

Analysis of the effects of oil pollution of soils

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ABSTRACT :Acest articol face o introducere în efectele poluării cu petrol a solurilor și de asemenea tratează modul de determinare a nivelului de poluare. De asemenea sunt prezentate și modul de comportare a poluantului asupra vieții microroganismelor. Un subcapitol este dedicat analizei unei poluării accidentale cu petrol din zona Dobrogea, Romania.

KEYWORDS oil pollution, soil, rehabilitation.

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I. INTRODUCTION

The development of the oil industry, both extractive and processing, including the transportation of oil and oil products, is sometimes accompanied by unforeseen secondary phenomena with more or less harmful effects on the environment and human life.

One of these phenomena is soil pollution with oil residues with or without salt water and other residual oil products resulting from the exploitation and extraction of crude oil.

Accidents involving the breaking of pipelines that transport salt water or oil products are also very harmful, especially when pipelines pass through arable land where agriculture is concentrated.

Knowing the changes produced in the soil by the pollution of salt water and petroleum materials is very necessary to explain these polluted soils' behavior and establish measures to restore their economic potential.

The comparative study of polluted and unpolluted soils presented in this paper provides some data in this regard [1].

Oil residues (we called them specific oil pollutants most often determined in ecological accidents in Romania) are complex mixtures of hydrocarbons with a relatively low content of impurities or polluting substances, such as oxygen, nitrogen, sulfur, and various metals [2].

Oxygen compounds are made up of naphthenic acids and phenols.

Nitrogen compounds are derivatives of pyridine, quinoline, and pyrrole.

Sulfur is found in crude oil in amounts from less than 0.08% to 1.5% in elemental sulfur, hydrogen sulfide, mercaptans, sulfides, and thiophene.

Oil residues are made up of hydrocarbons, which represent about 98% of the total. The elemental composition is expressed by the C/N ratio, which varies with the structure of the hydrocarbons.

The following classes of hydrocarbons are found in oil residues[3]:

- saturated acyclic hydrocarbons-alkanes;
- cyclic saturated hydrocarbons-cycloalkanes;
- mononuclear aromatic hydrocarbons;
- condensed polynuclear aromatic hydrocarbons.

The research shows that crude oil residues are complex mixtures of substances and that crude oil has not been separated into its components in the soil.

Analyzing these from the soil determines their total and sometimes fractional amounts.

The analytical methods used to determine oil residues consist of their extraction with a selective solvent (hot or cold), followed by the separation of the main phases through a silica gel column, elution according to their polarity with the heliotrope series hexane-benzene-benzene: methanol and gravimetric dosing.

The properties of oil, the most important from the point of view of interaction with soil and water, are:

- the state of aeration-mixture of liquid compounds in which solid and gaseous compounds are dissolved;
- hydrophobic character - repels water molecules, is immiscible in water, forms a superficial film;
- high content of organic carbon;
- very high C/N ratio.

Hydrocarbons enter the body via the respiratory route (the volatile ones) or the digestive or skin route. They affect the nervous system, digestion, skin conditions, fatigue, headache, liver disorders, nausea, folliculitis, and dermatitis.

Chemical complexation sometimes causes the appearance of particularly toxic compounds, such as organic compounds containing chlorine, arsenic, sulfur, etc.

Some hydrocarbons have a narcotic effect: chloroform, ethyl chloride, methyl chloride.

Polycyclic aromatic hydrocarbons hurt cell metabolism, cell reproduction, and hematopoiesis.

Nitro and amino derivatives of polycyclic hydrocarbons block hemoglobin in the form of methemoglobin, reduce work capacity and have carcinogenic action.

Pollution with aliphatic hydrocarbons is caused by the oil industry and the secondary use of products in the organic chemical industry (petrochemistry, rubber, plastics, pesticides) and metallurgy.

Water pollution, directly or through the soil, is a large-scale process caused by waste water spills or accidents and oil infiltration into underground water layers.

The methane content is approx. $500 \cdot 10^6$ t/year and its content is constantly increasing by about 1% per year.

The increase in methane content is due to the increase in the areas cultivated with rice, the increase in cattle, the losses from obtaining and distributing and using natural gas and oil; the other sources are natural and represent approx. 30% of total methane emissions.

Methane plays a significant role in thinning the ozone layer.

