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Soil remediation comparative analysis on two historically contaminated industrial sites in Romania

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Abstract. In the period after the Second World War the industry has known a continuous development and territorial expansion, quite essential for the restoration, increase and improvement of life standards and for the stimulation of the economic activity in all fields and all over Europe. Due to these activities, currently several industrial sites affected by historical contamination were inherited. One of the important aspects regarding these sites is represented by the remediation step and their reinsertion in the urban circuit, most of them being located nearby or in some cases inside cities. Nonetheless, based on the fact that a large number of methods, techniques and technologies for the remediation of contaminated sites are known and applied, and taking into account the principle that their treatment is done by reducing the risks while applying the best available techniques at feasible costs, the paper presents both technical aspects, the differences and particularities of the developed actions for the two case studies as well as aspects regarding the cost/benefit analysis. The paper proposes a comparative analysis regarding the development of remedial actions necessary for two former industrial sites on which the presence of contamination was confirmed, in different time periods, as follows: a) the remedial works for the first analysed site were developed between 2007-2008, before a specific legislation for contaminated sites field was implemented, respectively the legislative regulations Governmental Decision 1408/2007 and Governmental Decision 1403 /2007; b) the remedial works for the second analysed site will be developed between October-November 2012, as a result of geological environment investigations for the assessment of contamination in accordance with the provisions of Governmental Decision 1408/2007; the remedial methods were determined based on the analysis of conclusions in the previously mentioned study, respecting the provisions in Governmental Decision 1403/2007.
Key Words: contaminated site, remediation, soil monitoring, comparative analysis.

Introduction. Land contamination is a serious environmental and developmental problem in many countries, including developing countries as Romania. If managed well, some contaminated sites can be an opportunity for urban renewal and development. Conversely, if brownfields are not used due to legal concerns or lack of financial resources, or not properly remediated, they can present a serious threat to public health and the environment and become a barrier to local economic development (Bardos 2004; World Bank 2010). Contamination problems can stem from historical activities dating back hundreds of years, such as spoil heaps from Roman lead mines. The larger-scale problems started with the acceleration of processing, manufacturing and waste-disposal activities associated with the industrial revolution at the end of the 18th century (Siller et al 2004).

Managing land affected by contamination involves identifying any unacceptable risks posed by the presence of contaminants, then acting to reduce and control those risks to an acceptable level so that the land is “suitable for use” (Ozunu et al 2009). Risks presented by any given level of contamination are assessed for each site, bearing in mind the relevant site-specific factors. These might include proposed and current land use, proximity to groundwater or surface water or receptors in adjacent dwellings or habitats (Darmendrail & Muller 2011).

There are many different technical approaches to remediating land affected by contamination. They can be categorized as either civil engineering or process-based technologies. Each of the different technologies may be capable of treating either a wide range of contaminants, or specific ones, so the remedial strategy needs to be carefully selected on a site-specific basis. Historically, the most common form of remediation has been to remove the contaminated soil and dispose of it at a licensed landfill site (ICCL 2011; FRTR 2011). A number of process-based remediation technologies to treat contaminated soils and groundwater are commercially available worldwide, including Romania. These include bioremediation, air sparging, soil vapour extraction, soil washing, thermal treatment, permeable reactive barriers and other such. This is a developing market in developing countries as Romania. The new process-based technologies offer a more sustainable way of dealing with contaminants in soils (Khan et al 2004; US Sustainable Remediation Forum 2009).

Many owners of land affected by contamination have initiated work to investigate and undertake remediation on a voluntary basis. Incentives for voluntary remediation include the desire to increase the value of the land, perhaps with a view to selling it, and the removal of potential liabilities from the company ledger. At international level is encouraged the beneficial re-use of previously-developed land, and more specifically the remediation of land affected by contamination (World Bank 2010; Darmendrail & Muller 2011).

Purpose and objectives. This paper proposes a comparative study on the progress of remedial actions required on two former industrial sites, on which was confirmed the existence of contamination in different time periods, as follows:

a) remediation works on the first site were performed during 2007-2008, before the implementation of specific legislation in the field of contaminated sites, namely Governmental Decision (GD) 1408/2007 and GD 1403/2007;

b) remediation works to the second site will be conducted during October-December 2012, as a result of geological environmental investigations based on the provisions of GD 1408/2007, and GD 1403/2007.

