DYNAMIC OF PETROLEUM HYDROCARBON CONCENTRATION IN SOIL POLLUTED WITH CRUDE OIL

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Abstract: *Once with the establishment of the first human communities, with socio-economic development start, with exacerbation of the industrial and post-industrial revolution, etc., people, not content with nature itself, with his intelligence and creative spirit, started to adapt and transform it according to his needs. Unfortunately, there are harmful secondary effects of human activity, becoming more frequent and aggressive, some completely unforeseen and even irreversible impacts on environmental quality, plant and animal life, even on people existence. All this led to the appearence, development and progression in an accelerated rhythm of a phenomenon very new, very complex and more extensive and dangerous identified, on a global scale, by all countries and continents*, as environmental *pollution. Pollution is any undesirable change in physical, chemical and biological characteristics of water, air and soil that can affect harmful health, survival or activities of humans or other living organisms, whose term for clearer understanding is necessary to take into account also, the term "contamination", which refers to the presence, no matter the quantity of elements or unwanted hazardous substances in water, air or soil due to human activities that definitely are not harmful. The experimental scheme included two groups of experimental variants in which the soil was polluted by 5% and 10% crude oil. For each concentrations of crude oil were created 4 experimental variants, achieved by conditioning the soil with Ecosol in two doses, with or without inoculation with selected bacteria. Bioremediation protocol focused on the application of the two major technologies known in bioremediation method such as: soil biostimulation based on environmental conditions improvement for microorganisms multiplication and activity to degrade petroleum hydrocarbons, and bioaugmentation based on enriching the soil with specific biodegrading hydrocarbons microorganisms. The efficiency of bioremediation process in optimal variants was 92.6% in case of soil polluted with 5% crude oil and 80.5% for excessively polluted soil with 10% crude oil.*

Key words**:** *bioremediation, crude oil, soil pollution, total petroleum hydrocarbons.*

INTRODUCTION

Pollution phenomena cause significant changes in the zoosphere and phytosphere, as well as microorganisms, leading to the disappearance of many species, soil fertility decreasing, its most important property that allows support for plant and animal life, and thus the human (ENGELKING, 2000; PEPPER et al., 1996).

Although possessing self-regeneration capacity, specific forming conditions are that, once destroyed, the soil should not be able to recover as it was, because it can not reproduce the conditions and millennial history of the formation. It can be created a body with similar functions. Until recent decades, land was seen mainly due to fertility, for his ability to support plant life, just as the main means of production in agriculture, the last time recognizing that the existence and development of human society will be put in future abundance and quality of terrestrial higher plants, which should ensure people's food and raw materials for clothing, shelter, medicines and other requirements. Between elements with potential in polluting soil, the petroleum hydrocarbons are considered among the strongest soil pollutants (AYOTAMUNO et al., 2006). Given that crude oil pollution and waste oil products affects the soil capacity to

support life, improving remediation methodology, adapted to conditions in Romania is a necessity (BALBA et al., 1998).

For decontamination of soil polluted with petroleum hydrocarbons can be used physical, chemical and biological methods. From all these, research has shown that bioremediation, especially for crude oil pollution is a superior method, effective and much cheaper than physical or chemical methods. Indeed, in recent years, bioremediation of soils contaminated with petroleum hydrocarbons is a challenge to modern scientific research (RAHMAN et al., 2003). Bioremediation can be: in-situ and ex situ. Bioremediation is based on the ability of microorganisms to use hydrocarbons as carbon source and energy. It is considered to be the most effective because, in addition to lower cost, no irreversible effects on pedogenetical characteristics of affected soil (VOICULESCU et al., 2005).

MATERIAL AND METHODS

Experiments included two groups of experimental variants in which the soil was polluted by 5% and 10% crude oil. For each concentrations of crude oil were created 4 experimental variants, achieved by conditioning the soil with Ecosol in two doses, with or without inoculation with selected bacteria, and a control variant were the soil polluted was subject to natural attenuation. All data were reported at a variant with unpolluted soil achieved and maintained under the same experimental conditions.

Bioremediation protocol focused on the application of the two major technologies known in bioremediation method such as: soil biostimulation based on environmental conditions improvement for microorganisms multiplication and activity to degrade petroleum hydrocarbons, and bioaugmentation based on enriching the soil with specific biodegrading hydrocarbons microorganisms.

Biostimulation - the first technological link included a innovation element based on using an organic compound made from cellulose fibers for soil polluted conditioning with additives to optimize its structure, water and air circulation regime in soil, and not least achieving a protective interface between degrading microorganisms and pollutant. Ecosol compound was chosen for experiment by analysing a series of organic compounds suitable for conditioning soil contaminated with organic pollutants, especially because of its biodegradability properties.

Bioaugmentation - the second technology link was achieved by soil inoculation with bacterial bioproducts made from specific bacteria selected and tested in the laboratory for their ability to degrade petroleum hydrocarbons.

The values obtained by analyzing soil samples were processed using more specific methods of mathematical statistics. Analysis of variance for establishing Fischer and Tukey tests determined for $\alpha = 0.05$, which shows the changes produced on soil and plant characteristics, the effects of treatments applied. ANOVA method provides information allowing the calculation of limit differences used in multiple comparison methods and the mean average for each graduation of studied factor.