Other hydrocarbons, such as terpenes, are anthropogenically emitted in quantities lower than natural sources.

Fluoro-chloro hydrocarbons come from the industrial branches that use them as foaming agents, cooling agents, solvents, fire extinguishers because they are very effective, they are free of toxicity for living things, they are clean, non-flammable and non-aggressive towards materials, thermally stable. $400 \cdot 10^3$ t were used annually. Because the increase in their concentration in the atmosphere was 5%/year (the sum of all compounds containing chlorine is 3ppm in the stratosphere and 0.7ppm in the troposphere) and because of their effect of destroying the ozone layer, prohibition laws were issued of their use.

In 2024, the production of chlorofluorocarbons decreased by 50%; research is being done to obtain substitutes (eg cyclopentane as a refrigerant).

Aromatic hydrocarbon pollution occurs in numerous branches of the modern organic chemical industry (dyes, rubber, plastics, synthetic resins, detergents, explosives, antioxidants).

The organic substances generated by various industries are varied and produce pollution both in the industrial atmosphere and in the surrounding environment; these are alcohols, phenols, ethers, esters, aldehydes and ketones, carboxylic acids, nitriles, aromatic amines, azo compounds, polychlorinated biphenyls, and polycyclic aromatic hydrocarbons: 3,4 benzpyrene, methylchloranthrene, benzfluoranthrene, benzanthracenes, and tetramethyl phenanthrene.

II. OIL POLLUTION ASSESSMENT

Soil pollution means any action that causes the disruption of the normal functioning of the soil as a support and living environment within the various natural or artificial ecosystems, disruption manifested by the physical, chemical, or biological degradation of the soil and the appearance in the soil of some characteristics that reflect the deterioration of fertility its, respectively the reduction of the productive capacity both qualitatively and quantitatively[4].

In the first hours after the spill, the most volatile hydrocarbons (most of the C13 and C14 components) evaporate. Also, the solubilization of light hydrocarbons and the transformation by photooxidation under ultraviolet solar rays occur.

The immediate effect of soil pollution with oil residues is reflected in the disruption of microbiological activity in the soil. Thus, in the first weeks after the spill, depending on the intensity of the pollution, there is a reduction in microbial activity in the soil, a situation confirmed by the analysis of soil respiration (CO₂ release) and enzymatic (dehydrogenase) activity. Then, under optimal temperature conditions, there is a tendency to return to the regular microbiological activity of the soil because numerous bacteria, yeasts, and even algae can biodegrade the hydrocarbons.

Therefore, biodegradation appears as an operational means for combating oil pollution, and it is considered that the strategy of soil depollution affected by contamination with oil residues consists of favoring the natural means of their elimination.

Oil applied to the soil in small quantities can, after a few months of pollution, lead to bacterial degradation that can improve the soil's physical properties.

In soils polluted with drilling mud rich in oil, oil losses due to volatilization cannot be differentiated from those due to microbial degradation. It is estimated that in this case, the loss of oil due only to microbial degradation is 8.3 kg/m³ per month, regardless of the spilled quantities.

After 18 months after the pollution, the oil turned into naphthenic acids, which decomposed very slowly.

The degree of pollution also affects the microbial degradation. Thus, in the case of 5% pollution, ammonification decreases by 40-45% in the first 7 days and by 35% in the following days. Also, a content of 1% inhibits ammonification, which leads to the inhibition and death of bacteria.

Studies carried out on the process of biological assimilation for petroleum products showed an oil degradation rate of 1.425% in the first month; after three months, it rose to 61.4%.

In addition to natural means, research was done to intensify the biodegradation of hydrocarbons. For this purpose, two ways were considered. The first refers to the selection of hydrocarbon-consuming bacteria, then the creation of cultures and their preservation (by biophilization, for example) to be seeded in hydrocarbon-polluted environments.

Some products have given results for specific terrain and climate conditions.

However, their use in other conditions (marine environments, for example) has yet to yield satisfactory results.

The explanation is that when a foreign species is applied in a natural environment (even if it is native to this environment), a rejection phenomenon occurs, a well-known phenomenon in medicine. As a result, the use of microorganisms existing in the respective environment (bacteria) was considered to provide them with nutrients to be as efficient as possible in the biodegradation of oil residues.