The objectives of paper are:

- remediation process steps presentation and implementation for each industrial site historically contaminated;
- emphasizing the technical and economic aspects resulted from the comparison of the two remediation methods.

Methodology. In this paper are analysed and compared two industrial sites in Romania historically contaminated with total petroleum hydrocarbons (TPH) and heavy metals for which remedial works were required by applying different methods/technologies, namely:

- bioremediation in case of Bucharest site;
- stabilization technology for Brasov site.

The following criteria were considered in this paper to compare the performance of remedial actions necessary for the two industrial sites:

- general characterization of sites, including their past activities;
- analysis of pollution level for each site, based on environmental studies conducted previously, their conclusions, adjacent investigations, the nature of the identified pollutants and delineated areas of land identified as contaminated, remediation volumes etc.;
- analysis and monitoring of remedial works.

Characterization of the analysed sites

Bucharest Site. The Bucharest site is located in the North-West side of Bucharest and has an area of 6.1 hectares of which approximately 10% is open space, the rest being occupied by buildings and transportation routes.

Topographically speaking, the location and neighbourhoods are located in a flat area at an altitude of approximately 87 m. The nearest surface water is Herastrau Lake, located approximately 1.5 km North-West of the site (Figure 1).

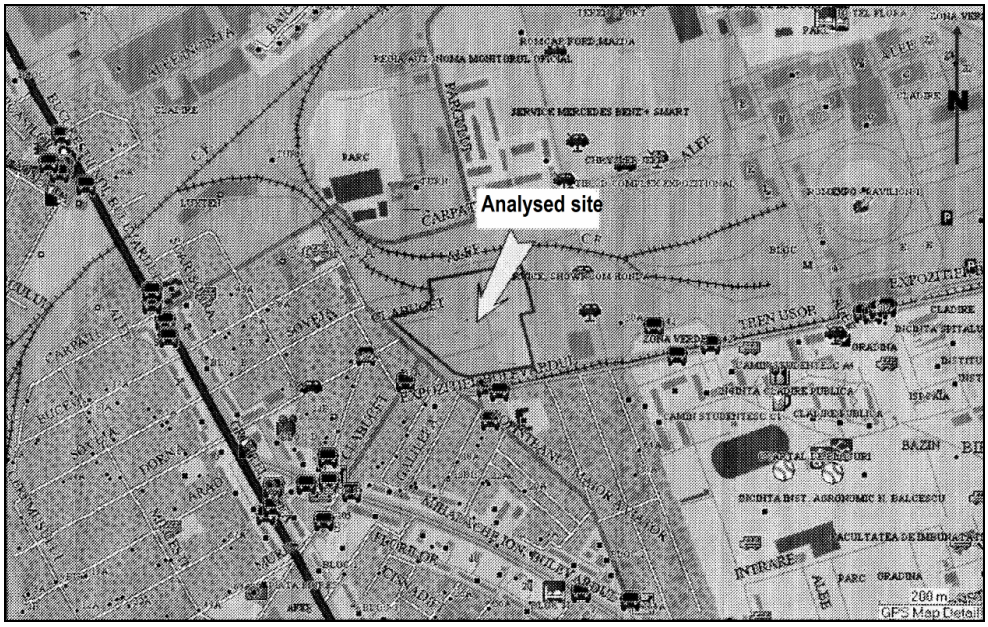

Figure 1. Areal location of Bucharest site.

Between 1940-2008, the site specific activities were auto industry - construction and repair of heavy vehicles. In December 2008 the company closed down all the work on site, the new owner of the land intending to change land use from industrial to residential.

According to the information and data from former studies, lithological strata sequence can be summarized as follows:

- concrete layer and/or fillers of up to 2 m thick (except for locations where infill layer has a thickness of 5 m);
- layers of clays and clay powder (plastic clay with rare Varta brown limestone, clay brown-black, yellowish grey clay with dusty limestone, hard plastic brown clay, brown clay dust) with thickness up to 7 m;
- alternating sands and gravels with clay area, arranged in layers with thickness of 1 to 4 m in layers of sand, gravel medium and large confined groundwater free level.

