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Assessment of Cleanup Tests of an Oiled Salt Marsh — The Golden Robin Spill in Miguasha, Quebec

Part 1.

Residual Bunker C Hydrocarbon
Concentrations and Compositions

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ASSESSMENT OF CLEANUP TESTS OF AN OILED SALT MARSH -
THE GOLDEN ROBIN SPILL IN MIGUASHA, QUEBEC

Part I. Residual Bunker C Hydrocarbon
Concentrations and Compositions

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FOREWORD

The Atlantic and Quebec regional offices of the Environmental Protection Service, Department of Fisheries and the Environment, conducted cleanup tests in 1975 on the uncleaned portions of the Miguasha Marsh, Quebec which had become contaminated by oil spilled from the GOLDEN ROBIN in 1974. This preliminary work led to the undertaking of a study one year later to evaluate the efficacy of the cleanup tests and the regrowth potential of the affected marsh vegetation. The preliminary work was made possible through the assistance of Messrs. P. Cejka and Y. Plunier (Quebec regional office) and Dr. F.C. Duerden and Mr. W. Pierce (Atlantic regional office), and this study through the technical assistance of Messrs. S. Vandermeulen and T.P. Ahern of the Bedford Institute of Oceanography. Also acknowledged is the assistance and advice of Dr. P.D. Keizer (Bedford Institute of Oceanography) in providing GLC data interpretation.





Plate 1. Miguasha Marsh, Quebec



ABSTRACT

Miguasha Marsh, Quebec, was heavily oiled in 1974 by Bunker C fuel oil spilled from the tanker GOLDEN ROBIN. This marsh was not cleaned until the spring of 1975, when a number of cleanup procedures were carried out and tested. Of these, both manual and mechanical methods were utilized, as well as the technique of burning oiled debris and vegetation.

This report represents the first portion of a two-part study examining the fate and biodegradation of residual Bunker C fuel oil in Miguasha Marsh sediments following the cleanup trials. Sediment analyses by gas liquid chromatography (GLC) and fluorescence were carried out on marsh sediments to determine the concentration and composition of residual hydrocarbons. Results were correlated with observations on survival and regrowth of marsh vegetation.

RESUME

En 1974, le pétrolier GOLDEN ROBIN a déversé accidentellement une grande quantité de fuel-oil Bunker C dans les marécages de Miguasha, au Québec. Il a fallu attendre au printemps de 1975 avant d'y éprouver diverses méthodes de nettoyage (entre autres, nettoyage manuel et mécanique, ainsi que brûlage de la végétation et des matières polluées) et d'en évaluer les résultats.

Le présent rapport n'est que le premier des deux volets d'une étude portant sur la biodégradation et les effets du fuel-oil présent dans les sédiments après le nettoyage. L'analyse par fluorimétrie et chromatographie en phase gazeuse des sédiments des marécages a été faite afin de déterminer la nature et la concentration des hydrocarbures résiduels. Les résultats ont été mis en corrélation avec les données recueillies sur la survie et la croissance nouvelle de la végétation des marécages.

<u>TABLE OF CONTENTS</u>	<u>PAGE</u>
FOREWORD	i
ABSTRACT	v
RESUME	vi
TABLE OF CONTENTS	vii
LIST OF FIGURES	viii
LIST OF TABLES	ix
LIST OF PLATES	x
1 INTRODUCTION	1
2 METHODS AND MATERIALS	1
2.1 Petroleum Hydrocarbon Residue Analysis	1
2.1.1 Sediment Sampling	1
2.1.2 Hydrocarbon Analysis	1
3 RESULTS	3
3.1 Mechanical Cleanup Effectiveness	3
3.1.1 Petroleum Hydrocarbon Concentrations in Sediments - Quantitative Analysis	3
3.1.2 Qualitative Analysis - Aliphatics	5
3.1.3 Qualitative Analysis - Aromatics	14
3.2 Revegetation of Oiled Plots	14
4 DISCUSSION	27
5 CONCLUSIONS AND RECOMMENDATIONS	29
REFERENCES	31

<u>LIST OF FIGURES</u>	<u>PAGE</u>
1 SKETCH OF MIGUASHA MARSH AND TEST SITES	2
2 GLC SPECTRUM OF ARROW BUNKER C	6
3 GLC SPECTRUM OF SEDIMENT, QUADRAT A2	7
4 GLC SPECTRUM OF SEDIMENT, QUADRAT A3	8
5 GLC SPECTRUM OF SEDIMENT, QUADRAT A4	9
6 GLC SPECTRUM OF SEDIMENT, QUADRAT A5	10
7 GLC SPECTRUM OF SEDIMENT, QUADRAT A6	11
8 GLC SPECTRUM OF SEDIMENT, QUADRAT A7	12
9 GLC SPECTRUM OF SEDIMENT, QUADRAT A7 (CONTROL)	13
10 GLC SPECTRUM OF SEDIMENT, QUADRAT A8	15
11 GLC SPECTRUM OF SEDIMENT, QUADRAT A9	16
12 GLC SPECTRUM OF SEDIMENT, QUADRAT B1 (SURFACE)	17
13 GLC SPECTRUM OF SEDIMENT, QUADRAT B1 (SUBSURFACE)	18
14 GLC SPECTRUM OF SEDIMENT, QUADRAT B2	19
15 SYNCHRONOUS FLUORESCENCE SPECTRUM OF ARROW BUNKER C	20
16 SYNCHRONOUS FLUORESCENCE SPECTRUM OF QUADRAT A2 (CONTROL)	20

<u>LIST OF TABLES</u>	<u>PAGE</u>
1 PETROLEUM HYDROCARBON CONCENTRATIONS IN SUBSURFACE SEDIMENTS FROM MIGUASHA MARSH, QUEBEC	4
2 SUMMARY OF MIGUASHA MARSH CLEANUP STUDY	28

<u>LIST OF PLATES</u>		<u>PAGE</u>
1	MIGUASHA MARSH, QUEBEC	iii
2	QUADRAT A2	21
3	QUADRAT A3	21
4	QUADRAT A4	22
5	QUADRATS A5 AND A6	22
6	QUADRAT A7	23
7	QUADRAT A7-b	23
8	QUADRATS A8 AND A9	24
9	QUADRAT B1	24
10	DETAIL OF QUADRAT B1	25
11	QUADRAT B2	25
12	DETAIL OF QUADRAT B2	26

1 INTRODUCTION

On September 14, 1974, the tanker, GOLDEN ROBIN, ran aground in the Chaleur Bay off Dalhousie, New Brunswick. Approximately 1,000 barrels (159,000 litres) of Bunker C fuel oil were released into the waters, resulting in extensive contamination of both New Brunswick and neighbouring Quebec shores. Although cleanup of the bay was effected immediately, one oiled salt marsh, the Miguasha Marsh in the province of Quebec, was left uncleaned to serve as a study area for future oil spill cleanup and research work (see Figure 1).

The spring of 1975 marked the initiation of cleanup in the back areas of the marsh where heavier oiling was evident. Manual and mechanical removal of oiled vegetation, as well as burning, represented some of the techniques employed and tested. Results and recommendations based on these tests have been described by Cejka (1975).

During the summers of 1976 and 1977, a follow-up program was initiated to evaluate the efficacy of the cleanup tests and the regrowth potential of the affected marsh vegetation. Part I of this program consisted of an analysis of marsh sediments from cleaned-up areas to reveal hydrocarbon concentrations and compositions. Results were correlated with regrowth of vegetation in an attempt to determine which cleanup methods were most effective in minimizing contamination by residual oil, thereby maximizing revegetation. More recently, further analyses of marsh sediments were carried out to determine the depth of penetration of Bunker C hydrocarbons in marsh conditions, and to evaluate the potential of the area's hydrocarbon-utilizing bacteria to respond to sediment-trapped Bunker C. Results of these latter studies form the basis of Part II of the program referred to above.

2 METHODS AND MATERIALS

2.1 Petroleum Hydrocarbon Residue Analysis

2.1.1 Sediment Sampling:

The marsh soil-sediment structure, which is composed of an extremely tough mat of silt, roots and rhizomes up to 30 cms thick, is not easily sampled by coring. Consequently, 20-cm square blocks were dug out of each study plot (quadrat) with a spade and broken open by hand. With the interior surfaces being exposed in this way, sediment samples were scraped from the blocks and collected in hexane-rinsed, sharp-edged 16-oz. metal cans (Canlab.). After subsequent freezing under dry ice, these cans were transported to the Bedford Institute of Oceanography for analysis.

2.1.2 Hydrocarbon Analysis:

For hydrocarbon analysis, 15 gms wet weight of sediment were mixed with 5 ml of hexane-extracted, distilled water. The resulting slurry was then extracted with two volumes of 50-ml pentane on a Burrell wrist-action shaker (15 minutes, maximum setting). After each extraction,

