

**A Process for the Simplified Biological Rehabilitation of Land Polluted
with Long-term Contamination Based on Mineral Oil**

As is now known and understood, the careless handling for decades of mineral oils, mineral oil fractions and products derived from them as their economic exploitation has grown has resulted worldwide in considerable damage which is now also known as long-term contamination. A large part of this long-term contamination formed by hydrocarbon compounds based on mineral oil (hereinafter referred to in shorts as MHCs) affects ground where, for example, corresponding MHCs have been stored, pumped, distributed and/or transported for decades. Corresponding storage tanks for the industrial exploitation of MHCs and/or pipelines for the transport of MHCs, possibly over long distances, are mentioned purely by way of example in this regard.

For some time, awareness of such irresponsible behaviour from the past has been increasing. On the one hand, it has led - at least in important industrialized countries - to far more stringent requirements in regard to the pollutant-free handling of the class of compounds in question. On the other hand, a large and extensive practical and printed "prior art" on the elimination of the damage inflicted, particularly in the form of corresponding long-term contamination, has grown up. Despite this meanwhile focussed image of the significance of the environmental problems involved here and despite considerable safety improvements in the handling of MHCs, there is still an urgent need today for the inexpensive and simple regeneration of wide areas of land which, in particular, are so heavily polluted by corresponding long-term contamination that simple rededication of the areas affected by local authorities is not possible. The polluted areas first have to be adequately cleaned to sufficient depths to enable them to be put to new uses.

Numerous proposals for solving the problems outlined above are known from the literature and from practice. The effectiveness of the particular cleaning procedures used is understandably a critical factor in determining the practicability of the particular method. However, it is not
5 the only determining factor. The cost involved is an at least equally important factor, particularly in the regeneration of relatively large areas of land.

Controlling the pollution of land and water by hydrocarbon compounds, for example MHCs, through bioremediation is acquiring
10 increasing significance. Microorganisms which consume hydrocarbon compounds are valuable tools in this technology providing their concentration and/or growth in the contaminated area can be sufficiently stimulated. Among the more recent literature, reference is made to the Articles in **Chemische Industrie** 5/91, 10-12 "Hunger auf Erdöl" and in
15 **Erdöl und Kohle - Erdgas** 44, April 1991, 197-200, Th. Höpner et al. "Die Ölkatastrophe im Persisch-Arabischen Golf" and the extensive literature cited therein.

Bioremediation is based on the principle of optimally promoting the growth of dirt-consuming microorganism populations. According to the
20 prior art, cf. for example **DE-A-42 18 243** and **PCT/EP92/02146**, two important auxiliaries are of paramount importance in this regard. First, growth-promoting elements have to be applied because they are generally not available in sufficient concentrations in the contaminated area. The growth promoters in question are primarily inorganic and/or organic
25 compounds of nitrogen and phosphorus which are available as nutrient concentrates for stimulation and as growth aids for the accelerated cultivation of the hydrocarbon-consuming microorganisms. Second, preformed concentrates of suitable hydrocarbon-consuming microorganisms which can be applied to the contaminated areas, for
30 example at weekly intervals, are often used according to the prior art

literature in question, in particular to accelerate biological degradation in the initial phase. However, depending on the particular situation, particularly the previous history of the area to be cleaned up, it may even be the case that there is no need for such inoculation with microorganism concentrates. In general, this will always be the case when the natural process of biological degradation has already resulted in the development of sufficient concentrations of microorganism strains, cf. in this connection the second of the literature references cited above.

The teaching according to the present invention is based on the fact that, particularly where long-term contamination is involved, the basically possible natural degradation process by remediation takes place to only a negligible extent, if at all, under local ground conditions, climatic conditions and the like. The problem addressed by the invention was to enable land to be cleaned up by remediation to considerable depths of the soil layers affected under the particular conditions prevailing without any need for labour-intensive and hence expensive measures, such as the known excavation of the earth affected and its treatment under optimized climatic conditions at a tip.

