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STUDY ON THE PRESENCE OF TOXIC SUBSTANCES IN WATERS FROM DISAFFECTED INDUSTRIAL AREAS

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ABSTRACT

The paper presents aspects regarding the water quality coming from drillings carried out in disused industrial areas, from the Brăila - Galați area. The studied parameters include the analysis of salts such as sulphates, nitrates, phosphates and carbonates and the analysis of toxic compounds such as phenols, copper, lead, iron and aluminium. The experiments revealed the presence of some of the studied toxic substances. Finally, the results obtained and the classification of the studied indicators within the admissible limits were discussed and potential methods and techniques for depollution of drilling waters and soil in derelict industrial areas were proposed.

KEYWORDS: industrial site, drilling water, salts, phenol, heavy metals

1. Introduction

Decommissioned industrial areas are characterized by a significant diversity of waste and residual toxic substances. The reuse of these areas requires a whole range of actions to carry out the transformation and revitalization of the environment, such as the determination of existing pollutants and the indication of ways and methods to remove or to minimize the negative impact of these substances on the environment [17]. There is currently a growing concern among policy makers and scientists about the effects of human and wildlife exposure to chemicals in the environment, especially in the aquatic environment. Phenolic compounds are among the chemicals of great concern in this regard, as they tend to persist in the environment for a long time, to accumulate and exert toxic effects on living organisms [2,6]. Phenolic compounds are present in the effluents of various industries, such as oil refining, petrochemicals, pharmaceuticals, coking operations, resin manufacturing, plastics, paint, cellulose, paper and wood products [15,20].

Discharge of these compounds without treatment can lead to serious health risks to humans, animals and aquatic systems [24]. Phenolic compounds are highly toxic pollutants even at low concentrations [25] and act as carcinogens, causing damage to red blood cells and liver [1]. Some phenolic compounds are abundant in nature and are associated with the colours of flowers and fruits [18], others are synthesized and are used in various aspects of daily life of the population. Phenolic compounds are used in low concentrations in disinfectants and are also present in many alcoholic beverages, pharmaceuticals and cosmetics [13]. Environmental pollution with heavy metals is also of significant importance. Heavy metals are harmful to humans and animals and tend to bioaccumulate in the food chain [6]. The threat posed by heavy metals to humans is exacerbated by their long-term persistence in the environment. Industrial and municipal waste generates a large amount of particulate emissions and slag waste enriched with heavy metals that contaminate the soil, water and surrounding air [6]. Such effects are particularly serious and present a severe ecological risk to human health when certain metallurgical processes take place in the vicinity of the urban environment [21].

Heavy metal pollution is due to both natural and anthropogenic activities, such as smelting procedures, mining, excessive use of wastewater and fertilizers in agricultural fields and atmospheric deposition from vehicles and industries [16,23]. The presence of heavy metals in soil and groundwater is very dangerous because they are transferred to the food chain through vegetables, and long-term exposure to heavy metals causes various fetal diseases, mental illness, kidney failure, lung cancer, bone fractures, kidney dysfunction and hypertension [19,22,26].

Table 1. *The maximum permissible loading limits of the ground waters [11]*

Parameters/ Indicators	**The number of states that have set limit values**	**Maximum allowed range**		
		From	**To**	**Units**
pH	1	6,5	9,5	[-]
Chlorides (Cl^-)	22	24	12300	mg/l
Bicarbonates (HCO_3^-)	2	10	500	mg/l
Phosphates (PO_4^{3-})	3	0,0414	4,4	mg/l
Sulphates (SO_4^{2-})	21	129,75	4200	mg/l
Nitrates (NO_3^-)	5	18	50	mg/l
Nitrites (NO_2^-)	8	0,05	1,4	mg/l
Ammonium (NH_4^+)	20	0,0837	52	mg/l
Phenol (C_6H_5OH)	3	7,5	10	μg/l
Total ionic iron (Fe^{2+} and Fe^{3+})	5	103	5000	μg/l
Copper (Cu^{2+})	10	10,1	2000	μg/l
Lead (Pb^{2+})	20	5	320	μg/l
Aluminium (Al^{3+})	7	100	200	μg/l
Cyanide (CN^-)	3	37,5	50	μg/l

Another important aspect regarding the groundwater in the disused industrial areas is related to their corrosive character. Corrosion control involves several parameters, including concentrations of calcium, bicarbonate, carbonate and dissolved oxygen, as well as pH. The pH value controls the solubility and reaction rate between metals that are involved in corrosion reactions. It is especially important to ensure a certain concentration of calcium in the water, in order to form a protective film on the metal surface. For certain metals, alkalinity (carbonate and bicarbonate) and calcium content (hardness) also affect the level of corrosion. The hardness of groundwater is greatly influenced by the composition of minerals in the soil. Dissolved natural calcium and magnesium salts (carbonates) determine the hardness of the water, which forms crusts on the surface of water pipes or water boilers [16].

