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Bioremediation Treatability Trials Using Nutrient Application to Enhance Cleanup of Oil-Contaminated Shoreline

Albert D. Venosa, John R. Haines, John A. Glaser, Edward J. Opatken
U.S. Environmental Protection Agency
Risk Reduction Engineering Laboratory
Cincinnati, OH 45268

Parmely H. Pritchard
U.S. Environmental Protection Agency
Environmental Research Laboratory
Gulf Breeze, FL 32561

Charles Costa
U.S. Environmental Protection Agency
Office of Radiation Programs
Las Vegas, NV 89114

INTRODUCTION

On March 24, 1989, the supertanker Exxon Valdez went aground in Prince William Sound, Alaska, releasing approximately 11 million gallons of Prudhoe Bay crude oil. The spilled oil spread over an estimated 350 miles of shoreline. The oil settled into the beach gravel and on rock surfaces and the faces of vertical cliffs. Contamination occurred primarily in the intertidal zone. Initial weathering of the oil resulted in a loss of approximately 15 to 20% of the oil by volatilization. Components lost through volatilization included normal aliphatic hydrocarbons of 12 carbon atoms and less and low molecular weight aromatic hydrocarbons (benzene, toluene, xylene, and some naphthalenes). The residual oil consisted of approximately 40 to 50% high molecular weight waxes and asphaltenes.

Biodegradation of oil has been extensively studied over the last 20 years[1]. As a result, the fate and microbial decomposition of oil in aquatic environments is well understood. Studies have shown that oil degradation can occur in cold-water environments.[2-12]

In response to the spill, the U.S. Environmental Protection Agency assembled a panel of experts to determine what could be done to accelerate the natural biodegradation process in Prince William Sound. The panel recommended the creation of a bioremediation research plan with the following major objectives:

- Examine the extent to which natural biodegradation of oil on the contaminated beaches was occurring.

- Determine if nutrient addition enhanced natural biodegradation of contaminated beaches in the field.[13-15]

- Develop methodology for full-scale application of nutrients to contaminated beaches.

FERTILIZER SELECTION AND CHARACTERISTICS

An important aspect of this project was the selection of fertilizers for the field test. The goal was to find fertilizer formulations that would release nitrogen and phosphorus nutrients over extended time periods or would hold nutrients in contact with surface microbial communities over extended time periods. It was essential that fertilizer formulations be practical and cost-effective for large scale application to contaminated shorelines and have minimal impact on eutrophication potential. Three types of fertilizer were selected:

- Solid, slow-release fertilizer, in which nutrients would be released slowly from a point source and tidal action and rainfall would distribute the nutrients over the beach surface.[16]

- Liquid oleophilic fertilizer, in which nutrients would partition to the oil covering the rock and gravel surfaces and would not be washed away by tidal fluxes.[4,17]

- Fertilizer solutions, in which inorganic nitrogen and phosphorus would be dissolved in seawater and distributed via fixed sprinkler systems.

Several commercially available fertilizer formulations that satisfied these requirements were selected and their nutrient-release characteristics determined.

IBDU Briquettes.

This fertilizer formulation is manufactured in the form of briquettes containing isobutylidene diurea (IBDU), a chemical that spontaneously hydrolyzes into isobutyl aldehyde and urea when released from the briquette matrix into water. Hydrolysis is temperature dependent, being slower at lower temperatures but still significant. The source of phosphorus is a citric acid soluble phosphate fertilizer. Each briquette weighs approximately 17 grams and has a specific gravity of 1.5 to 1.8. The N:P:K ratio is 14:3:3.

Granular Fertilizer.

This fertilizer formulation consists of inorganic nutrient sources (ammonium nitrate, calcium phosphate, and ammonium phosphate) contained in a vegetable oil coating (polymerized by reaction with a cyclic diene). The coating gives the fertilizer its slow-release characteristic. The N:P:K ratio is 28:8:0. The granules have a specific gravity of 1.8.

Oleophilic Fertilizer.

Inipol EAP 22 is a mixture of nutrients encapsulated by oleic acid. Oleic acid and surfactants in the product formulation cause the nutrients to become partitioned to the oil phase, preventing rapid release into the aqueous phase and subsequent washout. Inipol EAP 22 is a clear liquid with a specific gravity of 0.996 and a pour point of 11°C. The N:P:K ratio is 7.3:2.8:0. The chemical composition is given in Table 1.

Table 1. Chemical Composition of Inipol EAP 22.

INGREDIENT	CHEMICAL FORMULA	PURPOSE
Oleic Acid	$CH_3(CH_2)_7CH{=}CH(CH_2)_7COOH$	Oleophilic phase (continuous)
Lauryl Phosphate	$C_{12}H_{25}PO_4$	Phosphate source, surfactant
2-Butoxy-1-Ethanol	$HO\text{-}C_2H_4\text{-}O\text{-}C_4H_9$	Co-surfactant, emulsion stabilizer
Urea	$NH_2\text{-}CO\text{-}NH_2$	Nitrogen source
Water	H_2O	Hydrophilic phase

SITE SELECTION AND CHARACTERISTICS

Criteria for the selection of the test sites were based on the following:

- Typical shoreline of Prince William Sound; i.e., mixed sand and gravel and cobblestone beaches.
- Sufficient area with fairly uniform distribution of sand, gravel, and cobble for the test plots.
- Protected embayment with adequate staging areas and sufficient size to support several test and control plots.
- Uniform oil contamination.

Two test sites were selected for the field demonstration project, Snug Harbor and Passage Cove. Snug Harbor was selected to serve as an oiled beach that approximated the degree of contamination remaining after a heavily oiled beach had been physically washed. In July, a second site was selected that had been physically washed by the Exxon operations. This site, Passage Cove, served as the main reference beach for the large-scale application of fertilizers and as a means to evaluate a sprayer system for liquid fertilizer application.

SNUG HARBOR PROJECT SITE

Site Characterization.

Snug Harbor is located on the southeastern side of Knight Island. The shoreline used for the demonstration was located on the western side of this harbor (Figure 1). The area is surrounded by mountains, reaching an elevation of approximately 2,000 feet with steep vertical ascents. Major sources of freshwater runoff are from precipitation and snowmelt, which is typical of islands in Prince William Sound.

One of the primary reasons for selecting Snug Harbor as a project site was that it contained a long expanse consisting of sand and gravel and another of cobble. Table 2 identifies the plot dimensions and types of treatment used at Snug Harbor. Each plot was divided into 21 blocks, 7 across and 3 deep, with each row of blocks occupying a different tidal area: high, intermediate, and low.

Table 2. Description of Fertilizer Treatment Plots at Snug Harbor.