Before the measures taken by the teaching according to the invention to solve this problem are discussed in detail, reference is made to a special feature which is directly related to the understanding of the teaching according to the invention and the application of the measures according to the invention. Soil samples of typical long-term contaminated areas taken separately from one another and at vertically increasing depths often show a pollutant pattern which is evidently characteristic of the particular contaminated area. Particularly long-chain and/or cyclic hydrocarbon compounds of high molecular weight, which are distinguished by particularly difficult degradability, are concentrated in the upper soil layers. Depending on the particle structure of the soil affected, these upper layers are followed at sometimes considerable depths by zones where the

pollutants appear to migrate more readily. Considerable concentrations of unwanted pollutants may even be present at groundwater level. Depending on their molecular structure and size, these hydrocarbon compounds are evidently capable of migrating through the soil structure, even over prolonged periods. They then float as it were on the surface of the groundwater and, over decades, harden on the surfaces of the soil particles providing they are not discharged via the groundwater. The subsequent explanation of the inadequate rehabilitation of these areas of land affected by long-term contamination is interesting and, ultimately, also understandable: the degradability of the comparatively large HC molecules in the upper soil layers so seriously impeded by the hydrocarbon structure prevents the natural development of hydrocarbon-consuming microorganism strains although relatively large amounts of atmospheric oxygen are available there for promoting bacterial growth. This impeding of degradation is generally intensified by a lack of essential nutrients, more particularly P and N. The more readily migrating HC components are flushed to such depths in the soil that a serious insufficiency of oxygen and possibly even a lack of essential nutrient components exist.

Starting out from these fundamental observations, the teaching according to the invention now proposes for the first time an extremely simple and hence inexpensive process - suitable even for large areas of land - for eliminating MHC contamination with which in particular areas of virtually any size polluted by long-term contamination can be cleaned.

Subject of the invention

Accordingly, the present invention relates to a process for the simplified biological rehabilitation of land areas of virtually any size which are polluted by mineral-oil-based hydrocarbons (MHCs) and, in particular, by corresponding long-term contamination by treatment with nutrient concentrates for stimulation and growth promotion for the accelerated

cultivation of MHC-consuming microorganisms. The process according to the invention is characterized in that,

- 5 (a) after preliminary moistening/watering where necessary,
- (b1) on the one hand, the ground to be rehabilitated is broken up to a sufficient depth and turned over in at least one operation,
- 10 (b2) and on the other hand the nutrient concentrate is applied in the form of a water-diluted emulsion and/or dispersion, more particularly by spraying in substantially uniform distribution, again in at least one operation and
- (c) the area thus treated is intensively watered.
- 15

In one preferred embodiment of the invention, steps (a), (b2) and (c) at least are repeated at relatively long intervals. Preferred intervals are of the order of 1 to 6 months. Repetition at least 1 to 3 times is advisable.

20 In other preferred embodiments of the invention, concentrates of microorganisms capable of degrading MHC compounds are additionally applied to the contaminated area. The use of corresponding microorganism concentrates of natural origin is a particularly important embodiment in this regard. In one particular embodiment of the teaching according to the invention intended in particular for the elimination of the

25 MHCs present in relatively deep layers of soil, the relatively deep layers of soil are either additionally aerated and/or oxygen chemically combined and released in the soil is introduced. Particulars of this are discussed in the following.

Particulars of the teaching according to the invention

The foregoing definition of the process steps according to the invention makes it understandable that, for the first time to their knowledge, applicants are proposing such a technically uncomplicated approach to the elimination of MHCs by bioremediation that areas of land of virtually any size and virtually any level of MHC pollution in terms of amount and age can be subjected to biological rehabilitation under acceptable working and cost conditions. More specifically:

The area of land to be rehabilitated should first be sufficiently moistened or watered. This preliminary step can make use of the natural process of rainfall and of the simple method of artificial watering. The soil surface generally compacted by decades of mechanical compression should be broken up to a sufficient depth and turned over in another operation. Preferred depths are at least 15 cm, depths of $\geq$ about 30 cm, being particularly preferred. It will readily be appreciated that agricultural Instruments, such as harrows and especially ploughs, can be used to satisfy this requirement of modifying the soil surface to be rehabilitated. In some cases, it may be preferred to break up and turn over the surface soil to greater depths, for example of about 50 to 80 cm or more. In cases such as these, the modern technology of grubbing enables the above-mentioned requirement to be comfortably satisfied.