Nitrogen and phosphorus fertilizers are applied to favor the degradation rate, which doubles the degradation rate.

Over 200 species of microorganisms (bacteria, yeasts, microscopic fungi) capable of transforming oil residues into degradation products (metabolizing hydrocarbons) have been identified in the last decades.

These are primarily anaerobic bacteria, i.e., oxygen-consuming, the most common in the natural environment: *Pseudomonas*, *Acinetobacter*, *Flavobacterium*, *Corynebacterium*, and *Arthrobacter*[5].

In the soils of oil fields in Kuwait, it was isolated, along with other bacteria from those mentioned above, and *Rhodococcus rhodochrous*, which proved to be one of the most efficient bacteria that degrade oil.

Oil spilled on the ground, aquatic sediments, and surface water led to the disappearance of some invertebrates (*Chironomidae*, *Simuliidae*, and *Oligochaeta*), reappearing after a while (532 days).

Some plants' roots resist oil-polluted soils, only reducing their development and fruiting. This is the case of the vigorous *Spartina* that grows on sandy soils.

Oil degradation in the soil also depends on the amount spilled and the degree of pollutant loading. At particularly high loading levels, 1,3,5,10 or over 10%, the oil fractions susceptible to microbial attack are preferably degraded.

It was determined on a soil polluted with a variety of oils that contained 22% paraffin, 28% aromatic hydrocarbons, and 50% asphaltenes; after 22 months, following microbial degradation, it still had 18% paraffin, 40% aromatics, and 90% from asphaltenes. By adding a surplus of 50 ppm N and 20 ppm P to the initial soil content, this amount was found to be sufficient to ensure the degradation.

Soil respiration is also an indicator of degradation processes under the influence of microorganisms. Thus, dusty soil polluted in a proportion of 0.5 g/100 g soil suddenly changes its breathing after pollution. In the first week, there is a sudden drop in the CO₂ content emitted, maintained until the 4th week when the CO₂ content from breathing stabilizes or drops very little. This decrease in CO₂ emission lasts until the 12th week when it starts to increase again.

The fastest decomposition occurs in the first four weeks when the maximum weekly decomposition is 2.5-8 metric t/ha out of 11.2 and 112 metric t applied, respectively.

In an experiment carried out by marking hexadecane with C¹⁴, almost 93% of the organic carbon in the polluted soil was oxidized into carbon dioxide, and 7% was assimilated into the cellular mass of *Cladosporium resinae*. When some amounts of nitrogen are added, the activity of this bacterium takes on an appreciable development.

Oil pollution changes the ratio between the different chemical elements. Thus, the contents of total C, N, K, Fe, and exchangeable Mn increased, and the levels of nitric nitrogen, mobile phosphorus, and exchangeable calcium decreased.

Little is known about cyclic alkanes with saturated carbons (cyclooctane and cyclohexane with the formula C_nH_{2n}). However, it was possible to highlight an oxidation reaction with the formation of acids (adipic acid and pimelic acid) during the degradation of decalin (hydrocarbons $C_{10}H_{18}$) by Flavobacterium.

The case of aromatic hydrocarbons (benzene C_6H_6 and its higher counterparts) is more complex. Depending on the nature of the microorganism and the aromatic hydrocarbons (according to the type of oil), very diverse intermediate compounds can appear, and the heterocyclic hydrocarbons can be degraded to the same extent as the aromatic ones[6].

The first factor limiting hydrocarbons' biodegradability is the limited number of microorganisms capable of metabolizing them. Another limiting factor is the lack of oxygen necessary for the oxidation reactions of biodegradation. Thus, for the complete oxidation of one milliliter of hydrocarbon, 3-4 mg of oxygen is needed.

Thus, polluted soil in the warm season is degraded in six months.

The biodegradation activity is insignificant between November and March. Maximum biological degradation occurs at a temperature of 30 °C and with a corresponding water content. Under optimal conditions, the half-life of oil, determined according to CO_2 , was 130 days in the case of refinery sludge pollution and 600 days in the case of petrochemical sludge pollution.

