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THE WEST FALMOUTH OIL SPILL

Persistence of the Pollution Eight Months After the Accident

by

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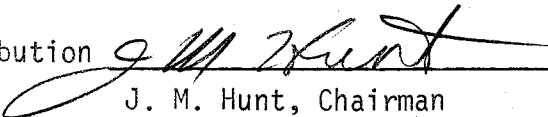
TECHNICAL REPORT

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13. ABSTRACT A spill of 650,000-700,000 liters of #2 fuel oil has contaminated the coastal areas of Buzzards Bay, Mass. The present report summarizes the results of our continuing chemical and biological study which were available at the end of May 1970, more than eight months after the accident.  The effects of environmental exposure on the composition of the oil are discussed; many analytical parameters are sufficiently stable to permit continued correlation of the oil remaining in sediments and organisms with the fuel oil involved in the spill.  Oil from the spill is still present in the sediments, inshore and offshore and in the shellfish. A further spread of the pollution to more distant offshore regions has occurred during mid-winter; as a result, the pollution now covers a much larger area than immediately after the accident. The first stages of biological (presumably bacterial) degradation of the oil are now evident especially in the least polluted regions; however, it has depleted predominantly the straight and branched chain alkanes. The more toxic aromatic hydrocarbons are resistant; as a result, the toxicity of the oil has not been diminished.  Where oil can be detected in the sediments there has been a kill of animals; in the most polluted areas the kill has been almost total. Shellfish that survived the accident have taken up the fuel oil. The 1970 crop of shellfish is as heavily polluted as was last year's crop. Oysters transplanted to unpolluted water for as long as 6 months retained the oil without change in composition or concentration.			

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WOODS HOLE OCEANOGRAPHIC INSTITUTION

WOODS HOLE, MASSACHUSETTS 02543

Phone (617) 548-1400

September 23, 1970

The Honorable Board of Selectmen  
Town Hall  
Falmouth, Massachusetts 02540

Gentlemen:

I take pleasure in forwarding to you a report describing the continuing investigation of the effects of the West Falmouth oil spill.

This work was done jointly with Mr. George Souza, Shellfish Warden of the Town of Falmouth by Dr. M. Blumer, Mr. J. Sass, Dr. H. L. Sanders, Dr. J. F. Grassle and Mr. G. Hampson. It is intended to publish the report.

As this work progresses we will submit to you additional reports. They will continue to outline the area where the pollution still persists and we hope that they may aid the Town in the decision where and when to restock the areas where the shellfish have been destroyed.

Sincerely yours,

  
Paul M. Fye

Enclosure

### ABSTRACT

A spill of 650,000 - 700,000 liters of #2 fuel oil has contaminated the coastal areas of Buzzards Bay, Mass. The present report summarizes the results of our continuing chemical and biological study which were available at the end of May 1970, more than eight months after the accident.

The effects of environmental exposure on the composition of the oil are discussed; many analytical parameters are sufficiently stable to permit continued correlation of the oil remaining in sediments and organisms with the fuel oil involved in the spill.

Oil from the spill is still present in the sediments, inshore and offshore and in the shellfish. A further spread of the pollution to more distant offshore regions has occurred during midwinter; as a result, the pollution now covers a much larger area than immediately after the accident. The first stages of biological (presumably bacterial) degradation of the oil are now evident especially in the least polluted regions; however, it has depleted predominantly the straight and branched chain alkanes. The more toxic aromatic hydrocarbons are resistant; as a result, the toxicity of the oil has not been diminished.

Where oil can be detected in the sediments there has been a kill of animals; in the most polluted areas the kill has been almost total. Shellfish that survived the accident have taken up the fuel oil. The 1970 crop of shellfish is as heavily polluted as was last year's crop. Oysters transplanted to unpolluted water for as long as 6 months retained the oil without change in composition or concentration.

### INTRODUCTION

A spill of 650,000 - 700,000 liters of #2 fuel oil in Buzzards Bay, Massachusetts, U.S.A., on September 16, 1969 has severely polluted the coastal waters of Falmouth and of Bourne, Mass. In a preliminary publication (Blumer et al., 1970 a,b) we reported that the hydrocarbons in the sediments and the shellfish of the polluted area corresponded analytically to the fuel oil involved in the spill. We were able to distinguish the pollutants from the natural hydrocarbons that occur normally in the sediments and organisms of this area. From this we concluded that the spill had contaminated the sediments and organisms and that it was remarkably persistent.

We now present additional results of our continuing study of the long term effect of this oil spill and we will discuss those analyses which were available to us at the end of May 1970, more than eight months after the accident. Future reports are planned to cover additional data over a longer time period.

Analyses of the #2 fuel oil that was involved in the accident have been published (Blumer et al., 1970 a,b). It has a boiling range of 170-370°C; the largest fraction distills between 200 and 300°C. The aromatic content by column chromatography is 41%. The gas chromatogram (Blumer et al., 1970 a, b, Fig. 3A) shows the predominance of normal paraffins, they range approximately from n-decane to n-docosane with a maximum in the C<sub>14</sub>-C<sub>15</sub> range. In addition to the straight chain alkanes several other homologous series are evident. Next in intensity are peaks which we assign

to the isoprenoid alkanes from  $C_{13}H_{28}$  to  $C_{20}H_{42}$  (phytane). Minor and only partly resolved peaks may represent other branched alkanes; in addition, a broad and unresolved background from cycloparaffins and aromatics is present. This high, unresolved background is typical for gas chromatograms of crude oil and oil products that are analyzed on columns of moderate separating efficiency.

### Weathering of Oil

Crude oil or oil products spilled in nature are altered by evaporation, by dissolution and by chemical or biological attack. Not all hydrocarbons are affected at the same rate; environmental exposure ("ageing", "weathering") therefore affects both the contamination level and the relative composition of the residual oil. From oil analyses obtained at different times after an accident we can estimate the relative degradation rates, and the observation of compositional changes can be interpreted in terms of the processes that are effective in the degradation.

Evaporation depletes the lower boiling components of an oil but leads to little or no fractionation between hydrocarbons of the same boiling point that belong to different structural series. Gas chromatograms on Apiezon columns (boiling point columns) show the result of evaporation as a uniform lowering of the lower molecular weight envelope.

Dissolution also removes preferentially the lower molecular weight components of an oil. However, aromatic hydrocarbons are more readily dissolved than alkanes of the same boiling point

because of their greater solubility. This effect is easily recognized in the gas chromatograms obtained on Apiezon columns.

Biochemical (microbial) attack affects compounds within a much wider boiling range than evaporation and dissolution. Hydrocarbons within the same homologous series are attacked roughly at the same rates. Normal paraffins are most readily degraded. In gas chromatograms this type of degradation manifests itself as a lowering of the ratios between straight chain and adjacent branched paraffins, e.g. between n-heptadecane and pristane or between n-octadecane and phytane. Extended biochemical degradation then results in gradual removal of the branched alkanes. Cycloalkanes and aromatic hydrocarbons are more resistant and disappear at a much slower rate.

In combination, dissolution and bacterial attack lead first to a relatively rapid depletion of the more soluble lower boiling aromatic hydrocarbons. Later, the overall aromatic content of the residual oil rises again, when the less resistant saturated hydrocarbons are removed by bacteria. Much of the toxicity of the oil is associated with the aromatic fraction; this implies that the oil is not readily detoxified but may, on an equal weight basis, be more toxic after considerable degradation than immediately after the spill.

Chemical degradation processes of oil during weathering are not well understood. However, the overall result appears analogous to that obtained in technical oxidation processes (e.g. "blowing" of asphalt). Oxidation affects most readily the aromatic hydrocarbons



of intermediate and higher molecular weight; its effects are recognized analytically by an increase in the insoluble polymer fraction ("asphaltenes").

#### Stable Parameters of Oil

Weathering processes modify spilled oils; however, many compositional parameters are relatively stable and are not obliterated readily until an advanced stage of decomposition is reached. These parameters may aid in the identification of a hydrocarbon pollutant and in the correlation with its source.

Boiling range. Evaporation and dissolution affect the molecular weight distribution of an oil. However, only the lowest boiling components in the gasoline range are lost rapidly and completely. Thus, oil recovered from the feathers of birds exposed to an oil slick off Marthas Vineyard Island, Massachusetts, U.S.A. in February 1970 still contained normal heptane and normal octane. (Blumer, 1970, unpublished). Similarly, a light paraffinic crude oil that washed ashore on Marthas Vineyard in June 1970 was sampled immediately and after eight days exposure on the beach. The gas chromatograms of both samples were identical and extended in boiling range as low as n-octane. Even in oil that has been exposed to weathering for several months (tar balls from the Mediterranean Sea (Blumer et al., 1970 a,b), "tar" from the beaches of Santa Barbara (Blumer, 1970, unpublished) hydrocarbons boiling as low as n-decane can still be recognized. Above n-tetradecane evaporation and dissolution is very slow and has little effect on the hydrocarbon concentrations. Chemical and biological degradation are even less dependent on the molecular weight than evaporation and dissolution, they tend to preserve

the original boiling point distribution of the pollutant.

Thus, it is possible, even after considerable weathering, to estimate the initial boiling range of an oil spill. This permits the classification of a hydrocarbon pollutant, for instance as a natural product or as a full range crude oil, as a processed product (e.g. a fuel oil) or as a residuum (a bunker oil or an asphalt). Characteristic features in the relative distribution of higher boiling hydrocarbons are stable and may permit the recognition of oils from different geographic areas or of modified crude oils (e.g. blends or tank sludges) (Brunnock et al., 1968, Ramsdale and Wilkinson, 1968).

Hydrocarbon Ratios. Crude oils of different age, geological and thermal history differ characteristically in their chemical composition. Oil from different oil fields, from different producing horizons and oil products that have undergone refining and chemical operations can thus be distinguished by chemical analysis. Even in spilled and partly degraded oil we can still recognize the more stable features.

Bacterial degradation and solubilization change the ratios between hydrocarbons belonging to different structural series. However, bacterial and chemical degradation exhibit little selectivity within the same homologous series. Thus, bacterial degradation alters the ratio between paraffins and isoparaffins or between paraffins and aromatics; however, ratios between adjacent paraffins, e.g. between heptadecane and octadecane or between heptadecane and hexadecane remain fairly constant. Other homologous series that are less readily degraded provide even more stable hydrocarbon ratios. The diagnostic use of stable hydrocarbon ratios is

best limited to the higher boiling ranges of crude oil, e.g. above C<sub>14</sub> where the effects of solubilization and volatilization are minimal.

Closely related homologous series respond to the combined effects of weathering in a very similar manner. Therefore, ratios between adjacent members of such series can be expected to be stable and of diagnostic value. For instance, isomeric isoalkanes of similar substitution type and isomeric alkylcycloparaffins or alkylated aromatics should provide stable markers for the characterization of oils and oil products.

The number of such characteristic stable parameters can be expected to be large; often simple analytical techniques may be sufficient for the reliable measurements of many stable parameters, but in the most demanding cases more sophisticated, but existing instrumentation, e.g. mass spectrometry coupled with gas chromatography, may have to be utilized.

## RESULTS

Our investigation of the long term physical and biological effects of the West Falmouth oil spill continues; we now report the data available at the end of May 1970.

### Samples and Techniques

The experimental techniques are identical with those reported previously. All gas chromatograms were obtained on new Apiezon columns. Their greater efficiency is evident from the better resolution of pristane from heptadecane and of phytane from octadecane.

The offshore or subtidal sediments were collected with a 1/25 m<sup>2</sup> Van Veen grab. The top 2-3 cm of the undisturbed samples were scraped off for analysis. At the inshore or intertidal stations the first three centimeters of the sediments were sampled for analysis. In the laboratory all samples were frozen and kept at -25°C in sealed glass containers.

The benthic, subtidal fauna was also collected from the 1/25 m<sup>2</sup> Van Veen grab. The entire grab sample was gently washed through a 0.295 mm mesh screen. The component retained on the screen was preserved in 5% formalin, then transferred to 70% ethanol and finally stained with Bengal. The animals were separated under the microscope from sediment and detritus, they were then sorted into species.

Sediments for chemical analysis were obtained through our sampling program at stations 7, 20, 31, 35 and 37 (Fig. 1). Inshore sediments were taken in Wild Harbor River at stations II, IV and V and at coring locations 1, 2, 4 and 5. Sediment samples from West Falmouth Harbor were provided by the Department of Natural Resources, Commonwealth of Massachusetts (Mr. Arnold Carr) (Fig. 2).

Shellfish were collected in West Falmouth Harbor and in the adjacent, uncontaminated Great Sippewissett Marsh. Shellfish samples from North Falmouth and Bourne (Megansett Harbor, Fiddlers Cove, Rands Canal and Squeteague Harbor, Fig. 2) were obtained through the cooperation of the Department of Public Health, Commonwealth of Massachusetts (Mr. M. Boschetti).

Contaminated oysters, taken in Wild Harbor River on November 12,

1969, after 57 days exposure to the polluted environment, were maintained in flowing sea water at this laboratory. One of these oysters was frozen on January 23, 1970, after 72 days in uncontaminated water. Two additional specimens were kept in clean water for 180 days until May 11, 1970.

#### Analytical Results - Offshore Sediments

Station 31 (Silver Beach Harbor), first analyzed in September 1969 (Blumer et al. 1970 a,b, Fig. 3B) continues to yield the most heavily contaminated offshore sediments (Fig. 3A - G). The chromatograms of the sediment extract are strikingly similar to those of the fuel oil involved in the spill and agree in the carbon number range and in the relative distribution of adjacent homologous hydrocarbons. The principal differences, just after the spill, between the fuel oil and the hydrocarbons in the sediment can be attributed directly to the partial dissolution of the lower boiling hydrocarbons, especially those in the alicyclic and aromatic fraction. Thus, we find a general decrease in the lower molecular weight envelope, below C<sub>15</sub>, and a relative increase in the abundance of the lower boiling paraffins over the background from cycloalkanes and aromatics. At the elution position of n-tridecane, the background level has decreased almost twofold, compared to the fuel oil.

Some degradation of the higher boiling normal paraffins, presumably by bacterial attack, can be noticed. The ratio of n-heptadecane to pristane (n-C<sub>17</sub>/Pr.) in the fuel oil is 2.6; in the sediments sampled on September 28, 1969 twelve days after the accident, this has decreased to 1.5.

The large peak after eicosane ( $C_{20}H_{42}$ ) in the September sample can be attributed to heneicosahexaene (HEH), an olefinic hydrocarbon that is abundant in algae and serves as a marker of the biological hydrocarbon contribution to these sediments (Blumer et al., 1970 c).

The chromatograms show only minor changes in the fuel derived hydrocarbons between September and December 1969. The slow rate of solubilization and bacterial degradation is evident, for instance, from the nearly constant content of low boiling hydrocarbons and from the constant heptadecane to pristane ( $n-C_{17}/Pr.$ ) ratio. However, the amount of the algal derived HEH decreases and remains low through May 1970. In other, less polluted areas this olefin persists throughout this period. This suggests that the high pollution level in Silver Beach Harbor may have affected the normal algal flora.

A slight increase in the oil content of the sediments occurs between September and October 1969, (Table 1) it is followed by a much more dramatic increase between December 1969 and March 1970. At the same time, the composition of the oil within the sediments is altered; the  $n-C_{17}/Pr.$  ratio increases from 1.3 in December 1969 to 1.6 in March 1970. More dramatic is the increase in aromatics and cycloparaffins (equivalent to the background level) at lower molecular weights. The general chromatographic pattern and the ratio between adjacent isomeric and homologous hydrocarbons demonstrate convincingly that the pollution is still derived from the fuel oil spill in September. Continued bacterial degradation again lowers the heptadecane to pristane ratio from 1.6 in March to 1.2 in May 1970.