The mechanical breaking up of the soil surface described here leads in several ways to optimization of the cleaning results to be achieved in accordance with the invention by biological rehabilitation. The earth broken up and turned over is accessible to an increased air supply and existing seepage channels for rainwater in this upper layer of soil are destroyed so that uniform moistening of this layer is guaranteed. At the same time, however, it is the layer of the soil to be rehabilitated in which non-readily degradable, comparatively large MHC components have concentrated and hardened. The described restructuring creates a totally new starting situation for the following biological remediation.

According to another aspect of the teaching according to the invention, the nutrient concentrate required for the biological rehabilitation is introduced into the soil surface in at least one operation. This nutrient concentrate is applied in particular in the form of water-diluted solutions, emulsions and/or dispersions. Application by substantially uniformly distributed spraying can be particularly useful.

A concluding step of the process according to the invention comprises intensive watering. In this way, the nutrient concentrate sprayed on is uniformly distributed throughout the broken-up soil. In addition, parts of the nutrient concentrate are able to seep into deeper layers of soil untouched by the mechanical breaking-up treatment.

The above-described sequence of process steps is distinguished in particular by the fact that steps (b1) and (b2) can be carried out one or more times in various combinations. Particularly important combinations of steps (b1) and (b2) are characterized by one of the following sequences:

(b1) $\Rightarrow$ (b2), i.e. the surface to be rehabilitated is first broken up and the nutrient concentrate is subsequently applied. However, (b2) $\Rightarrow$ (b1) is also possible, i.e. the nutrient concentrate is first applied to the as yet untreated surface to be rehabilitated, after which the surface is broken up.

In important other embodiments, step (b1) and/or step (b2) is/are repeated several times, preferably twice. This provides for the other possible and possibly preferred combinations of steps (b1) and (b2) in the following sequences: (b1) $\Rightarrow$ (b2) $\Rightarrow$ (b1) or (b2) $\Rightarrow$ (b1) $\Rightarrow$ (b2).

In another embodiment of the invention, the watering steps (a) and (c) may be carried out in one and/or more stages. As mentioned above, step (a) is redundant where the soil is wet with rain. Similarly, intensive watering may be unnecessary where application of the nutrient concentrate in step (b2) is immediately preceded or accompanied by rainfall.

Although the details of the multistage procedure according to the invention are influenced in each case by the particular soil formation and/or

the level of the MHC contamination, the repetition one or more times of at least the application of the nutrient concentrate for the biological rehabilitation under suitable moisture conditions of the ground to be treated and, in particular, subsequent watering in one or more stages (step c) are
5 generally advisable and preferred. The time interval between the individual applications of the nutrient concentrate has to be adapted to the particular conditions prevailing. In general, it is at least about 1 to 2 weeks although application at longer intervals, for example 1 to 6 months, can be particularly useful.

10 The number of repetitions of the rehabilitation measure according to the invention is in turn determined by the particular circumstances prevailing. The now standard testing of the various vertical layers of the contaminated soil at regular intervals for pollutant levels provides information as to whether and to what extent further repetition of the
15 measure according to the invention is desirable or even appropriate. Generally, it has been found that repetition of these measures once to three times is sufficient for rehabilitating even deeper layers of soil providing barrier layers within the soil are not an obstacle.

It is known that the growth of suitable MHC-consuming
20 microorganism strains under aerobic conditions is also controlled by the available oxygen. Additional measures may be necessary here, particularly in the deeper layers, when on the one hand considerable MHC pollution levels are present and, on the other hand, these pollution levels are unacceptable. The teaching according to the invention can avail itself in
25 this regard of a number of known technical measures which provide assistance in special cases such as these. For example, the oxygen required can be introduced into the critical area in chemically combined form, more particularly in the form of hydrogen peroxide. Corresponding commercial products have been developed by industry for the purpose in
30 question and are freely available (cf. for example the product marketed by

Degussa AG under the registered name of "RENATox". However, atmospheric oxygen may also be brought into the critical area, for example by laying sufficiently deep exhaust shafts from which the gas phase is pumped off so that air is forced to flow through the surrounding soil formation, cf. for example the publication of the Bundesministerium für Forschung und Technologie, Förderkennzeichen 1460505, **"Technologieregister zur Sanierung von Altlasten"**, B. Böhnke and K. Pöppinghaus, December 1990, in particular pages 31 and 32.