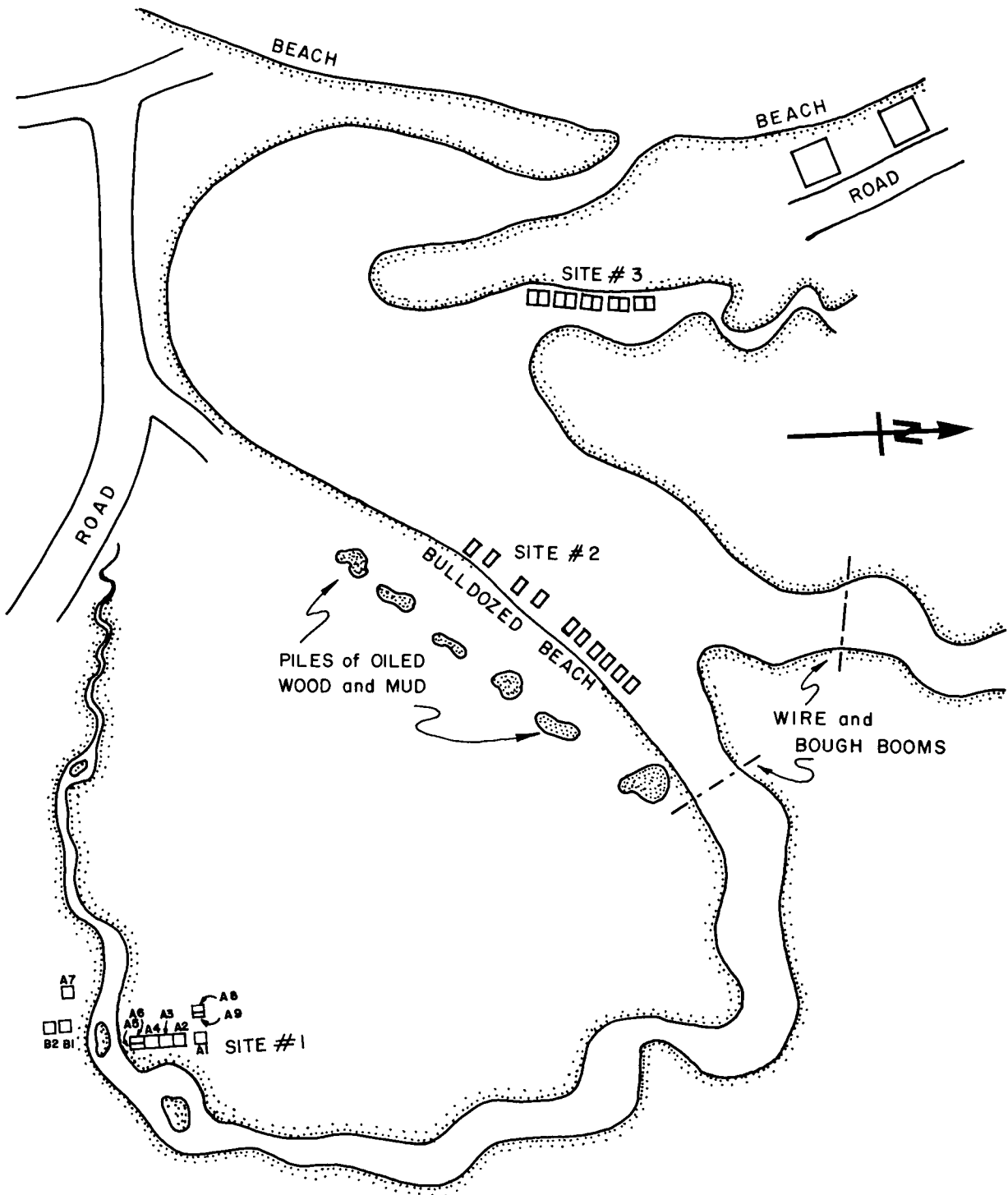


Fig.1 Sketch of MIGUASHA MARSH and TEST SITES

the mixture was centrifuged and the pentane phase decanted. The pellet was dried to constant dry weight and the weight was recorded. The pentane phase was concentrated to 10 mls under a nitrogen flow (N-Evap, Organomation Assoc.) in preparation for analysis by fluorescence spectroscopy.

Hydrocarbon concentrations were determined by fluorescence, using ARROW Bunker C fuel oil as a standard. Synchronous emission spectra were obtained by the method of Lloyd (1971a, 1971b), as used by Gordon and Keizer (1974). Spectroscopy was carried out with a Perkin-Elmer MPF-2A fluorescence spectrophotometer.

For GLC analysis, the pentane extract was further reduced to 500 μ ls, and put onto a silica gel-alumina column, and the saturate fraction was eluted with pentane. If sulfur was present, as indicated by a white precipitate, the extract was first passed through an activated Cu-column (Blumer, 1957). The eluate was concentrated to 100 μ ls and 1 to 20 μ l volumes were injected into a Perkin-Elmer 3920 gas chromatograph coupled to a Hewlett-Packard 3380-A integrator.

All solvents were redistilled in glass from ACS-grade reagent stock.

3 RESULTS

3.1 Mechanical Cleanup Effectiveness

3.1.1 Petroleum Hydrocarbon Concentrations in Sediments-Quantitative Analysis:

Quantitative results of the subsurface sediment hydrocarbon analysis are shown in Table 1. Concentrations of petroleum hydrocarbons range widely, that is, from a low of 5.77 ppm in quadrat A9, where the oiled vegetation and surface mat had been removed by saw cutting, to a high of 179.62 ppt in a surface sediment sample taken from a heavily tracked area.

Lowest hydrocarbon concentrations were found in quadrat A2, a heavily oiled control plot left uncleaned; in quadrats A5, A6 and A7, plots cleaned by burning; and in quadrats A8 and A9, the oiled vegetation of which was removed by saw cutting or digging by spade.

Intermediate concentrations (616-1,1,429 ppm) were found in quadrats A3, A4 and B2. The oiled vegetation of quadrats A3 and A4 had been removed either by hand or sickle and pitchfork, whereas in quadrat B2, the oiled mass had been turned under by plow.

Highest concentrations (13.6-179.62 ppt) were found in quadrats A7-b and B1, and also in a surface sample from a heavily tracked, oiled plot. Quadrat A7-b was a heavily oiled, untreated area adjacent to quadrat A7, which had been burned off. Quadrat B1 was a cleaned plot in which a commercial sod-cutter had been tested.

Sediments from the creek bed and shoreline were intermediate in hydrocarbon concentrations (560.3-1,278.76 ppm) with oil found in all samples.

TABLE 1. Petroleum Hydrocarbon Concentrations in Subsurface (10 cm) Sediments From Miguasha Marsh, Quebec. (For quadrat description and location, and cleanup methodology, refer to Figure 1, Plates 1-12, and Cejka (1975))

Quadrat	Cleanup Method	Hydrocarbon concentration ¹ (ppm)
A2	control plot	7.21
A3	sickle & pitchfork	819.15
A4	pulling up vegetation	616.53
A5	burning with blowtorch	6.39
A6	burning with varsol	11.34
A7	burning with gasoline and oil	18.29
A7-b	control plot	13,599.86
A8	digging out vegetation	8.60
A9	cutting out vegetation and surface mat	5.77
B1-surface	sod-cutter test	115,122.86
B1-subsurface	sod-cutter test	145,860.57
B2	plowing test	1,429.82

Concentrations determined by fluorescence, based on ARROW Bunker C fuel oil as a standard.

3.1.2 Qualitative Analysis - Aliphatics:

The GLC spectrum of ARROW Bunker C fuel oil, similar to the Bunker C spilled in Miguasha Marsh, is shown in Figure 2. This spectrum shows the characteristic unresolved envelope peaking at C_{25} , the peaks for C_{17} /pristane and C_{18} /phytane diagnostic of petroleum-derived hydrocarbons, and the absence of the odd-Carbon preference found in non-petroleum terrigenous hydrocarbons.

The aliphatic qualitative analysis of each quadrat is summarized as follows:

1. Quadrat A2 (see Figure 3): The spectrum shows a strong odd-C number preference with a small unresolved envelope. The pattern is probably due to plant material indigenous to the marsh, and there is little or no indication of oil.
2. Quadrat A3 (Figure 4): Low organic material content is evident. The presence of some material under the unresolved envelope, together with the shape of the envelope peaking at C_{25} , suggests some oil presence.
3. Quadrat A4 (Figure 5): Pattern is due to oil, as indicated by the presence of a C_{18} /phytane peak, the lack of odd-C dominance, and an envelope maximum at C_{25} . Degradation of the n-alkanes suggests microbial activity.
4. Quadrat A5 (Figure 6): Strong odd-C number preference suggests presence of terrigenous n-alkanes. Unresolved envelope is in part masked by solvent tail, but the shape suggests some petroleum-derived material. The two peaks with retention times 25.56 and 26.22 are unique, as they are not found in preceding spectra nor in ARROW Bunker C.
5. Quadrat A6 (Figure 7): Strong odd-C preference and the shape and maximum of the unresolved envelope indicate a mixture of petroleum and terrigenous-derived n-alkanes. The two peaks at retention times 25.52 and 26.20 are unique.
6. Quadrat A7 (Figure 8): The odd-C preference, together with the small but characteristically peaking (at C_{25}) unresolved envelope, suggests a mixture of indigenous and oil-derived hydrocarbons. The peaks with retention times 25.53 and 26.19 are unique.
7. Quadrat A7-b (Figure 9): The GLC spectrum is characteristic of weathered Bunker C fuel oil with the typical unresolved envelope, the C_{18} /phytane peak and the progression of n-alkanes to C_{30} . Some degradation of n-alkanes at the low end suggests microbial activity.