The rise in oil content by March 1970 in Silver Beach Harbor and the observed compositional change strongly suggests that oil from the spill has been remobilized and trapped in the sediments of this area, during early 1970. This oil has been less affected by bacteria (higher  $n\text{-C}_{17}/\text{Pr.}$  ratio) but more changed by solubilization (less low boiling aromatics) than the oil found in situ between September and December.

Oil Films in Wild Harbor (near Station 28, Fig. 1) were collected and analyzed on November 21, 1969 (Blumer et al., 1970 a,b, Fig. 3C). The chromatographic distribution pattern of the #2 fuel oil is easily recognized. In its  $n\text{-C}_{17}/\text{Pr.}$  ratio and in the low aromatic content in the  $\text{C}_{12}$  to  $\text{C}_{16}$  range the oil in these films strongly resembles the fuel oil that had been incorporated into the sediments. The presence of these oil films in Wild Harbor in the latter part of November 1969 proves that macroscopic quantities of free oil, derived from the oil spill in September, were still being released into the environment at that time.

Station 7 (off Nye's Neck). The chromatogram of the oil recovered from this location in September (Fig. 4A - E) strongly resembles that from Station 31, except for a slightly greater degree of bacterial degradation ( $n\text{-C}_{17}/\text{Pr.} = 1.25$ ). During the following months, degradation proceeds faster than at Station 31 and the ratio  $n\text{-C}_{17}/\text{Pr.}$  reaches 0.6 in November. A slight increase in this ratio and in the ratio of octadecane to phytane ( $n\text{-C}_{18}/\text{Pr.}$ ) is apparent in December and still evident in April 1970. Again, this change must be explained as the

result of an influx of relatively undegraded fuel oil into this area. The more rapid degradation at Station 7, compared to the more highly contaminated Station 31, in the months before this new influx, indicates that degradation proceeds more slowly in the more heavily polluted area. The olefinic hydrocarbon HEH, which is derived from algae, is present in all samples at this location in contrast to the more heavily contaminated station 31.

Station 20 (off Old Silver Beach) was originally selected as an uncontaminated control station for biological studies.

In the New England Region, the crustacean amphipods of the family Ampeliscidae are among the most characteristic elements of the offshore sandy sediments with some admixture of silt and clay. On September 24, 1969, eight days after the oil spill, replicate 1/25 m<sup>2</sup> Van Veen samples yielded 24 and 11 ampeliscids. No dead or decaying specimens were present. Three weeks later, the situation was altered radically. In a single sample only two living representatives were present, while the decaying remnants, detected by the more refractory eye lenses, numbered thirty. By mid-November, a single living individual was present in each of two replicate samples; the dead component numbered two fragments in one and none in the other sample. This condition remained essentially unaltered in December. On March 19, 1970 the same low density was evident in one of the paired samples, one living and three dead specimens; the other sample contained nine living and no dead ampeliscids.

The chemical analysis revealed the presence of fuel oil already



in the September samples (Fig. 5A - F).

As in the fuel oil, the hydrocarbons recovered from the sediments range from about C<sub>11</sub> to C<sub>23</sub>. Considerable biochemical alteration is evident. The n-C<sub>17</sub>/Pr. ratio is unity in September and becomes progressively lower in October and November. By December 1969 this ratio begins to climb again and reaches unity in April.

A slight increase in the oil content, measured in April 1970 (Table 1) and the reversal in the trend observed in the n-C<sub>17</sub>/Pr. ratio during the previous months, suggests again an influx of less degraded oil from a more heavily contaminated area, sometime between December and April. Heneicosahexaene, the olefin derived from algae, is evident in all samples and reaches a high relative concentration in May, together with heneicosane, its saturated equivalent.

Station 37 (off Nye's Neck) was selected as an uncontaminated biological control station after the oil pollution had reached station 20.

This station inherently supported larger populations of ampeliscids than station 20. The initial sample taken in mid-November yielded 142 living and five dead specimens. By January 6, 1970, there was a precipitous drop in numbers; only three living and 18 dead ampeliscids were recovered on that date. The February and March samples remained largely unchanged, with 13 living and ten dead and with eight living and 12 dead specimens, respectively.

The gas chromatographic data agree with the biological survey. No evidence for oil pollution was found in the November samples (Fig. 6A) but in January the fuel oil contamination is evident; the boiling range of the oil recovered from the sediment ranges from C<sub>11</sub> to C<sub>23</sub>, as in the original spill. Biological alteration has proceeded farther at this station than at the more contaminated stations closer to shore. By February 1970 the n-C<sub>17</sub>/Pr. ratio had dropped to 0.4. However, by March we observe the return of the ratio to 0.65 and in April to 0.8. At the same time, an increase in the oil content of the sediment (Table 1) becomes apparent. By May the n-C<sub>17</sub>/Pr. ratio had again dropped to 0.47. A similar trend is observed in the hydrocarbon pair n-octadecane/phytane. Phytane predominates in the February samples, in April octadecane has again increased considerable, but by May phytane is again more abundant than the straight chain hydrocarbon.

Again, these data indicate that oil derived from the accident moved into an area that had remained unpolluted for several months after the accident. Bacterial degradation is more rapid at this more remote station, however, the station experienced a new influx of less degraded oil between February and April. It is evident from the last chromatogram of this series (Fig. 6F) that the progressive degradation which we observe in the straight chain hydrocarbon fraction, does not deplete rapidly the cyclic saturate and aromatic fraction. The residual oil in May at this location is highly aromatic. Thus, the biological degradation of the fuel oil observed during the first eight months after the accident has not resulted in a detoxification of the oil.

The concentration of heneicosahexaene, which is derived from algae, is higher here than at the stations closer to shore, also we observe other biogenic hydrocarbons; as at station 20 n-eicosane which elutes just before HEH is present.

Station 35 (Near Cleveland Ledge). This station was also selected for biological reference when the previously uncontaminated control station 20 came under the influence of the pollution from the West Falmouth oil spill.

Like station 37, this station normally supports high densities of ampeliscid amphipods. The initial sampling in October yielded 73 living and two dead specimens. Densities remained high in the November, December, March and May samples, with 270 living and two dead, 112 living and no dead, 247 living and no dead and 137 living and three dead specimens, respectively.

Chemical analysis of the hydrocarbons recovered from this station in December 1969 shows the presence of fuel oil pollution (Fig. 7). Biological degradation has been considerable and has resulted in the removal of a large fraction of the straight chain hydrocarbons. It is interesting that the ampeliscid amphipods have been able to survive a concentration of oil at this station (Table 1) that proved lethal for them at the other stations closer to shore. Future work will have to decide whether this is due to the higher degree of biodegradation of the oil at this more remote location or to more favorable environmental conditions.

Results - Inshore Surface Samples

Station II (Wild Harbor River). The pollution level at this station is high compared to most offshore samples (Fig. 8A - C, Table 2). Again, pollution with #2 fuel oil is evident from the boiling range, extending from C<sub>11</sub> to C<sub>23</sub>, and from the relative abundances of adjacent homologues and isomers. The oil recovered from the sediments in December shows evidence of considerable biodegradation but only a minimal further change is observed between December 1969 and May 1970, as indicated by the detectable reversal in the ratio of octadecane to phytane. The slow rate of degradation during these five months suggests that the principal alteration of the oil has occurred before it was incorporated into the sediments.

Station IV (Wild Harbor River). The pollution at this upstream location is more severe than at Station II; (Fig. 9A - D; Table 2), the agreement in composition with the oil from other offshore and inshore locations is excellent and indicates that the fuel oil spill has affected this location. The alicyclic-aromatic background is high, this suggests modification through partial dissolution, in addition to the bacterial degradation which is evident from the n-C<sub>17</sub>/Pr. ratio. However, bacterial degradation has not proceeded as far as at the less contaminated Station II. The consequence of the persistence of the aromatic hydrocarbons is a high toxicity level of the residual oil.

Station V (Wild Harbor River, Fig. 10A - D). Biodegradation of the fuel oil in this heavily contaminated upstream area proceeds at a lower rate than in the less contaminated inshore regions. The n-C<sub>17</sub>/Pr.

ratio rises from 1.2 in December 1969 to 1.7 in April 1970; it then returns to 1.2 in May. This suggests a possible influx of less degraded oil in the intervening period and renewed bacterial degradation by May.

Our analyses of offshore and inshore samples show that the biodegradation of the oil is retarded in areas of high contamination. As a result, heavily polluted regions retain their oil content and may remain a source of relatively undegraded oil for the outlying areas; thus, secondary pollution may continue for a long time after the initial accident.

#### Results - Inshore Core Samples

Several core samples from Wild Harbor River were analyzed in order to study the penetration of the marsh sediments by the spilled fuel oil.

Core Location 1. Sediments at 43 to 51 cm below the surface at this location contain a complex mixture of hydrocarbons, extending at least from C<sub>13</sub> to C<sub>26</sub> (Fig. 11). A mixture of plant-derived, indigenous hydrocarbons with fuel oil components appears indicated. Characteristic for the fuel oil are the presence of and the ratios between phytane, pristane and the C<sub>18</sub> isoprenoid homologue, which elutes immediately after n-hexadecane.

Core Location 2. The chromatogram of the oil from the top 10 cm of this core show the presence of partly degraded fuel oil; (Fig. 12A, Table 2) the n-C<sub>17</sub>/Pr. ratio is 1.07. Hydrocarbons derived from algae are evident at and above n-eicosane.

A sandy layer of the same core at 51 - 58 cm below the surface yielded a larger extract than the surface sediment (Fig. 12B, Table 2). Again, the boiling point distribution and the ratios between adjacent homologues and isomers demonstrate convincingly that we are dealing with pollution derived from fuel oil. The chemistry of the oil at this level is different from that of the surface oil. The biogenic hydrocarbons are absent and the  $n\text{-C}_{17}/\text{Pr.}$  ratio is higher (1.44). This suggests that the oil in the deep sample has not been entrained by the sampling but that we are dealing with fuel oil pollution that has penetrated the marsh to this depth. The chemical differences between this oil and the surface oil suggests that both entered the sediment at different times and from sources representing different degrees of biodegradation of the same fuel oil. Another explanation, assuming more rapid degradation of the oil at the surface, seems unlikely in view of the observed low degradation rates of the oil contained in the surface samples in the Wild Harbor River estuary.

Core Location 4. The oil recovered from the surface sediments (Fig. 13A) agrees in composition with that from the adjacent River Station II. A clay sample at 36 - 43 cm below the marsh surface yielded hydrocarbons similar in quantity and composition (Fig. 13B, Table 2) to those at core location 1. Again, a mixture of biogenic hydrocarbons with fuel oil components is present; here the natural hydrocarbon background predominates.

Core Location 5. The highly polluted surface sample at this location agrees in hydrocarbon content and composition with other

surface samples from Wild Harbor River (Fig. 14A, Table 2). A core section at 41-51 cm below the surface also contains hydrocarbons derived from the fuel oil (Fig. 14B); the low n-C<sub>17</sub>/Pr. ratio indicates considerable biodegradation.

These samples were taken on a mud flat, adjacent to the river bank. An additional core was taken near this location, but starting at the river bank, 43-51 cm below the surface level of the mud flat and penetrating the sediment in a horizontal direction. The face of the river bank contains fuel oil (Fig. 14C), slightly fresher than the top of the mud flat. The innermost section of the core (46 cm below mud flat, 23-30 cm inward from river bank) was sliced and a sample from the center of the core was analyzed. The gas chromatogram (Fig. 14D) shows the presence of indigenous hydrocarbons and pollutants. The large peak preceding the heptadecane-pristane pair may correspond to one of the olefinic n-C<sub>17</sub> olefins, which are common in many algae (Blumer et al., 1970 d).

#### Analytical Results - Shellfish

Oysters and scallops that were exposed to the West Falmouth Oil Spill incorporated the hydrocarbon pollutants into their tissues (Blumer et al., 1970 a,b). The oil recovered from the shellfish shows the evidence of a much more intense biodegradation than the oil remaining in the inshore and offshore sediments. The normal paraffins are severely depleted, especially in the scallops, and the more resistant branched alkanes are also attenuated. The residual oil in the shellfish has a rather smooth boiling point

distribution, covering the range of the fuel oil, and it consists essentially of cyclic saturates and aromatics.

We now examined shellfish that had remained in the polluted area for seven months; additional specimens were removed from the polluted environment after two months and then maintained in clean water.

Polluted Oysters Maintained in Clean Water. Contaminated oysters from Wild Harbor River were removed to clean laboratory tanks on November 12, 1969. (Analyses of the shellfish taken at this time in: Blumer et al., 1970 a, b, Fig. 4B - D). The gas chromatograms (Fig. 15A, B) of the oil from oysters that were maintained in clean water for 72 and 180 days demonstrate that little, if any, further biodegradation of the hydrocarbons took place during this period. The hydrocarbon quantities (Table 3) remaining within the shellfish agree well with those present in November, especially if we allow for the apparent dilution of the oil by the growth of new tissue between November and May.

If the oil were localized solely within the digestive tract of the shellfish, elimination would be possible; the persistence of the pollutant, its presence in the adductor muscle of the scallop and the lack of further biodegradation show that the hydrocarbons have become part of the lipid pool of the animals. The persistence of natural hydrocarbons in the lipids of marine animals has been well established (Blumer, 1967; Blumer 1970 c); it now appears that hydrocarbon pollutants are equally well preserved within the lipids of marine animals.



Scallops and Quahaugs from West Falmouth Harbor. Mature scallops from West Falmouth Harbor were analyzed on November 13, 1969 (Blumer et al., 1970 a, b, Fig. 4). New specimens of scallops and quahaugs were collected from the same stations within the Harbor on April 17, 1970. These animals had been exposed to the spill as juveniles and would have reached maturity in summer 1970. The gas chromatograms (Fig. 16A - D) demonstrate the presence in these animals of hydrocarbons within the approximate C<sub>12</sub> to C<sub>23</sub> range, in agreement with the original boiling point distribution in the fuel oil. Additional evidence for the degradation over that observed in November 1969 appears now. The normal paraffin peaks remain barely recognizable and the isoprenoid peaks have been reduced further. The more soluble hydrocarbons of lower molecular weight have been partly depleted; as a result, the maximum amplitude of the hydrocarbon envelope now lies at about C<sub>18</sub>, compared to about C<sub>16</sub> to C<sub>17</sub> in last years shellfish crop and to C<sub>14</sub> to C<sub>15</sub> in the spilled oil. Surprisingly, the pollution level, (Table 3) on a weight basis, is nearly identical with that of the animals taken late in 1969 at the corresponding locations.

The severe alteration of the hydrocarbons contained in the shellfish may have occurred external to the animals, or within their digestive tract or in their lipids. In view of the great stability of the hydrocarbons in oysters that are maintained in clean water we favor the first or second alternative.

The persistence of the pollution in the new shellfish crop should be of concern to the public health authorities. Oil is

present now at the same concentration level as last year, but the higher aromaticity implies that the 1970 shellfish crop is more severely contaminated and more toxic than the 1969 crop had been.

To complement our analyses of shellfish from West Falmouth Harbor we have now also analyzed the bottom sediments of this area for their hydrocarbon content (Table 1 and Figure 16F - G). The presence of the fuel oil contaminant is evident, the low boiling hydrocarbons extend to n-dodecane. Biogenic hydrocarbons are also present, among them eicosane and higher boiling straight chain paraffins and olefinic hydrocarbons of 17 and 21 carbon atoms.