Assigned name	Beach type	Fertilizer Treatment	Length, m	Depth, m
Eagle	Sand, gravel	None, control	21	12
Otter	Sand, gravel	Oleophilic fertilizer	21	12
Otter	Sand, gravel	IBDU briquettes	35	12
Seal	Cobble	IBDU briquettes	28	12
Seal	Cobble	Oleophilic fertilizer	28	12
Seal	Cobble	None, control	21	8

Fertilizer Application.

Slow-release fertilizers used in this project were briquettes that were applied in mesh bags and granules that were broadcast. The following paragraphs describe the methods used to place these fertilizers.

Large mesh bags composed of herring seine filled with slow-release fertilizer briquettes (IBDU) were placed on the beach in a manner that was intended to provide complete exposure of the beach material to nutrients leaching from the bags. Each bag contained approximately 33 pounds of briquettes. Application of the briquette bags occurred on June 11, 1989. The total quantity of briquettes applied to the 35 m x 12 m plot (Otter Beach) was 800 pounds, representing approximately 100 pounds nitrogen and 24 pounds phosphorus (as P_2O_5). The bags were tethered to 3-foot sections of steel rods that were buried 6 inches below the surface of the beach. Figure 2a indicates the positioning of the 24 bags in the experimental plot.

On June 20 and 21, 1989, the bags were repositioned according to the layout in Figure 2b, as the bags located at the top most row were not being submerged consistently by the high tide (see below). Additionally, preliminary data indicated that the nutrients were being channelled vertically down the beach. Four more bags were added to the beach, resulting in 920 pounds of fertilizer (130 pounds N).

The same arrangement and repositioning was used for the briquette bags on Seal Beach. This beach was 7 m narrower than Otter beach, and the weight of briquettes applied per bag was proportionately reduced.

Figures 3a and 3b represent the significant tidal fluctuations typical of Snug Harbor. These tidal fluctuations affected the amount of time each zone was under water, which in turn affected nutrient dissolution and transport. For example, in the sand and gravel plot treated with the fertilizer briquettes, the top row of fertilizer bags was placed at a relative tidal height of 13 feet. As shown in Figure 2a, the top row of bags was only underwater approximately one-fourth of the days in June. Consequently, precipitation was the primary factor controlling the dissolution and transport of the nutrients in this zone.

Oleophilic fertilizer (Inipol EAP 22) was first applied to Otter Beach in Snug Harbor (mixed sand and gravel) on June 8, 1989. A total of 10 gallons (83 pounds) was applied, which represented approximately 5% of the estimated weight of the oil on the treated beach. A second application of 10.5 gallons of Inipol was made on June 17, 1989 to the Otter Beach plot based on recommendations from Elf Aquitaine representatives.

An application of 14 gallons at Seal Beach in Snug Harbor (cobble) occurred on June 9 and a second application of 13 gallons on June 18.

Inipol was applied to the plots in the evening as the tide was ebbing. Application was initiated at the top of the beach, an hour after the tide was past the lowest zone in the plot. A backpack sprayer with a capacity of four gallons was used to apply the liquid Inipol. The product was warmed to a temperature higher than the pour point to ensure uniform application and to prevent clogging of the spray nozzle.

The weather during the first applications was rainy and cool. During the second application, both days were clear and sunny with temperatures around 60°F. Examination of the plots the day after the second application revealed a gelatinous sheen on the surface of the treated rocks. The sheen lasted for two days, during which time wave action was minimal. This sheen was not observed with the first application.

PASSAGE COVE PROJECT SITE

Site Characterization.

Passage Cove is located on the northwestern side of Knight Island. This site was originally heavily contaminated with oil and was subjected to physical washing by Exxon. Even after physical washing, considerable amounts of oil remained at this site, distributed on the surface of rocks and in beach material below the rocks. Pools of oil and mousse-like material were minimal on the surface. Contamination extended about 50 cm below the beach surface. The shoreline area and the designated beaches in Passage Cove are shown in Figure 1b. All of the beach areas tested consisted of cobblestones set on a mixed sand and gravel base. Table 3 lists beach types, plot dimensions, and fertilizer treatments at Passage Cove.

Table 3. Description of Fertilizer Treatment Plots at Passage Cove.

Assigned name	Beach Type	Fertilizer Treatment	Length, m	Depth, m
Raven	Cobble over mixed sand and gravel	None, control	28	21
Tern	Cobble over mixed sand and gravel	Inipol and water soluble	35	21
Kittiwake	Cobble over mixed sand and gravel	Nutrient soln. sprinkler system	28	21
Guillenot	Mixed sand/gravel with patchy cobble	Inipol and slow-release granules	21	7

Fertilizer Application.

Slow-release granules (Sierra Chemical Co.) and Inipol EAP 22 were both applied to Tern Beach in Passage Cove. The Inipol was applied at the same rate used on the Snug Harbor plots. The granular fertilizer was applied using a commercial broadcast fertilizer spreader at a rate of approximately 0.0033 lbs/ft^2. The total application of nitrogen and phosphorus by slow-release granules in Passage Cove was approximately 400 lbs and 40 lbs, respectively. The granules stuck to the oil on the rock surfaces and were not easily displaced from the beach or redistributed by the tidal action.

Kittiwake Beach in Passage Cove was used to evaluate the effectiveness of application of nitrogen and phosphorus by spray irrigation. Inorganic salts of nitrogen and phosphorus were dissolved in seawater and sprayed onto the beach daily. The spray irrigation system used sprinkler heads typical of lawn sprinklers. The fertilizer solution was pumped by a gasoline-driven well pump to four sprinkler heads set on each side of the plot. Typical applications were about 0.4 inch of water per day. Application rates were established to supply 7 mg/L of nitrogen and 4 mg/L of phosphorus to pore water in the saturated beach material to a depth of 2 m.

ANALYTICAL PROCEDURES

Oil Chemistry.

Beach samples of either mixed sand and gravel or cobble were preserved by freezing. The samples were thawed and mixed thoroughly prior to the initiation of oil analysis. A weighted 100 g subsample was removed and mixed thoroughly with 300 mL of methanol in a separatory funnel. The slurry was shaken for five minutes, and the methanol was decanted into a 2 L separatory funnel. The samples were similarly re-extracted two times with 300 mL HPLC grade methylene chloride. The three organic fractions were combined and back-extracted with 100 mL of 3% aqueous sodium chloride. The phases were separated and the aqueous portion was extracted with 50 mL of fresh methylene chloride. This aqueous extraction in methylene chloride was added to the combined organic fraction.

