Acta Agrophysica, 2002, 70, 163-172

UTILIZATION OF THE COMPOSTS BY DECONTAMINATION OF POLLUTED LANDS BY PETROLEUM PRODUCTS^{*}

J. Hrubý, B. Badalíková, J. Nedělník

Research Institute for Fodder Plants, Ltd. Zahradni 1, 664 41 Troubsko, Czech Republic e-mail: agro@vupt.cz

Summary. In a model trial the presumption that the course of decontamination of petroleum contaminated soil was positively influenced by the effect of incorporated organic matter with high microbial activity (compost) in the soil and the plants seeded was tested. During vegetation a decrease in NES content under the limit of hygienic safety was recorded. Simultaneously, it was found that with the increasing level of petroleum contamination of the soil the yields of dry and green matter reduced (treatments 3-5) in all the plants under study, which was caused by different conditions for plant emergence and growth in the initial stages of the contaminated soil.

Keywords: decontamination, composting, polluted soils, petroleum products, crops.

INTRODUCTION

The main cause of long-term damage to petroleum-contaminated soils is predominantly the destruction of soil structure by colloid peptisation and disturbance of air: water ratio. For successful decontamination of each locality it is necessary to have a good knowledge of geology, geochemistry and hydrogeology. On the other hand, physical and chemical properties of petroleum products have a marked effect on the technology of decontamination, which can be used to reduce the content of hydrocarbons in the soils [7]. Bioremediation of

^{*}The present study was conducted as part of the international research project EUREKA E! 2190 (Revital) entitled "Ecological Recycling of Organic Wastes with Respect to Remediation of Petroleum Polluted Soils and Areas" undertaken in collaboration with a Hungarian partner BIO-Gén, Soil Biological Research Company (Tapolca).

petroleum-contaminated soils is based predominantly on the capacity of microorganisms to utilize these products as a source of energy and nutrients. Previous results suggested that the optimum conditions for the acceleration of humification of organic substances are also the optimum conditions for the acceleration of decontamination of soils impacted by organic pollutants. According to Raclavská, [7], biodegradation of petroleum hydrocarbons is significantly dependent on microorganisms, which are primarily present in the soil, and on specific environmental conditions. Aerobic biodegradation is related predominantly to the density of microbial population, presence of oxygen, nutrient concentrations, temperature of the environment, pH, and soil water content. The rate of biodegradation of petroleum products is also dependent on the structure of their molecules. For example, Gough et al. [2] suggested rapid degradation of hydrocarbons with simple structure. Římovský et al. [8] pointed out that the level of contamination of soil environment by petroleum products is determined by the total content of nonpolar extractable substances (NES in mg kg⁻¹ dry matter). The authors also carried out model experiments to study biodegradation of petroleum products in the soil. They also tested potential utilization of the stimulating effect of pig slurry. The values reported for the parameters of petroleum-contaminated soil (expressed mostly in NES) showed the positive effect of slurry on the biodegradation of petroleum products. Slurry, applied after stimulation of soil microorganisms, accelerated this process in the soil and reduced the negative effect on the production of biomass of hay oats and winter rye at harvest. The effect of slurry on biodegradation of petroleum products was also confirmed by other authors [1].

One of the goals of the research project was to study potential utilization of forced composts with subsequent growing of crops (predominantly with high DM content) for biodegradation of petroleum products in the soil. A promising technology of compost production based on processing of plant and animal residues and specially grown crops is called "Ekobioprogres". This technology provides optimum conditions for the development of soil microorganisms, creating aerobic environment by regular automatic digging of compost material, maintaining optimum C:N ratio and supplying optimum water content to the material by continual mixing of solid and liquid bio-wastes. These findings were used to develop a technology of the so-called "two-phase decontamination" of petroleum-impacted soil (phase 1 = compost activity, phase 2 = rhizosphere activity), which was already described [3-5]. The technology of *in situ* decontamination of petroleum-impacted soils observes the ecological aspects, which are essential for these technologies, and also meets requirements for economic efficiency.