Scallops, Clams and Quahaugs from North Falmouth and Bourne

At the request of the Department of Public Health, Commonwealth of Massachusetts, shellfish from the adjacent harbors and estuaries of North Falmouth and Bourne were analyzed. These regions had remained open to the taking of shellfish in Fall 1969. The recent spread of the pollution in the sediments posed the question whether these locations might also be contaminated. Analyses were carried out to decide whether the shellfish in these regions constituted a public health hazard that would require closing of the shellfish beds to the public.

The gas chromatograms (Fig. 17A - E) and the weights of the hydrocarbon extracts (Table 3) demonstrate that all the shellfish analyzed were as severely contaminated by the fuel oil as the shellfish in the West Falmouth area. Numerous peaks in the gas chromatograms correlate with hydrocarbons present in the original

fuel oil. This and the boiling point distribution of the extracted hydrocarbons proves conclusively the presence of fuel oil hydrocarbons in these shellfish. As a result, these areas have now been closed to the taking of shellfish.

Uncontaminated Clams from Sippewissett Marsh. Scallops from Waquoit Bay, an estuary on the South Shore of Cape Cod that was not affected by the West Falmouth oil spill had been analyzed previously (Blumer et al., 1970 a,b). The hydrocarbon level in these animals was low and individual hydrocarbons could be fully accounted as derived from biological sources. We now have analyzed clams from Great Sippewissett Marsh, West Falmouth, Mass. U.S.A., an area adjacent to the two harbors that were affected by the spill. The hydrocarbon content of these animals is low (Table 3) and the gas chromatogram (Fig. 18) does not display the broad envelope from about C<sub>12</sub> to C<sub>22</sub> which is characteristic for the fuel oil contamination. The principal peaks in the chromatogram in the C<sub>20</sub> to C<sub>21</sub> region stem from saturated and olefinic hydrocarbons in organisms.

#### CONCLUSIONS

The present findings confirm and extend those reported two months after the accident in West Falmouth. Chemical analysis is well suited for the distinction between the natural hydrocarbons in the environment and the pollution derived from the fuel oil. The presence of the fuel oil in sediments and organisms can still be demonstrated conclusively eight months after the accident. Alteration

and degradation, though slow, is now recognizable; it is primarily the result of dissolution and bacterial degradation. However, the "ageing" or "weathering" of the spilled oil has not yet resulted in a detoxification. Specifically, we arrive at the following conclusions.

#### Persistence and Spread of the Pollution

Oil from the accident has been incorporated into the sediments of the tidal rivers and marshes and into the offshore sediments, down to 42 feet, the greatest water depth in the sea.

The fuel oil is still present in inshore and offshore sediments, eight months after the accident.

The pollution has been spreading on the sea bottom and now covers at least 5000 acres offshore and 500 acres of marshes and tidal rivers. This is a much larger area than that affected immediately after the accident.

Bacterial degradation of the oil is slow; degradation is still negligible in the most heavily polluted areas and the more rapid degradation in outlying, less affected areas has been reversed by the influx of less degraded oil from the more polluted regions. Thus secondary pollution from heavily affected areas continues for a long time after the accident.

The kill of bottom plants and animals has reduced the stability of marshland and sea bottom; increased erosion results and may be responsible for the spread of the pollution along the sea bottom.

Bacterial degradation first attacks the least toxic hydrocarbons. The hydrocarbons remaining in the sediments are now more toxic on an equal weight basis than immediately after the spill.

Oil has penetrated the marshes to at least 1-2 feet depth; after deposition, bacterial degradation within the marsh sediment is still negligible eight months after the accident.

#### Biological Effects of the Pollution

Where oil can be detected in the sediments there has been a kill of animals; in the most polluted areas the kill has been almost total. Control stations outside the area contain normal, healthy bottom faunas.

The kill associated with the presence of oil is detected down to the maximum water depth in the area.

The Ampeliscid amphipods are highly sensitive to small concentrations of oil and serve as an excellent biological indicator of this type of pollution.

A massive, immediate kill occurred offshore during the first few days after the accident. Affected were a wide range of fish, shellfish, worms, crabs and other crustaceans and invertebrates. Bottom living fishes and lobsters were killed and washed up on the beaches. Trawls in 10 feet of water showed 95% of the animals dead and many still dying. The bottom sediments contained many dead clams, crustaceans and snails.

Fish, crabs, shellfish and invertebrates were killed in the tidal Wild Harbor River; and in the most heavily polluted locations of the river almost no animals have survived.

The affected areas have not been repopulated, nine months after the accident.

Mussels that survived last year's spill as juveniles have developed almost no eggs and sperm.

#### Effect on Commercial Shellfish Values

Oil from the spill was incorporated into oysters, scallops, softshell clams and quahaugs. As a result, the area had to be closed to the taking of shellfish.

The 1970 crop of shellfish is as heavily contaminated as was last year's crop. Closure will have to be maintained at least through this second year and it has now been extended to areas more distant from the spill than last year.

Oysters that were removed from the polluted area and that were maintained in clean water for as long as 6 months retained the oil without change in composition or quantity. Thus, once contaminated, shellfish cannot cleanse themselves of oil pollution.

The tidal Wild Harbor River, a productive shellfish area of about 22 acres, contains an estimated 4 tons of the fuel oil. This amount has destroyed the shellfish harvest for two years. The severe biological damage to the area and the slow rate of biodegradation of the oil suggest that the productivity will be ruined for a longer time.

The presence or absence of an "oily smell" is no clue for the presence of oil pollution in fish or shellfish. Only a small fraction of petroleum has a pronounced odor; this may be lost while

the more harmful long-term poisons are retained. Boiling or frying may remove the odor but will not eliminate the toxicity.

Shellfish areas in North Falmouth and Bourne which were not closed last Fall had to be closed now on the basis of analysis that showed the presence of hydrocarbons derived from the West Falmouth oil spill. In view of the persistence of the oil in shellfish, closure may have to be maintained for a prolonged time.

#### ACKNOWLEDGMENTS

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Table 1

Hydrocarbons in Offshore Sediments

Month 1969-1970	Sediment Station, Hydrocarbons in mg/100 g dry Sediment				
	7	20	31	35	37 West Falmouth Hbr.
September	6.9	3.9	55		
October	19	4.4	110		
November	14	2.4	110		4.0
December	12	4.5	113	6.0 <sup>1)</sup>	
January					4.4
February					3.8
March			1240		
April	4.8	6.2 <sup>1)</sup>	450		5.9
May		3.5 <sup>1)</sup>	214		7.0
June					1.8

1) single analysis, otherwise average of two-four samples.

Table 2

Hydrocarbons in Inshore Sediments

Month 1969-1970	Wild Harbor River, Station #, Hydrocarbons in mg/100 g dry sediment		
	II	IV	V
December	76	145 <sup>1)</sup>	175
February	51	117	15
April		140	187
May	52 <sup>1)</sup>	40 <sup>1)</sup>	77 <sup>1)</sup>

1) single analysis, otherwise averaged value

Hydrocarbons in Cores, Wild Harbor River

Core #	Sediment Type	February 24, 1970	
		Depth, cm	Hydrocarbons, mg/100 g dry sediment
1	coarse sand	43-51	3.2
2	sand	Top 10	4.2
2	coarse sand	51-58	18
4	clay	Top 10	70
4	clay	36-43	5.5
5 vertical	clay	Top 4	280
5 vertical	clay	43-51	9.3
5 horizontal	clay	43-51	93 exposed face of river bank
5 horizontal	clay	43-51	19 23-30 cm horizontal penetration

Table 3

Hydrocarbons in Shellfish - 1970

Date	Location	Specimen	Number of Animals	Net Weight Grams	Hydrocarbons mg/100 g wet wt.
April 17	West Falmouth Hbr. Station 1 <sup>1)</sup>	Scallops	30	22.5	0.40
April 17	West Falmouth Hbr. Station 2 <sup>1)</sup>	Scallops	26	39.0	1.2
April 17	West Falmouth Hbr. Station 3 <sup>1)</sup>	Scallops	27	48.0	1.1
April 17	West Falmouth Hbr. Station 4 <sup>1)</sup>	Scallops	33	68.0	0.75
April 17	West Falmouth Hbr. Station 2 <sup>1)</sup>	Quahaugs	8	64.5	0.83
May 13	Wild Harbor River <sup>2)</sup>	Oysters	1	17	12.6
May 13	Wild Harbor River <sup>3)</sup>	Oysters	2	33	3.8
June 8	Megansett Harbor	Scallops	10	36	1.1
June 8	Squeteague Harbor	Scallops	10	31	0.8
June 8	Rands Canal	Soft Shell Clams	4	64	2.1
June 8	Fiddlers Cove	Soft Shell Clams	2	57	3.6
June 8	Squeteague Harbor	Quahaugs	3	45	1.2
<u>Controls</u>					
May 13	Sippewissett Marsh	Soft Shell Clams	4	81.5	0.17

1) Same stations as in Blumer et al. 1970 a,b.

2) Removed from Wild Harbor River on November 12, 1969, after 57 days in polluted environment. Maintained in clean water until January 23, 1970, (72 days).

3) Removed from Wild Harbor River on November 12, 1969. Maintained in clean water until May 11, 1970, (180 days).

LEGEND

- Fig. 1 Chart of the affected area off West Falmouth. X marks site of accident. Numbers correspond to offshore sediment samples.
- Fig. 2 Sediment stations in Wild Harbor River. Location of shellfish samples. WF 1 - WF 4: West Falmouth Harbor stations 1-4; WH: Wild Harbor River; Mg: Megansett Harbor; Sq: Squeteague Harbor; R: Rands Canal; FC: Fiddlers Cove; Si: Sippewissett Marsh.
- Fig. 3 A-G Offshore Station 31, September - December 1969, March - May 1970.
- Fig. 4 A-E Offshore Station 7, September - December 1969, April 1970.
- Fig. 5 A-F Offshore Station 20, September - December 1969, April - May 1970.
- Fig. 6 A-F Offshore Station 37, November 1969, January - May 1970.
- Fig. 7 Offshore Station 35, December 1969.
- Fig. 8 A-C Wild Harbor River Station II, December 1969, February and May 1970.
- Fig. 9 A-D Wild Harbor River Station IV, December 1969, February, April and May 1970.
- Fig. 10 A-D Wild Harbor River Station V, December 1969, February, April and May 1970.
- Fig. 11 Wild Harbor River, Core #1
- Fig. 12 A-B Wild Harbor River, Core #2, top and bottom
- Fig. 13 A-B Wild Harbor River, Core #4, top and bottom
- Fig. 14 A-B Wild Harbor River, Core #5, vertical, top and bottom  
C-D Wild Harbor River, Core #5, horizontal, face of riverbank and innermost section.
- Fig. 15 A-B Polluted Oysters, maintained in clean water for 72 and 180 days.
- Fig. 16 A Scallops, West Falmouth Harbor, Station 2
- Fig. 16 B Same as Fig. 16 A, internal standard added, n-C<sub>14</sub>, n-C<sub>16</sub>, n-C<sub>18</sub> and n-C<sub>20</sub>.
- Fig. 16 C-D Scallops, West Falmouth Harbor, Station 3 and 4.

- Fig.16 E      Quahaugs, West Falmouth Harbor, Station 2.
- Fig.16 F-G    Sediments, West Falmouth Harbor, taken just N and W of  
scallop station 1
- Fig.17 A-E    Shellfish, June 8, as follows: Scallops, Megansett;  
Scallops, Squeteague; Soft shell clams, Rands Canal;  
Soft shell clams, Fiddlers Cove; Quahaugs, Squeteague.
- Fig.18        Controls, Soft shell clams, Sippewisset Marsh, May 13.

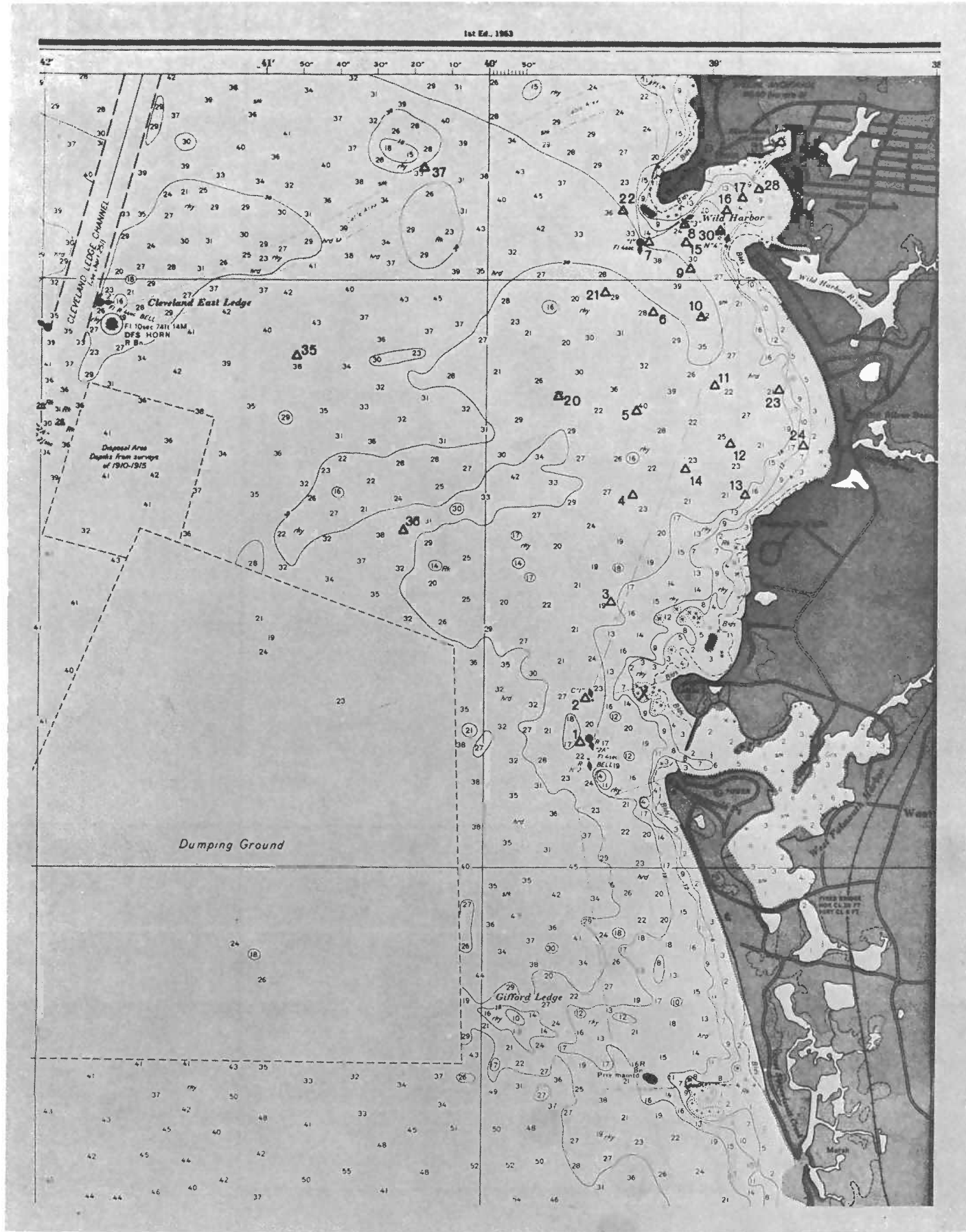


Figure 1. Chart of the affected area off West Falmouth. X marks site of accident. Numbers correspond to offshore sediment samples.