Taking into account these aspects, the development, rationalization, reuse and reconversion of industrial areas play a fundamental role in the processes of urban transformation [9].

2. Purpose and objectives of the work

The purpose of this study is to assess the real situation of the drilling waters concerning their contamination with heavy metals, phenols and various salts, in industrial sites located in the Brăila-Galaţi area, before resuming the activity of these industrial landmarks. In order to determine the quality parameters of the waters and soils from the studied derelict industrial areas, five drillings were made marked with F1, F2, F3, F4 and F5. For an overview of the loading degree of these waters with polluting compounds, for each of these boreholes the following indicators were determined:

- The fixed residue, which represents the totality of mineral and organic solids in the water and which is obtained by heating the water to 105 °C, when complete evaporation is carried out.
- Salts of calcium and magnesium in solution, that can be in the form of carbonates, chlorides, sulphates, nitrates, phosphates or silicates. Temporary hardness is determined by carbonates, which precipitate by boiling, and permanent hardness is determined by the other calcium and magnesium salts (sulphates, chlorides, etc.), which do not precipitate by boiling.
- Iron, which is found in groundwater in the form of compounds such as ferrous bicarbonate. Water that contains large amounts of iron is opalescent, with a sour, astringent taste, stains laundry and can not be used in the paper, pulp, dyes, and others;
- Chlorine, which is found in water in the form of chlorides and is most often of a mineral nature. The presence in large quantities of chlorides gives the water an unpleasant, characteristic taste (salty, bitter).
- Copper, lead and aluminium can be found in the form of oxides and indicate corrosion of the pipes. In surface waters they can be found downstream of wastewater discharges from extractive and processing industries or from galvanizing operations. Their compounds are very poisonous.

In order to establish the degree of pollution for the studied drilling waters, the determined values were compared with the admissible limits imposed by the legislative regulations. The Romanian legislation establishes the maximum allowed concentrations for wastewater discharge in natural receptors [10] and in sewerage networks, but in the case of groundwater the limits are regulated for a limited number of indicators (such as nitrates, pesticides).

Table 1 shows the permitted limit values for the waters subjected to this study, as set out in the Commission report in accordance with Article 3.7 of the Groundwater Directive 2006/118/EC, for the establishment of groundwater thresholds [11]. In this report there are centralized the ranges of limit values established by all member states of the European Union, through the domestic legislation of each state.

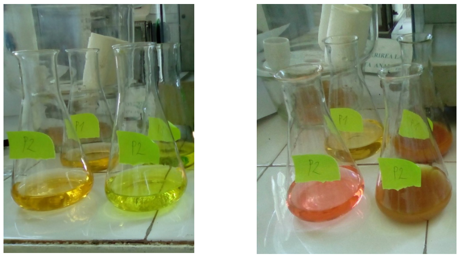

Fig. 3 Determination of bicarbonate and chloride anions before titration (left photo) and after titration (right photo)

3. Materials and working methods

In this study, five water samples were analyzed, collected from five drillings carried out in different points of an industrial site in the Brăila - Galaţi area.

The drillings were carried out at depths of about 7-8 meters and were provided by the representatives of the disused industrial area. In order to maintain the groundwater rate at a level of 6 m (necessary for the establishment of heavy equipment and constructions within the industrial area), a system of discharging wells was made. Through a siphon system, the wells discharge water from the first aquifer into medium depth aquifer located at 62 - 64 m from the earth's surface. The medium depth aquifer has ascending tendencies up to 7-8 m from the earth's surface.