However, it is again emphasized in this regard that the teaching according to the invention is not confined solely to the elimination of MHC-based long-term contamination to unlimited depths of the soil formations affected. The invention seeks primarily to open up and clean the soil layers which are affected by the impact of everyday life, such as plant growth, erection of buildings and the like. In these layers of comparatively limited depth, very simple and hence inexpensive and effective cleaning is necessary and is the primary objective of the invention.

In this connection, reference is made to another possibility which the elements of the teaching according to the invention support through an adapted (intermediate) ground-care measure involving the areas of land to be regenerated:

For example, it can be useful according to the invention to plant the loosened surface of the soil to be treated with deep-rooting plants, a typical example being the cultivation of sunflowers. At the same time as or instead of this example, seeds of N-forming plants can be sown in the soil to be treated. Examples of suitable plants are alfalfa, clover and oil radish. Basically, representatives from the class of pulses are suitable in this regard.

The nutrients or nutrient mixtures used in accordance with the invention for cultivating the MHC-consuming microorganism strains are discussed in more detail in the following. Basically, general expert

knowledge as described, for example, in DE-A-42 18 243 mentioned at the beginning and the literature cited therein is applicable in this regard. The disclosure of DE-A-42 18 243 on the characteristics of suitable and, in particular, preferred nutrients or nutrient mixtures is hereby expressly
5 included in the disclosure of the present invention. Accordingly, extracts from that disclosure are expressly discussed once more in the following.

First, the fact known among experts that the nutrient media may consist both of conventional synthetic and/or natural fertilizers and, in particular, of at least substantially hydrophilic components also applies to
10 the teaching according to the invention. However, mixtures containing at least partly hydrophobic components and, in particular, corresponding compounds of P and/or N can afford particular advantages in the field of application in question. Thus, corresponding compounds of phosphorus (P) and nitrogen (N) can be provided with hydrophobic molecule
15 components so that these molecules attach themselves to the MHC-polluted soil. The advantage of such an interaction between the contamination to be eliminated from the soil and the nutrient or nutrient mixture for promoting the required bacterial growth will be readily appreciated: the growth of the required bacteria and hence the degradation
20 of the MHC-based contamination is optimally promoted. In general, the hydrophobicized components of the nutrient mixture are not washed out downwards into the groundwater whereas any standard inorganic and/or organic, natural and/or synthetic fertilizer components used are easily but undesirably washed out in this way.

25 The nutrient concentrates containing hydrophobicized components are used in known manner together with emulsifying and/or suspending aids. However, the same function can also be at least partly performed by one or more components of nutrient character. Suitable emulsifying and/or suspending aids are the known wetting agents based on alkyl

oligoglucosides of which further particulars can be found in DE-A-42 18 243 cited above.

Preferred nutrient concentrates according to the invention contain the water- and/or oil-soluble compounds of P and/or N in admixture with
5 other water- and/or oil-soluble organic components which are at least partly nutrients for the growth of hydrocarbon-consuming microorganisms. Water-based preparations containing an ester of phosphoric acid as emulsifier and P source and, if desired, one or more water-soluble or water-dispersible N sources are particularly suitable. It can be useful to apply
10 nutrient concentrates containing 10 to 40% by weight of emulsifying phosphoric acid ester, 10 to 40% by weight of the N source and, for the rest, water. Nutrient concentrates containing 20 to 30% by weight of the emulsifying phosphoric acid ester, 15 to 30% by weight of the N source, 0.5 to 5% by weight of anionic surfactant compounds and, for the rest, water
15 may be used with particular advantage. Nutrient concentrates containing phospholipids, alkyl phosphates and/or alkylether phosphates as phosphoric acid esters are suitable representatives in the context of the teaching according to the invention. The N source is generally present in the form of inorganically and/or organically combined nitrogen, a preferred
20 N source being urea which may be present in the nutrient mixture in quantities of, in particular, 10 to 50% by weight. Besides the above-mentioned alkyl glycoside compounds - preferably obtained from straight-chain C₈₋₂₄ fatty alcohols and mono- and/or oligoglucosides - suitable nonionic surfactant compounds are sugar partial esters of C₈₋₂₄
25 monocarboxylic acids, sorbitan esters and/or biosurfactants of biological origin. Examples of such nonionic surfactants are sophorose lipid, trehalose lipid and lipopeptides.