The first groundwater aquifer is confined to depths between approximately 8.5 and 10 m, varying according to rainfall. Since the location is near Herastrau Lake, it is estimated that the predominant groundwater flow is towards the NE of the lake.

Surfaces on which were executed remedial works were free of construction (buildings were demolished) and there were only the foundations.

Braşov Site. The site is located inside Brasov City, in the NE part of it and has an area of 120 ha. Topographically speaking, the location and neighbourhoods are located on a flat lowland with an altitude of approx. 560 m; the nearest surface water is Timis river, located immediately adjacent to the Eastern boundary of the site (Figure 2).

Industrial processes on Brasov site date since 1925 when it was started the production of aircrafts. After 1945 and until 2007 the work focused on the production of agricultural tractors. In 2007 production ended all activity on the site following the sale and purchase agreement, and the land use was planned for commercial, residential purposes.

Surfaces on which remedial works will be executed are free of constructions and covered 60-70% by concrete platforms. From geological point of view, the site is located in the Braşov Depression, the largest mountain depression of tectonic origin inside Carpathians in Romania, formed at the end of Pliocene and Quaternary age in the crash

early tectonic plates that are related to the Eastern Carpathians in the immediate vicinity (Străinescu 2010). Soil structure has been investigated in depth by drilling and excavations carried out by a specialized company. It was generally observed that for the depth of 0-1.5 m the soil structure consists of fillers and brown-grey sandy clay, plastic consistency, gravel and boulders. The first aquifer is confined groundwater at depths ranging from approximately 15 to 20 m, varying according to rainfall. Given that the site is near the Timis River, groundwater flow is from SW to NW.

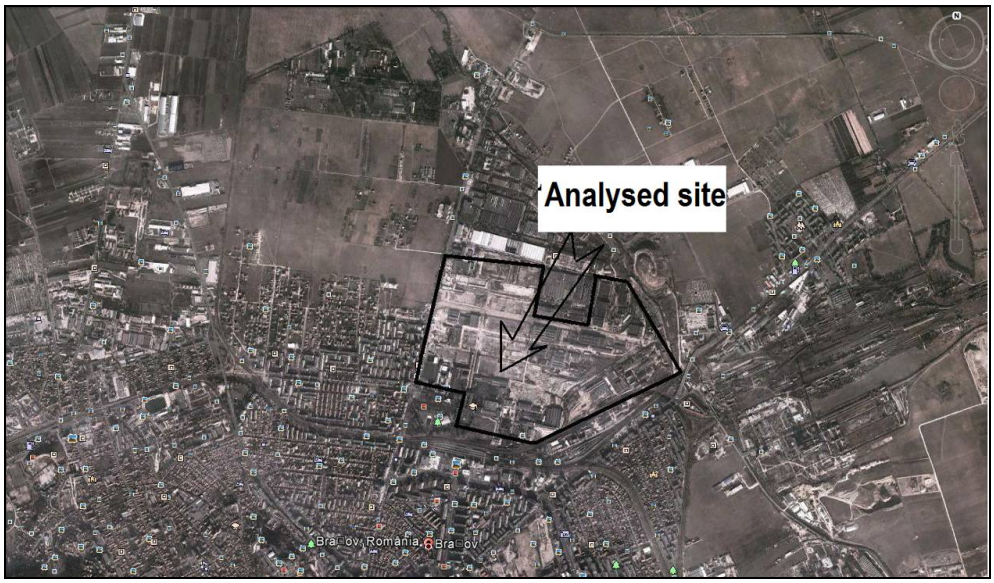

Figure 2. Areal location of Braşov site.

Pollution level

Bucharest Site. Soil investigations on the site were conducted in 2007, for the entire surface of 6.1 ha, and consisted in the construction of 13 boreholes to a depth of 10 m and the collection of 50 soil samples. For the samples were performed chemical analyses, determining heavy metals concentrations (cadmium, copper, manganese, nickel, lead, zinc) and TPH.

These investigations revealed concentrations above the intervention threshold values for sensitive uses in case of TPH and heavy metals (cadmium, lead, zinc) in some areas of the site, leading to the conclusion that remediation works are needed.