The quality parameters for the drilling water samples studied are analyzed according to the following methods [3]:

- pH, by the electrometric method (fig. 1);
- Mineral residue, by conductometric method (fig. 2);
- Carbonates – by the volumetric method, by titration with sulfuric acid, in the presence of phenolphthalein;
- Chlorides – by the volumetric method, by titration with silver nitrate in the presence of potassium chromate (fig. 3);
- Bicarbonates – by the volumetric method, by titration with sulfuric acid, in the presence of methyl-orange (fig. 3);

Fig. 1 Determination of pH

Fig. 2 Determination of mineral residue

- Sulphates – by the volumetric method, by titration with barium chloride, in the presence of sodium rhodizonate;
- Phenol (fig. 4), nitrates, ammonium, phosphates and dissolved oxygen – by the colorimetric method with UV-VIS Nanocolor spectrophotometer (fig. 5);

Total iron – by the colorimetric method, with Jenway colorimeter (fig. 6);

- Copper, aluminium, lead, cyanide – by the colorimetric method with analysis kits (fig. 7–10);
- Nitrites – by the Griess colorimetric method, with sulfanilic acid and alpha naphthyl amine (fig. 11);

Fig. 4 Determination of phenol

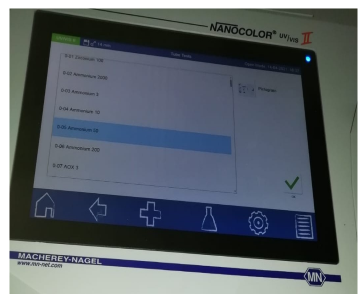

Fig. 5 UV-VIS Nanocolor spectrophotometer

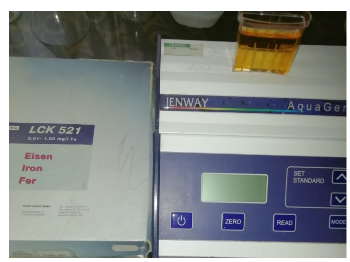

Fig. 6 Determination of total iron

Fig. 7 Determination of copper

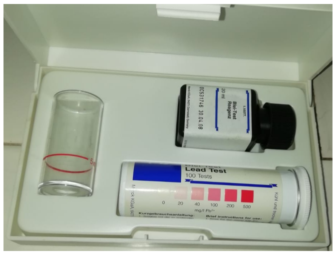

Fig. 8 Determination of lead

Fig. 9 Determination of aliminium


Fig. 10 Determination of cyanide

Fig. 11 Determination of nitrites

4. Results and discussions

The results of the conducted experiments, for all the five samples collected from the drillings of the disused industrial areas and for all the types of performed analyzes, are presented in Table 2.

The results of the pH values determination, for the 5 samples of ground water are shown in the diagram of the figure 12. The basic values determined for the pH are within the permissible limits for all the studied drillings.

Also, the obtained concentrations for the filtered residue are constant for all the studied boreholes (fig. 13). The determined low values fall even within the norms imposed for the discharge in natural receptors [10].

Table 2. The concentrations determined for the studied parameters (mg/l)

Studied parameters	Studied boreholes				
	F1	F2	F3	F4	F5
pH	8,01	8,03	8,09	9,06	8,12
Mineral residue	1486	1462	1196	1496	1498
CO_3^{2-}	0	21	17,4	30	7,2
HCO_3^-	793	713	695	707	750
SO_4^{2-}	498	492	240	432	430
NO_3^-	10,6	4	4	4	6,44
NO_2^-	0	0	0	0	0
NH_4^+	< 4	< 4	< 4	< 4	< 4
PO_4^{3-}	0,06	0,05	0,05	0,05	0,05
Cl^-	28,4	24,8	24,8	24,8	39,1
Phenol	0,2	0,2	0,2	0,2	0,4
Total Iron	0,5	0,5	0,4	0,5	0,6
Cu^{2+}	0	0	0	0	0
Pb^{2+}	0	0	0	0	0
Al^{3+}	0	0	0	0	0
CN^-	0	0	0	0	0

Fig. 12 The pH values of the boreholes

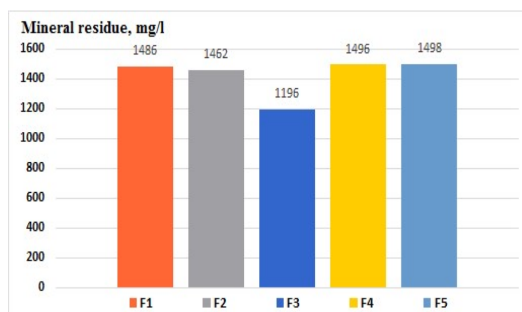

Fig. 13 The mineral residue of the boreholes

The concentrations of carbonates and bicarbonates are shown in figure 14, respectively in figure 15, and the determinations of sulphates, nitrates and phosphates are shown in figures 16 - 18. The presence of all these salts gives alkalinity to the analyzed groundwater. Mineralization and saline soils require permanent monitoring in the derelict industrial areas. Such soils and groundwater may not be suitable for agricultural use for a certain period of time since the cessation of the industrial activity.