Other preferred elements of the invention are summarized in the following with reference to the specific disclosure of DE-A-42 18 243:

Particular significance is attributed to the phosphoric acid esters used which are preferably selected so that, on the one hand, they act as a P source and, on the other hand, have an emulsifying effect. Preferred phosphoric acid esters are taken up by the microorganisms to be cultivated. Accordingly, among the compounds which may be used, the phospholipids are particularly important. Phospholipids are amphiphilic substances which are obtained from vegetable or animal cells. Preferred phospholipids are the glycerophospholipids which, normally, are also referred to as lecithin. Known and suitable phospholipids are the diacyl phospholipids, phosphatidyl cholines, phosphatidyl ethanolamines, phosphatidyl inositols, phosphatidyl serines, phosphatidyl glycerols, phosphatidyl glycerophosphates, diphosphatidyl glycerol, N-acyl phosphatidyl ethanolamine and phosphatidic acid. Monoacylphospholipids, lysophosphatidyl cholines, lysophosphatidyl ethanolamines, lysophosphatidyl inositols, lysophosphatidyl serines, lysophosphatidyl glycerols, lysophosphatidyl glycerophosphates, lysophosphatidyl glycerols, lyso-n-acylphosphatidyl ethanolamines and lysophosphatidic acid are preferred. By virtue of their accessibility, the expert will always opt for commercially available phosphatidyl glycerides which are marketed as vegetable or animal lecithins or cephalins. These preparations are mostly obtained from oils, such as corn oil or cottonseed oil. The enzyme-hydrolyzed glycerophospholipids (enzyme-hydrolyzed lecithin), which are relatively hydrophilic in character through the elimination of a fatty acid residue, are preferred for the purposes of the invention. The only exception are products which have lost their phosphoric acid residue through the enzymatic hydrolysis.

In addition to or instead of the phospholipids mentioned, partial esters of phosphoric acid with fatty alcohols, more particularly with straight-chain fatty alcohols, may be used as the P source of the nutrient

concentrates. Relevant particulars can be found in the disclosure of DE-A-42 18 243.

As their N source, the nutrient concentrates contain corresponding compounds in the form of inorganically and/or organically combined
5 nitrogen. N sources containing only organically combined nitrogen may be preferred. Inorganic salts, such as alkali metal nitrate or nitrite, or even ammonium salts are suitable N sources. Organic N sources are, for example, aminocarboxylic acids, for example of the glutamic acid type. Urea has proved to be particularly accessible for controlling and promoting
10 microorganism growth. At the same time, it has a preserving effect against unwanted microbial contamination so that the nutrient concentrates show high stability in storage. Where phospholipids are used as the P source, there is no need to use an additional N source because phospholipids contain both P and N.

15 So far as the surface-active mixture components optionally used are concerned, reference is made to the foregoing observations, particularly on the alkyl glycoside compounds, and to the special disclosure of DE-A-42 18 243. The surfactant compounds are normally present in the nutrient concentrate in quantities of 0.5 to 5% by weight. In general, quantities of
20 up to about 1% by weight of surfactant are sufficient to initiate and promote the intended impact stimulation of microorganism growth. Thus, surfactants based on alkyl glycoside compounds may be used in quantities of about 0.5 to 1% by weight, based on the mixture as a whole, for effectively improving microorganism growth. Further particulars,
25 particularly in regard to suitable working parameters, can be found in DE-A-42 18243.

For normal use, the nutrient concentrates are preferably diluted with water in a ratio of 10 to 50:1 and introduced into the contaminated soil one or more times in quantities of 5 to 200 mg per g of contamination.

Examples

Four test plots of ca. 30 m² on a site contaminated over decades by surface pollution with fuels were selected.