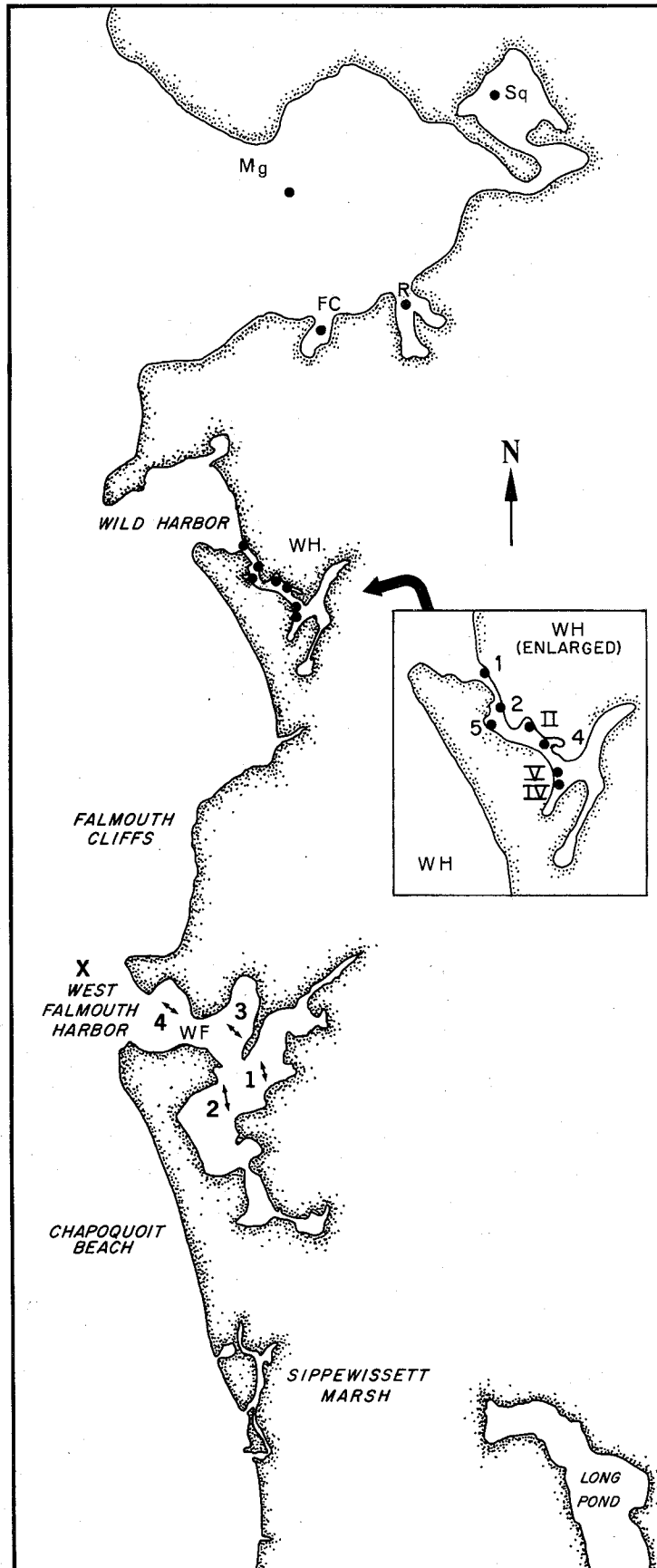
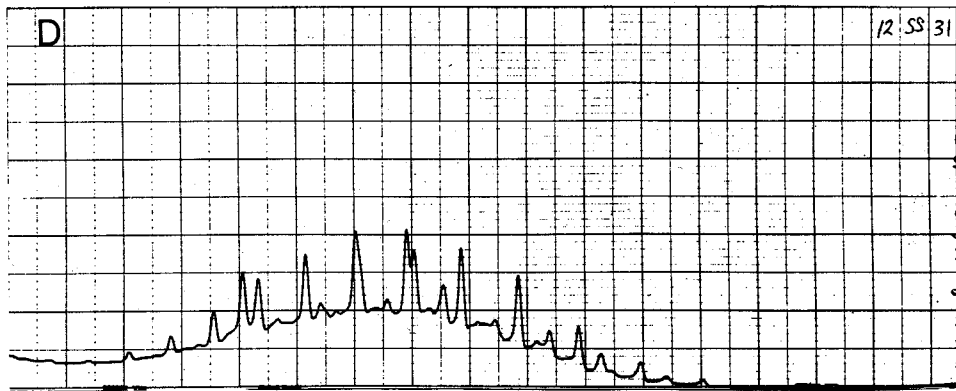
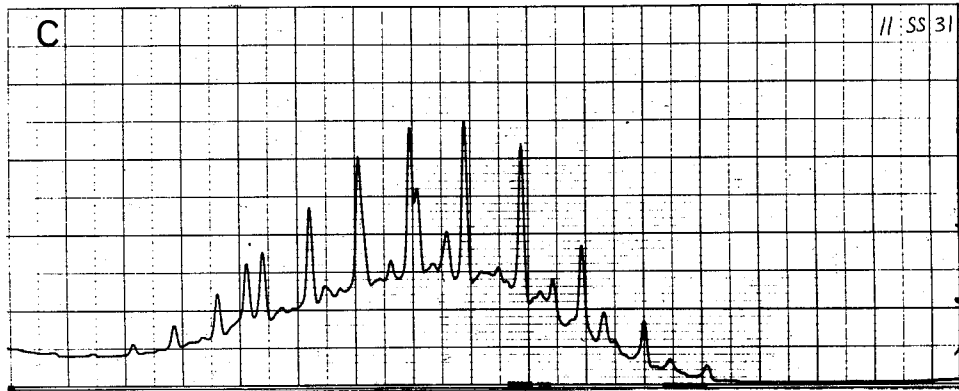
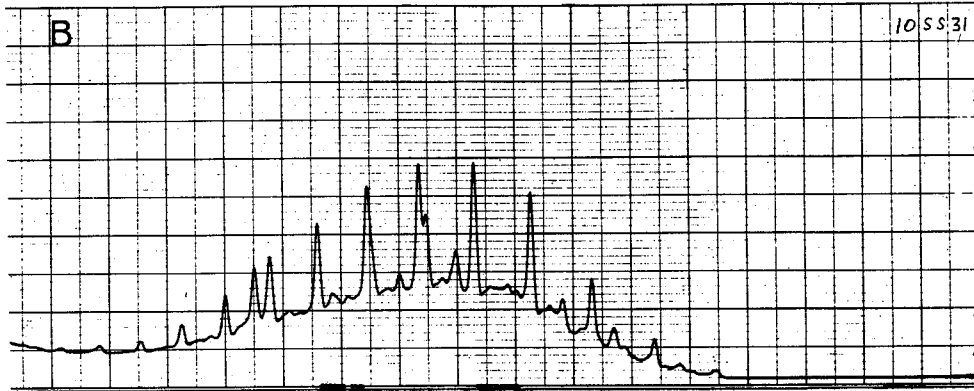
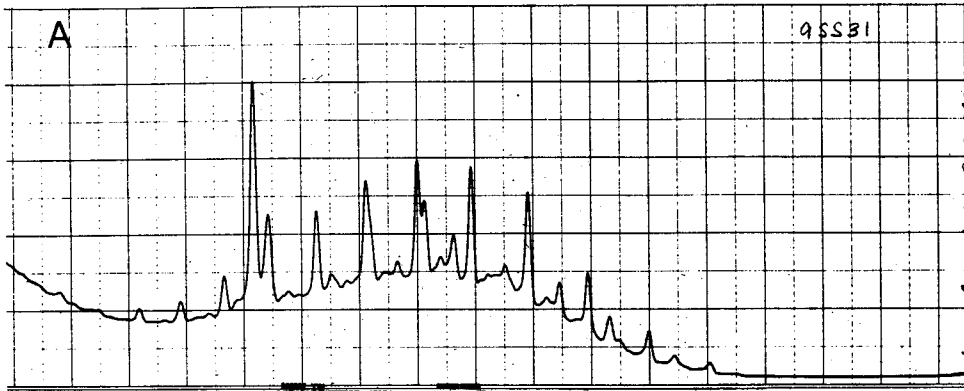


Figure 2. Sediment stations in Wild Harbor River. Location of shellfish samples. WF 1 - WF 4: West Falmouth Harbor stations 1-4; WH: Wild Harbor River; Mg: Megansett Harbor; Sq: Squeteague Harbor; R: Rands Canal; FC: Fiddlers Cove; Si: Sippewissett Marsh.





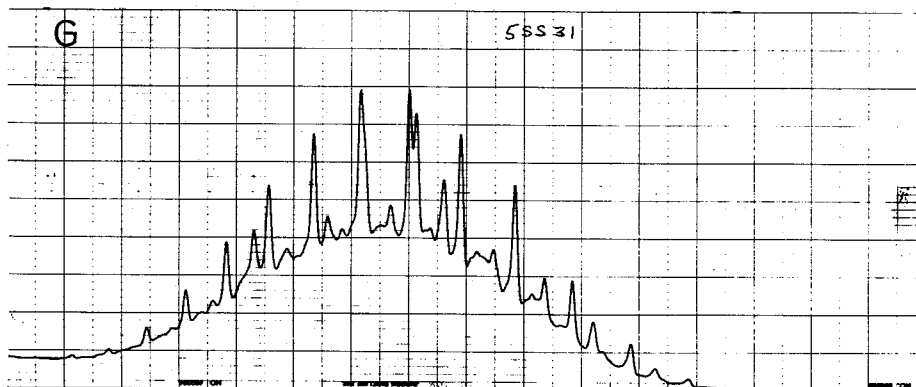
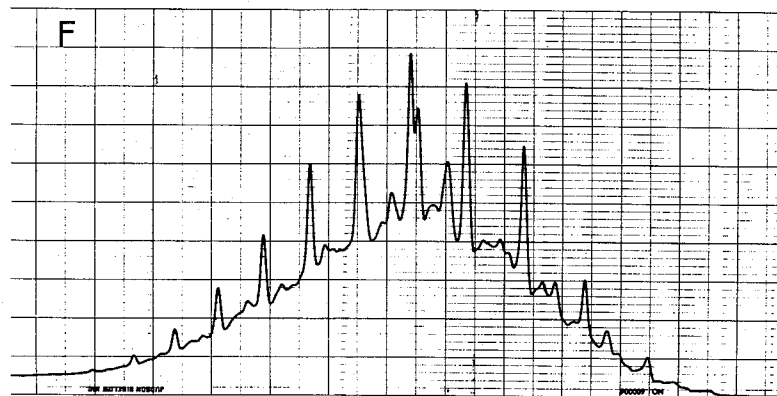
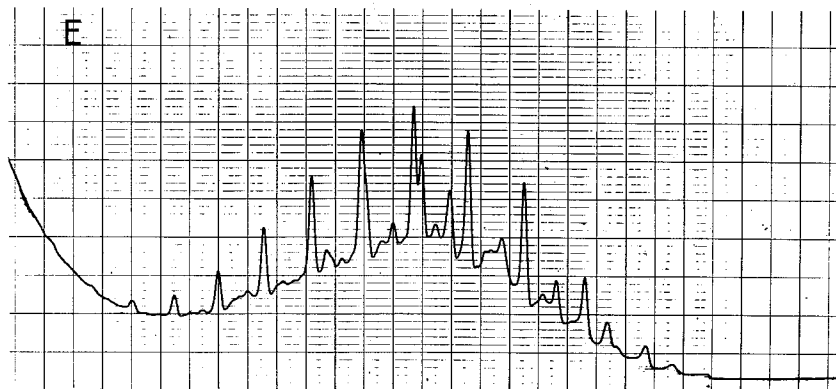
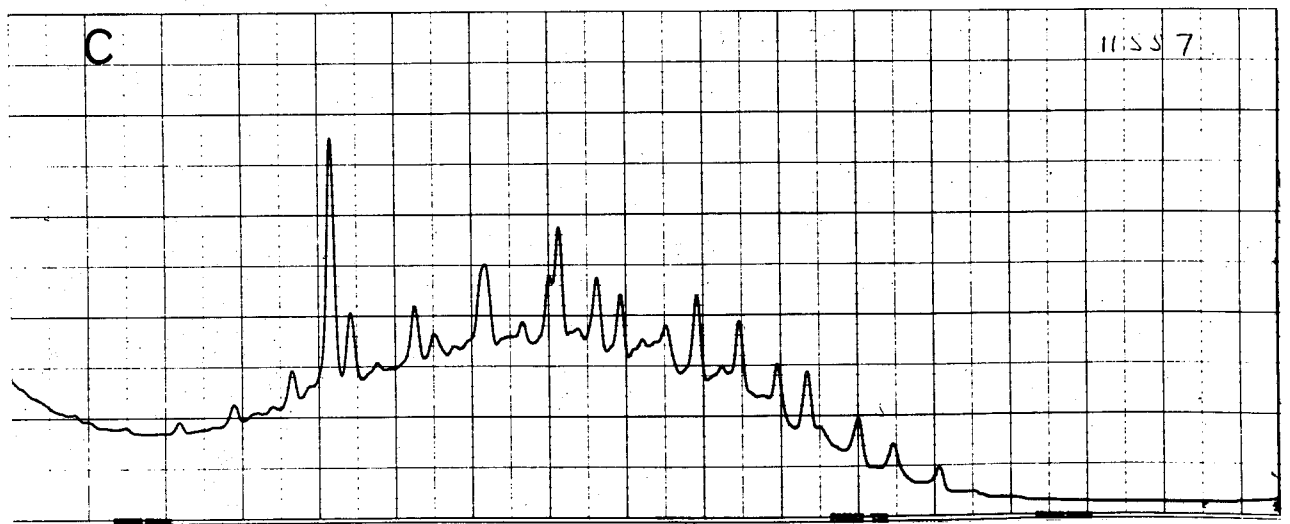
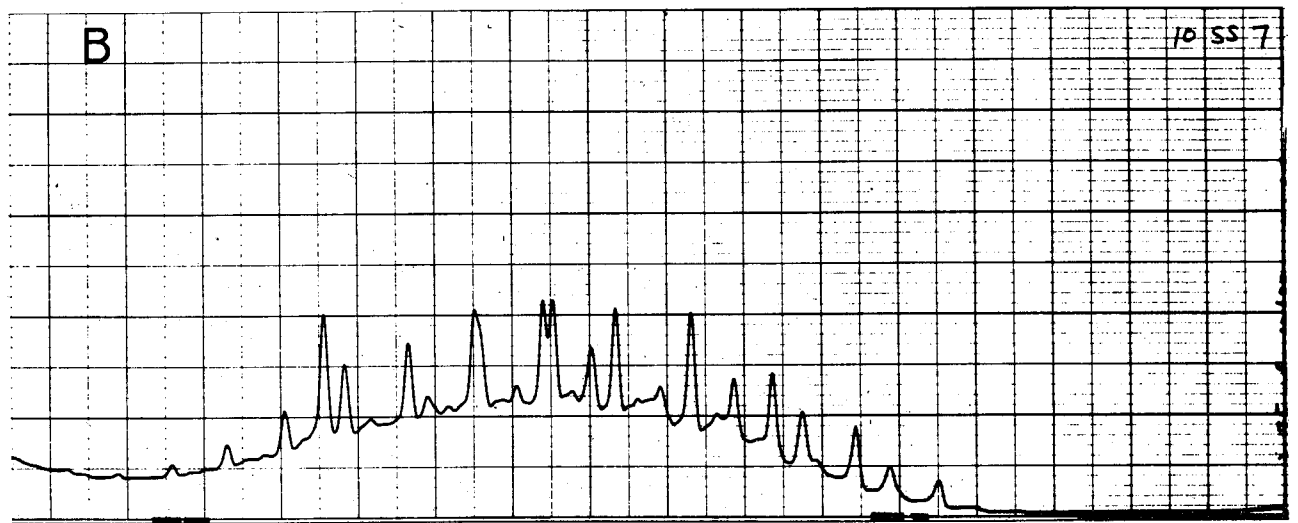
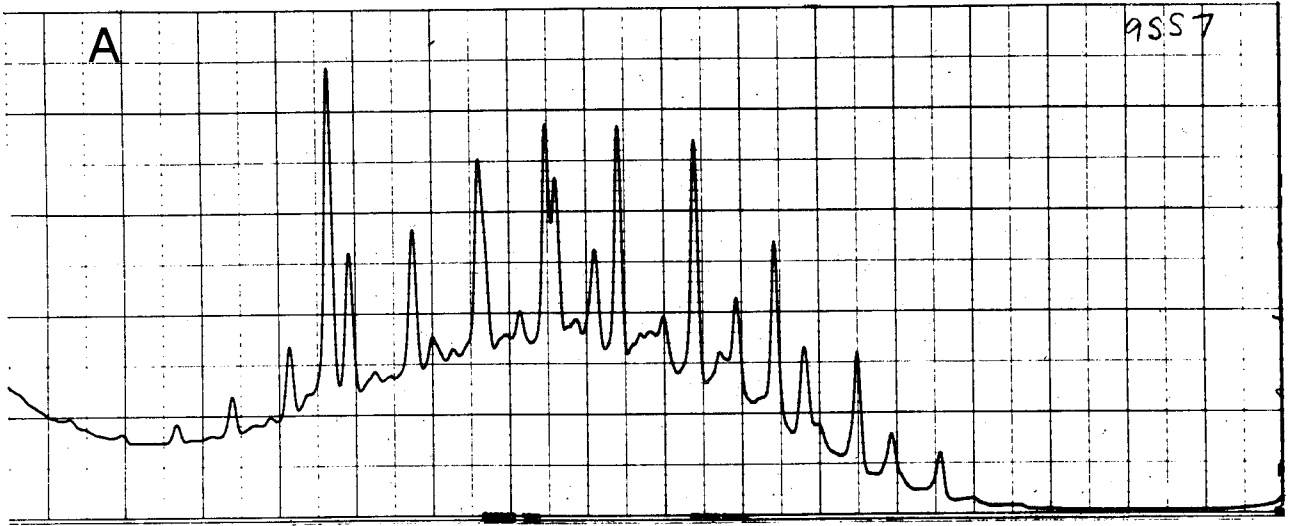