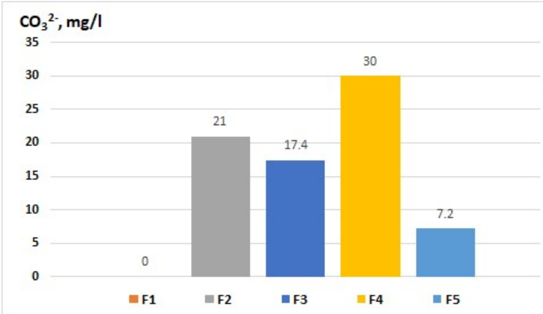

Fig. 14 The carbonate content of the boreholes

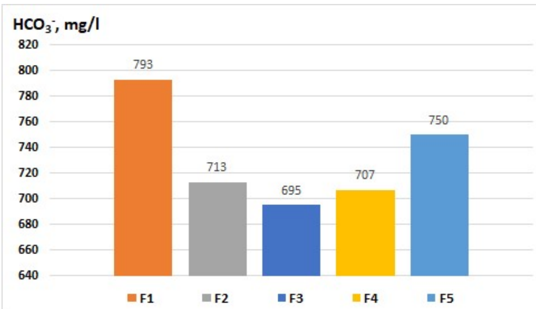

Fig. 15 The bicarbonate content of the boreholes

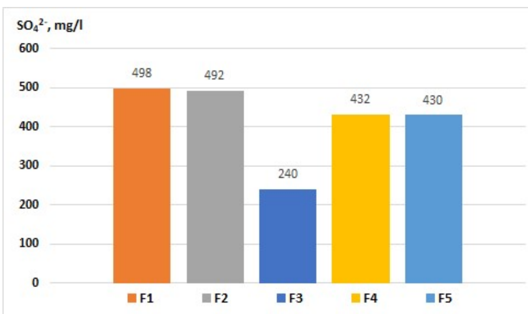

Fig. 16 The sulfate content of the boreholes

The slightly alkaline character of these waters is also explained by the determined low chloride content, shown in figure 19.

The values obtained for the total iron concentration are presented in figure 20. These values fall within the range of maximum allowed concentrations, between 0.1 ÷ 5 mg/l depending on the rigors imposed by different European countries [11]. Regarding the phenol content, presented in figure 21, it can be found that the permissible limit range has been exceeded, the registered values being 2 to 4 times higher. This excess of phenol in groundwater can be a danger of soil pollution as they can rise and seep into the soil being very volatile. Phenols are compounds with high toxicity, because they tend to persist in the environment for a long time, and once accumulated in the soil they can exert toxic effects on living organisms [6]. The presence of heavy metals was not indicated by the analyzes performed for the groundwater samples.