The test plots consisted to a depth of three metres of readily permeable medium sands with some fine sand components. The upper 20 cm thick layer was humous. The seasonally fluctuating groundwater level was on average at 2 metres.

Various pollutant concentration levels were determined downwards through the layers of sand as far as the groundwater level, the zones of highest contamination being situated at a depth of 0.8 to 1.5 metre.

The following treatment schedule was selected for land rehabilitation:

Treatment	Test Plot 1	Test Plot 2	Test Plot 3	Test Plot 4
Pretreatment	Watering	• Uprooting existing vegetation • Watering	Watering	No treatment
Fertilizing measure	Nutrient concentrate acc. to DE-A-42 18 243	Nutrient concentrate acc. to DE-A-42 18 243	Fertilization with agricultural mineral fertilizer	No treatment
Working-in	Ploughing	Ploughing	Ploughing	No treatment
Aftertreatment	Watering	Watering	Watering	No treatment
Repetition of fertilizing measure after 6 weeks	Nutrient concentrate acc. to DE-A-42 18 243	Nutrient concentrate acc. to DE-A-42 18 243	Fertilization with mineral fertilizer	No treatment

The natural biological degradation process had been monitored for the previous three years and, as expected, was hardly progressing because of the lack of nutrients in the soil. The starting contamination levels at the beginning of the test were as follows:

Contamination levels	Test Plot 1 in mg/kg	Test Plot 2 in mg/kg	Test Plot 3 in mg/kg	Test Plot 4 in mg/kg
Upper soil layer	2,300	1,300	2,300	800
Unsaturated region at depth 0.02-0.8 m (outside the range of variation of the water table)	3,800	10,200	2,100	4,300
Unsaturated region at depth 0.8 - 1.50 m (outside the range of variation of the water table)	3,700	20,000	4,000	10,200
Saturated region (in the range of variation of the water table)	360	2,700	1,800	2,400

After a reaction time lasting from April to August, the following pollutant reductions were observed:

Contamination levels	Test Plot 1 in mg/kg	Test Plot 2 in mg/kg	Test Plot 3 in mg/kg	Test Plot 4 in mg/kg
Upper soil layer	1,100	700	2,400	700
Pollutant reduction by	52%	46%	None	None
Unsaturated region at depth 0.02-0.8 m (outside the range of variation of the water table)	1,800 53%	10,200 None	2,300 None	None
Unsaturated region at depth 0.8 - 1.50 m (outside the range of variation of the water table)	2,600 30%	13,800 31%	4,100 None	10,200 None
Saturated region (in the range of variation of the water table)	160 55%	2,100 22%	1,900 None	2,600 None

The results show that the natural degradation process can be considerably accelerated by the use of suitable nutrients, such as the nutrient mixture described in DE-A-42 18 243. As expected, this effect is particularly clear in the unsaturated soil region and satisfactory in the saturated region. The uprooting of existing vegetation - in this case weeds - does not produce a significant advantage and is only recommended for impassable terrain. In addition, it was found that conventional agricultural fertilizers are unsuitable for stimulating degradation because the nutrients are washed out very quickly.