Figure 3 A-G. Offshore Station 31, September - December 1969, March-May 1970.



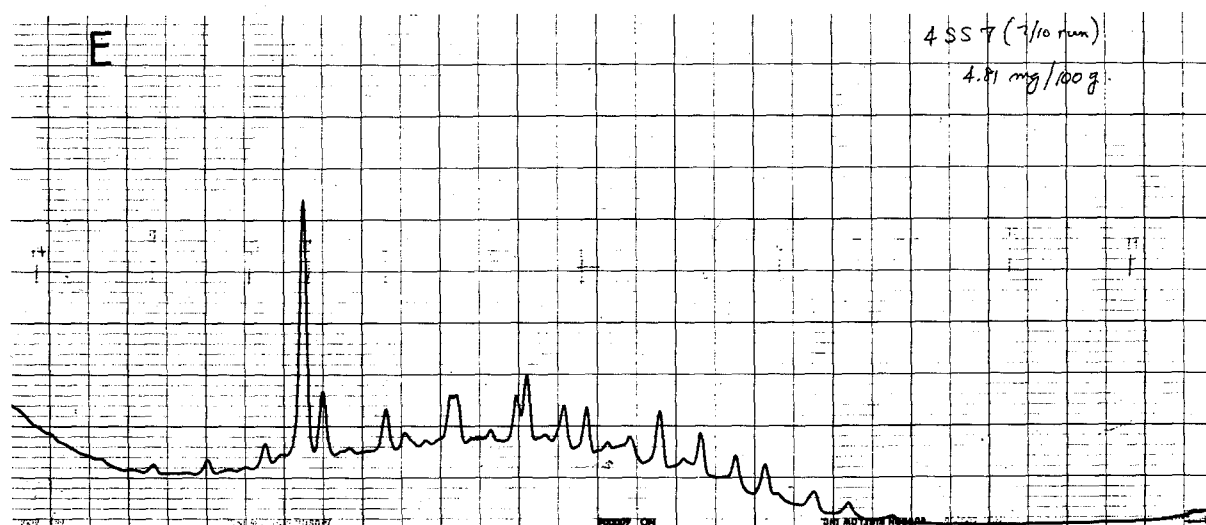
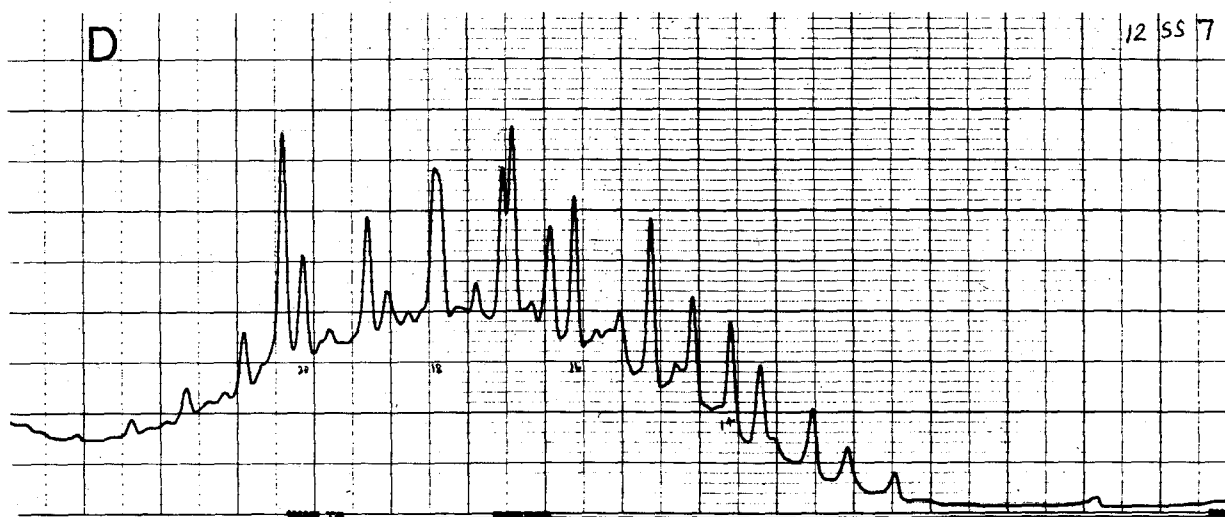
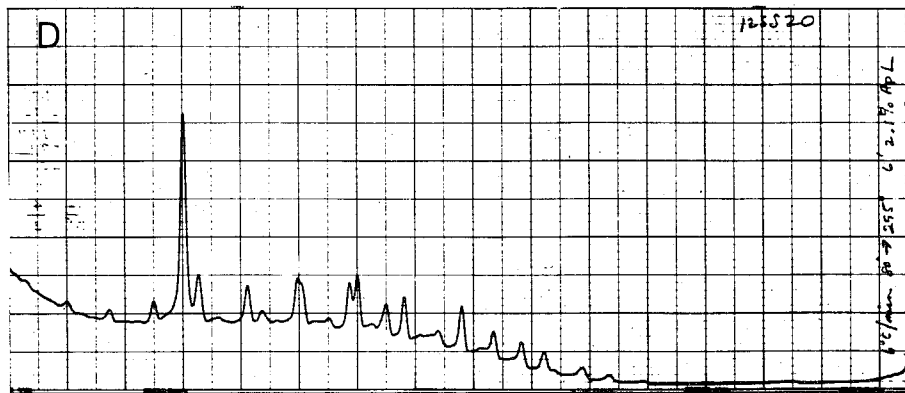
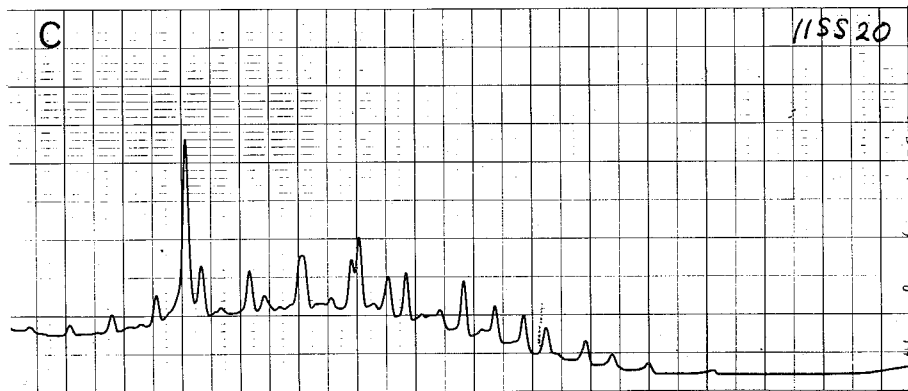
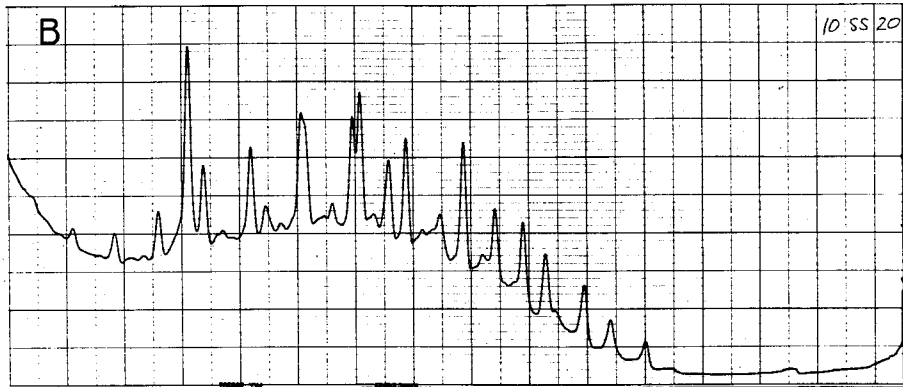
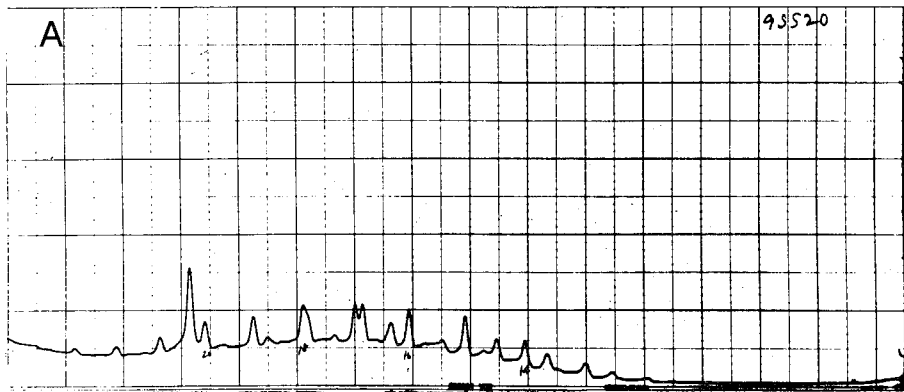


Figure 4 A-E. Offshore Station 7, September - December 1969, April 1970.



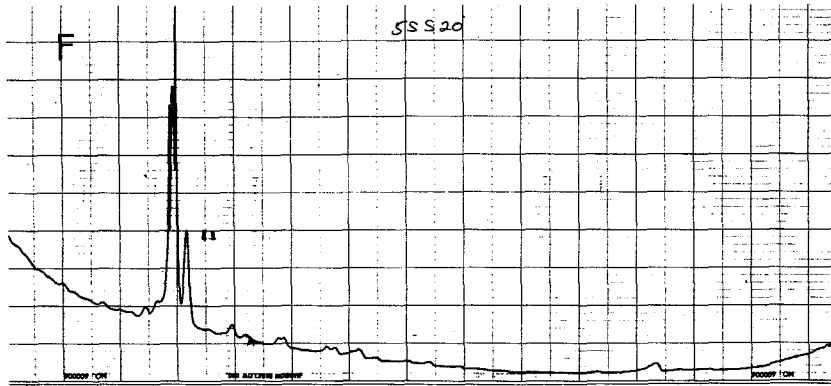
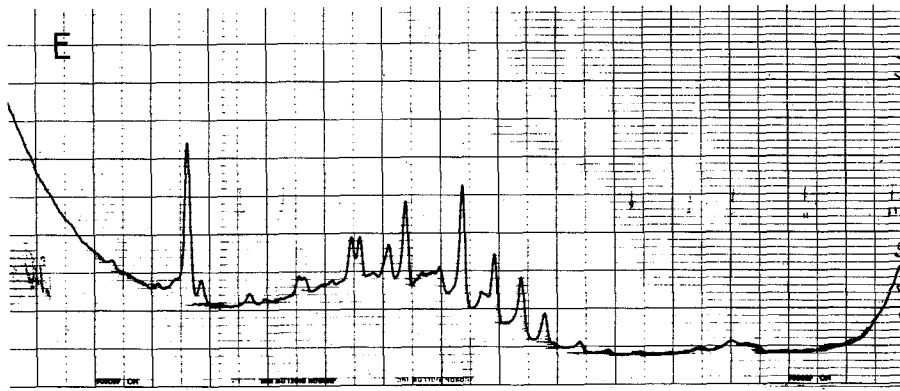
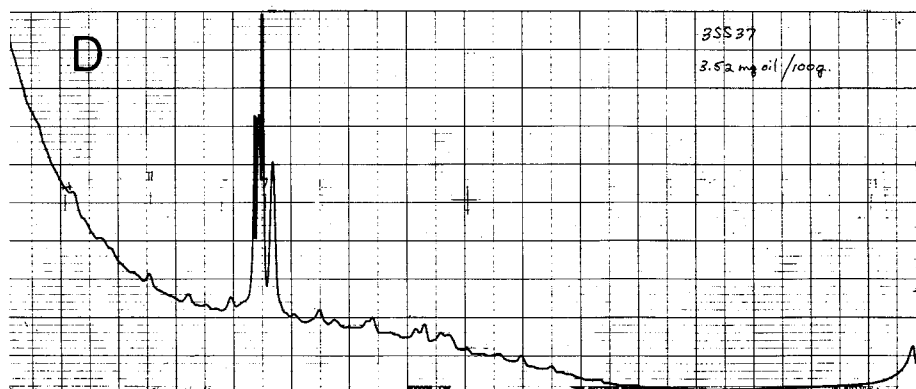
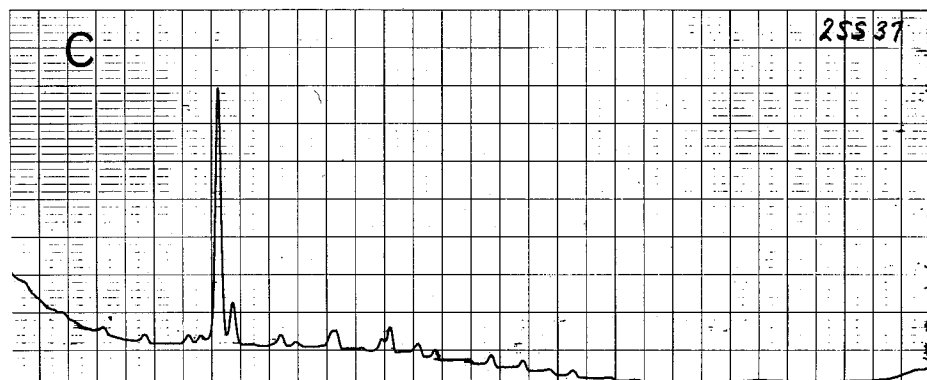
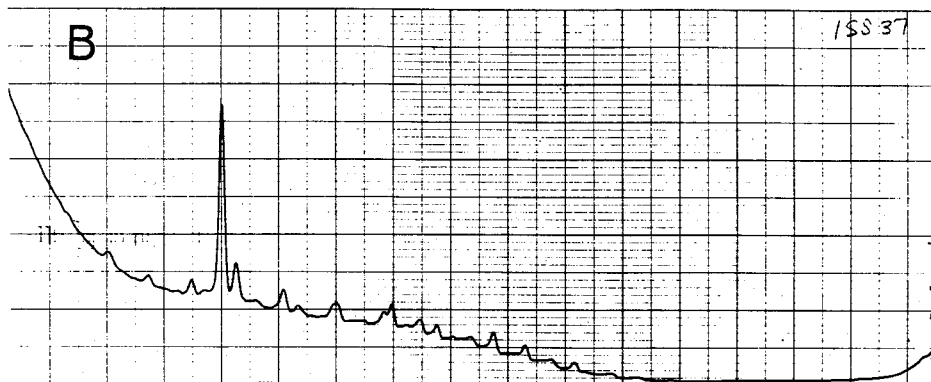
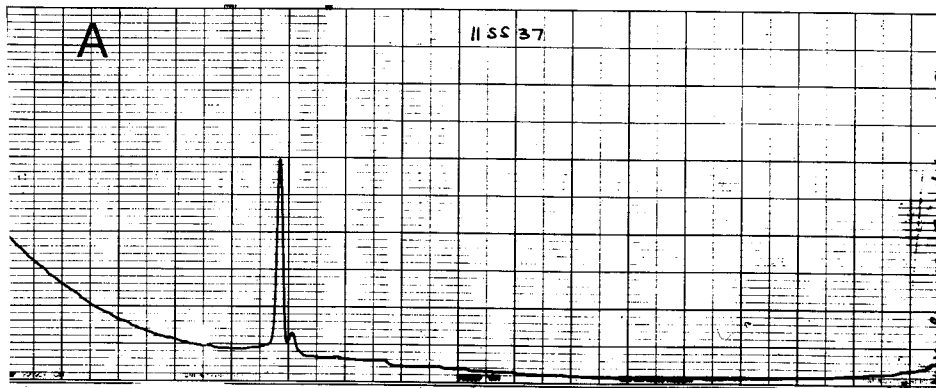