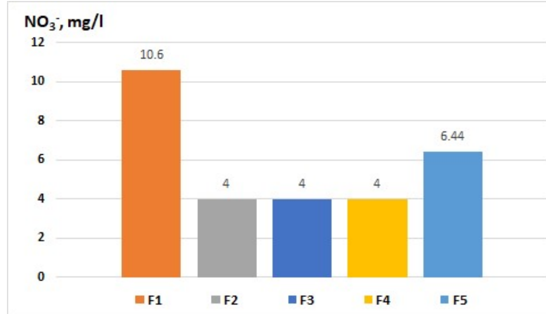

Fig. 17 The nitrate content of the boreholes

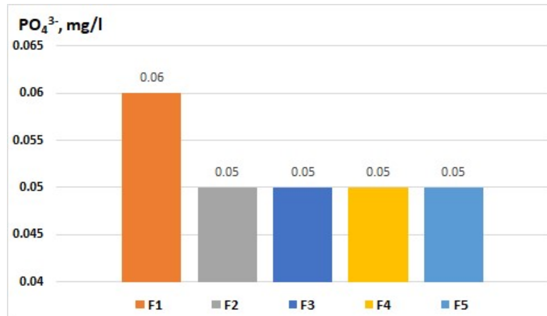

Fig. 18 The phosphate content of the boreholes

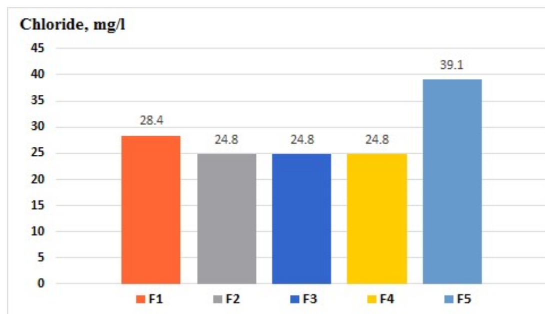

Fig. 19 The chloride content of the boreholes

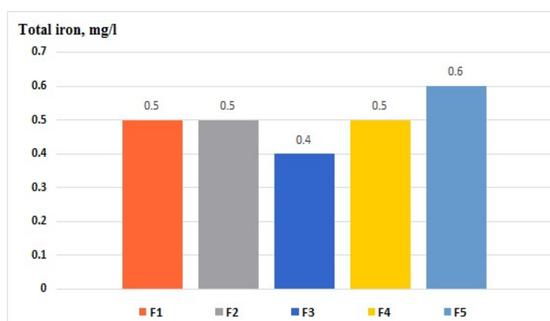

Fig. 20 The total iron content of the boreholes

However, for a clearer picture of the pollution level of the studied disused industrial area, the analyzes were compared with soil samples collected in previous years in this area, where some heavy metals such as chromium, cadmium, lead, nickel, copper, manganese were determined according to figure 22 [7].

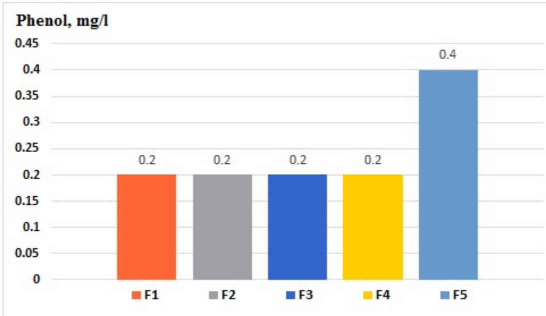

Fig. 21 The phenol content of the analyzed boreholes

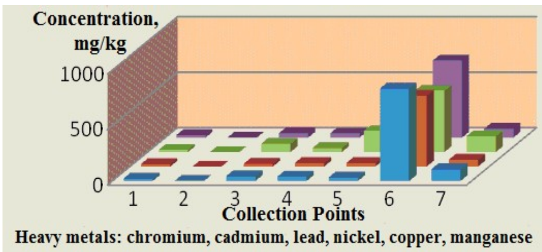

Fig. 22 The heavy metals content determined for soil samples from the disused industrial area [7]

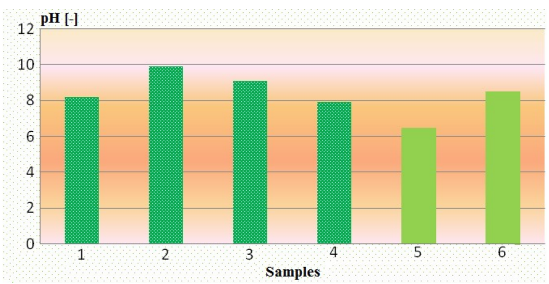

Fig. 23 The pH values determined for soil samples from the disused industrial area [7]

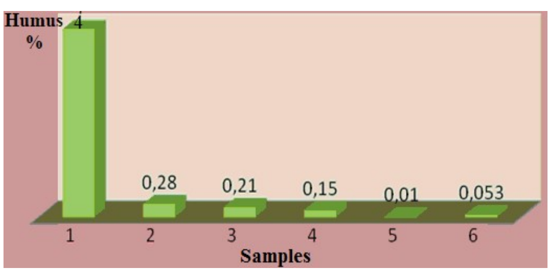

Fig. 24 The humus content determined for soil samples from the disused industrial area [7]

In parallel, for these soil samples, the pH value was determined, which was found to be between 6.5 - 9.8, according to figure 23. The presence of heavy metals in these soils can be observed, with maximum values of over 500 mg/kg [7]. The presence of heavy metals in the soil is very dangerous, because they can be transferred to the food chain through vegetables and fodder, and long-term exposure to heavy metals causes various diseases [6]. Also, according to the study performed on soils [7], the presence of total hydrocarbons can be noticed, in a concentration of 20 mg/l. This concentration exceeds the value of the permissible limit range given in Table 1, by about 20 times. However, the concentration determined for total hydrocarbons does not exceed the alert threshold required by law, which implies permanent monitoring of the area without the need for intervention [4,5]. The total hydrocarbons content recorded may be due to heavy road traffic within the decommissioned industrial area. Figure 24 shows the humus content of the soils in the studied industrial areas [7]. The low values of the humus content may be due to the scrapings and the excavations carried out in these areas. These low values are an indicator that the soils in the studied derelict industrial areas are not suitable for agricultural use.