EXPERIMENTAL PROCEDURES

The technology of two-phase decontamination of petroleum-impacted soils was studied in detail in pot and small-plot trials and also under pilot conditions, for example, on reclaimed soil of the abandoned coal mine in northern Moravia as part of the international project Eureka E! 2190 – Revital.

The aim of the present experiment with petroleum-contaminated soil under model conditions was to test the hypothesis that the rate of decontamination was positively affected by the action of organic matter with high microbial activity (i.e. forced compost) incorporated into the soil and subsequently grown crops.

The response of crops to different levels of petroleum contamination of soil and the decontamination of soil by biological processes were assessed in a model small-plot trial established in a sugar-beet growing region in the Research Institute for Fodder Crops, Ltd. Troubsko. The trial was established on non-agricultural land (Chernozem) with very favourable soil structure (to a depth of 0.20 m the coefficient of texture was 2.22). The long-term mean annual temperature on the experimental site was 8.4° C, mean annual precipitation was 547 mm. The patterns of precipitation and temperature in the experimental period of the years 2000 – 2001 are given in Tables 1 a, b.

Table 1a. Pattern of climatic conditions throughout the experimental years (mean temperatures ($({}^{0}C)$

	Ι	II	III	IV	V	VI	VII	VIII	IX	Х	XI	XII	Mean
2000	-2.1	2.6	4.7	12.9	15.9	18.8	17.2	20.0	13.7	11.6	6.5	1.0	10.2
2001	-0.6	1.1	4.4	8.2	15.7	16.2	20.3	20.4	13.1	12.4	2.5	-3.6	9.2

Table 1b. Pattern of	of climatic conditions	throughout th	ne experimental	vears	(mean pred	cipitation sum	(mm)
A PROTE ANT A PROPERTY .				1	(P		. (

	Ι	II	III	IV	V	VI	VII	VIII	IX	Х	XI	XII	Sum
2000	33.3	20.9	56.7	3.0	31.3	24.9	115.4	56.4	27.2	22.9	52.4	31.0	475.4
2001	36.7	20.6	50.0	31.2	57.0	32.7	76.4	60.9	103.9	14.9	25.2	27.3	536.8

The principle of two-phase *in situ* decontamination of petroleum-impacted soils is characterized as follows:

a) the first stage includes production of forced compost in the fermentation trough with daily regular digging of fermented material, which provides optimum conditions for the development of aerobic microorganisms. The resultant forced compost characterized by rich microbial activity and high nutrient content is incorporated into the petroleum-contaminated soil, which is gradually decontaminated due to the microbial activity of the compost. After mixing the petroleum-contaminated soil with the compost intensive the development of aerobic microorganisms occurs. The process is dependent on the presence of oxygen. Primarily, oxygen comes to the soil environment during soil tillage operations. The development of aerobic microorganisms in soil gradually decreases but it can be fully restored only after further incorporation of compost into the contaminated soil.

b) as for the reduction of the concentration of petroleum products the second stage of decontamination seems to be economically more important, i.e. subsequent growing of plants and utilization of their rhizosphere. This process can gradually reduce the concentration of nonpolar extractable substances in the soil below 100 mg·kg⁻¹ dry matter, i. e. the level of natural background (i. e. soil is considered decontaminated).

Treatments: 1) control (without compost) + crop, 2) soil + compost + crop, 3) soil + compost + petroleum $(0.6 \text{ l}\cdot\text{m}^{-2})$ + crop, 4) soil + compost + petroleum $(1.2 \text{ l}\cdot\text{m}^{-2})$ + crop, 5) soil + compost + petroleum $(1.5 \text{ l}\cdot\text{m}^{-2})$ + crop.

In the first stage of the experiment, forced compost produced by "Ekobioprogres" technology in the fermentation trough in the compost producing company ZEMSPOL a. s. Sloup in Moravský Kras, was applied to the petroleumimpacted soil on small plots. The application rate of compost was 40 Mg-ha⁻¹ and a rotary implement was used to incorporate compost into the soil.