Figure 5 A-F. Offshore Station 20, September - December 1969, April - May 1970.



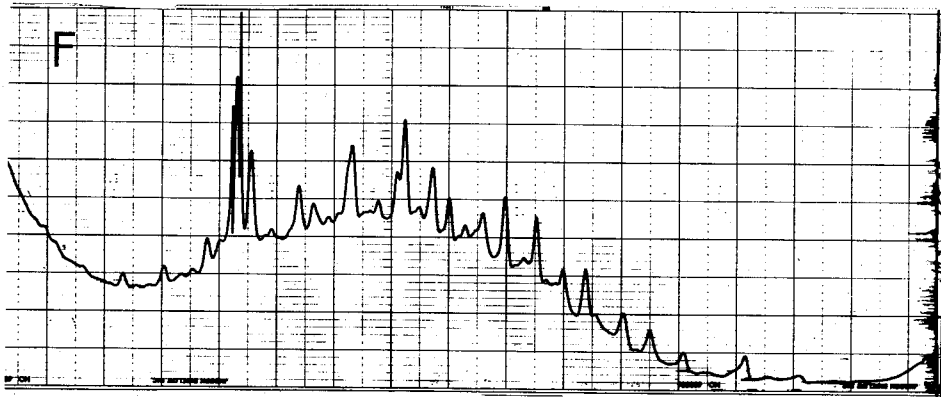
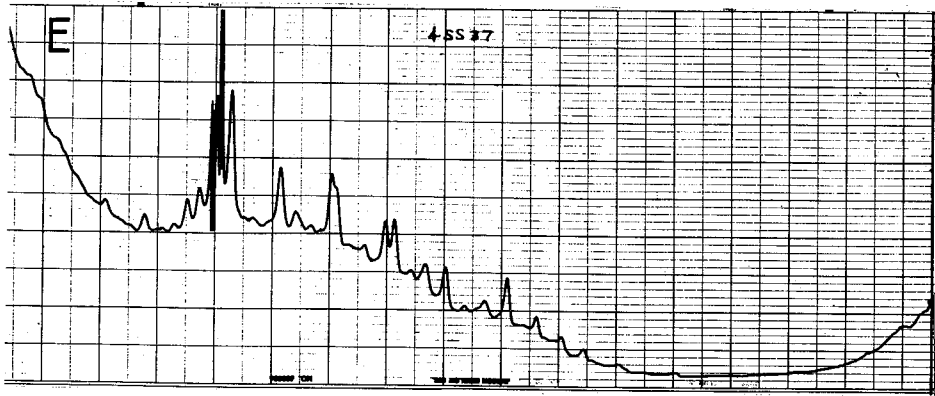


Figure 6 A-F. Offshore Station 37, November 1969, January - May 1970.

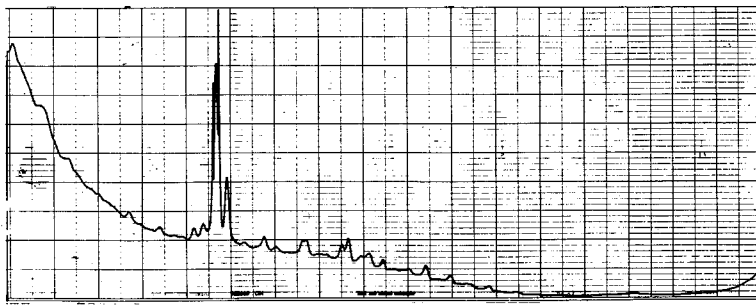


Figure 7. Offshore Station 35, December 1969.

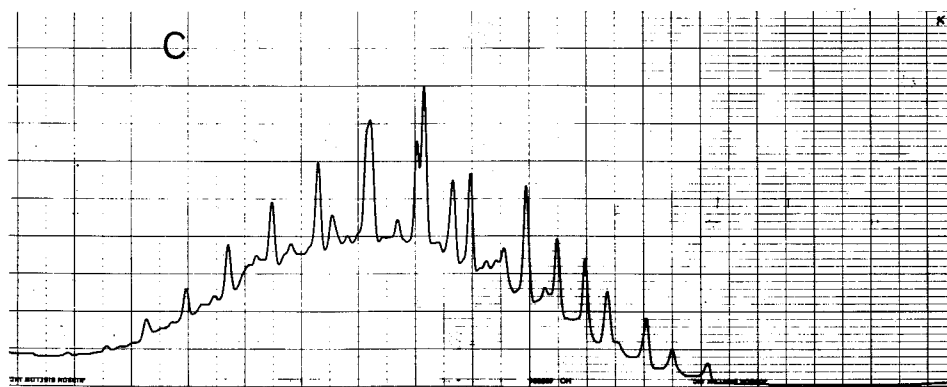
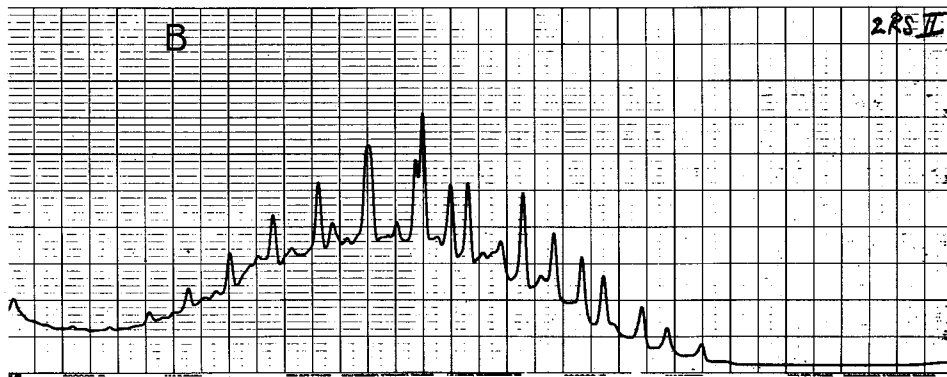
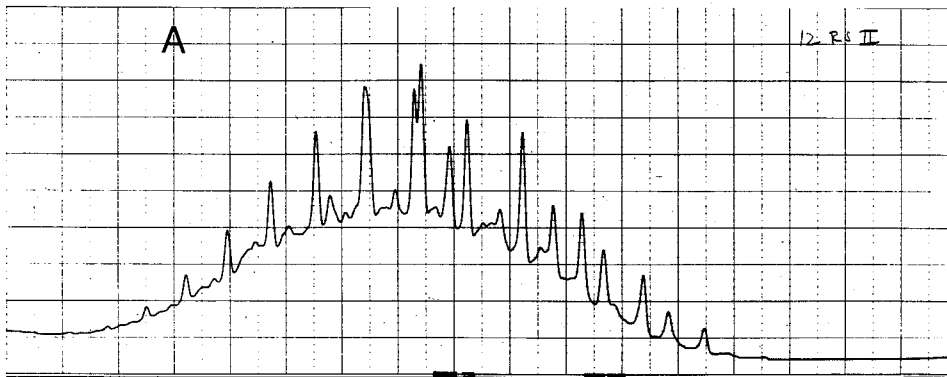


Figure 8 A-C. Wild Harbor River Station II, December 1969, February and May 1970.



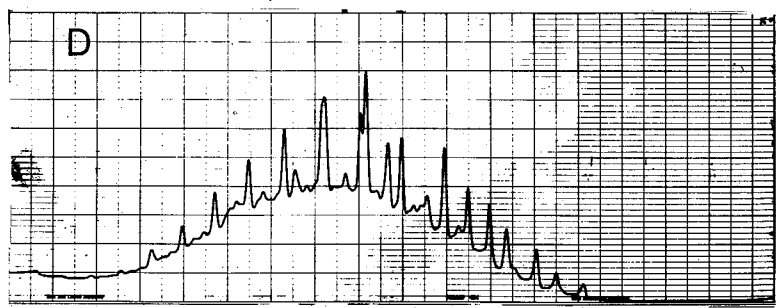
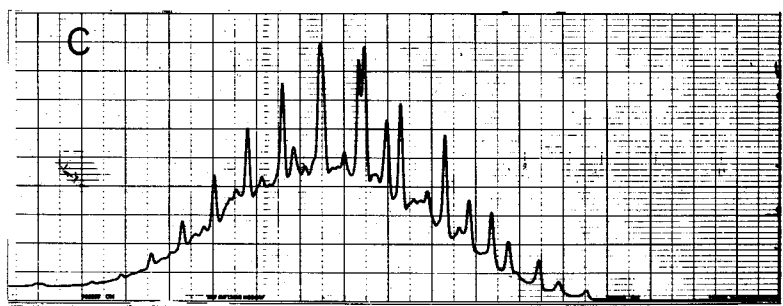
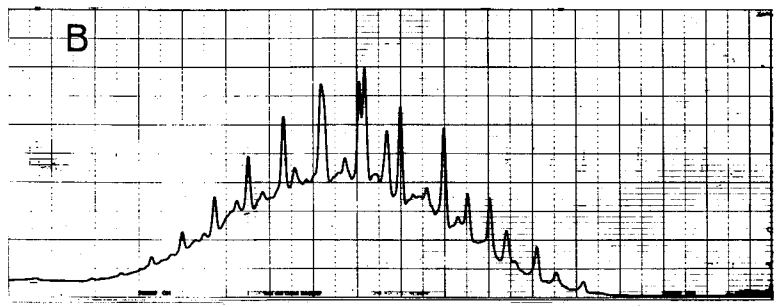
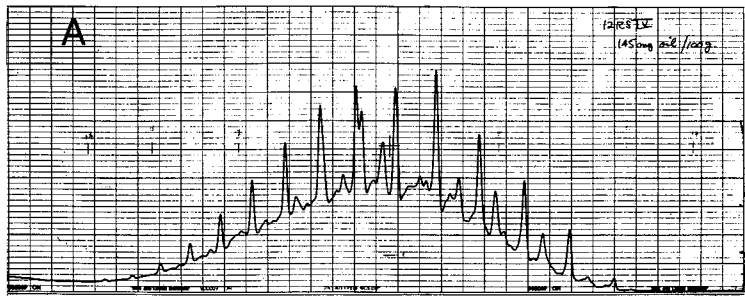


Figure 9 A-D. Wild Harbor River Station IV, December 1969, February, April and May 1970.

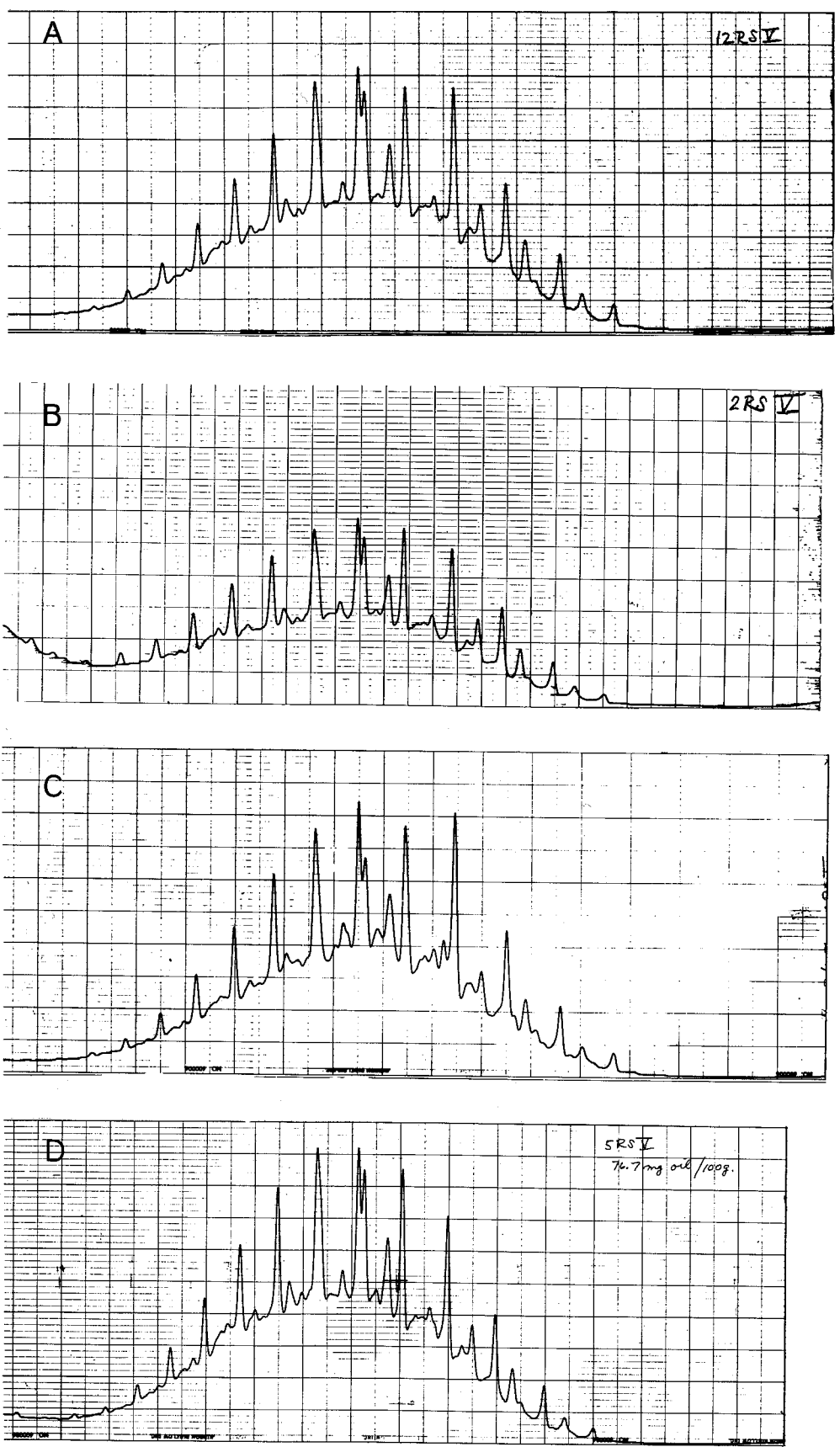


Figure 10 A-D. Wild Harbor River Station V, December 1969, February, April and May 1970.