Also, due to the presence of phenols and heavy metals both in drilling waters and soil, for the future rehabilitation of the studied derelict industrial area, only the development of new industries or commercial activities can be recommended, but these areas can not be indicated for agriculture or housing construction.

4.1. Measures to prevent pollution in the industrial sites

In order to avoid contamination of soils and groundwater in active or decommissioned industrial areas, measures may be taken to prevent the pollution and the infiltration of toxic substances. Below are some recommendations regarding the environmental factors: water, air and soil [8].

a. For the environmental factor - water, it is recommended:

- the compliance with the technologies of transfer, handling, transport and evacuation of the effluents that can lead to the pollution of the sewerage and the groundwater in these areas;
- the observance of the dismantling conditions of the equipment containing oils, in order to avoid accidental leaks on the floor of the buildings and their discharge into the sewerage network;
- the avoidance of the temporary storage of waste on non-concrete and uncovered platforms due to the danger of precipitation runoff that washes the surface of the waste in the groundwater.

b. For the environmental factor - soil, it is recommended:

- the avoidance of the temporary storage of waste on non-concrete and uncovered platforms due to the danger of washing the surface of the waste and runoff directly into the soil;
- the wetting operations of the asbestos-cement slabs must be carried out carefully, taking measures to insulate the space under the work area by covering them with plastic foil, so that there is no asbestos infiltration into the soil.

c. For the environmental factor - air, it is recommended:

- the cover with protective foils of the area that is being decommissioned in order to avoid the scattering of the resulting dusts in the environment;
- for interior demolition - the protection of the windows with nets, for the deposition of sedimentable dusts on a small area in the immediate vicinity of the buildings, in order to reduce their degree of dispersion;

Also, in the case of the dismantling of the industrial sites, it is recommended [8]:

- for the demolitions by controlled explosion - the covering in wire mesh or rubber carpets of the constructive elements, in order to completely stop any pieces of material and to prevent the throwing of small material under the effect of the explosion;
- to prevent as much as possible the dust rise during the downfall, these halls will be cleaned beforehand or water sprayed before the explosion;

Table 3. *The groundwater treatment matrix [7,12]*
(• - *applicable;* O – *partially applicable;* x - *not applicable)*

	Neutralization	Precipitation	Co-precipitation/coagulation	UV/Ozone	Chemical oxidation	Reduction	Distillation	Air stripping	Vapor stripping	Activated carbon adsorption	Evaporation	Gravity separation	Flotation	Membrane separation	Ion exchange	Filtration	Biological treatment	Electrochemical treatment
Heavy metals	x	•	•	x	x	O	x	x	x	O	•	•	x	•	•	•	x	•
Cr VI	x	•	x	x	x	•	x	x	x	O	•	x	x	O	•	x	x	•
As	x	O	•	O	O	x	x	x	x	O	x	O	x	•	•	•	x	x
Hg	x	•	•	x	x	•	x	x	x	•	x	O	x	O	•	•	x	x
Cyanide	x	x	x	•	•	x	x	x	x	x	•	x	x	•	•	x	O	O
Corrosive elements	•	•	x	x	x	x	O	x	x	x	x	x	x	x	x	x	x	x
VOC, VOCH	x	x	x	O	•	x	•	•	•	•	x	x	x	O	O	x	O	x
Ketones	x	x	x	O	•	x	•	•	•	x	x	x	x	x	x	x	•	x
Pesticides	x	O	O	•	•	x	•	x	O	•	O	O	O	•	•	•	O	x
PCB	x	•	•	•	•	x	•	x	x	•	•	•	•	•	•	•	O	x
Dioxins	x	•	•	•	O	x	•	x	x	•	•	•	•	•	•	•	O	x
Oils and greases/ floating products	x	•	•	x	x	x	•	x	x	x	•	•	•	•	•	O	O	x

