ekologiniam kriterijui, tinkamiausiu remediacijos metodu reikia laikyti fitoekstrakcinį metodą, kuris yra vienas pirmųjų pagal ekologinius ir ekonominius kriterijus.