The combined organic fractions were reduced in volume within a 1 L round bottom flask fitted with a three-ball Snyder column. The volume of solvent was reduced until the color was approximately the color of dilute weathered oil (ca 15 mg/2 ml methylene chloride). The final volume of the extract was measured with a syringe having an appropriate graduated cylinder, and an aliquot was transferred to a GC autosampler vial.

All of the cobblestones were extracted using the same procedure (methanol, followed by methylene chloride), except that shaking was replaced by gentle swirling to remove oil from the rock surfaces.

Gas chromatographic (GC) analysis was accomplished with an instrument capable of reproducible temperature programming with a flame ionization detector and a reliable autosampler. The GC conditions were:

Column: DB-5, 30 m X 0.25 mm, film thickness 0.25 um
Initial Temperature: 45°C, 5 min. hold
Temperature Rate: 3.5°C/min
Final Temperature: 280°C, 60 min. analysis
Injector: splitless, 1 minute valve closure
Injector Temperature: 285°C
Injection: 2.0 microliter
Detector: FID, 350°C

Those samples that demonstrated significant evidence of biodegradation were fractionated to allow separate determination of aliphatics and aromatics. Extracts selected for fractionation were solvent-exchanged to hexane. A volume of 50 microliters of hexamethylbenzene (80 ng/microliter) and 25 microliters of n-decyclohexane (1 microgram/micriliter) was added to each sample extract prior to fractionation. The fractionation was accomplished using a 60/200 mesh silica gel activated at 210 C for 24 hours. The aliphatic fraction was eluted with 30 mL of hexane and the aromatic fraction was eluted with 45 mL of hexane/benzene (1:1). Aliphatic and the aromatic fractions were analyzed using the GC methods described above.

Subsamples of the final concentrated extract were subjected to mass spectral analysis. The analytical procedure is given in the Fucus oil analysis protocols.

Subsamples (5-15 mL) of the final concentrated extract were also removed, filtered through sodium sulfate, and placed in tared watch glasses. After passive evaporation of the solvent, the oil residue weight was determined.

Changes in oil composition were determined by comparing the total weight of all alkanes appearing on the chromatograph, normalized to the total residue weight of oil, on a sample by sample basis.

FIELD TEST RESULTS -- SNUG HARBOR

Visual Observations.

Test beaches at Snug Harbor were moderately contaminated. Visually, the cobble plots had a thin coating of dry, sticky, black oil covering rock surfaces and gravel areas under the cobble. Oil did not penetrate more than a few cm below the gravel surface. In mixed sand and gravel plots, oil was well distributed over exposed surface areas and commonly found 20-30 cm below the surface. In many areas of the test plots, small patches of thick oil and mousse (emulsified oil) could be found. This material was very viscous and mixed with extensive amounts of debris.

Approximately 8-10 days following oleophilic fertilizer application to the cobble beach plot, reductions in the amount of oil on rock surfaces were visually apparent. It was particularly evident from aerial observations where the contrast with oiled areas surrounding the plot was dramatic. A clean rectangle was etched onto the the beach surface. The contrast was also impressive at ground level, where a precise demarcation between fertilizer treated and untreated areas was clearly visible.

Close examination of the treated cobble plot showed that much of the oil on the surface of the rocks was gone. There were still considerable amounts of the oil under rocks and in the mixed gravel below the rocks. The remaining oil was not dry and dull as was the case with oil in other areas of the beach, but appeared softened, more liquid, and sticky to the touch. It had little tendency to come off the rocks. At the time of these observations, oil slicks or oily materials were observed leaving the beach during tidal flushing.

The mixed sand and gravel beach treated with oleophilic fertilizer exhibited visually reduced amounts of oil in an 8-10 day period. Differences between treated and untreated plots, however, were not as dramatic as on the similarly treated cobble beach. Loss of subsurface oil in treated areas was visually apparent.

All other plots looked as oiled as they did at the beginning of the field study. There were essentially no visual indications of oil removal on plots treated with slow-release fertilizer briquettes.

Over the next 2-3 weeks, the cleaned rectangle on the cobble beach remained clearly visible. Oil below the rocks remained but was less apparent.

Six to eight weeks after fertilizer application, the contrast between the treated and untreated areas on the cobble beach narrowed. This was due to reoiling from subsurface material concurrent with the slow removal of oil on the beach material surrounding the plot. It was evident, however, that the total amount of oil on the treated plots had decreased substantially relative to control plots. The corresponding mixed sand and gravel plot was also reoiled but to a lesser extent. All of the other plots still had observable oil contamination but generally lesser than that seen at the beginning of the study.

Toward the end of the summer season, the area used for the nutrient application study became steadily cleaner, including most of the area surrounding the test plots as well. This was attributed to several storms and more frequent rainfall. A heavily contaminated area to the south, which was never treated, remained heavily contaminated by all visual criteria.

Changes in Oil Composition.

Data analysis for oil residue weight and chemistry has not yet been completed. Over 1100 samples have been analyzed and the resulting information is being incorporated into the data base. Six different approaches for analyzing trends in the data are being used. These involve analysis through time of the following:

- Oil residue weights (methylene chloride extractable material),
- Ratios of C17/pristane and C18/phytane,
- Gas chromatographic profiles of aliphatic hydrocarbons,
- Total concentration of aliphatic hydrocarbons,
- Average individual aliphatic hydrocarbon concentrations, and
- Relationship of degradation extent to oil residue weight.

Only the gas chromatographic profiles of aliphatic hydrocarbons are being presented in this paper. The data are still preliminary in nature and are subject to ongoing statistical review.

Gas chromatographic profiles from the beach plots at Snug Harbor were recreated from computer data files. All data have been normalized to oil residue weight and plotted on the same scale. Figures 4a and 4b present the profiles from samples of oil extracted from the mixed sand and gravel under the cobble prior to and four weeks after application of the oleophilic fertilizer, respectively. There were clearly marked compositional changes in the oil over the four week time period. Almost every block in the plot shows significant disappearance of alkane fractions. Figures 5a and 5b are the respective profiles for the plot prior to and after treatment with IBDU briquettes. Although the changes were not as dramatic as with the Inipol treated plot, they were still significant. Chromatographic profile data from the control plots were not available for inclusion.

FIELD TEST RESULTS -- PASSAGE COVE

Visual Observations.

Original contamination in Passage Cove was heavy. Following complete physical washing, oil was well distributed over most of the surface of all cobble and all gravel under the cobble. The oil appeared black, dry, and dull with considerable stickiness. It was spread as a thin layer over the beach material. Relatively few patches of pooled oil or mousse were present, but, where they were present, the oil was thick and viscous. Oil was also found at depth in the beach, generally 30 to 40 cm below the surface.