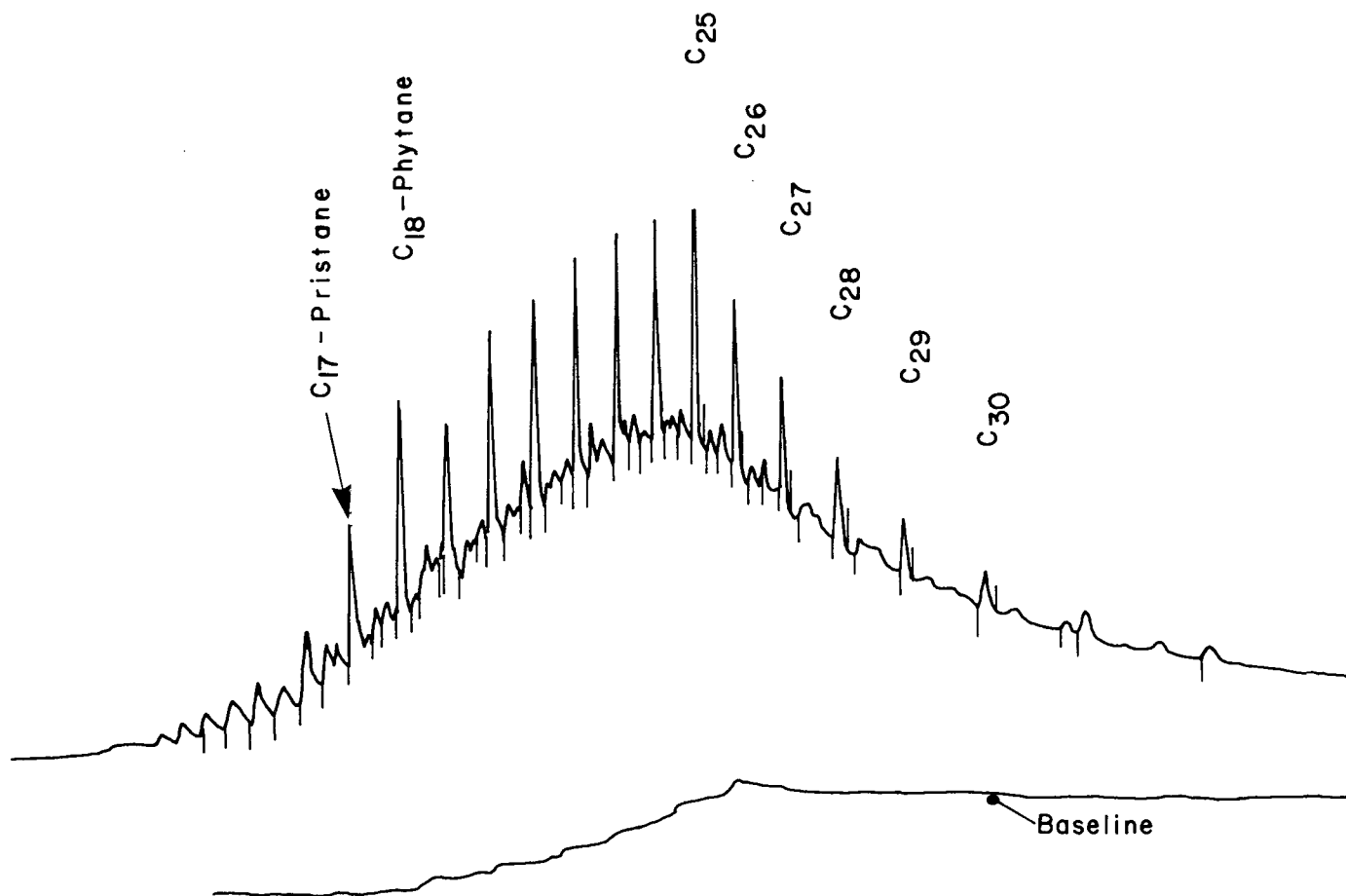


FIG. 2 GLC spectrum of ARROW Bunker C standard showing characteristic C₁₇/pristane and C₁₈/phytane peaks, and absence of odd-Carbon presence.

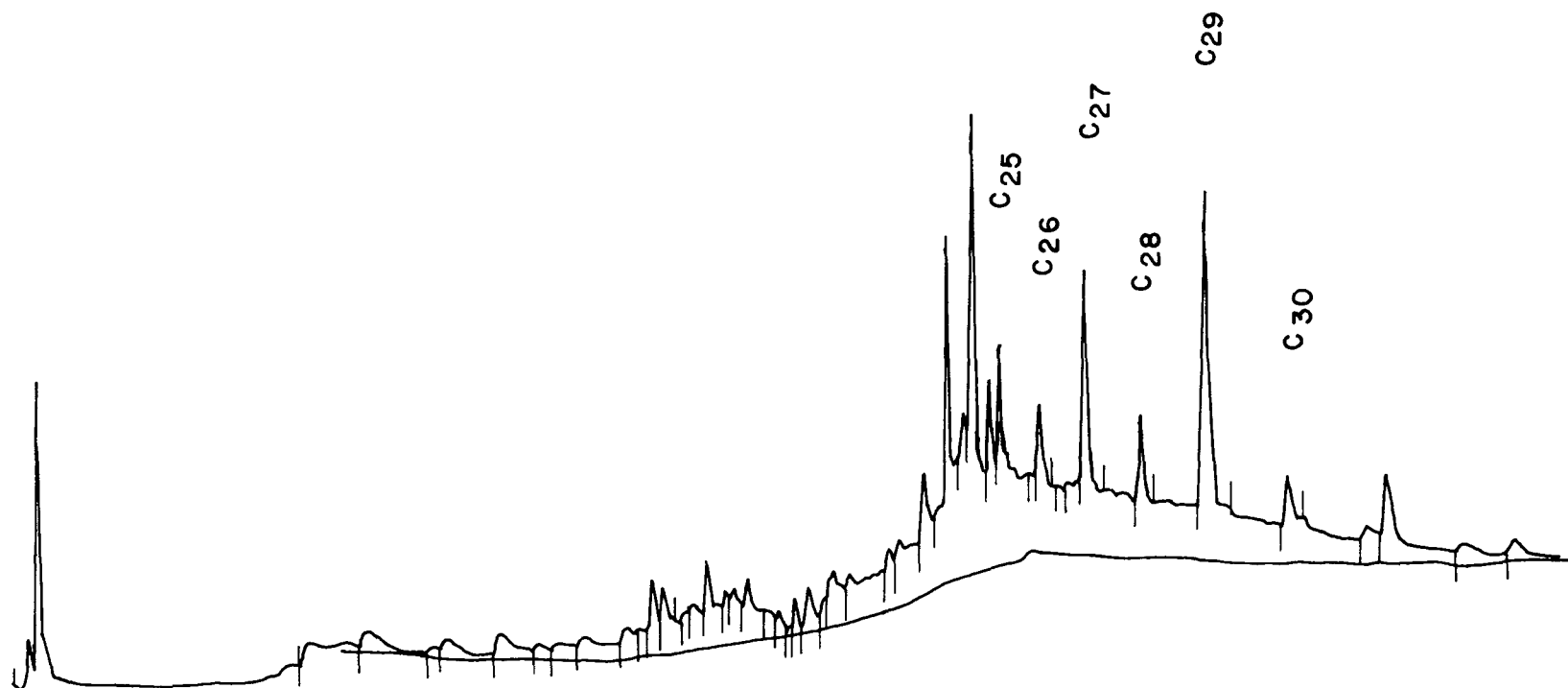


FIG. 3 GLC spectrum of subsurface (10 cms) sediment from quadrat A2 ,
a non-cleaned, heavily oiled plot (Bunker C, 1974).

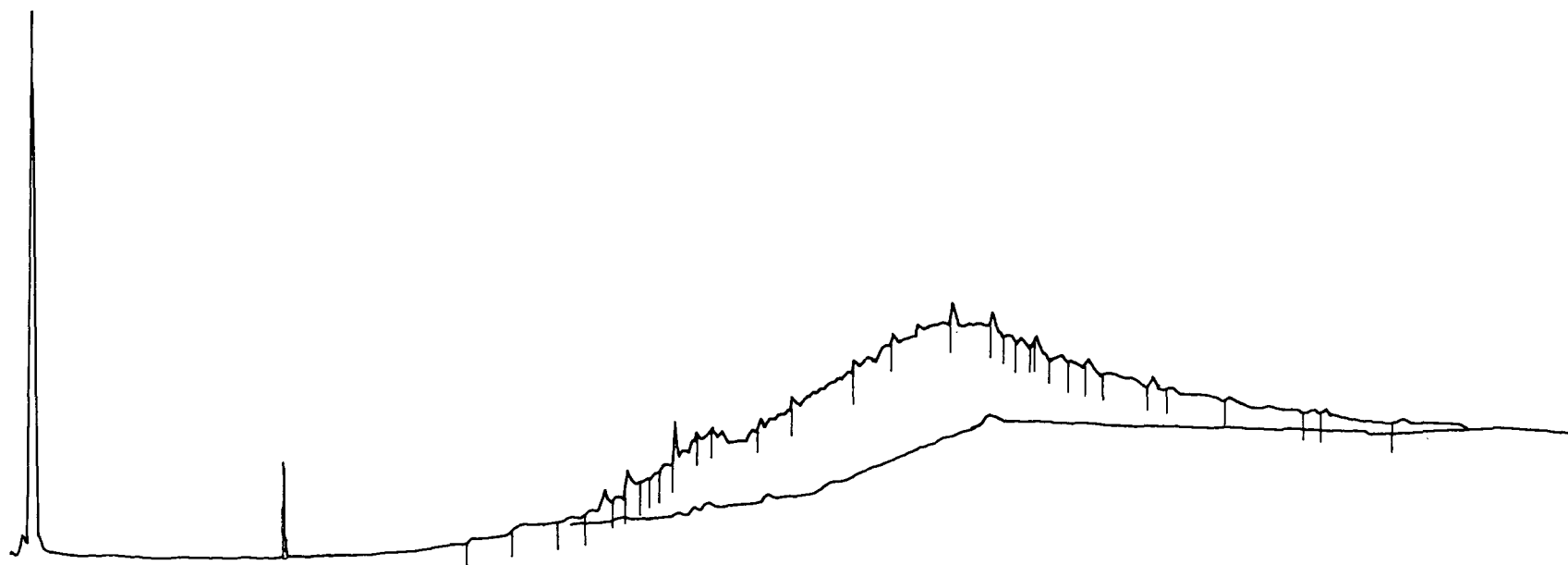


FIG.4 GLC spectrum of subsurface (10 cms) sediment sample from quadrat A3, an oiled plot cleaned by removal of vegetation by sickle and pitchfork.