Compost parameters: content of organic matter in solids – 46.8%, N–1.50%, P–0.54%, Ca–0.95%, Mg–0.33%, C:N ratio – 31.2. Analysis for the presence of nonpolar extractable substances in the soil – the extracts were measured by IR–spectrometer under usance - determined wave numbers. The results are converted to sample dry matter and are given in mg-kg⁻¹.

In the second stage each treatment involved the following crops: rye (*Secale cereale L.var.multicaule METZG.ex ALEE*), sweet clover (*Melilotus albus*), canary grass (*Phalaris canariensis L.*) and safflower (*Carthamus tinctorius L.*).

For each crop and treatment evaluations were made of herbage yields at harvest, yields in dry matter $(g \cdot m^{-2})$ and basic phenological data.

Characteristics of experimental crops:

Secale cereale L. var.multicaule METZG.ex ALEE): biennial species tolerant to a wide range of soil and climatic conditions, suitable for forage and food industry, suitable for autumn and spring sowing.

Melilotus albus: biennial form, grows on less fertile and non-fertile soils, does not tolerate heavy and waterlogged soils, it is utilized as a forage crop and soil-improving crop. The stem is erect, reaches a height of 150 cm.

Phalaris canariensis l.: annual, single-cut species, productive on a great variety of soil conditions, suitable even for monoculture, the stem is straight and smooth, the inflorescence is compact panicle.

Carthanus tincorius L.: has low requirements for soil conditions, grows in warm and dry regions, good crop for calcareous soil. It belongs to the family Compositeae, 100-130 cm high, it is mostly grown for seed, as an intercrop it is suitable for green manure and green food.

The small-plot experiment was established in autumn of the year 1999 (seedbed preparation). In the following year petroleum was applied to the soil at determined rates (13 April 2000) and forced compost was incorporated into the soil by a rotary implement (17 April 2000). Sowing and harvest of the test crops is given in the following chart:

sowing - 12 May 2000 (all crops)	harvest – 25 September 2000 (all crops)
3 May 2001 (except sweet clover)	20 June 2001 (sweet clover, 2nd cutting)
	20 August 2001 (all the other crops)

The size of the experimental plot (in four replications) was 2.6 m^2 . The results were statistically treated by analysis of variance.

RESULTS AND DISCUSSION

According to Raclavská [7], biodegradation of petroleum products is dependent on microorganisms, which are primarily present in the soil and on specific environmental conditions. The effective degradation of hydrocarbons requires a joint action of several different populations of microorganisms (Litchfield, [6]). From this aspect, the incorporation of forced compost with high microbial activity into the soil is very important. It is necessary to emphasize that the soil on the experimental plot at Troubsko location was only surface-contaminated by petroleum products (to a depth of ca 0.30 m) and compost was incorporated with a rotary implement.

The results of analysis for the content of nonpolar extractable substances $(mg \cdot kg^{-1} DM)$ showed that even in the first experimental year the concentration of petroleum products in the soil markedly decreased, reaching a level of hygienic safety according to valid legal standards of the Czech Republic. A marked decrease in nonpolar extractable substances is evident in all treatments. When petroleum products were applied at a rate of 0.6 $1 \cdot m^{-2}$ the content of nonpolar extractable substances was below 100 mg \cdot kg⁻¹ DM. In other cases of soil contamination by petroleum products (1.2 and 1.5 $1 \cdot m^{-2}$, resp.) the level of nonpolar extractable substances was above this level, being on average 20 and 30 mg \cdot kg⁻¹ DM higher, resp. After the first experimental year, soil contamination by petroleum products decreased on average to 6.6–11.6% (depending on treatments) of the initial level. Soil contamination by petroleum products (depending on treatment) before and after crop harvest in the year 2000 is given in Table 2.