Within approximately two weeks following application of Inipol and slow-release granules, it became apparent that the treated beach was considerably cleaner relative to the control plots. In contrast to the observations at Snug Harbor, not only did the rock surfaces look cleaner, but the oil under the rocks and on the gravel below was also disappearing. In another two weeks, oil could be found only in isolated patches and below 10 cm in the subsurface. At no time were oil slicks or oily material seen leaving the beach area.

Unexpectedly, the beach treated with fertilizer solution from the sprinkler system appeared as clean as the Inipol/granule treated plot. The only difference was that it lagged behind the Inipol/granule treated beach by approximately 10-14 days. By the end of August, both beaches looked equally clean. In contrast, the control plot appeared much as it did at the start of the field study. Oil in the subsurface still remained in all plots.

Changes in Oil Composition.

Over 600 samples have been analyzed and the resulting information has been incorporated into the data base. The approaches for evaluating trends in the data are the same as those used for Snug Harbor. Only a few chromatographic profiles were available for this paper.

Figures 6a and 6b present the respective profiles of the untreated control plot (Raven) prior to and three weeks after sprinkler application began in Passage Cove. Blocks with no profiles indicate that data for such blocks were unavailable for plotting. Since a floating concentration scale was used in these plots, changes in relative concentrations for the hydrocarbons can be visualized by comparing the overall profile of the peaks to a profile typical of a relatively undegraded but weathered oil. This is shown as the solid line in the figures. From comparison with the control plots, it is clear that significant degradation has taken place within two weeks even with no fertilizer treatment.

Figures 7a and 7b present the profiles of the treated beach (Kittiwake) before and three weeks after sprinkler application of dissolved nutrients, respectively. In contrast to the control plots, the biodegradation appears to have been substantially more extensive, and this corresponds to the visual disappearance from the rock surfaces. This suggests that degradation of other fractions (aromatics, waxes, asphaltenes, polars) of the oil may be degrading. As additional chemical and mass spectral analyses of the oil are completed, more insight into this supposition will be provided.

CONCLUSIONS

Information presented is preliminary in nature. Conclusions drawn are done so purely from a speculative standpoint and may change once all the data are tabulated, evaluated, and analyzed statistically.

Visual observations suggest enhanced biodegradation occurred on the beaches treated with Inipol, slow-release briquettes, and dissolved solutions of inorganic nutrients. Clean-up was especially visual on the Inipol plot at Snug Harbor and the sprinkler plot at Passage Cove. It is unclear that the differences between the treated and control plots were statistically significant. Analysis of oil from control plots showed that changes in oil composition were substantial and progressed steadily through time. This suggested that natural biodegradation of the oil occurred at a surprisingly rapid rate.

Samples of oil from fertilizer-treated beaches, particularly from cobble surfaces, taken at about the time when the oil was visually disappearing, showed substantial changes in hydrocarbon composition, indicating extensive biodegradation. This suggests that biodegradation was effecting removal of the oil, both through direct decomposition and possibly through the production of biochemical products (bioemulsifiers) known to be produced by bacteria as they consume oil and hydrocarbons as sources of food.

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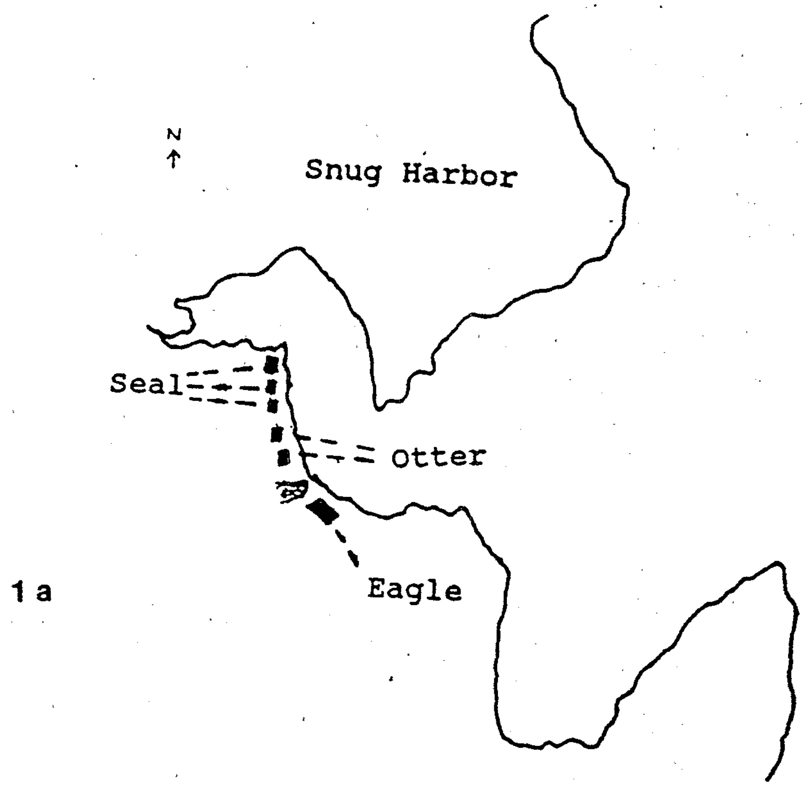

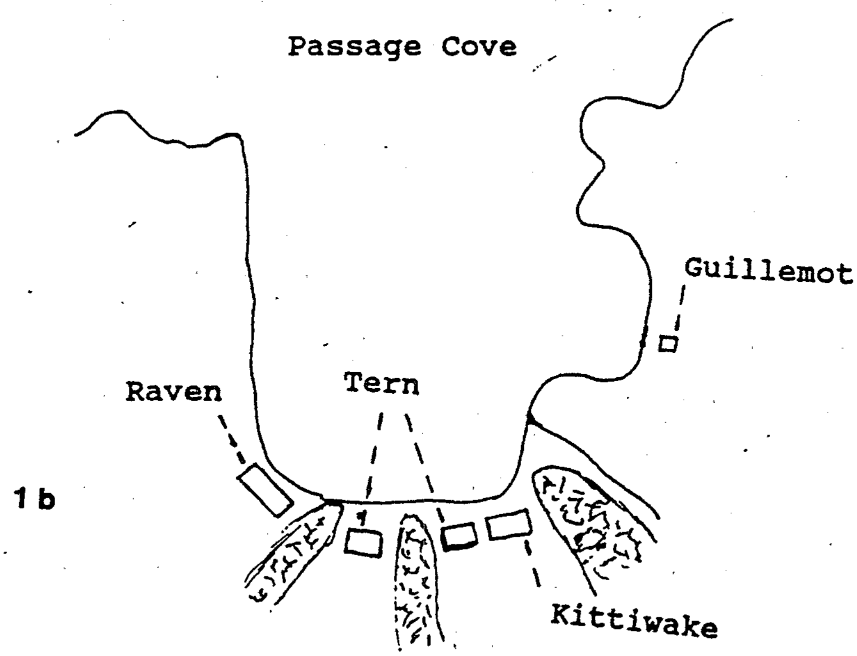

Figure 1a. Snug Harbor, Knight Island, with treatment beaches identified.