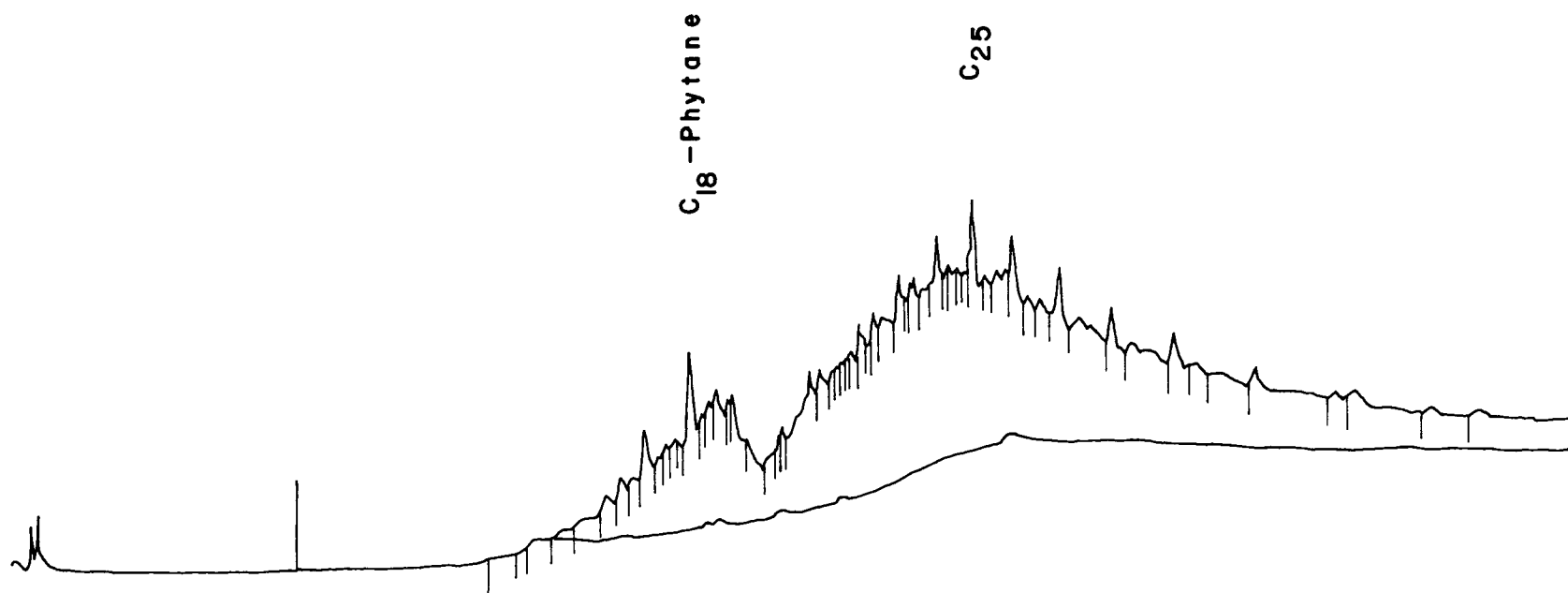


FIG. 5 GLC spectrum of subsurface (10 cms) sediment sample from quadrat A4, an oiled plot cleaned by pulling up oiled vegetation.

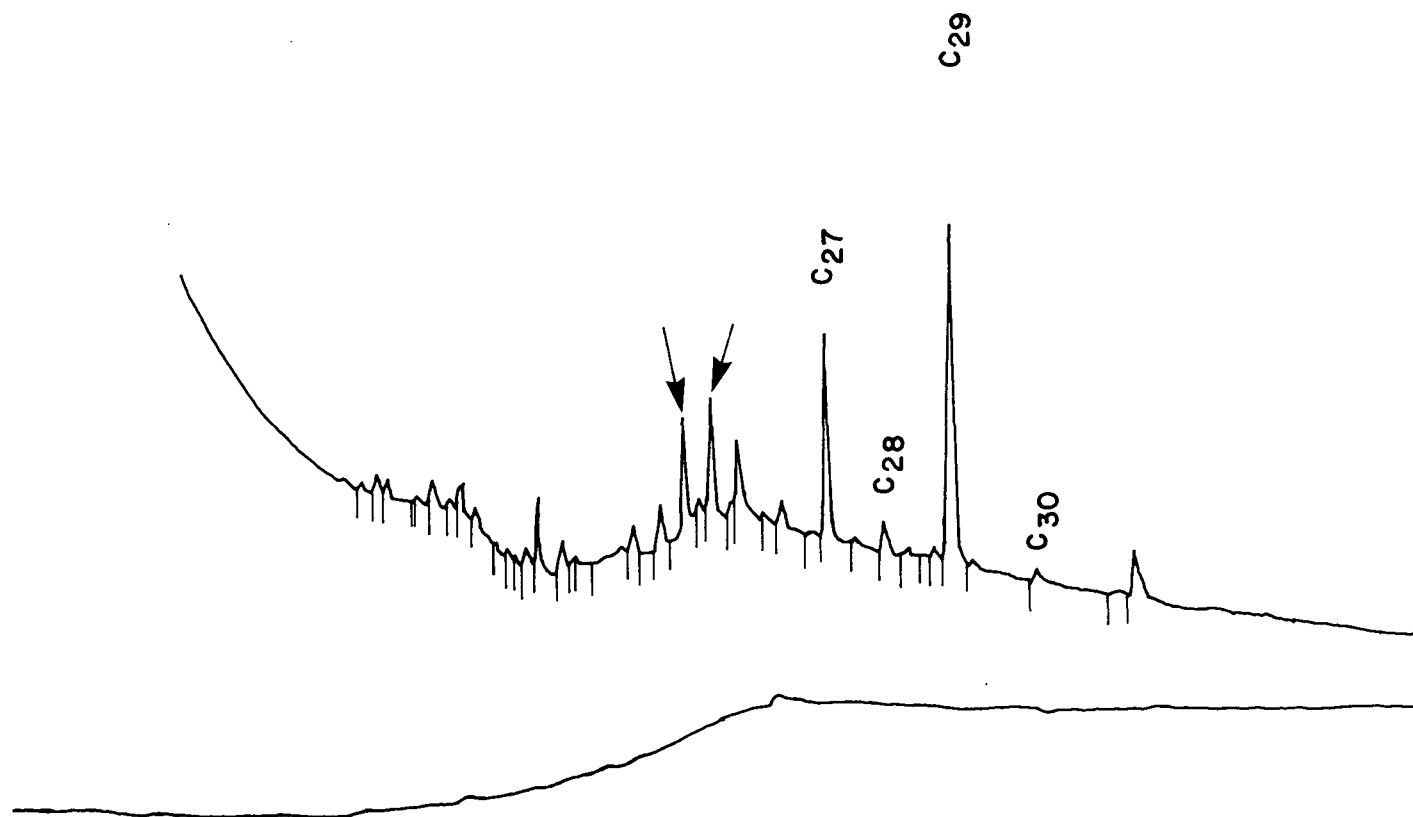


FIG.6 GLC spectrum of subsurface (10 cms) sediment sample from quadrat A5, an oiled plot cleaned by burning oiled vegetation with a blow-torch.

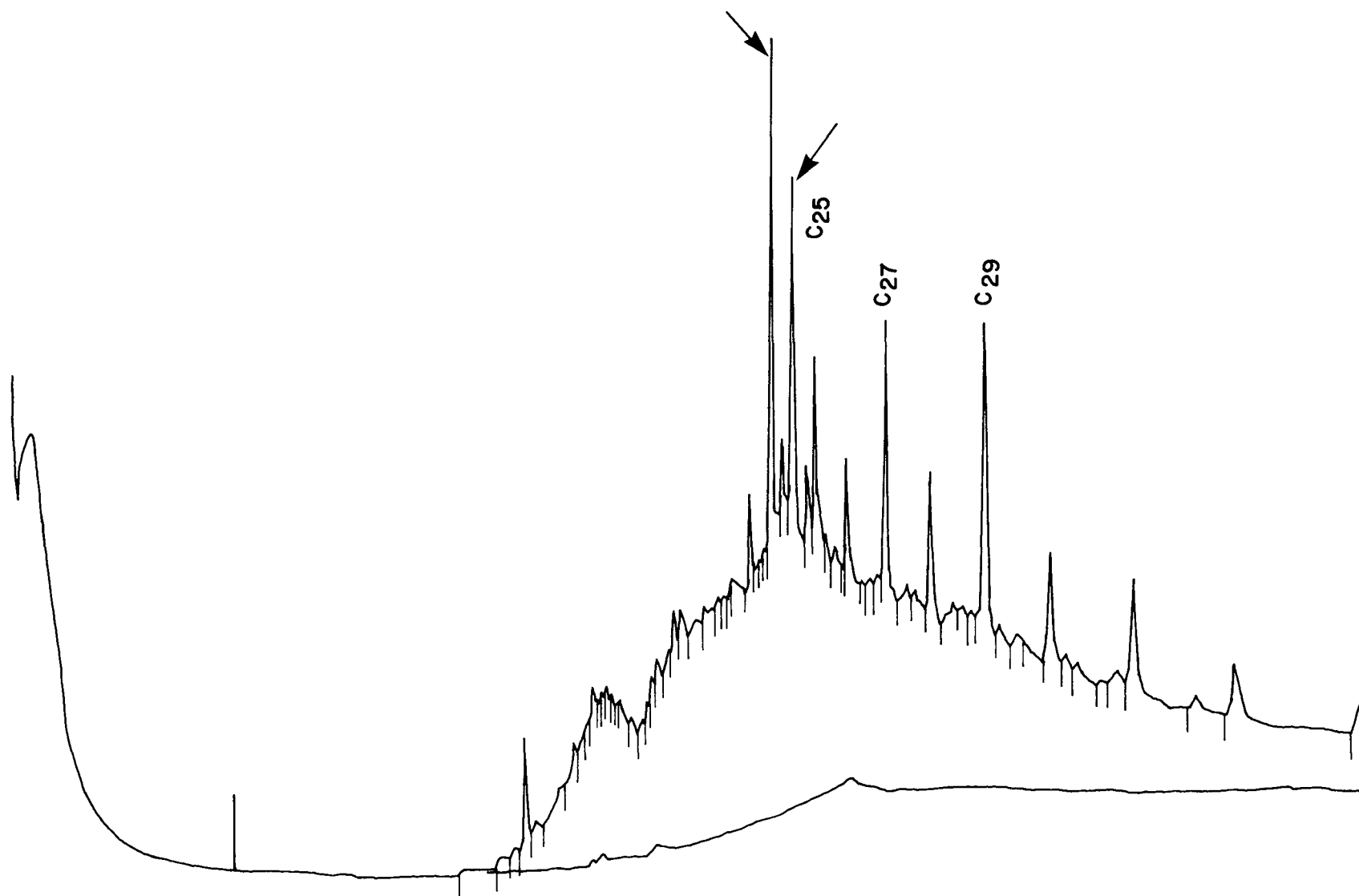


FIG. 7 GLC spectrum of subsurface (10 cms) sediment sample from quadrat A6, an oiled plot cleaned by burning oiled vegetation with varsol.