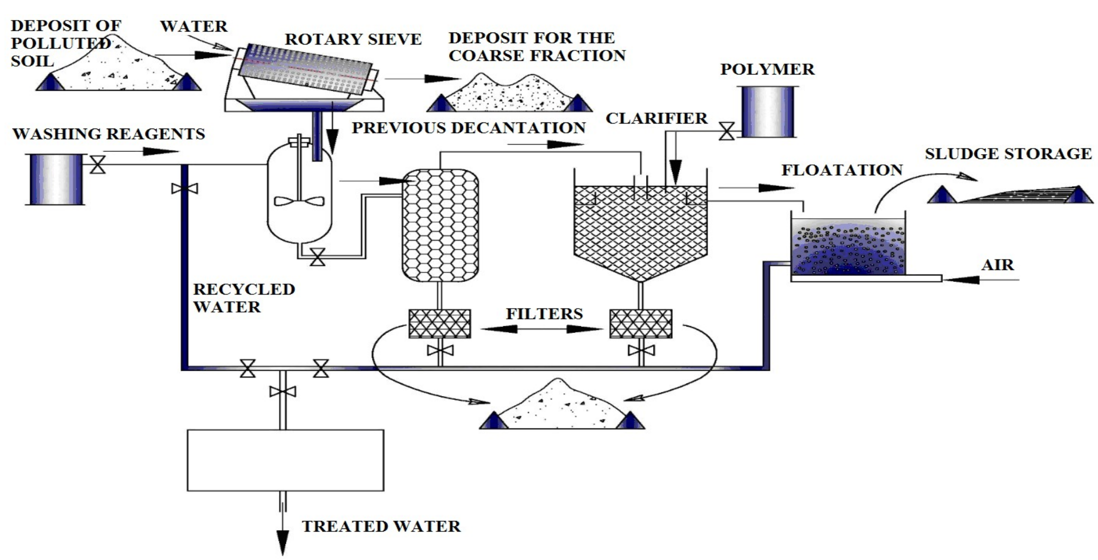

Fig. 25 The chemical depollution by in situ washing [8]

- during waste shipments the use of tarpaulins is recommended, placed so as to cover the entire surface of transported waste to prevent the wind from scattering the dust;
- washing the wheels of the means of transport to prevent the dust from being picked up by the vehicles and spread on the road to the destination.

4.2. Methods for depollution of the industrial sites

Beyond the involvement for determining the quality parameters of the groundwater in derelict areas, this study also aims to contribute to the process of decontamination and of relaunch into the industrial circuit of these areas without economic efficiency. Also, this study aims to identify the possibilities for the revaluation and the applicability of greening and continuous monitoring methods of the areas exposed to contamination with various pollutants.

The choice and selection of the techniques for greening, control and monitoring of the depollution works can be done according to several site-specific criteria of technical, organizational, economic, socio-political, or environmental nature [8]. For the depollution of groundwater, the suitable methods are mentioned in Table 3 [7,12]. The depollution of soils in disused industrial areas can be achieved by a number of methods, of which we mention [7,8]:
- Physical methods for the pollution evacuation: ventilation of the unsaturated area, extraction in double phase, bubbling in situ, pumping and treatment, pumping-skimming, excavation of the soil;
- Physical methods for the pollution blocking: confinement by coating and sealing, hydraulic trap or hydraulic coating;
- Chemical methods: the use of chemical reagents to destroy pollutants or to transform them into less toxic and/or easily biodegradable compounds, by processes such as "in site washing", implemented according to Figure 25;
- Biological methods: metabolism, cometabolism, biostimulation and biogrowth, dynamic biodegradation, controlled natural attenuation;

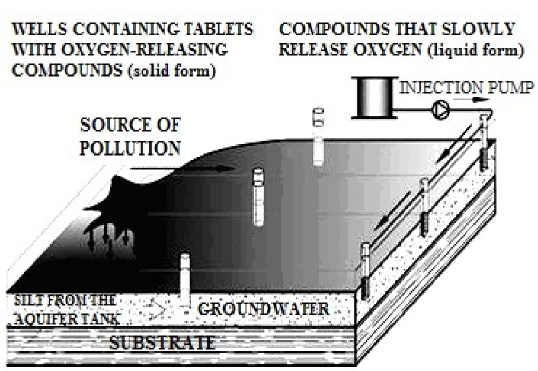

Fig. 26 The dynamic biodegradation process [7]