Figure 1b. Passage Cove, Knight Island, as above.

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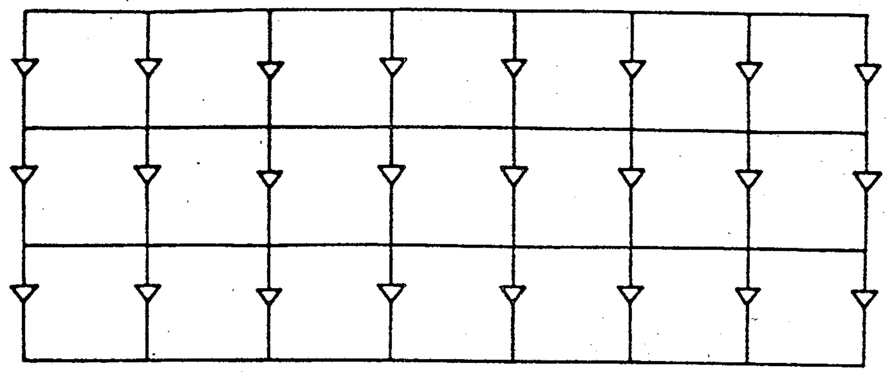

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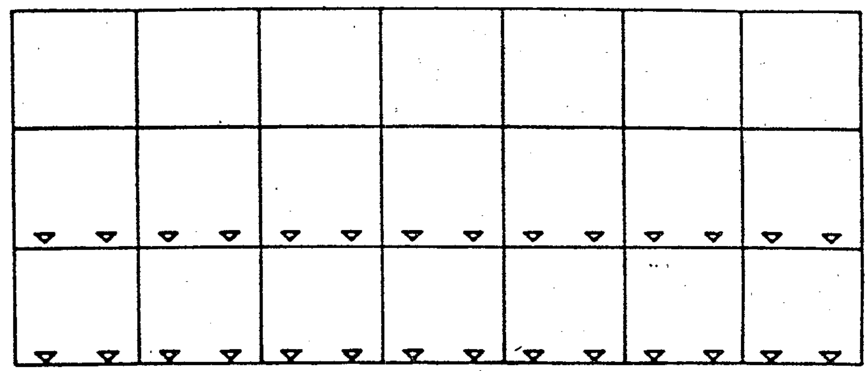

Figure 2a. **Placement of the Bags of Water-Soluble Fertilizer on Otter and Seal Beaches**

Figure 2b. **Repositioning of the Bags of Water-Soluble Fertilizer on Otter and Seal Beaches**

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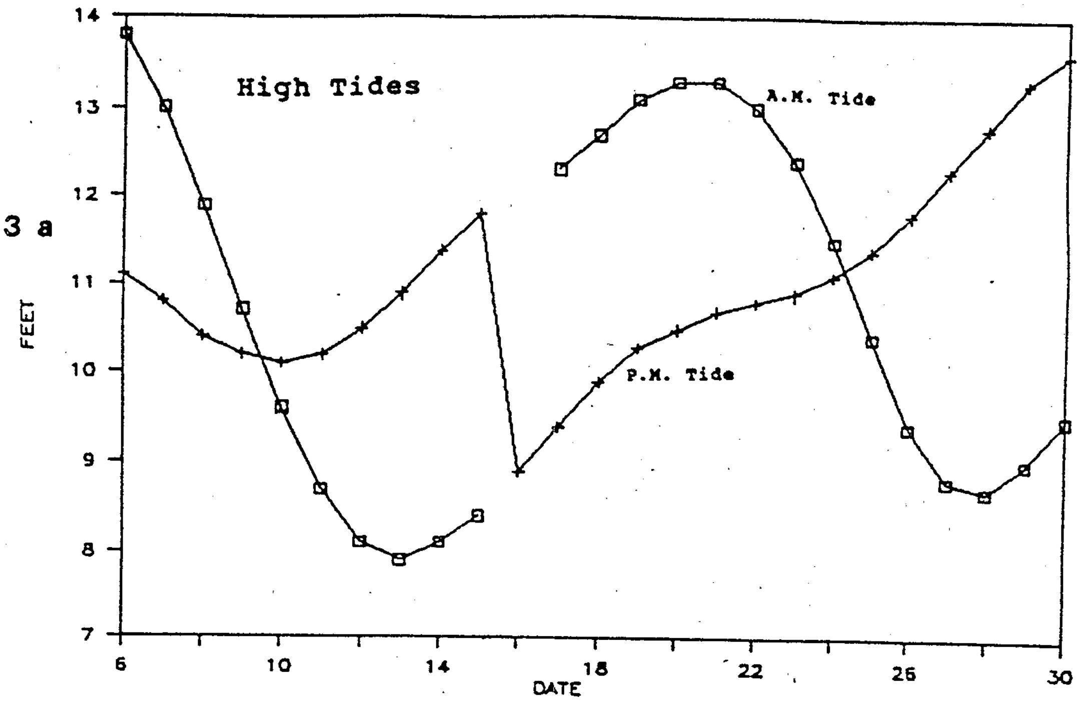