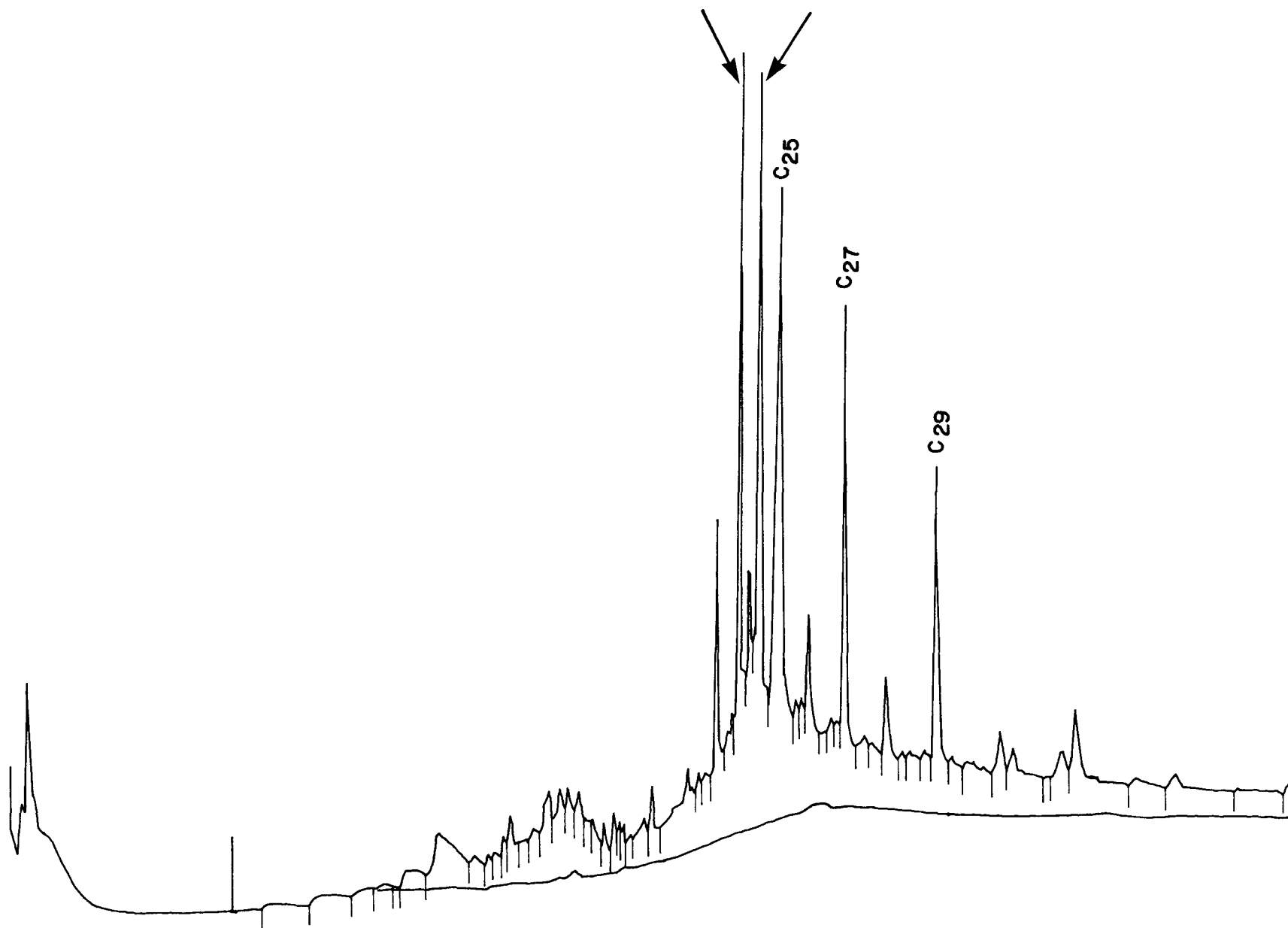


FIG. 8 GLC spectrum of subsurface (10 cms) sediment sample from quadrat A7, an oiled plot cleaned by burning vegetation with the aid of gasoline and oil.

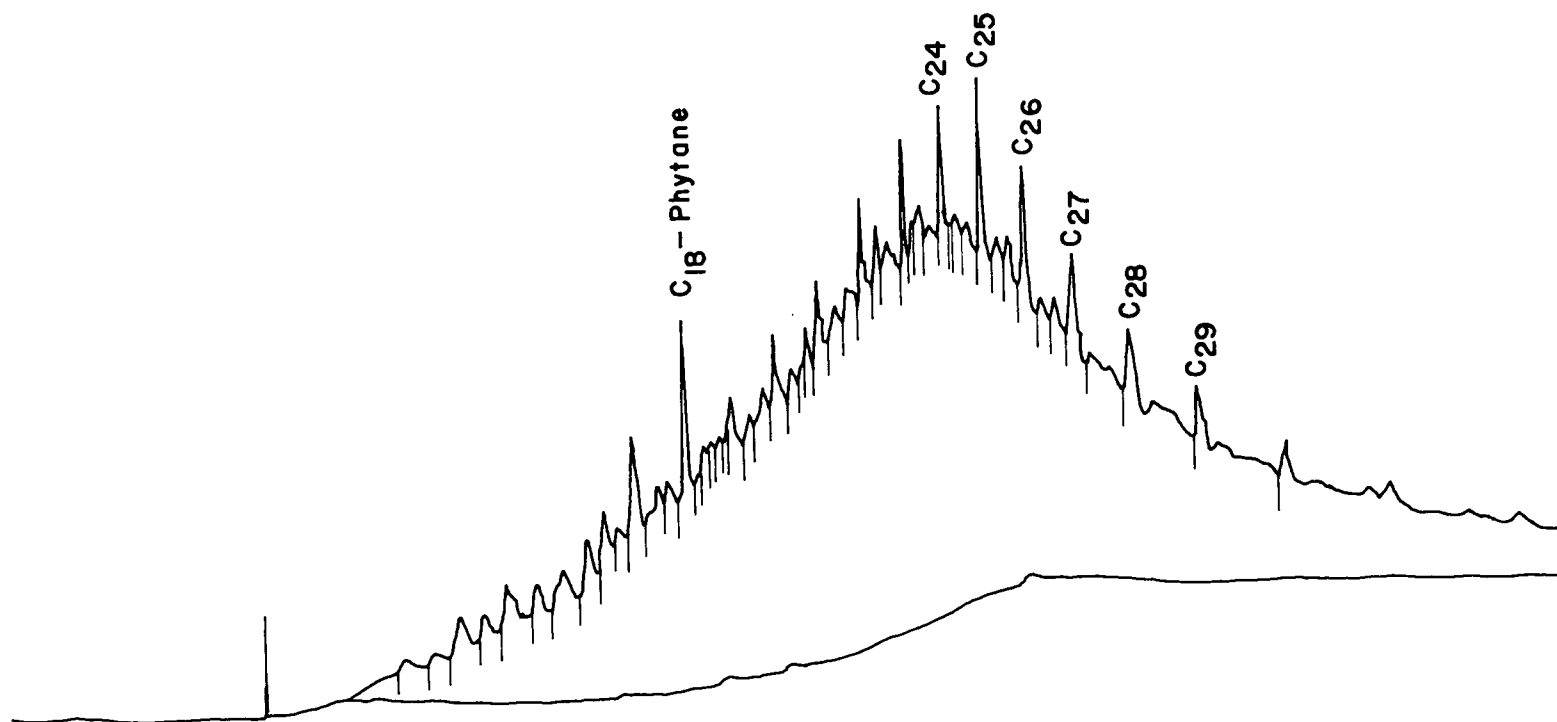


FIG. 9 GLC spectrum of subsurface (10 cms) sediment sample taken from quadrat A7 (control), an oiled plot adjacent to A7 but left uncleaned in 1974.

8. Quadrat A8 (Figure 10): The strong odd-C preference indicates most of the material is of terrigenous origin. The small C_{18} /phytane peak indicates the presence of some petroleum-derived hydrocarbons.
9. Quadrat A9 (Figure 11): The qualitative findings are similar to those of quadrat A8.
10. Quadrat B1-surface (Figure 12): The peaks are not representative of oil-derived n-alkanes. The small unresolved envelope suggests a small amount of oil-derived material.
11. Quadrat B1-subsurface (Figure 13): The spectrum is characteristic of weathered Bunker C fuel oil, with some degradation at the low end probably due to microbial activity.
12. Quadrat B2 (Figure 14): The unresolved envelope may be due to Bunker C oil, but nearly complete degradation of n-alkanes has occurred.

3.1.3 Qualitative Analysis - Aromatics:

From all the samples examined, it can be seen that the synchronous fluorescence emission spectra closely resemble that of ARROW Bunker C (viz. Figures 15 and 16). All spectra show the major peak at 360 nm, with the minor peaks or shoulders at 320, 330 and 400 nm.

Specific compositional differences or changes due to weathering could not be determined by this method.

3.2 Revegetation of Oiled Plots

The marsh sites sampled are shown in Plates 2 through 12. All quadrats were covered to some extent with a thin layer or crust of tar 3 to 4 mm thick. These tar crusts were broken up into large scales or flakes, ca. 100 cm² each, displaying a grey appearance of weathered oil on the upper exposed sides and a blacker, oilier appearance on the undersides (viz. Plates 6 and 10). Even those plots which appeared to have undergone considerable revegetation, such as quadrats A3 (Plate 3) and A7-b (Plate 7), were found to have tar packed down between the bases of the marsh grasses.

Revegetation ranged from nil to considerable. While some plots remained nearly denuded of grasses, others experienced regrowth with areal coverage over 50%. Those with revegetation included the control quadrats A2 and A7-b (Plates 2 and 7), and quadrats A3 (Plate 3) and A4 (Plate 4) from which oiled vegetation was removed by sickle and pitchfork and by hand respectively.