RESULTS AND DISCUSSION

After 14 days from soil pollution with 5% crude oil, TPH values of the improved variants begin to decrease compared with polluted variant, untreated, where the soil decontamination was achieved through natural attenuation processes. After 21 days, begins to be obvious that the variant with the soil conditioned with high dose of Ecosol, respectively 0.5% (100 g/vas) and inoculated with selected bacteria, soil pollutant disappearance rate was much higher, compared with other experimental variants, results having a high degree of statistical assurance.

In case of variants in which the soil was polluted with 10% crude oil was found a lower rate of crude oil degradation between 30-300 days. The significance of this long period of slow (bio)degradation processes of petroleum hydrocarbons, in case of excessive crude oil polluted soil may consist in adapting difficulty of biodegrading microorganisms, from soil and those introduced by adding the bacterial inoculum at excessive pollution. This is one of the major reasons for which bioremediation procedures are not recommended for soils polluted with petroleum hydrocarbons at concentrations higher than 5%. Biodegradation processes, though, happens to take place after a very long and unprofitable period of time.

In last experimental year between 660-810 days, the end of petroleum hydrocarbons degradation process by intrinsic capacity for self-cleaning of polluted soil, which was highlighted by a perfect linearity of the %TPH regression curve in control variant, subject to natural attenuation. The analysis of regression curves revealed the continuation of process in experimental variants treated with Ecosol, maximum decreasing of TPH concentration being registered in the variant treated with 0.5% (100 g/pot) Ecosol and inoculated.

In case of excessive polluted soil with 10% crude oil, dynamics and pattern of regression curves corresponding to the control and four treated variants are very similar to those registered in experimental variants polluted with 5% crude oil. The difference lies in the initiation speed of (bio)degradation processes being lower in excessively polluted variants manifesting after 21 days from pollution, microorganisms require a longer adaptation period to severe conditions of pollution.

In the second experimental year, the determinations achieved between 300-450 days, was very obvious the quantitative differentiation between experimental variants, so the regression curve calculated in the variants treated with 1% Ecosol (200 g/pot) and inoculated detaching the calculated regression curves platoon from treated variants.

Experimental data analysis revealed the positive effect exerted by doubling the Ecosol dose in uninoculated variants on petroleum hydrocarbons biodegradation, in case of soil polluted with 10% crude oil, conditioned with 1% (200 g/pot) Ecosol may supply the application of the inoculation with selected bacteria. However, should take into account the costs imposed by this operation and the large amount of required product.

At the end of the experimental period, TPH concentration in soil polluted with 5% crude oil still present a value of 1.81%, which means that it can only own detoxification mechanisms yield of soil pollutant without application of any treatment is 63.8%. This may seem very high efficiency for a process of natural attenuation if we take into account the huge interval of 810 days, which may mean no more than three years miss for land use.

In the variant with soil polluted with 5% crude oil, conditioned by 0.5% (100 g/pot) Ecosol and inoculated with selected bacteria, petroleum hydrocarbon content at the end of the experiment was only 0.37%, representing a yield by 92.6%.

In soil polluted with 5% crude oil, doubling Ecosol dose from 0.25% (50 g/pot) to 0.5% (100 g/pot) has exercised the most spectacular effect, causing an increase of yield to 45.7% in absence of bacterial inoculation. At low Ecosol dose, inoculation with selected bacteria induced a strong stimulation effect; the bioremediation yield is 24% higher than that determined in uninoculated variant.

In soil polluted 10% crude oil where no ameliorative treatments were applied, the crude oil concentration determined at the end of experiment was 3.5% TPH, and represents a yield of 65% attributed to natural attenuation process.

In soil polluted with 10% crude oil, the bioremediation process yield increased due to the complexity of treatments applied as follows: in soil conditioned by 0.5% (100 g/pot) Ecosol, petroleum hydrocarbon content determined in the pot at the end of the experiment was 2.82% TPH, corresponding to a yield of 71.8%, in the variant conditioned by 0.5% (100 g/pot)

Ecosol and inoculated with selected bacteria, final petroleum hydrocarbon content was 2.59%, corresponding to a yield of 74.1%, the variant in which the soil was conditioned with 1% (200 g/pot) Ecosol, the final content of petroleum hydrocarbons was 2.10%, representing a yield of 79% and, finally, the variant in which soil was conditioned with 1% (200 g/pot Ecosol) and inoculated with selected bacteria, petroleum hydrocarbon content at the end of the experiment was only 1.95%, representing a yield of 80.5%

In variants excessively polluted with 10% crude oil, microorganisms with biodegradation capacity of petroleum hydrocarbons have required a more or less period of time to adapt to pollutant presence in excess. Necessary period to adapt to environmental changing, microorganisms was significantly decreased by soil conditioning with Ecosol at 1% concentration.

CONCLUSIONS

Optimal variant of bioremediation in both experimental groups proved to be one in which soil was conditioned with Ecosol maximum dose (0.5% for soil polluted with 5% crude oil and 1% for soil polluted with 10% crude oil) and inoculated with selected bacteria, the two bioremediation technology work synergistically to remove the pollutant from the soil.

At the end of the experiment, the efficiency of bioremediation process in optimal variants was 92.6% in case of soil polluted with 5% crude oil and 80.5% for excessively polluted soil with 10% crude oil.

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