Previous studies show the following data regarding contamination aspects:

- contaminated surfaces: 2500 m^2 area A; 1500 m^2 area B;
- total volume of contaminated soil: 8000 m^3;
- maximum depth of contamination: 1 m in area A, 3.5 m in area B.

Taking into account the fact that between 2007-2009 on the site have been developed several decommissioning/demolition works, thus appeared the risk of accidental contamination, and thereby it was considered necessary to undertake further assessment of soil quality on the site, on the one hand to confirm the results of previous environmental studies and, secondly, to show any new potentially contaminated surfaces.

Thus, repeated sampling on the shallow surface (1m) was possible to make a general characterization of soil on the entire site of 6.1 ha, valid at the start of decontamination work. These included:

- 20 soil sampling points at 0.3 m and 1 m deep for general characterization of the site;
- 10 soil sampling points at 0.1 m and 1 m deep for the characterization of Zone A;
- 10 soil sampling points at 1 m deep for the characterization of Zone B.

The laboratory activities for the bioremediation process included the following works:

- analysis of parameters - pH, TPH, metals (cadmium, lead, zinc, copper, manganese, nickel) in relation to threshold values for sensitive land uses presented in the Decree no. 756/1997 (OG 1997) to determine the nature and establish more accurate hydrocarbon contaminated zones: position, area, depth (Table 1);
- timely analysis of leachate samples to determine the appropriate treatment variant and steps necessary for bioremediation: + wetting aeration stabilization/solidification with OXIZIN (frequency ventilation + number of applications OXIZIN/series).

Table 1

Threshold values for sensitive land use -Decree no. 756/1997

No.	*Parameter*	*Threshold values for sensitive land use in the Decree no. 756/1997; mg/kg dry substance*	
		Alert	*Intervention*
1	pH	-	-
2	TPH	200	500
3	Cadmium	3	5
4	Lead	50	100
5	Zinc	300	600
6	Copper	100	200
7	Manganese	1500	2500
8	Nickel	75	150

Depending on each sample location were selected for analysis only the most representative indicators in relation to potential sources of contamination.

The results of soil analysis confirm overcoming of the intervention thresholds for sensitive uses in case of petroleum products and heavy metals from areas A and B, while highlighting the existence of two zones, marked C and D in excess of intervention levels for sensitive uses petroleum products and heavy metals, requiring bioremediation works (Figure 3).

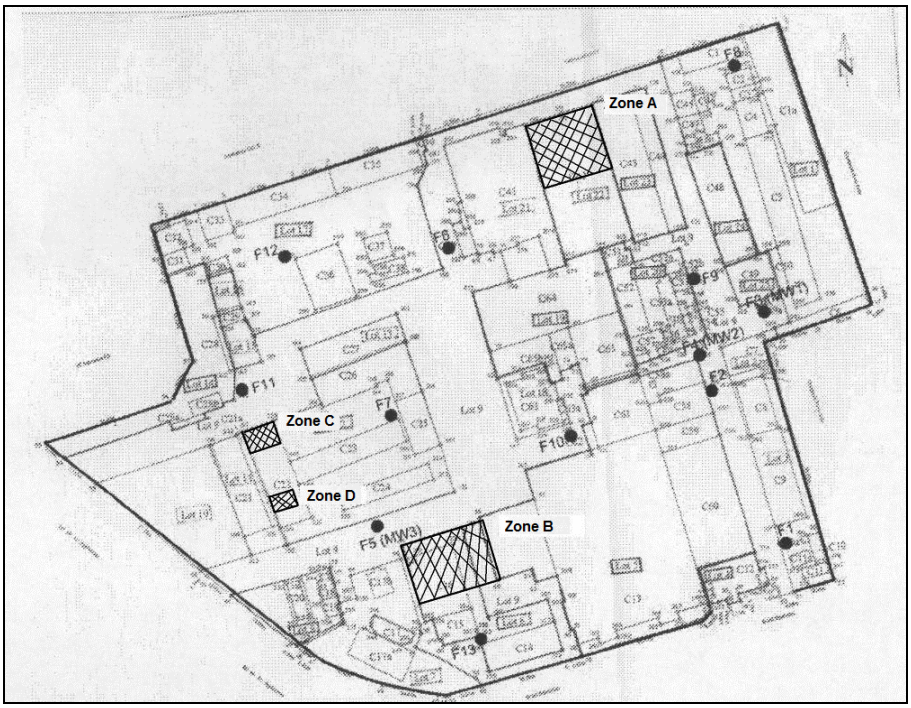

Figure 3. Contaminated land areas on which remedial measures are necessary- Bucharest Site.