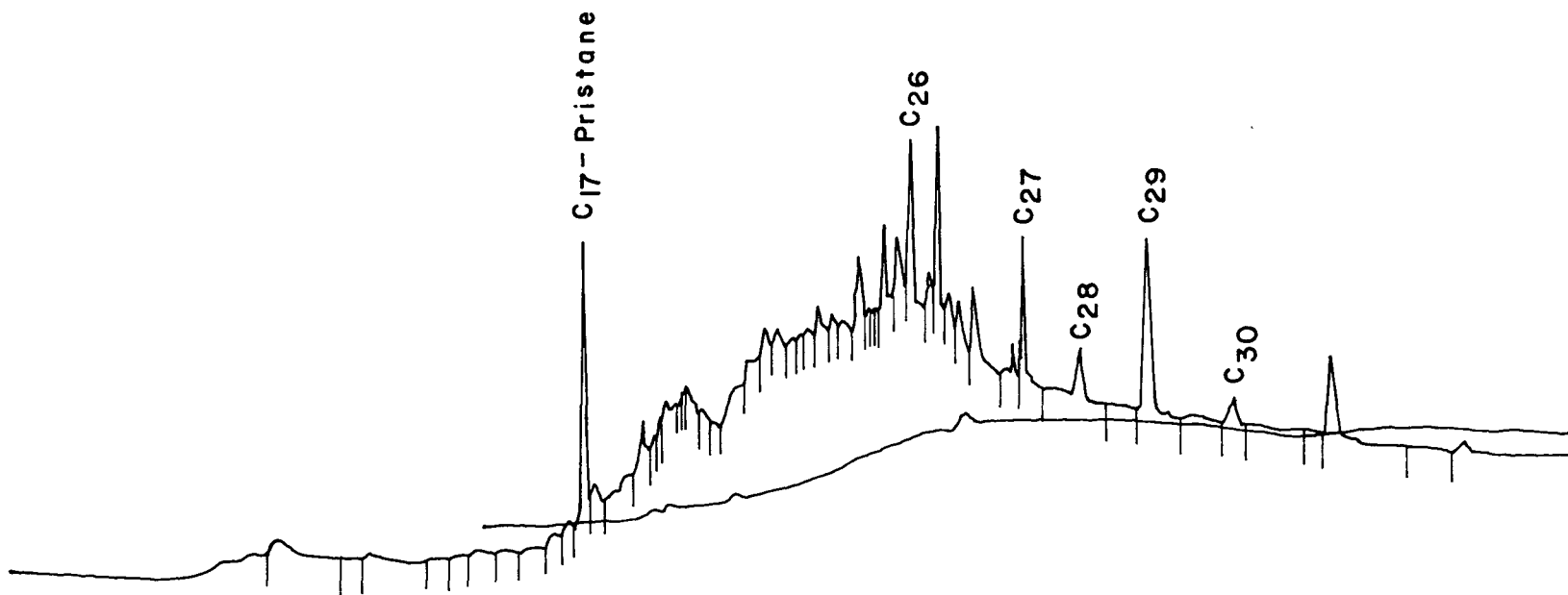


FIG.10 GLC spectrum of subsurface (10 cms) sediment sample from quadrat A8, an oiled plot cleaned by digging out oiled vegetation with a spade.

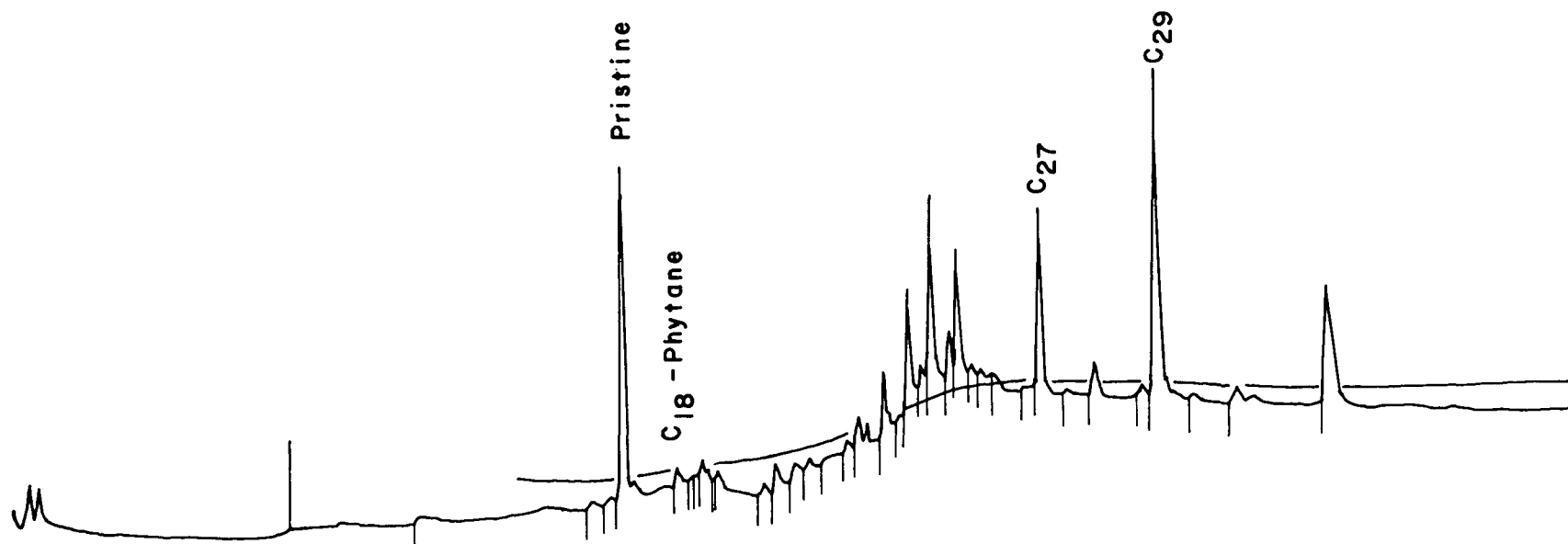


FIG. 11 GLC spectrum of subsurface (10 cms) sediment sample from quadrat A9, an oiled plot cleaned by cutting out oiled vegetation and surface root structure by saw.

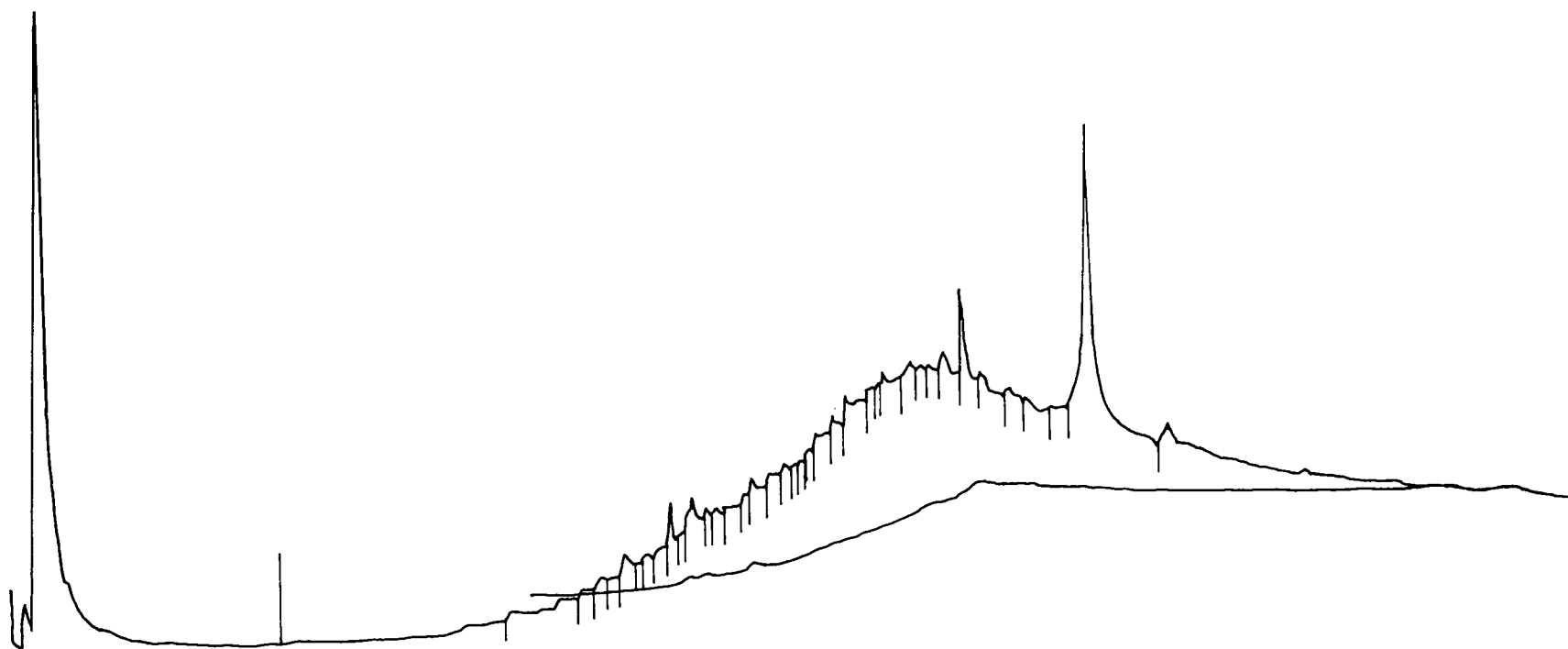


FIG. 12 GLC spectrum of surface sample from quadrat B1 , an oiled plot cleaned by removing surface vegetation with a sod - cutting machine .