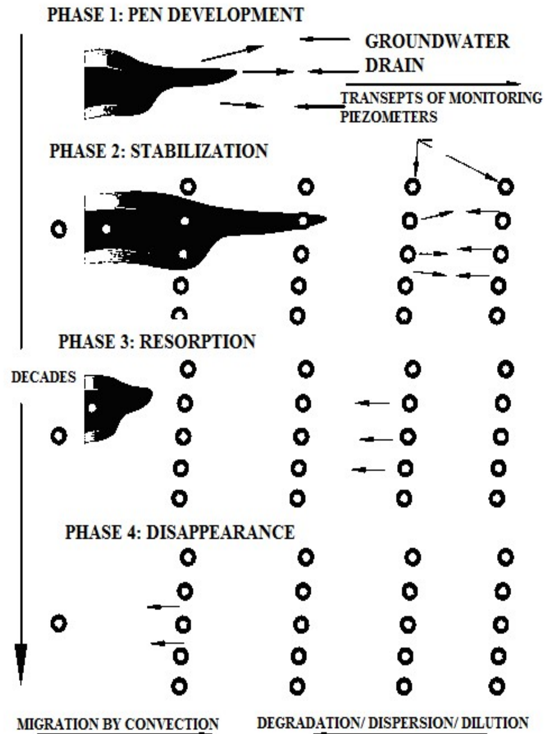

Fig. 27 The controlled natural attenuation process [7]

The dynamized biodegradation (fig. 26): or the dynamized natural attenuation in situ requires the addition of specific compounds in soil or into groundwater, to create favourable conditions for the activity of microorganisms responsible for the biodegradation of the pollutants. To do this, the microorganisms are maintained in optimal conditions (oxygen, pH, temperature, redox potential) and nutrients are added, especially in liquid form. This technique is commonly used on sites that exhibit biodegradable hydrocarbon from volatiles to semi-volatiles forms, as well as on sites that exhibit chlorinated solvents. The method is undergoing advanced development in terms of more recalcitrant compounds (like PAHs).

Controlled natural attenuation (fig. 27): process that occurs naturally in soil and groundwater, without human intervention, whose purpose is to reduce the mass, toxicity, mobility, volume or concentration of contaminants in these environments. The processes considered are: dispersion, dilution, volatilization, adsorption, mechanisms for stabilization or destruction of pollutants, whether physical, chemical or biological. It is necessary to specify that natural attenuation must include at least one destructive process and one sequestration process (example: adsorption).
- Other depollution methods: reactive permeable barrier, sorption barrier, precipitation barrier, degradation barrier.

5. Conclusions

- In this paper, the presence of salts, such as carbonates, phosphates, nitrates and the presence of toxic compounds, such as phenol and heavy metals was studied, in certain boreholes of an industrial site located in the area of Brăila - Galaţi.
- The determined analyzes reveal a degree of loading within admissible limits, for all the studied indicators and parameters at the groundwater level and the absence of toxic compounds such as copper, aluminium, lead and cyanides. However, the analyzes performed on the soil samples in the studied area, revealed the presence of heavy metals, qualitatively and quantitatively.
- The determination of phenol in groundwater indicated values up to 4 times higher than the range of permissible limit values in European countries. Being toxic compounds that persist for a long time, phenol can accumulate in the soil and can exert toxic effects on living organisms.
- It was found that pollutants migrate from water to soil and vice versa, perpetuating the degradation of the area depending on the climatic conditions.
- The development and the reuse of the industrial areas play a fundamental role in the processes of urban transformation. The aim is to restore these industrial surfaces in use, but given the degree of salting and pollution with phenol and heavy metals of the groundwater and soil, these areas can be recommended in the future only for the development of new industries or as commercial spaces and not for agriculture or housing construction.
- In order to stop or at least to reduce the pollution of the environmental factors during the period of activity or the decommissioning of the industrial areas, it is necessary to adopt measures to prevent pollution related to the handling, transport and storage of the toxic substances used in the production process.
- In addition to determine the quality parameters of groundwater in the studied areas, this work contributes to the process of decontamination and to the re-entry of the industrial areas without economic efficiency.
- Other aim of the study was to identify the possibilities for the valuation and the application of methods of greening and continuous monitoring of areas exposed to pollution with various toxic substances.
- The study can be continued with the monitoring of waters and soils in several derelict areas and several toxicity parameters.

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