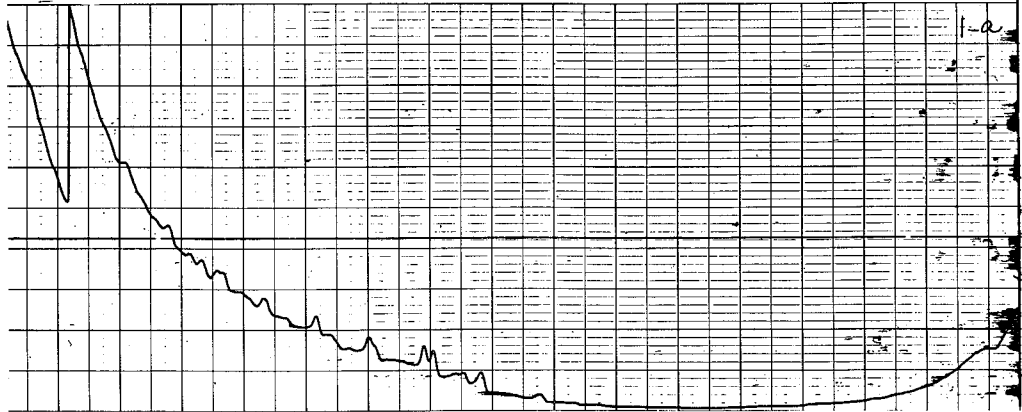


Figure 11. Wild Harbor River, Core #1.

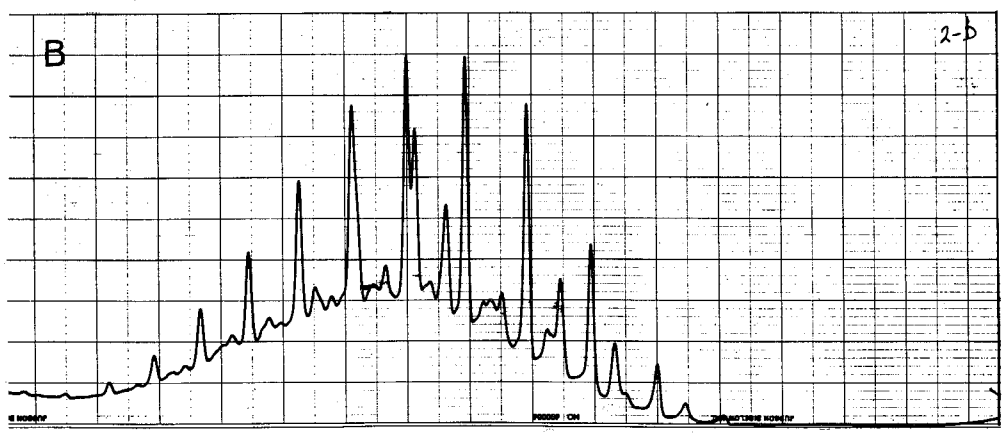
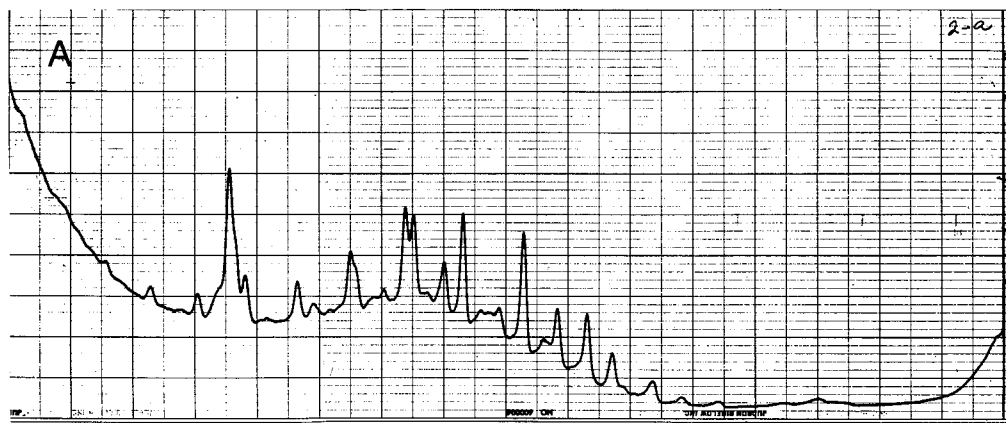


Figure 12 A-B. Wild Harbor River, Core #2, top and bottom.

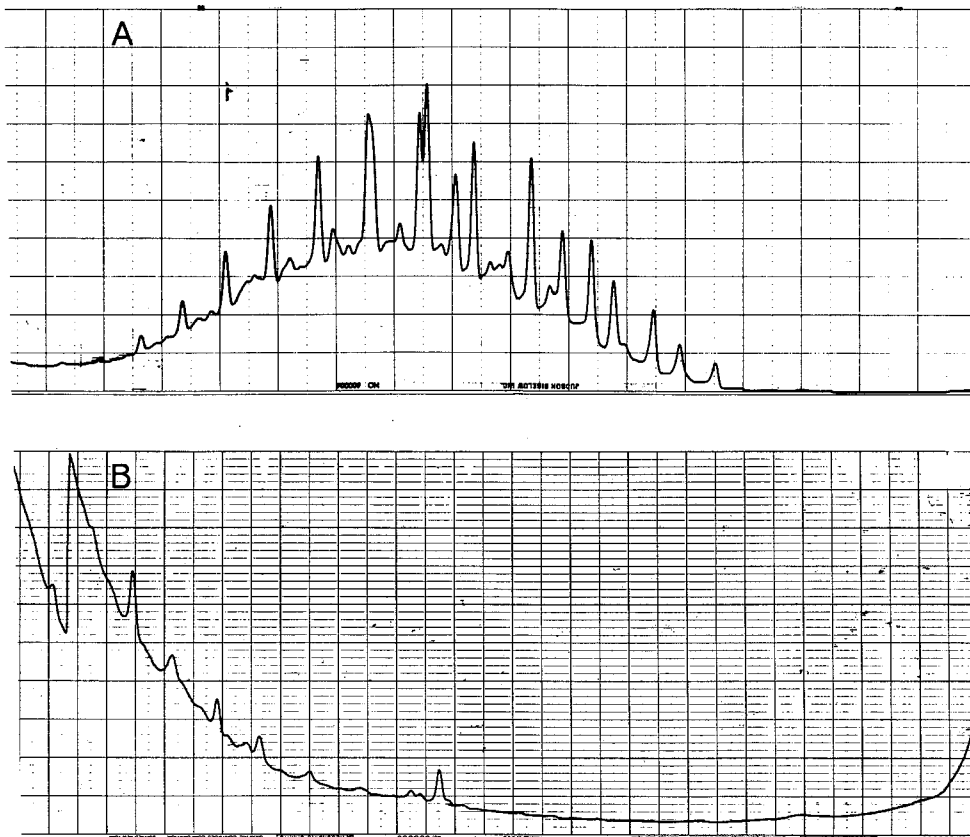


Figure 13 A-B. Wild Harbor River, Core #4, top and bottom.

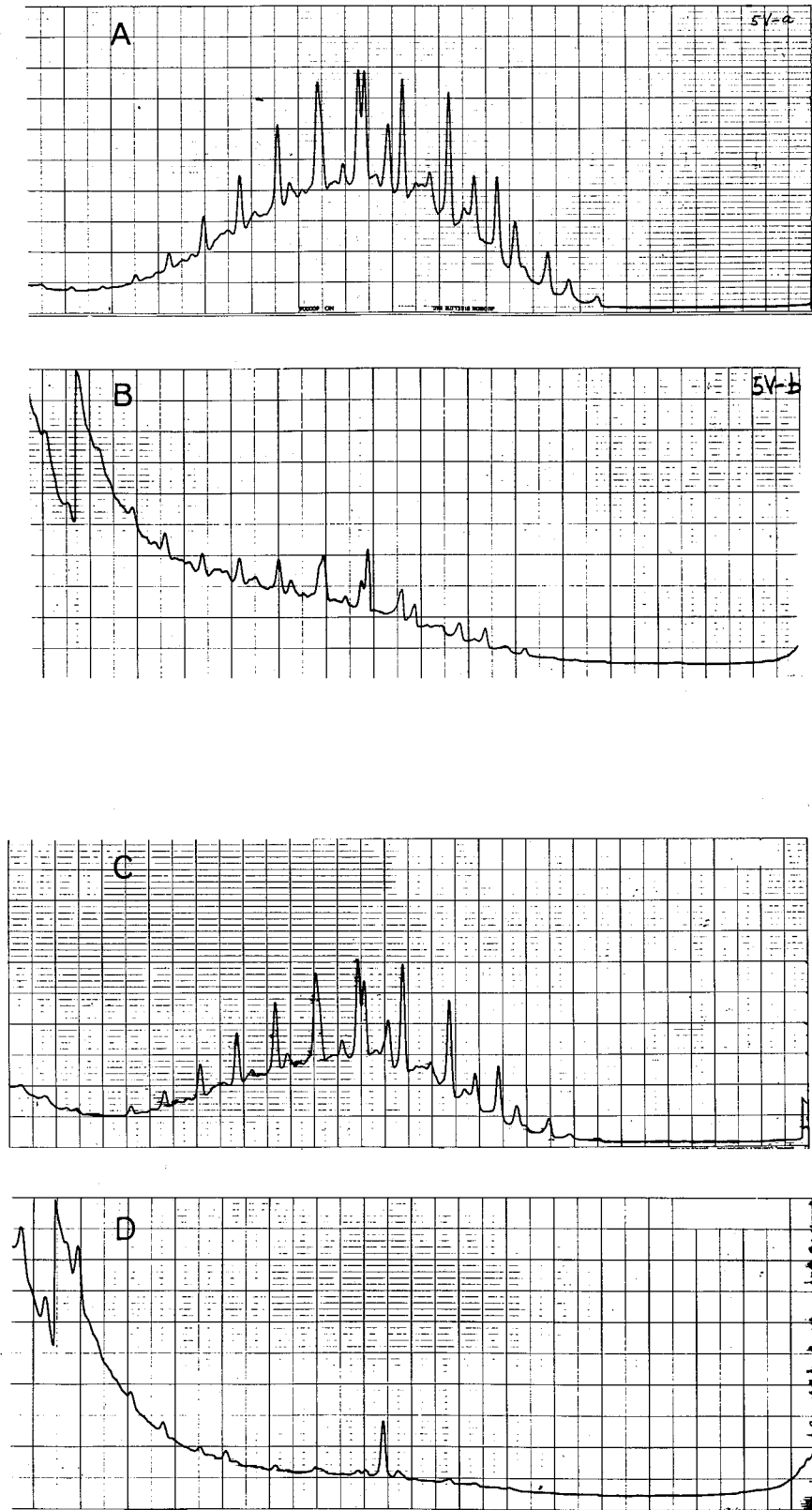


Figure 14 A-B. Wild Harbor River, Core #5, vertical, top and bottom.  
C-D. Wild Harbor River, Core #5, horizontal, face of riverbank and innermost section.

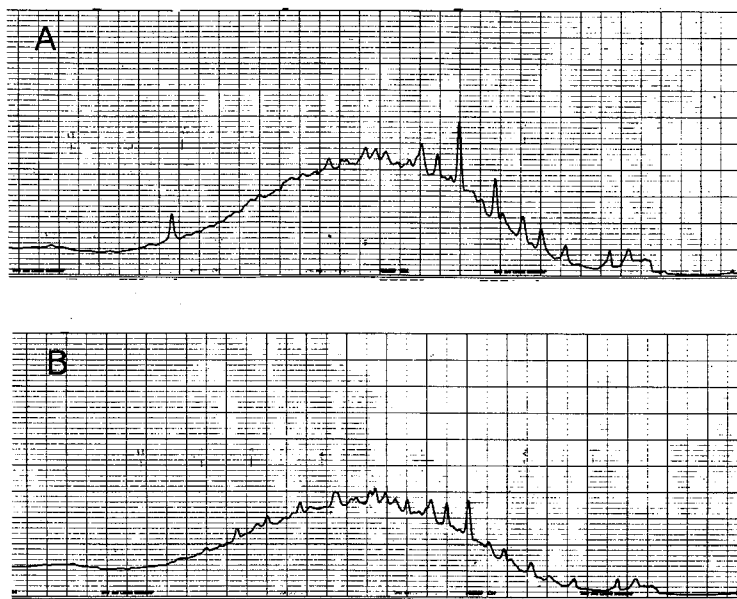
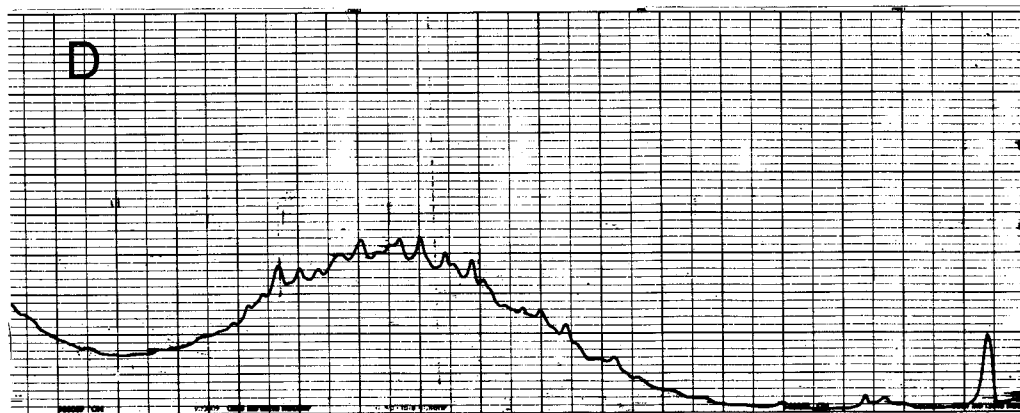
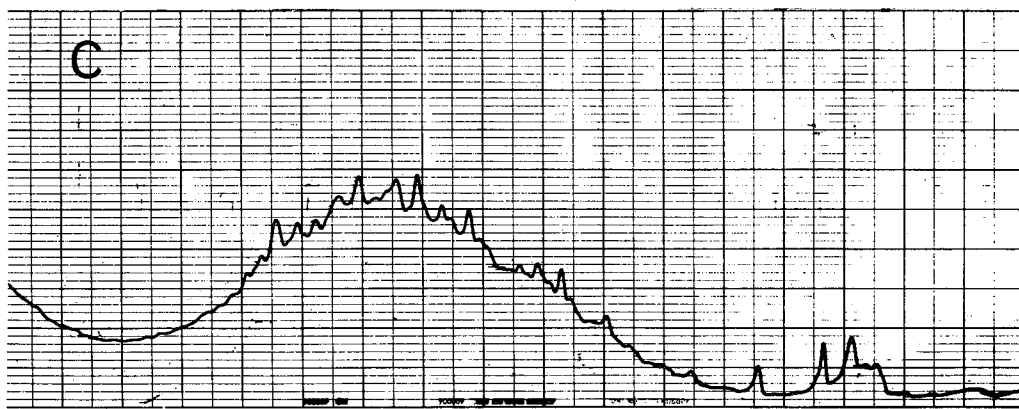
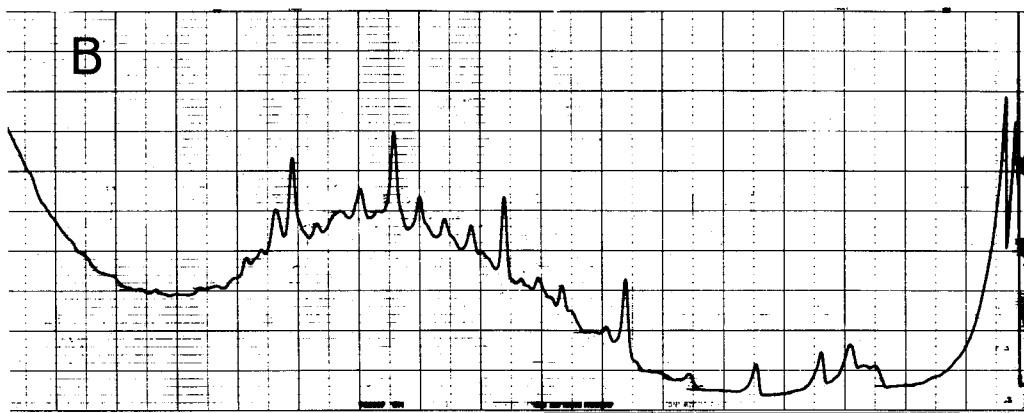
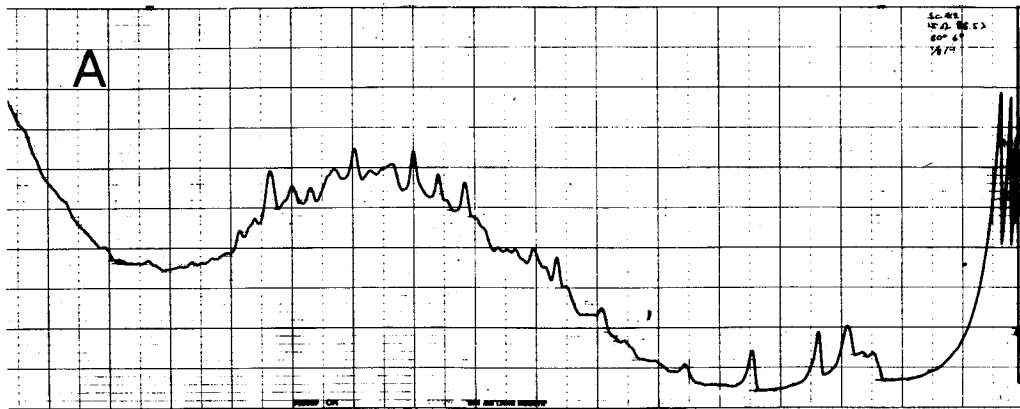