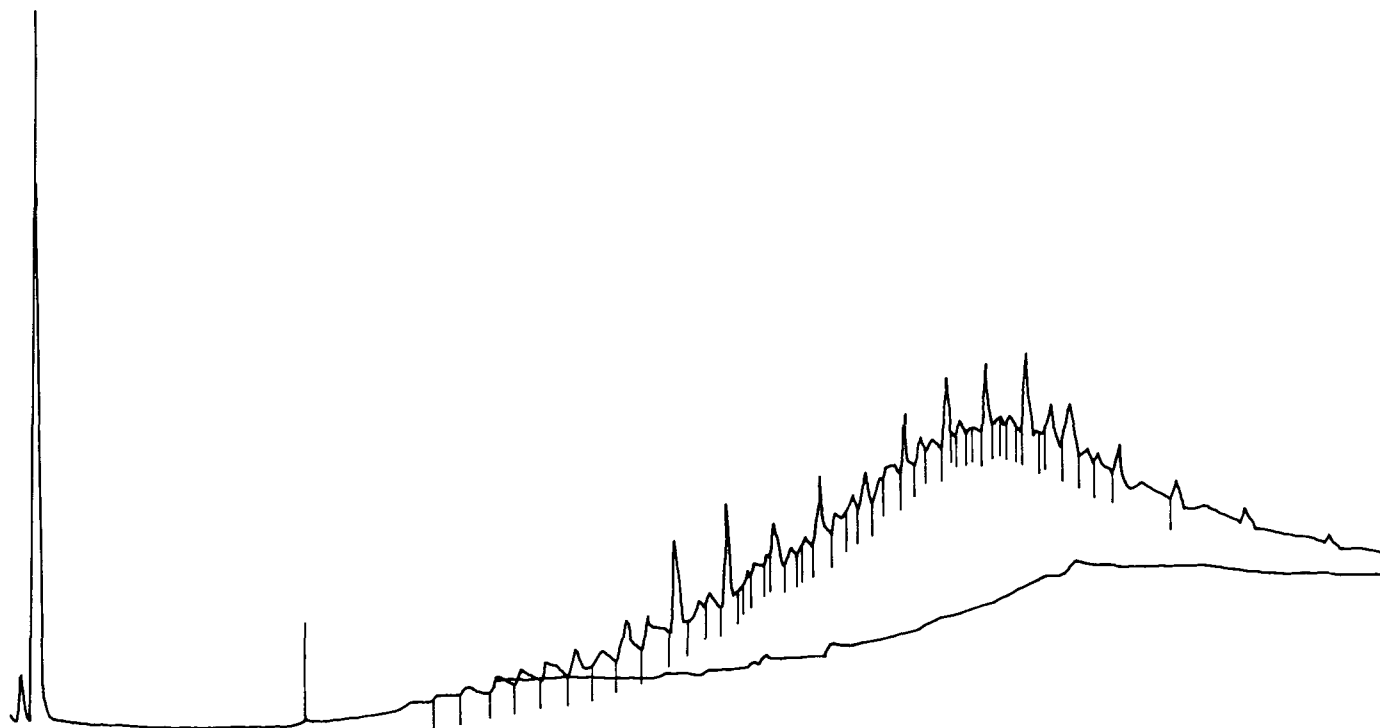


FIG.13 GLC spectrum of subsurface (10 cms) sediment sample from quadrat B1 (viz. FIG. 11), an oiled plot cleaned by removing oiled vegetation with a sod-cutting machine

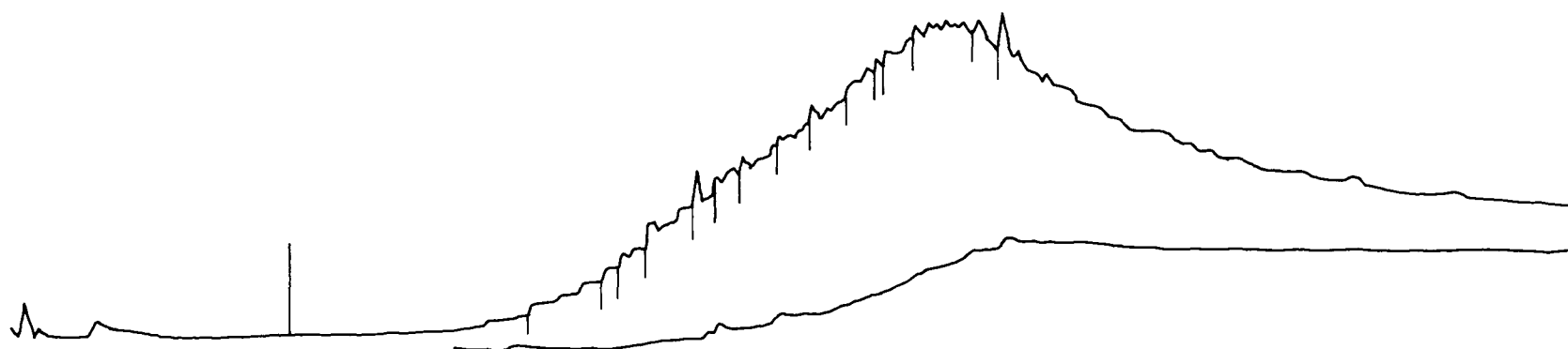


FIG. 14 A GLC spectrum of subsurface (10 cms) sediment sample from quadrat B2 , an oiled plot treated by plowing under the oiled vegetation .

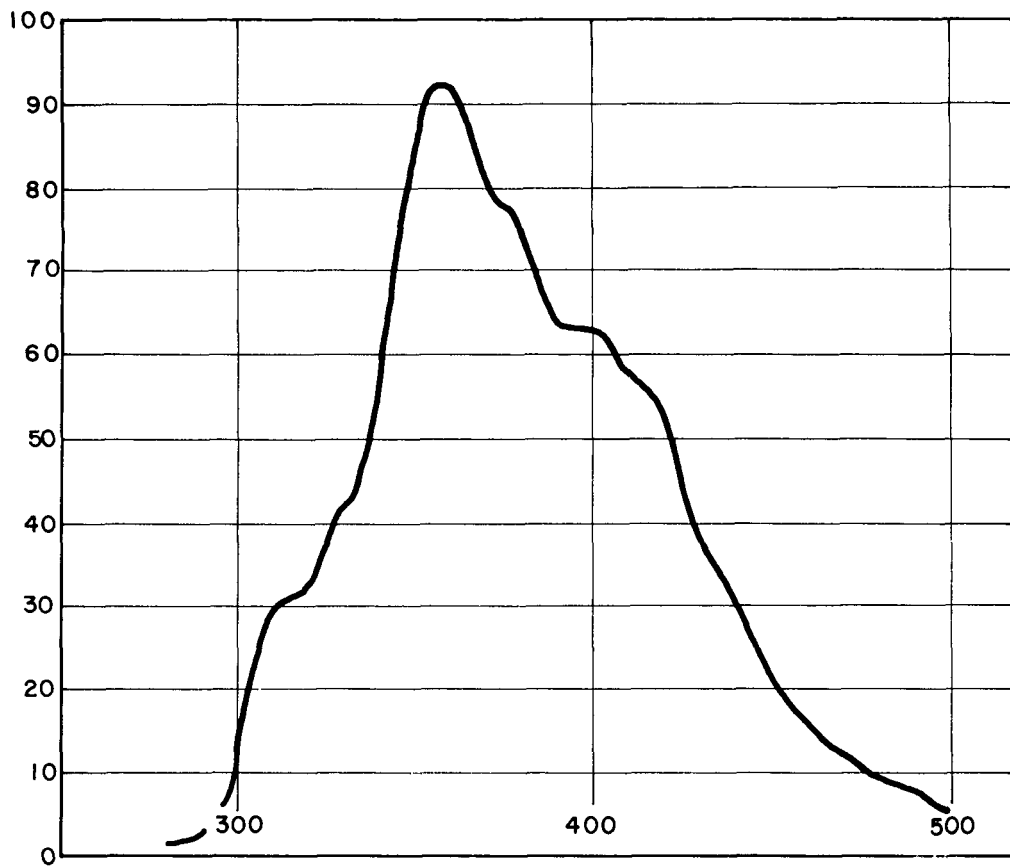


FIG.15 Synchronous fluorescence spectrum of ARROW Bunker C oil.

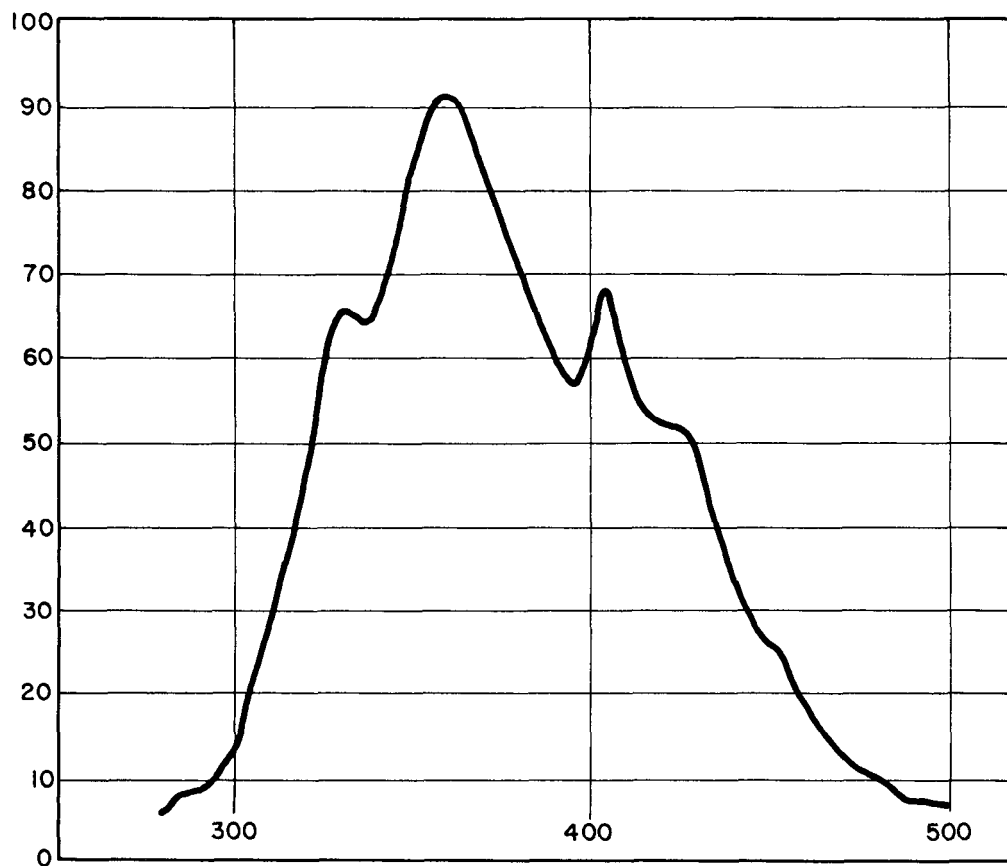


FIG.16 Synchronous fluorescence spectrum of heavily oiled non-cleaned control plot A2.



Plate 2. Quadrat A2, a non-cleaned, heavily oiled plot; rephotographed on August 20, 1976.



Plate 3. Quadrat A3, heavily oiled plot cleaned by sickle and pitchfork in 1975; rephotographed on August 20, 1976.