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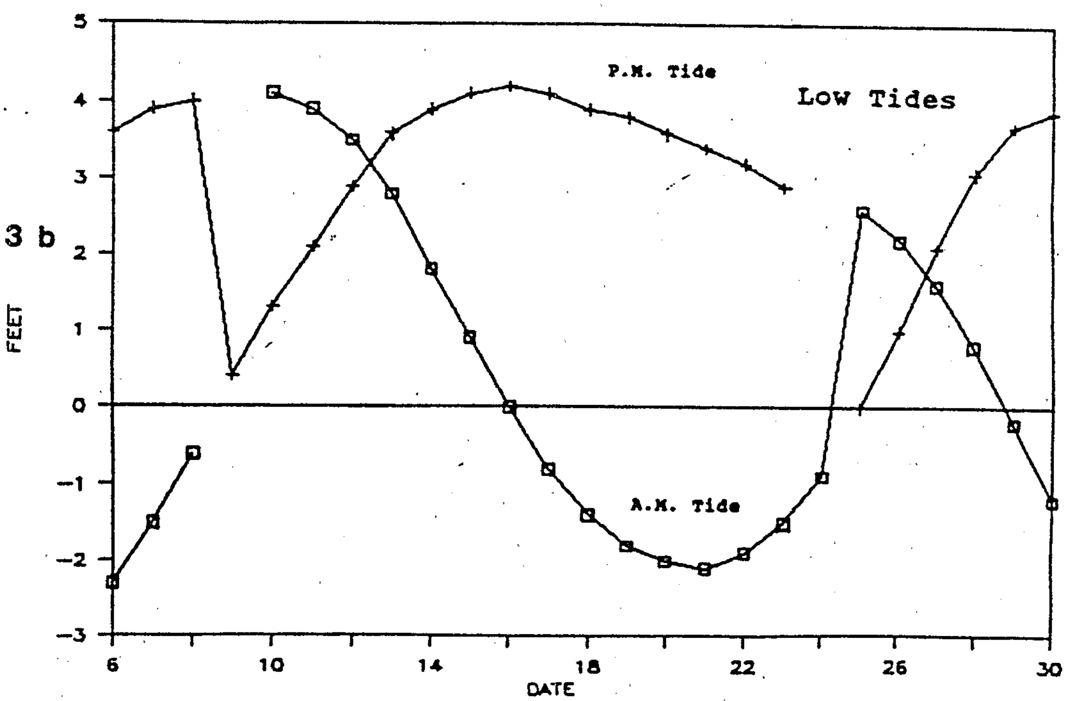
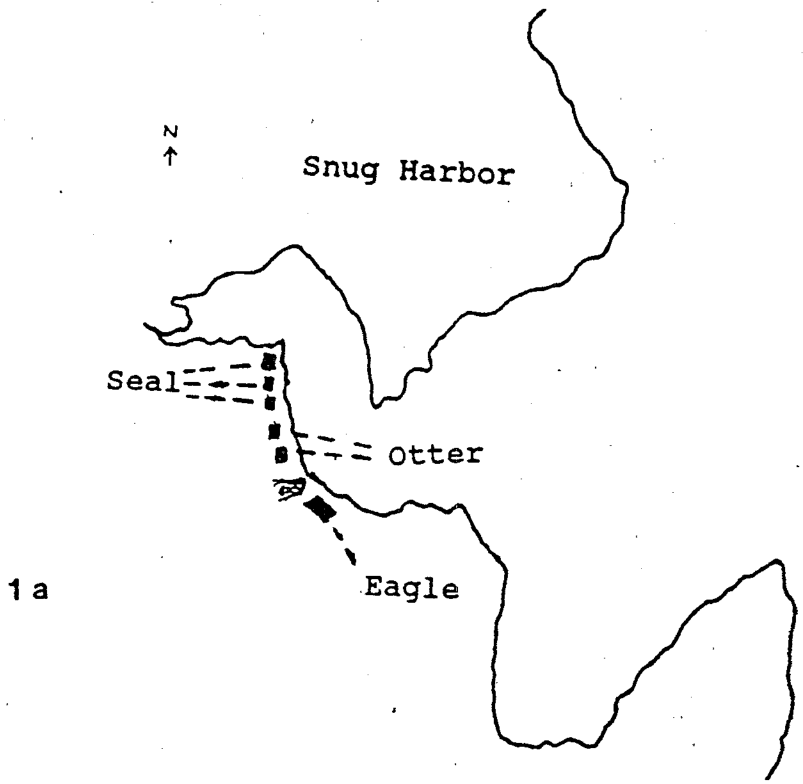
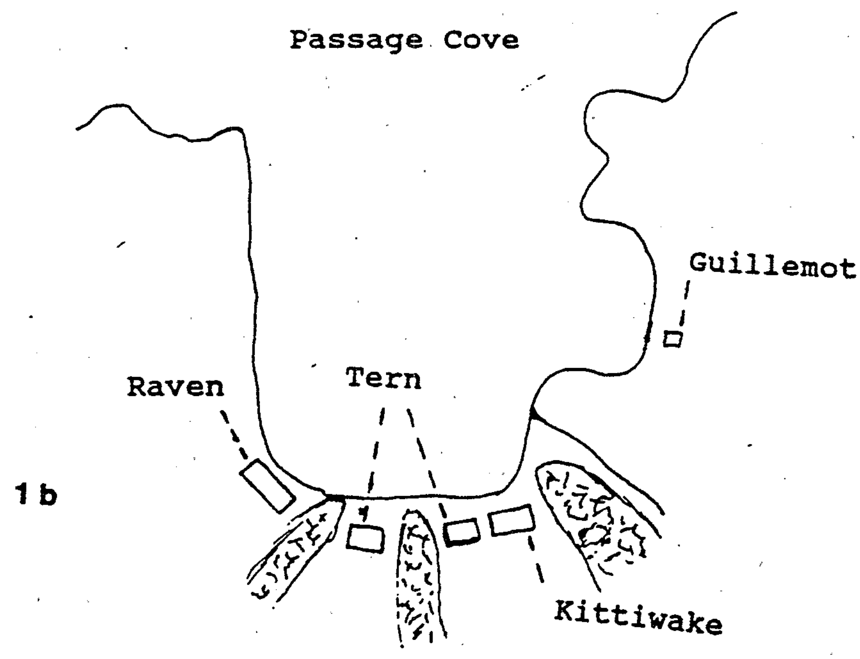


Tidal Fluctuations for Snug Harbor, June 6-30, 1989.

Figure 3a. High tides.

Figure 3b. Low tides.





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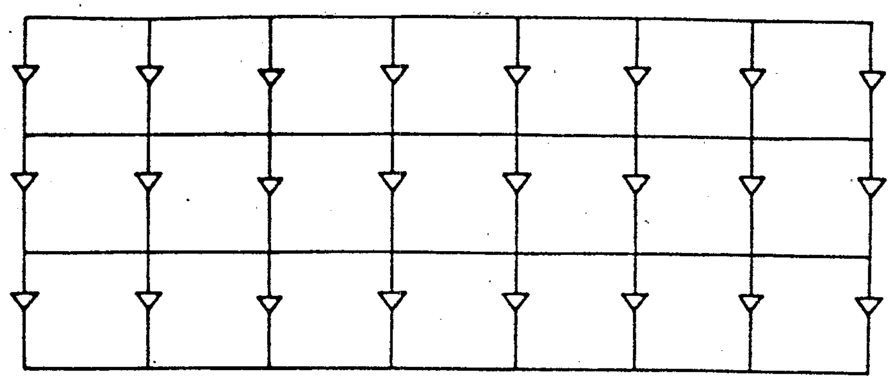