	Petroleum rates							
Sample collection	0.6 l·m ⁻²		1.2 ŀ	m ⁻²	1.5 l·m ⁻²			
	range	mean	range	mean	range	mean		
I. initial soil contamination (17 Apr.) NES (mg·kg ⁻¹ DM)	200-890	545	470-1600	1035	940-2400	1670		
II. soil contamination after crop harvest (16 Oct.) NES (mg·kg ⁻¹ DM)	10-53	36	23- 240	120	27- 300	130		

 Table 2. Content of nonpolar extractable substances (NES mg.kg⁻¹ DM) in soil related to treatments-Troubsko 2000

In the second harvest year (until 31 August 2001) NES concentrations in individual treatments mostly decreased below 100 mg·kg⁻¹ DM, i.e. to the level of natural background. Only in canary grass NES concentrations were 130 mg·kg⁻¹. DM, especially in the treatment, in which petroleum was applied at a rate $1.5 \text{ l}\cdot\text{m}^{-2}$. Soil contamination by petroleum products (in NES) in individual experimental treatments is presented in Table 3.

The results of statistical analysis suggested that the yield differences between treatments with different rates of soil contamination by petroleum products in the test crops were highly statistically significant Tables 4 a, b - Bartlett test with sources of variability (A – experimental treatments, B – years).

Table 3. Soil contamination by petroleum products (NES – $mg \cdot kg^{-1}$ DM) in relation to test crops before and after termination of experiment (Troubsko, 2000 – 2001)

Treatment	Sampling date	Rye	Sweet clover	Canary grass	Safflower
0.6 l·m ⁻² -	17 Apr. 2000	545	545	545	545
	31 Aug.2001	< 47	33	29	20
	17 Apr.2000	1035	1035	1035	1035
1.2 1.11	31 Aug.2001	< 20	40	33	59
1.5 l·m ⁻² –	17 Apr.2000	1370	1670	1670	1670
	31 Aug. 2001	< 20	< 20	135	23

Tables 4a. Statistical evaluation of crop yields by analysis of variance

Source of variability		Degree	Ry	/e	Sweet clover			
		of	of MS		MS			
		freedom	at harvest	in DM	at harvest	in DM		
Treatments	А	4	5215.788**	2844.462**	1011162.650**	89223.462**		
Years	В	1	133287.025**	43560.000**	627252.025**	731.025*		
Interaction	AB	4	3203.212**	211.438	198132.775**	34817.088**		
Technical error		30	66.092	123.517	85.208	110.925		

Tables 4b. Statistical evaluation of crop yields by analysis of variance

Source of variability		Degree	Canary	/ grass	Safflower MS			
		of	М	S				
		freedom	at harvest	in DM	at harvest	in DM		
Treatments	А	4	197297.212**	64035.038**	1174375.475**	204385.438**		
Years	В	1	28249.225**	42510.400**	19788048.900**	5939014.225**		
Interactions	AB	4	143276.913**	23529.587**	582675.275**	57317.663**		
Technical error		30	103.642	91.183	1258.867	175.225		

*LSD=0.05

**LSD=0.01

Herbage production (at harvest) and DM yields - $g \cdot m^{-2}$ (Tab. 5) decreased with the increasing level of soil contamination by petroleum products, whereas a considerable decrease was evident predominantly in the first harvest year. In the second harvest year when "two-phase" *in situ* soil decontamination was performed the yields of individual test crops became gradually consistent, even though in this year the growth and development of stands, especially in the initial stages, were also negatively affected by contaminated soil.