A series of experiments carried out on polluted soils in Dobrogea [3,4,5] show the following:

- from the initial amount added to the soil, the amount of oil that degraded after one year varied between 7% and 33%, depending on the type of soil and the treatments applied;
- more than 50% of the total amount of residue degraded in the first three months;
- the amount of CO_2 released in the first 4 weeks after incubation represented more than 25% of the amount of CO_2 released during one year;
- the addition of chemical fertilizers with nitrogen to some variants significantly increased the amount of degraded oil compared to the non-fertilized variants;
- salinity had a pronounced effect in reducing oil degradation, the degradation being about 2 times lower in the case of salinized soil;
- Waterlogging (excess water) reduces degradation.

In the warm season, the oil moves vertically in the areas unsaturated with water in the soil mass and laterally in the regions where it reaches the capillary strip. In the cold season, the soil's primary way of oil migration is through lateral movement. Usually, the oil does not exceed 20 cm deep in clayey soil, and microorganisms degrade all oil fractions.

Oil pollution has some effects on soil's physical properties. Oil pollution decreases the apparent density and increases the water holding capacity. In the long term, the situation improves, but in the short term (2-8 weeks), many physical properties can be negatively influenced.

Due to the effect of pollutants made up of various petroleum products, the state of aggregation of the soil increases, and the soil aggregates consolidate. For example, in the case of gasoline spilled on sandy-clay soil, the volume of residues was 6.5% of the soil volume, and the number of stable aggregates in water increased from 12.9 to 64%.

Field capacity and water retention capacity in the soil are also affected. Saturating soil with gasoline at a 6—and 12-cm depth increases the available water capacity. As the oil degrades, the effect of the structure and water retention capacity also decreases.

Due to oil flow through porous and permeable soils, surface tension forces can be predominant compared to gravitational forces.

The equation for the movement of oil through sandy-muddy soils is[7,8]:

$$Q * V = K * t^C, \quad (1)$$

Where: Q= volume growth rate of the soil in cm^3/min (after flooding with oil);

K=constant ;

$$C=(n+1-(1/x))/(n+2-(1/x)) \quad (2)$$

n=constant depending on the properties of the soil and is included between 3,4;

x= constant depending on the properties of the soil and is included between 0,75-0,82;

Soils polluted with oil show a dense, compact, glossy layer of oil on the surface, which prevents to a great extent the processes of infiltration of water into the soil, its circulation, reversible exchange of gaseous substances (oxygen, carbon dioxide, hydrogen sulfide, etc.) between the soil and the atmosphere, thus favoring the intense development of the reduction processes, with the release of some toxins, which cause the

asphyxiation of the roots and the death of the plants. The immediate effect of natural pollution or oil application on land is reducing air and water exchange between the soil and the atmosphere.

No agricultural or forestry plant species grow on the polluted lands, not weeds. The life of the plants is affected when the quantity of oil is greater than 1kg/m².

After the spill, when the oil comes into contact with the soil, there are changes in the physical, chemical, and biological properties of the soil, with the following phenomena taking place:

1. Volatilization of light compounds;
2. Stratification on the soil profile; the more polar components, especially the asphaltenes, remain on the surface of the soil, forming a compact film that prevents gas exchange between the soil and the atmosphere and that does not allow the normal circulation of water, thus creating unfavorable conditions of anaerobiosis and reduction.
3. Migration on the soil profile: the penetration of oil and oil residues on the soil profile, the depth to which they reach depends on the intensity of the pollution, its texture, and its apparent density. During the migration, a selective distribution takes place on the profile depending on the polarity of the components; saturated hydrocarbons penetrate to greater depths, followed by aromatic ones, and asphaltenes, as shown before, remain at the top of the soil profile.

Since oil and oil residues have a high content of organic carbon and a low content of nitrogen, the C/N ratio in the soil increases well above the normal value, making nitrogen the limiting factor for both plants and microorganisms.

Without any treatments, an essential part of the hydrocarbons slowly disappears under the action of natural phenomena such as evaporation, photooxidation, dissolution, and biodegradation.

If the spill is of short duration with a small flow, the influence on the soil is minimal, the agricultural works, the precipitation and the dryness of the summer succeeding in breaking down the hydrocarbons. As the frequency and flow rate of discharges in the same place increases, the balance of processes changes in the sense of decreasing soil aeration, development of anaerobic processes and damage to the root system of cultivated species. Where the puddles become persistent, there is a strong alkalization of the soils and/or the formation of a thick layer of up to 1 m of oil-soaked soil in which anaerobic and intense acidification processes develop.