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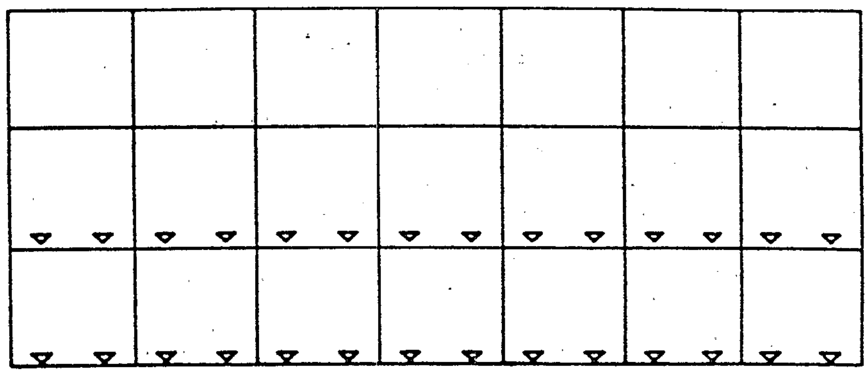

Figure 2a. **Placement of the Bags of Water-Soluble Fertilizer on Otter and Seal Beaches**

Figure 2b. **Repositioning of the Bags of Water-Soluble Fertilizer on Otter and Seal Beaches**

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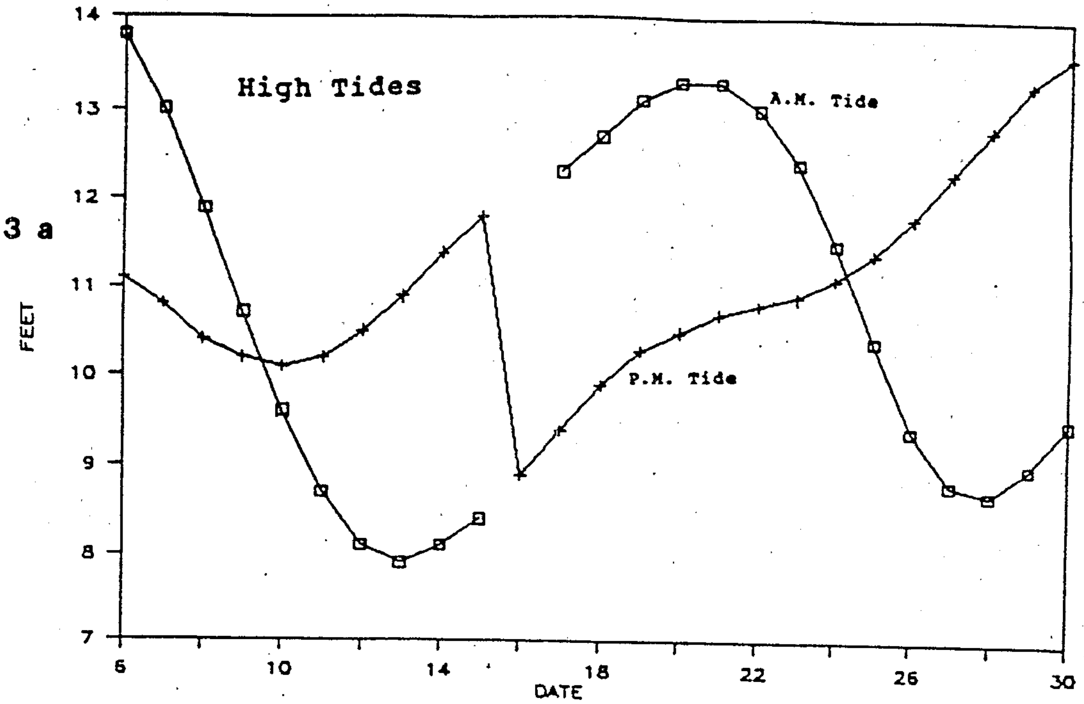

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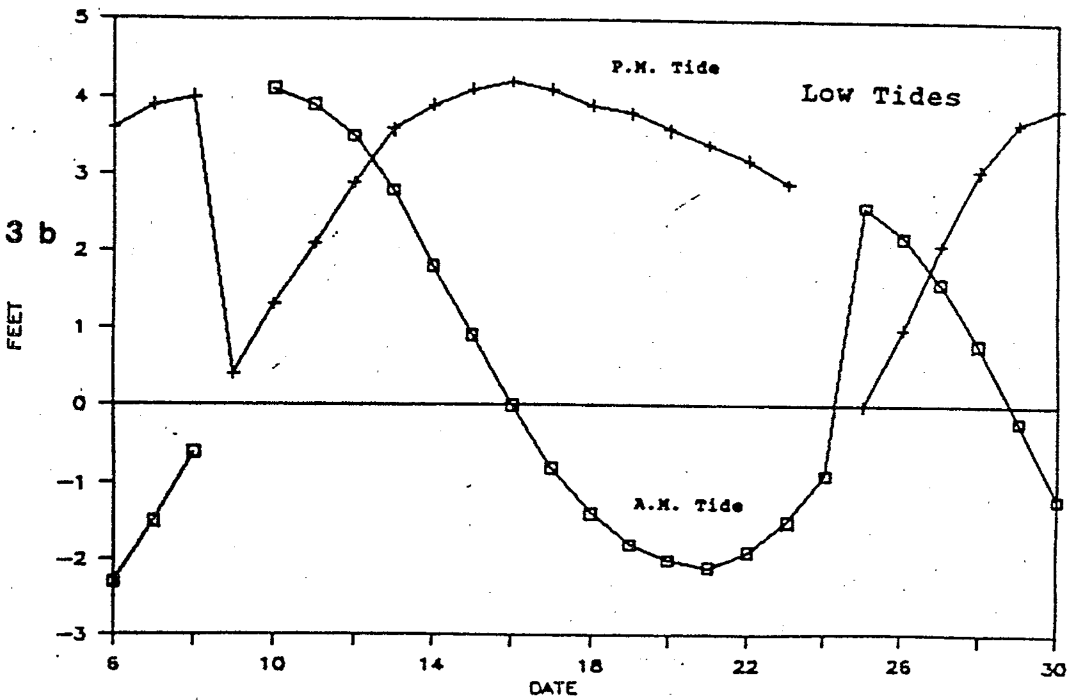


Tidal Fluctuations for Snug Harbor, June 6-30, 1989.

Figure 3a. High tides.

Figure 3b. Low tides.