During further investigations made in 2008-2009 were identified contaminated areas (A, B, C, D) where pollutants exceeded alert thresholds and intervention values for sensitive uses, respectively:

- total contaminated area: approx. 4250 m^2 which represents 6.9% of the total area of the site;
- total volume of contaminated soil: approx. 6850 m;
- maximum depth of contamination: 1 m.

Braşov Site. Over the years, several studies have been conducted on the premises of Brasov site. Between 2010-2012, on the site were carried out investigations and assessment of soil and subsoil pollution according to the provisions of GD 1408/2007.

Investigation and assessment phase was conducted in 2010 for the entire area of the site, or approx. 120 ha. Investigative works together with preliminary risk assessment results identified and delineated on Brasov site, two areas with high concentrations of contaminants associated with unacceptable risk located in upcoming residential area, with a total area of approx. 15,570 m^2, representing 12% of the total area of the site.

Detailed investigation and evaluation phase was conducted in 2012 for the two land areas over the surface of approx. 15,570 m^2. Mandatory minimum package of methods provided by the current legislation were used as follows: geological investigations, geophysical, geological investigation and geochemical investigation (OG 2007a). It was adopted a systematic sampling methods in soil samples of 20 x 20 m grid. Pollutants identified in the studied areas are: TPH, Pb, Zn, Cu, Ni, Cd, Mn. These pollutants exceed established alert and intervention threshold values for sensitive uses.

Two zones (I and II), each with two plots (A and B), presented levels of contamination and risk assessment determined that remediation measures are required (Figure 4), respectively:

- contaminated surface: approx. 3912 m^2 which represents 3.2% of the total area of the site (of which 2500 square meters in depth profile from 0.5 to 1.5 m;
- total volume of contaminated soil: 5282 m;
- maximum depth of contamination: 1.5 m.

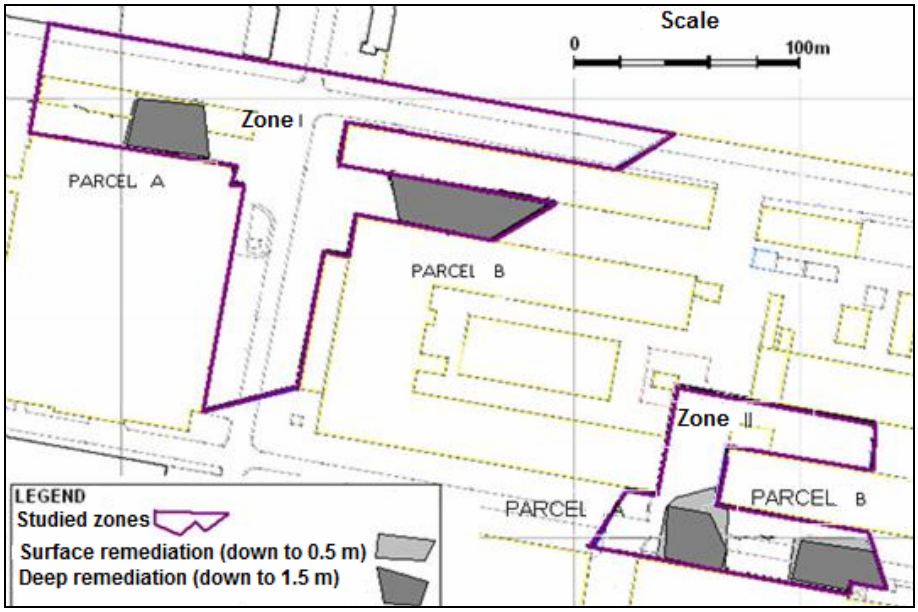

Figure 4. Contaminated land areas on which remedial measures are considered necessary - Braşov site.