Figure 15 A-B. Polluted Oysters, maintained in clean water for 72 and 180 days.



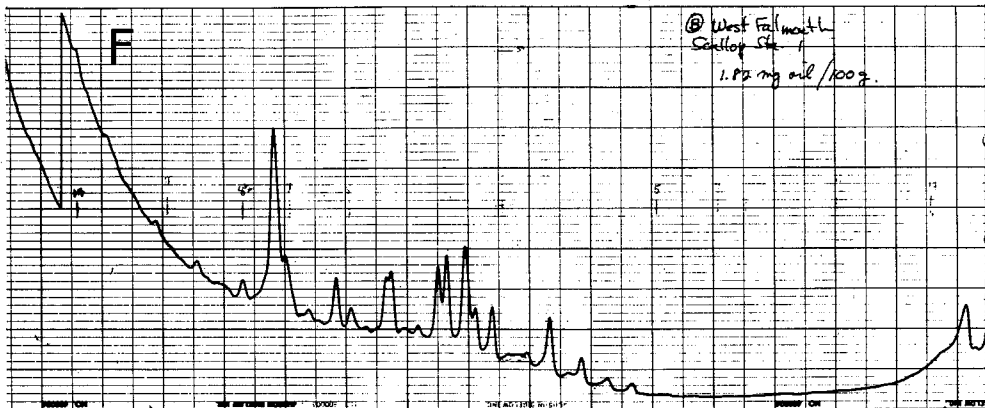
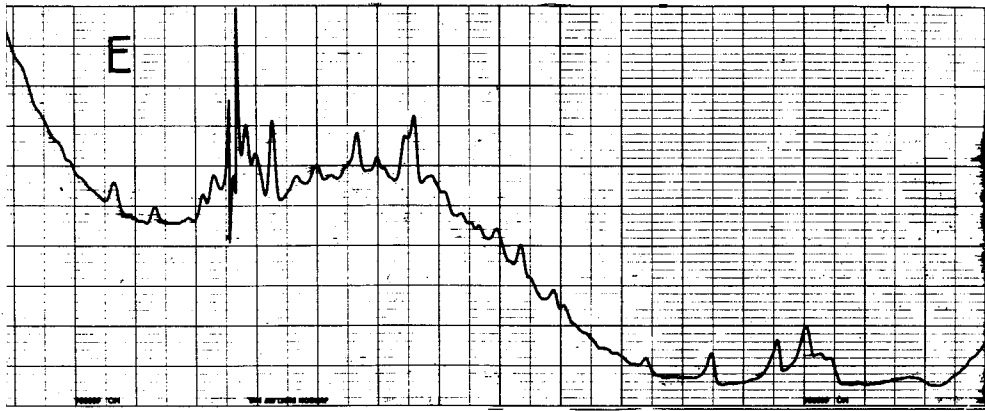


Figure 16 A. Scallops, West Falmouth Harbor, Station 2.

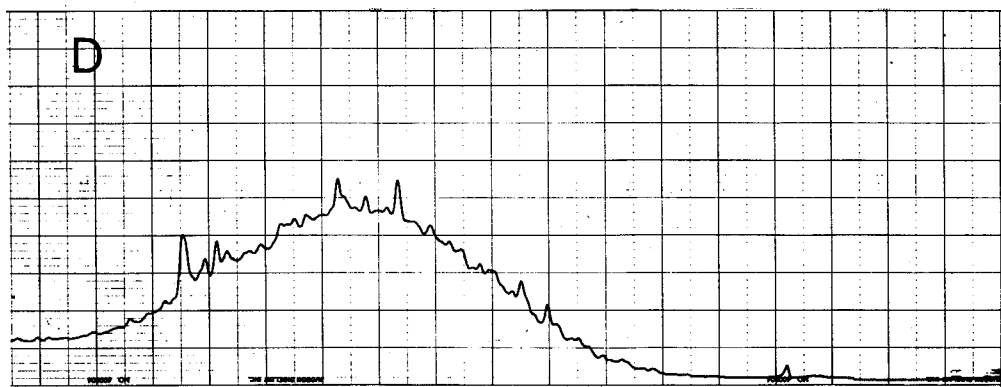
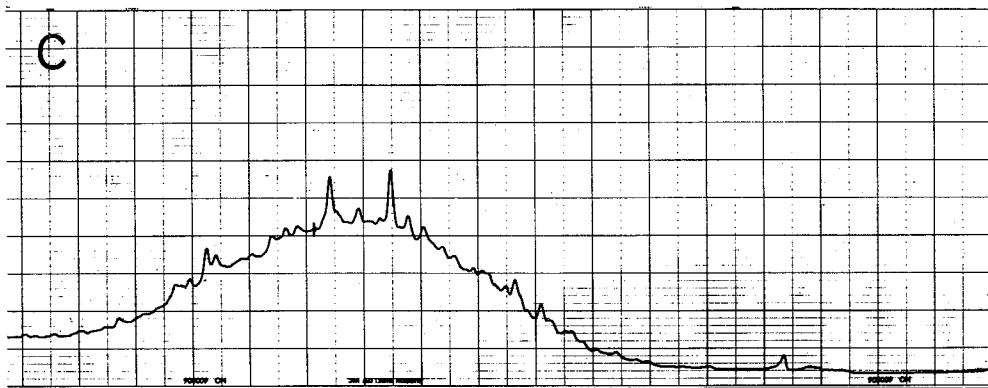
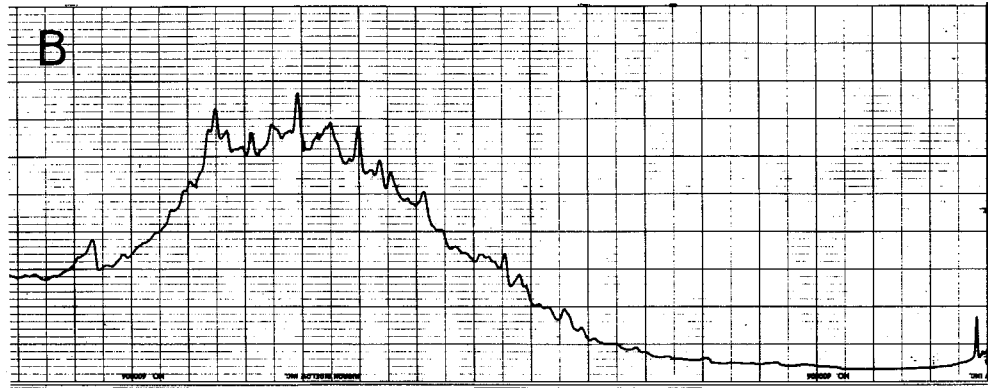
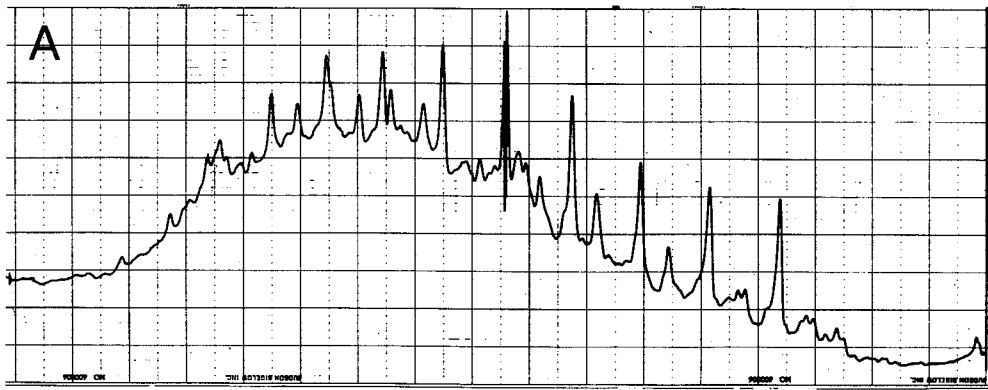
B. Same as Figure 16 A, internal standard added, n-C<sub>14</sub>, n-C<sub>16</sub>, n-C<sub>18</sub> and n-C<sub>20</sub>.

C-D. Scallops, West Falmouth Harbor, Station 3 and 4.

E. Quahaugs, West Falmouth Harbor, Station 2.

F-G. Sediments, West Falmouth Harbor, taken just N and W of scallop station 1.





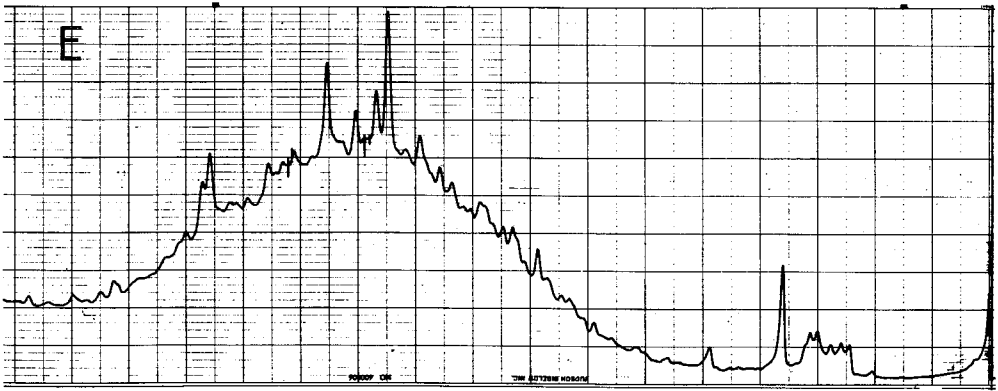


Figure 17 A-E. Shellfish, June 8, as follows: Scallops, Megansett; Scallops, Squeteague; Soft shell clams, Rands Canal; Soft shell clams, Fiddlers Cove; Quahaugs, Squeteague.

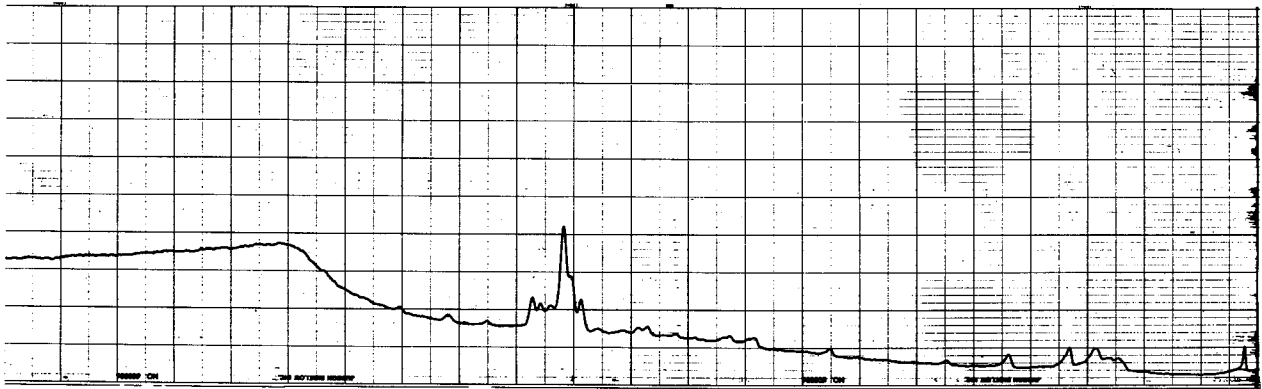


Figure 18. Controls, Soft shell clams, Sippewisset Marsh, May 13.

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The effects of environmental exposure on the composition of the oil are discussed; many analytical parameters are sufficiently stable to permit continued correlation of the oil remaining in sediments and organisms with the fuel oil involved in the spill.

Oil from the spill is still present in the sediments, inshore and offshore and in the shellfish. A further spread of the pollution to more distant offshore regions has occurred during mid-winter; as a result, the pollution now covers a much larger area than immediately after the accident. The first stages of biological (presumably bacterial) degradation of the oil are now evident especially in the least polluted regions; however, it has depleted predominantly by the straight and branched chain alkanes. The more toxic aromatic hydrocarbons are resistant; as a result, the toxicity of the oil has not been diminished.

Where oil can be detected in the sediments there has been a kill of animals; in the most polluted areas the kill has been almost total. Shellfish that survived the accident have taken up the fuel oil. The 1970 crop of shellfish is as heavily polluted as was last year's crop. Oysters transplanted to unpolluted water for as long as 6 months retained the oil without change in composition or concentration.

1. Oil Pollution

2. Shellfish
3. Sediments

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II. Sass, J

III. Souza, G

IV. Sanders, H.

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