**CLAIMS**

1. A process for the simplified biological rehabilitation of land areas of virtually any size which are polluted by mineral-oil-based hydrocarbons (MHCs) and, in particular, by corresponding long-term contamination by treatment with nutrient concentrates for stimulation and growth promotion for the accelerated cultivation of MHC-consuming microorganisms, characterized in that,
- 5
- (a) after preliminary moistening/watering where necessary,
- 10
- (b1) on the one hand, the ground to be rehabilitated is broken up to a sufficient depth and turned over in at least one operation,
- (b2) and on the other hand the nutrient concentrate is applied in the form of a water-diluted emulsion and/or dispersion, more particularly by spraying in substantially uniform distribution, again in at least one operation and
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- (c) the area thus treated is intensively watered.
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2. A process as claimed in claim 1, characterized in that steps (b1) and (b2) are carried out in one of the following sequences: (b1) $\Rightarrow$ (b2); (b2) $\Rightarrow$ (b1); (b1) $\Rightarrow$ (b2) $\Rightarrow$ (b1) or (b2) $\Rightarrow$ (b1) $\Rightarrow$ (b2).
3. A process as claimed in claims 1 and 2, characterized in that, in step(s) (b1), the soil surface is broken up and turned over to a depth of at least 15 cm and preferably $\geq$ 30 cm, depths of 50 to 80 cm being particularly preferred.
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4. A process as claimed in claims 1 to 3, characterized in that watering steps (a) and (c) may be carried out in one and/or more stages, step (a) being redundant where the soil is wet with rain.
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5. A process as claimed in claims 1 to 4, characterized in that watering in one and/or more stage in step (c) is intensified so that at least the soil layer loosened in step (b1) is penetrated by the nutrient mixture applied, although parts of the multicomponent mixture are preferably introduced into deeper soil layers.
6. A process as claimed in claims 1 to 5, characterized in that steps (a), (b2) and (c) at least are repeated at relatively long time intervals, preferred time intervals being 1 to 6 months and at least one to three repetitions being useful.
7. A process as claimed in claims 1 to 6, characterized in that the nutrient components used and, in particular, the corresponding compounds of phosphorus (P) and nitrogen (N) are provided with hydrophobic molecule components so that these molecules attach themselves to the MHC-polluted soil.
8. A process as claimed in claims 1 to 7, characterized in that the nutrient concentrates used in step (b2) contain emulsifying and/or suspending aids, the same function also being performed at least partly by one or more components of nutrient character, although corresponding added components are at least partly present in the nutrient concentrate.
9. A process as claimed in claims 1 to 8, characterized in that alkyl oligoglucosides are used as emulsifying and wetting aids in the nutrient concentrate.
10. A process as claimed in claims 1 to 9, characterized in that the nutrient concentrates used contain water- and/or oil-soluble compounds of phosphorus (P) and nitrogen (N) in admixture with other water- and/or oil-soluble organic mixture components which are at least partly nutrients for the growth of hydrocarbon-consuming microorganisms, water-based preparations containing an ester of phosphoric acid as emulsifier and P source and optionally one or more water-soluble or water-dispersible N sources being preferred.



11. A process as claimed in claims 1 to 10, characterized in that nutrient concentrates containing 10 to 40% by weight of emulsifying phosphoric acid esters, 10 to 40% by weight of N source and, for the rest, water are used.
- 5 12. A process as claimed in claims 1 to 11, characterized in that nutrient concentrates containing 20 to 30% by weight of the emulsifying phosphoric acid ester, 15 to 30% y weight of N source, 0.5 to 5% by weight of nonionic surfactant compounds and, for the rest water, are used.
- 10 13. A process as claimed in claims 1 to 12, characterized in that nutrient concentrates containing phospholipids, alkyl phosphates and/or alkylether phosphates as phosphoric acid esters are used.
14. A process as claimed in claims 1 to 13, characterized in that the N source is present in the nutrient concentrate in the form of inorganically and/or organically combined nitrogen, urea being a preferred N source
- 15 which may be present in the nutrient mixture in quantities of, in particular, about 10 to 50% by weight.
15. A process as claimed in claims 1 to 14, characterized in that nutrient concentrates containing alkyl glycoside compounds obtained from preferably straight-chain C₈₋₂₄ fatty alcohols and mono- and/or
- 20 oligoglycosides, sugar partial esters of C₈₋₂₄ monocarboxylic acids, sorbitan esters and/or biosurfactants of biological origin, such as sophorose lipid, trehalose lipid and lipopeptides, as nonionic surfactants are used.
16. A process as claimed in claims 1 to 15, characterized in that the nutrient concentrates are diluted with water in a ratio of 10 to 50:1 and are
- 25 introduced into the contaminated soil one or more times in quantities of 5 mg to 200 mg per g of contamination.
17. A process as claimed in claims 1 to 16, characterized in that concentrates of microorganisms which are capable of degrading hydrocarbon compounds and which are preferably of natural origin are
- 30 additionally applied to the soil surface.



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18. A process as claimed in claims 1 to 17, characterized in that it additionally comprises the forced aeration of deeper layers of soil and/or the introduction of combined oxygen which is released in the soil.