Remediation and monitoring activities

Bucharest Site. For the industrial site on the Bucharest location the proposed solution was to build a platform for bioremediation of contaminated soil to treat it to achieve

acceptance criteria for disposal according to legislative regulations. In order to reduce hydrocarbon and heavy metals content, the bioremediation method chosen is based on excavating contaminated soil, aerating it with a machine and sprinkling water for wetting the material deposited on the platform. The major advantage of this technology is that it presents good results in cleaning low levels of hydrocarbons. The disadvantage is the relatively long duration of the process, high costs associated with the process.

Work carried out on site to decontaminate polluted soil and identifying trace chemicals in soil assumed several stages, as follows:

- approval of the site operations by authorities;
- development of the bioremediation platform area of 1400 m^2;
- fast bioremediation process (excavation, transport, bioremediation, disposal);
- bioremediation process monitoring (monitoring excavation limits, monitoring bioremediation);
- removal of treated soil to a hazardous waste landfill.

Bioremediation works were made during 18 months through successive excavation on several areas (areas A, B, C, D), which were identified to exceed the intervention thresholds for the analysed indicators.

After conducting excavations to verify the quality of soil remaining on site, there have been taken samples from the bottom and walls of each excavated area. If laboratory results still indicated the presence of contaminants in the soil above the intervention thresholds, further excavations to identify pollution were done, until the intervention thresholds for soils, with sensitive use, were not exceeded.

Overview of the main landmarks of bioremediation:

- total volume of excavated soil + platform: 6850 m^3;
- total volume of soil bioremediation platform OXIZIN intake (natural catalyst degradation rapid biological organic matter): 5500 m^3;
- total volume of soil bio-remediated on the platform exclusively by aeration: 1350 m^3;
- bioremediation serial number: 5;
- bio-cell dimensions: 70m x 14m x 1.5m;
- the reversal frequency of each bio-cell aeration: a rollover/2 days;
- number of applications with OXIZIN: 2/series (series 1, 2), 1/series (series (3, 4), 0/series (Series 5);
- during bioremediation / range: 3 to 10 weeks.

Results for composite samples show that at the end of the bioremediation process both heavy metals concentrations and the TPH were lower than the limits for inert waste. Upon reaching the limits of acceptability in accordance with the provisions of the Decree 95/2005, treated land was discharged on the platform and temporarily stored in the vicinity, from where it was gradually evacuated to a non-hazardous landfill.

Braşov Site. For Braşov site the in situ Solidification/Stabilization method with cement additives has been chosen, using a device for contaminated soil mixing cement with additives where encapsulation occurs in situ to stabilize pollutants in the cement matrix, to treat it, and also to establish the category of waste and achieve acceptance criteria for storage.

The major advantage of this technology is that the additives used are relatively cheap and available, not considering the special logistics issues (productivity, cost acceptable). The downside is that technology cannot be applied to high soil contamination.

Contaminated soil remediation operations will be performed by inverting technology by a specialized company and consists of mixing until smooth vitrified soil contaminated with inset range in proportions that can vary between 3-15 % depending on humidity and concentration pollutants in soil.

During the inversion process, contaminants are chemically bonded and mechanically embedded in the matrix that do not allow their migration, so treated soils can be classified, following leaching tests under the provisions of the Decree 95/2005 (OG 2005), storage in landfills as a coating material with the following features:

- chloride as 15000 mg/kg;
- sulphides below 20,000 mg/kg;
- total dissolved solids (TDS) in the 60,000 mg/kg DM;
- dissolved organic carbon (DOC) in the 800 mg/kg DM; heavy metals as hazardous waste code limits.

Treatment will be performed in situ on a concrete platform near the areas identified as contaminated of approx. 1930 m^2.

The monitoring plan for pollutant concentrations in the soil will involve the following steps:

- collection of samples and laboratory testing, where appropriate, on decontaminated areas;
- collection of samples and laboratory tests of soils initially excavated. The operation will be performed to determine the initial state of contamination but also for a better delineation of areas with high concentrations of pollutants and likely not lend itself to treatment by stabilization;
- collection of samples and laboratory analysis for soil stabilization tests in order to determine the optimal formulation;
- collection of samples and laboratory analysis at the end of soil handling operations to certify completion and approval as filling material.