III. SOIL POLLUTION ANALYSIS

The purpose of this case study is the analysis of a pollution in a crude oil transport pipeline and consists of:

- a. creating a mathematical model that provides concrete data about oil pollution,
- b. creating a mathematical model for estimating the amount of oil spilled from pipelines.

The crude oil transport pipeline has the following characteristics:

- a. pipe diameter 500 mm,
- b. crude oil in the pipeline with a density of 0.8263,
- c. water content 0.001,
- d. salt content 0.001,
- e. sulfur 0.01,
- f. pipe pressure 6 bar,
- g. time of damage detection 3.00,
- h. drain stop time 8.00.

In the analysis of this case, we tried to give answers to the following questions:

- a. what is the amount of crude oil from the pipeline that leaked into the soil and onto the land,
- b. what is the impact on the soil.

The only data available are the measurements in the field and the estimate made by the operator and the pictures taken at the scene of the accident.

The simulation of crude oil losses is given by the relations [7,8]:

- a. the absolute pressure at the failure site is the relative pressure x 1.01325
- b. the flow rate through the hole is given by the formula:

$$v = \sqrt{200000 * (A - 1,01325) / P \alpha c} \quad (3)$$

- a. orifice surface:

$$A = \pi * \varphi^2 / 4 \quad (4)$$

- b. flow coefficient between 0.6 and 0.7

- c. flow rate through the orifice:

$$Q = 3,6 * \mu * A * v / 1000000 \quad (5)$$

- d. the amount of lost product is given by the relationship:

$$Cs = Q * t \quad (6)$$



Figure 1. Pollution pipeline area



Figure 2. Soil pollution by oil



Figure 3. Pollution by oil



Figure 4.Wheat agricultural culture pollution by oil



Figure 5. Soil pollution by oil

In order to determine the degree of pollution, we took a number of samples, namely on the surfaces under 1000 sq m - 4 samples and on the surfaces under 5000 sq m - 8 samples.

The samples were collected as follows in table 1 and taken from From 0-30 cm and From 30 cm to 50 cm.

Table 1. Sample collected by oil soil pollution

Pollution area	Area polluted, m ²	Samples number
I	1900	8
II	220	4
III	780	4
IV	2016	8
V	504	4

The analyzes performed were as follows:

- the amount of pollutant in the soil,
- soil characteristics,
- characteristics of polluting crude oil

Soil analysis:

- Content in humus between 2 and 6%;
- the degree of saturation in bases 85-95%;
- pH: 6.5-8.3;
- apparent density: 1.25-1.45 g/cm³;
- soil type - dust and fine sand,

Oil analysis

- density of crude oil -0.8263 at 15 °C,
- viscosity of crude oil 1.3 degrees E at 20 °C, i.e. 5.2 cP,
- content in volatile fractions 8.3%-contains condensate.

Polluted soil analysis

The amount of pollutant in the soil was extracted using a Soxhlet extractor. No traces of oil were found on the samples collected from 30 cm to 50 cm.

Table 2.Quantities of oil were found on the samples collected between 0 and 30 cm

Pollution area	Area polluted, m ²	The amount of sample analyzed	The amount of oil product
I	1900	50 grams per sample x 8 probe=400 grams	3 grams
II	220	50 grams per sample x 4 probe=200 grams	20 grams
III	780	50 grams per sample x 4 probe=200 grams	40 grams
IV	2016	50 grams per sample x 8 probe=400 grams	80 grams
V	504	50 grams per sample x 4 probe=200 grams	20 grams

Table 3. Modeling of soil pollution by oil

Soil type	Boulders-coarse gravel	Gravel-coarse sand	Coarse and medium sand	Fine and medium sand	Dust and fine sand
Evaporated amount (to)	1,25904	1,25904	1,25904	1,25904	1,25904
The amount of oil left on the ground (to)	13,38096	13,38096	13,38096	13,38096	13,38096
Soil infestation area (m ²)	5398	5398	5398	5398	5398
The volume of crude oil left on the ground (m ³)	16	16	16	16	16
Soil pollutant retention capacity (l/m ³)	5	8	15	25	40
Penetration depth unsaturated zone (m)	0,30	0,19	0,10	0,06	0,04
The amount retained in the unsaturated zone (m ³)	16	16	16	16	16
Permeability of the layer for water (m/s)	10	1	0,01	0,001	0,0001
Permeability of the layer for the pollutant (m/s)	3,423	0,3423	0,0034	0,000342	0,0000342
Average pollutant transfer time (s)	-	-	-	-	-
The final amount of pollutant retained by the soil (mc)	16	26	49	81	130