A significant yield decrease especially in the first experimental year was recorded in sweet clover. The yield fell down to 19.8 % of the yield of Treatment 1. This crop also greatly responded to changes in the soil environment, even in the year 2001. The results of statistical analysis showed that in both years the least responsive crop to the soil contaminated by petroleum products was rye. In this crop, however, the incorporation of forced compost into the soil, in spite of significant yield differences in treatments with petroleum products, positively stimulated herbage yields in the year 2000 (the maximum yield difference in these treatments was 29.6%). In the following year the yields of these treatments became consistent and the maximum difference in herbage yields at harvest was only 5.4%. Mass and DM yields of safflower were most consistent in both experimental years, compared with other test crops. Canary grass responded negatively to compost application, which resulted in yield decrease of 24.9%. This was also significant in treatments where petroleum products were applied (Mass yields at harvest in the year 2000 decreased by another 7.8 to 27.2% in these treatments).

CONCLUSION

In the model small-plot experiment a hypothesis was tested that with the so-called "two-phase decontamination" of petroleum-impacted soil "*in situ*" (applications of 0.6, 1.2 a 1.5 $1 \cdot m^{-2}$) the process was positively affected by the incorporation of organic matter with high microbial activity (of compost) into the soil and by the rhizosphere of seeded crops. In the experimental years 2000 and 2001 the content of NES fell below the level of hygienic safety in all treatments, i.e. to the level of natural background. Only in canary grass NES concentration was above 100 mg·kg⁻¹ DM when the highest rate of petroleum products (1.5 $1 \cdot m^{-2}$) was applied. Simultaneously, it was found that with the increasing soil contamination by petroleum products at harvest and DM yields statistically significantly decreased in all test crops, especially in the first experimental year. A negative response to the increasing rates of petroleum products in the soil was most evident in sweet clover. The least responsive crop to the changes in the soil environment was rye.

		Yields (g·m ⁻²)							
Crop	Treatment	at ha	rvest	in D	M				
		2000	2001	2000	2001				
	1	300	484	220	300				
	2	405	506	280	344				
Rye	3	389	481	265	321				
	4	381	462	256	315				
	5	340	456	242	316				
	1	1355	1220	562	383				
	2	1080	1440	460	485				
Sweet clover	3	925	890	412	350				
	4	330	846	212	363				
	5	270	810	187	294				
	1	1377	992	661	584				
Conomi	2	1035	1085	501	522				
Canary	3	925	989	444	507				
grass	4	714	962	397	494				
	5	660	951	271	491				
	1	3185	1385	1695	698				
	2	3495	1346	1570	674				
Safflower	3	2360	1261	1296	596				
	4	2290	1154	1246	577				
	5	1946	1092	1099	510				

Table 5. Mass yields of test crops related to soil contamination by petroleum products (Troubsko 2000-2001)

REFERENCES

- 1. Aggarwal P.K., Hlinchee R.E.: Monitoring in situ biodegradation of hydrocarbons by using stable carbon isotopes. Envir. Sci. Technol., 25, 1178-1180, 1991.
- Gough M.A., Rhead M.M., Rowland S.J.: Biodegradation studies of unsolved complex mixtures of hydrocarbons. Org. Geochem., 18, 1, 17-22, 1992.

- Hrubý J., Badalíková B., Prášek J., Ševčík V.: Bioremediation of polluted non-agricultural land by petroleum products. In: Biotechnology 2001, Č.Budějovice, 165, 2001.
- Hrubý J., Badalíková B., Ševčík V.: Ecological recycling of biowastes by the fast composting aiming at the revitalization of polluted soils. Folia Univ. Agric. Stetin. 209 Agricultura, 83, 127-132, 2000.
- Hrubý J., Ševčík V., Kučera A., Badalíková B.: Recycling of biological wastes in agriculture technology Ekobioprogres. In: Waste – Abfälle, Praha, 78-80, 1998.
- Litchfield C.D.: In Situ Bioremediation: Basis and practices. In: Biotreatment of industrial and hazardous wastes. New York, 197-197, 1993.
- 7. Raclavská H.: Znečištění zemin a metody jejich dekontaminace. Ostrava 1998.
- Římovský K., Bauer F., Boháček Z., Linhartová M., Toul J.: Effect of pig slurry on increase of biodegradation of petroleum products in soil. Rostl. Výr., 44, 325-330, 1998.