After the soil remediation project will end, an off-site transport company will implement the following measures:

- will limit access to areas by limiting excavation void;
- will secure the holes left after excavating the contaminated soil, in order to avoid the risk of accidents and the risk of pollution, either voluntary or accidental;
- supervise the excavation void in regular intervals to monitor the terrain conditions;
- make sure the filling material to be used for filling in excavation voids will meet the quality requirements for sensitive land use.

Comparative analysis of remedial activities. Comparative analysis, implementation of remedial action necessary on two former industrial sites - Bucharest and Braşov, on which was confirmed the existence of contamination in different time periods, are presented in Tables 2 and 3, as similarities and differences.

Table 2

Comparative analysis - similarities

Similarities
The previous activities carried out on the two sites were car manufacturing, and demolishing/decommissioning activities were developed with the purpose of changing land use in residential area;
Treatment goal: bringing soil to the parameters of acceptability for disposal in landfills of non-hazardous waste according to the Decree no. 95/2005;
The identified pollutants were the same for both sites: hydrocarbons and heavy metals;
Treatment in-situ concrete platforms handling, excavation, monitoring;
Remove and transport soil treated to a non-hazardous landfill;
Verification and validation done by the local environmental protection agency.

Table 3

Comparative analysis - differences

Differences	
Bucharest site	*Braşov site*
Limitations related to data and information on soil, lack of investigations	Existing investigations following the provisions of GD 1408/2007
Identification and delineation of contaminated areas was achieved by repeated sampling	Identification and delineation of contaminated areas was performed using specific methods provided by GD 1408/2007 together with associated risk assessment
Treatment of 1400 m^2 on a built platform	Existing processing platform
Project duration: 18 months	Project duration: 1, 5 months
Costs: Large	Costs: Acceptable
Residual water resulting from bioremediation - the treatment process using water, results in wastewater	No waste of stabilization, nor wastewater
Depending on the location of sampling sites, for each sample were selected for analysis only the most representative indicators in relation to potential sources of contamination	Were analysed the same quality indicators in all sampling locations
Required repeated monitoring before and during bioremediation to determine areas contaminated by leachate monitoring, point samples	Estimated self-monitoring to determine the initial state of contamination but also for a better delineation of areas with high concentrations of pollutants
It was considered a backup for when bioremediation results were not satisfactory; method efficiency can be improved by stabilization / solidification by use of additives, mainly to neutralize metal concentrations.	Monitoring during treatment works for a better delineation of areas with high concentrations of pollutants and likely not lend itself to treatment by stabilization. Monitoring technical project site after remediation.

Conclusions. Remediation of contaminated sites should be considered in the context of urban regeneration of contaminated land, and to reduce the risk from the presence of contaminants.

In the two presented cases, remedial project objectives were:

- to reduce the mobility of contaminants in soil and groundwater, leaving soil to an appropriate standard for public and environmental use and minimize the impact of past activities;

- to bring contaminated soil by bioremediation works and stabilization in the parameters of acceptability for disposal in non-hazardous landfill according to the decree no. 95/2005;

Remedial actions for the two industrial sites have been undertaken as a result of land owners intention to change the use in residential. The remediation works for the first analysed site were performed between 2007-2008, before the implementation of specific legislation in the field of contaminated sites (OG 2007a, 2007b) based on physico-chemical or repeated sampling of soil and leachate, for the surface and shallow (1 m) soil, in accordance with the Decree no. 756/1997, having high costs of repeated monitoring, long construction period, design, planning of the bioremediation platform, excavation and transportation of waste, and repeated attempts during the process of bioremediation treatment.

Remedial works on the second site considered will be made during October-December 2012, based on the geological investigations for assessing environmental contamination under the provisions of GD 1408/2007. The knowledge of significant pollution on the site allowed human health risk assessment associated to future land use,

and development of the conceptual model of the site, more accurate delineation of contaminated areas, leading to much lower costs for remedial works. Ways to restore the soil were established after analysing the findings of the aforementioned studies, observing the provisions of GD 1403/2007.

Analysing these findings, it is found that identifying contaminated areas and establishing rehabilitation works to reduce the risk associated with future land use generates significantly lower costs for beneficiaries and advisable to carry out studies to investigate soil and geological environment before starting remediation works for historically contaminated sites.

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