Table 4. Retention Capacity

Soil types	Soil retention capacity, R, l/m ³
Coarse gravel	5
Gravel and coarse sand	8
Medium and coarse sands	15
Fine and medium sands	25
Fine sands	40

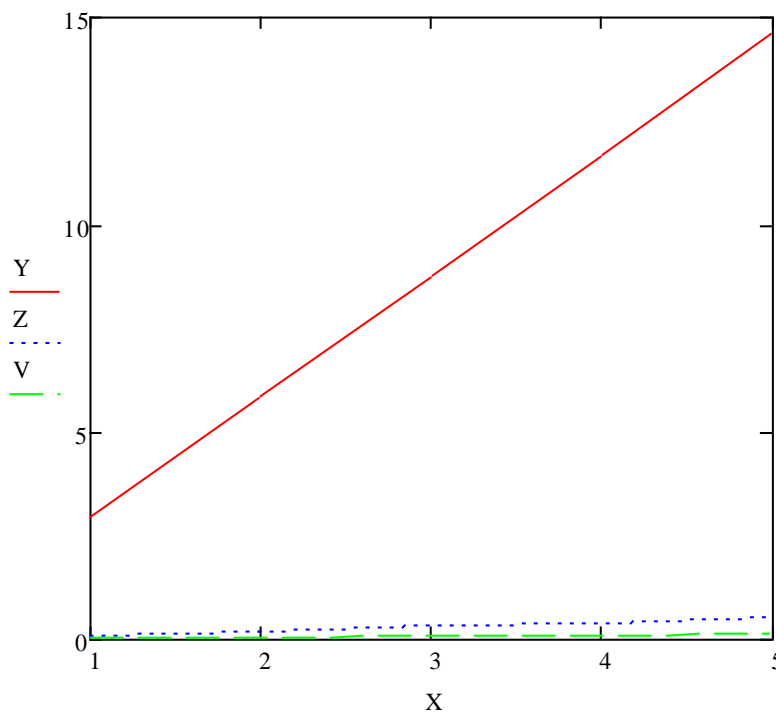


Figure 5. The evolution of the amount of crude oil spilled in tons (Y), the infested surface in ha (Z) as well as the amount of infested soil in thousands of tons (V) against the time of spillage in hours (X)

The depth of penetration into the unsaturated zone is equal to the product of 1000 x the volume of crude oil remaining on the soil / (infestation area x pollutant retention capacity in the soil x viscosity coefficient).
 The amount of pollutant retained in the soil is given by the relationship: the area of infestation x the depth of penetration into the soil x the capacity to retain pollutants in the soil x the viscosity coefficient / 1000.
 The total amount of pollutant is given by the relationship: the area of infestation x the maximum depth of penetration into the soil measured x the capacity to retain pollutants in the soil x the viscosity coefficient / 1000.

Table 5. The simulation of pollution as a function of time

Time elapsed since the accident	1 hour	2 hour	3 hour	4 hour	5 hour
The amount of oil spilled tons	2,96	5,85	8,77	11,70	14,64
The infested area, sq	1091	2157	3234	4134	5398
The final amount of pollutant retained by the soil, mc	26	40	78	104	130

IV. CONCLUSION

In conclusion:

- the postponement of the intervention led to the increase of both the polluted surface and the amount of oil spilled,
- the numerical models made are quite consistent with reality,
- the actual polluted surface is 5420 sq m and the calculated one is 5398 sq m, i.e. 99% real results,
- the amount of polluted soil was determined to be 130 mc compared to 805 mc (it should be noted that in the calculations of the commercial company, all the amount of possibly infested land is eliminated),
- The depollution method chosen by the transport company is to take over the entire volume of soil and transport it to an ecological treatment base,
- It is noted that if an in situ method was used then the required amount of soil would have been a maximum of 130 mc.

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