Plate 4. Quadrat A4, oiled plot cleaned by hand removal of oiled vegetation; photographed August 20, 1976.



Plate 5. Quadrat A5 (on right) and A6 (on left); A5 cleaned in 1975 by burning off vegetation with a blowtorch; A6 by igniting vegetation with varsol; each photographed on August 20, 1976.



Plate 6. Quadrat A7 cleaned by burning oiled vegetation with gasoline and oil in 1975; photographed August 20, 1976.



Plate 7. Quadrat A7-b, non-cleaned oiled area adjacent to quadrat A7; photographed in August 20, 1976.

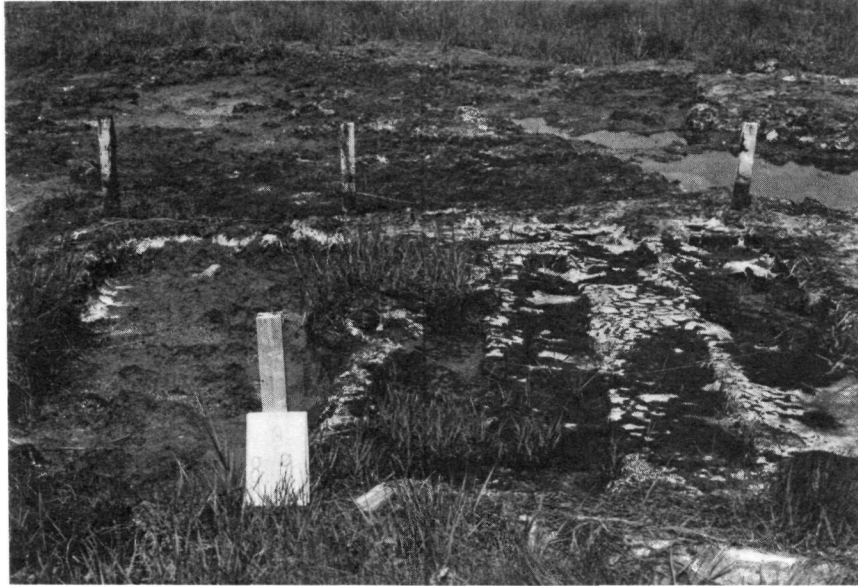


Plate 8. Quadrats A8 (on left) and A9 (on right); A8 cleaned by digging out oiled vegetation, and A9 by cutting out root mat with a saw; photographed August 20, 1976.

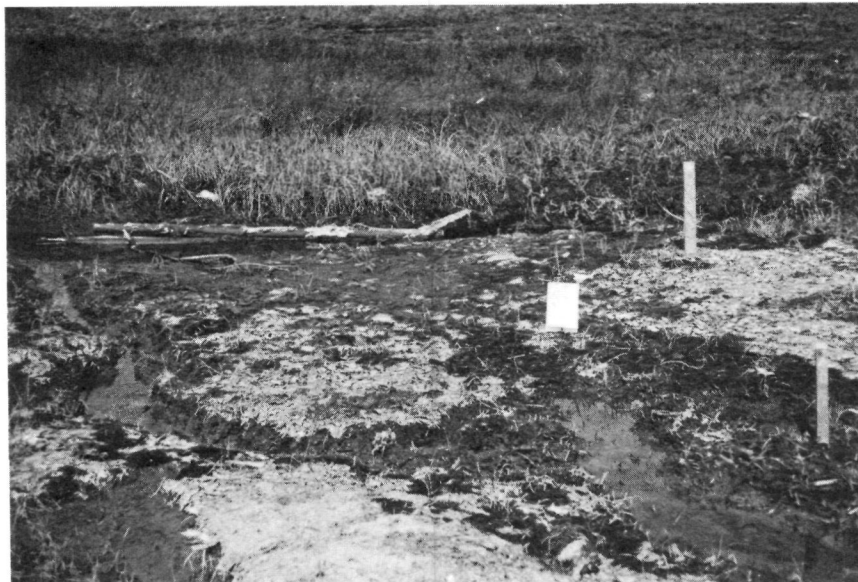


Plate 9. Quadrat B1, a heavily oiled plot cleaned by removing vegetation with a sod-cutter; photographed on August 20, 1976.



Plate 10. Detail of quadrat B1 (see Plate 8) showing tar crust overlying the sediments; photographed on August 20, 1976.



Plate 11. Quadrat B2, a heavily oiled marsh area in which oiled vegetation was turned under by plow; photographed on August 20, 1976.



Plate 12. Detail of quadrat B2 showing tar crust overlying the sediments; black appearance of underside of tar lump visible next to note-pad; photographed August 20, 1976.

The test plots still generally devoid of revegetation include those quadrats which were cleaned by burning off oiled vegetation (Plates 4 and 5), those from which some of the root mat, together with oiled vegetation, had been removed either by digging or sawing (Plate 8), those cleaned by a sod-cutter (Plates 9 and 10) and those in which oiled vegetation had been turned under by plow (Plates 11 and 12).

The vegetation which recolonized the test plots did not appear any different from that found in the other parts of the marsh, i.e. reed and grass types. Measurements were not made, however, with respect to growth, shoot size, seedbearing, etc. for the purpose of determining whether differences existed between grasses from oiled and non-oiled areas.

4 DISCUSSION

The cleaned areas of the Miguasha Marsh have undergone re-contamination, most likely through the raising and lowering of the water level by tidal action. This was evidenced by an examination of the tar crust which covered such thoroughly cleaned plots as A8 and A9 from which some of the root mat and all of the oiled vegetation had been physically removed. Although such re-oiling influences the data and analysis, it also points up the necessity for thorough cleaning of the entire oiled area, otherwise the threat of recontamination remains.

As was found in all samples, oil had penetrated the sediments at least ten centimetres, and in several quadrats the presence of oil was evident at 15 and 20-cm depths. At such depths, significant degradation of the n-alkane (aliphatic) component of the spilled oil had occurred during the 2-year period following the spill incident. The aromatic character of the oil, however, appears to have remained persistent, not having differed demonstrably from the original oil. Measurements of hydrocarbon concentrations, particularly those taken from plots cleaned by machinery, were high and likely resulted from this aromatic component in Bunker C oil. This tenacity of the aromatic fraction of spilled oil is a common feature of oil degradation in sediments, and therefore not unexpected.

From an oil cleanup point of view, remarkable differences are observed in both the success of cleanup operations and in subsequent marsh revegetation (Table 2). The most effective methods of oil cleanup appear to be those involving the removal of oiled vegetation by hand, whereas those involving machinery resulted in the highest residual hydrocarbon concentrations.

Of the manual cleanup methods applied, those which proved to be most effective were the removal of contaminated vegetation by burning and the removal of both oiled vegetation and root mat portions by way of spade or saw. Only small amounts of Bunker-C-derived hydrocarbons were found in subsurface sediments following the application of these techniques.

Table 2. Summary of Miguasha Marsh Cleanup Study
(Vegetation and residual oil/sediment analysis incorporated).

Cleanup Technique	Quadrat	Hydrocarbon Concentration	Revegetation	Results of GLC Analysis
Non-cleaned	A1	7.2 ppm	+	traces of Bunker C n-alkanes
Controls	A7-b	13,600.0	+	weathered Bunker C
Hand removal of oil vegetation	A3	819.2	+	traces of Bunker C
	A4	616.5	+	some Bunker C
Burning	A5	6.4	-	mixture of terrigenous and Bunker C, unusual GLC peaks
	A6	11.3	-	mixture of terrigenous and Bunker C, unusual GLC peaks
	A7	18.3	-	mixture of terrigenous and Bunker C, unusual GLC peaks
Digging out vegetation and root mat portions	A8	8.6	-	mostly terrigenous n-alkanes
	A9	5.8	-	mostly terrigenous n-alkanes
Machinery	B1	115-145,860	-	eroded Bunker C
	B2	1,429.8	-	eroded Bunker C

Samples from the burned plots differed significantly from all others in one detail, however, in that the GLC spectra contained two unknown and unique n-alkane peaks. These peaks were found in the three burned plot samples only, and not in any of the other sediment samples, whether from oiled or non-oiled areas, nor in the raw Bunker C oil, whether original or weathered. The compounds giving rise to these peaks have not been identified. It appears likely that they represent some combustion products of the burned tar, since they were found in both blowtorch-burned and solvent-ignited quadrats. This points out an unknown and important factor in cleanup techniques involved burning, i.e. the possible introduction of further unknown products into the environment.

Of medium effectiveness were those manual labor methods whereby oiled vegetation was either pulled up by hand or cut out by sickle or scythe and carried away. Although considered incomplete in terms of cleanup generally, these methods did result in significant removal of spilled tar.

Least effective from an oil cleanup point of view was the utilization of heavy mechanical devices, such as sod-cutters and plows, whereby large amounts of oil were forced into subsurface sediments through churning up of marsh structures. Although most of the aliphatic fraction of this subsurface oil readily degraded in the relatively short period of two years, the aromatic, and allegedly more toxic component of the oil, persisted and apparently remain unaltered.

From these observations, and from a revegetation viewpoint, the best response to a marsh spill would appear to be either manual cleanup by the simple removal of oiled vegetation by sickle and by hand or leaving it untouched. The latter is, of course, less desirable in that re-oiling will undoubtedly occur with tidal flushing, which will, over time, result in high concentrations of oil penetrating the marsh root structures to considerable depths. As was the case with the Miguasha Marsh, the other cleanup methods would appear to either burn out or remove the regenerative root structure. Over the 2-year study period, very little reseeding from nearby non-oiled vegetation was observed.

5 CONCLUSIONS AND RECOMMENDATIONS

In the cleanup of oil spills in marshes, the use of machinery or burning techniques is to be avoided. Where possible, cleanup should be done by hand through the simple cutting and removal of oiled vegetation, and through the use of sorbent materials.

Mechanical cleanup by devices such as sod-cutters or plows results in increased penetration of oil into marsh sediments. Burning of oiled debris results in marked decreases in revegetation. This cleanup technique also makes possible the introduction of unknown combustion products into the environment.

Reseeding cleaned marsh areas should be investigated, particularly with respect to areas cleaned by plowing or burning.



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