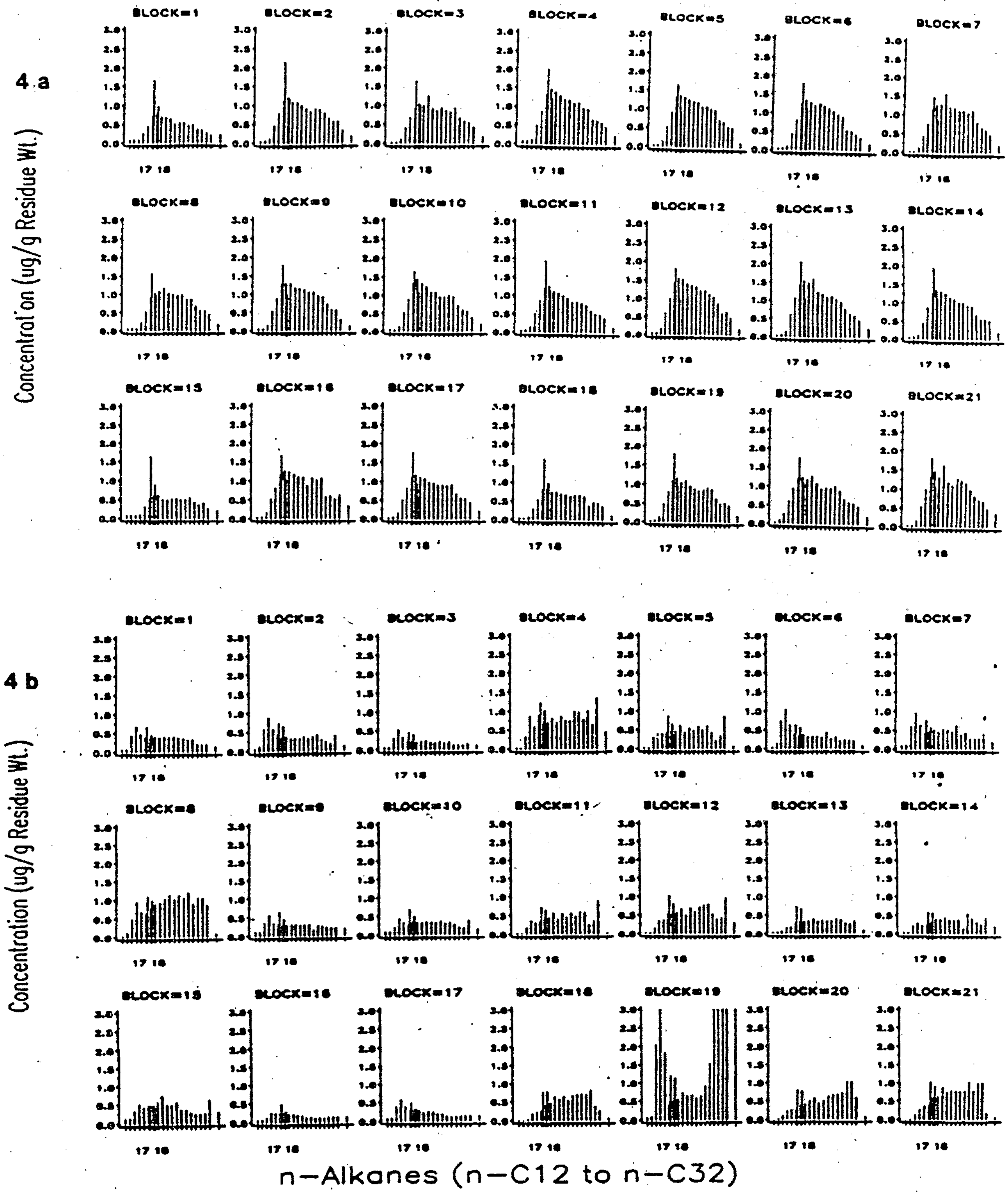
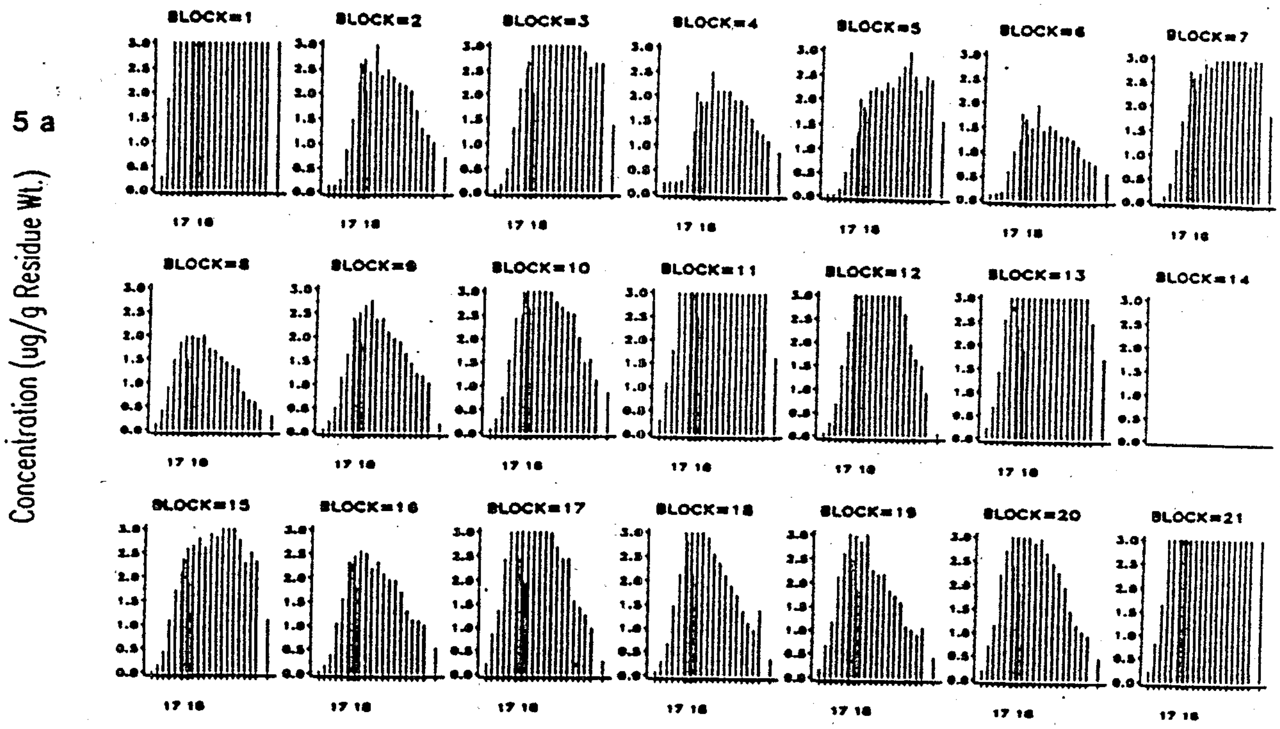


Figure 4a. Seal beach, cobble, before fertilizer application.

Figure 4b. Seal beach, cobble, four weeks after oleophilic fertilizer application.



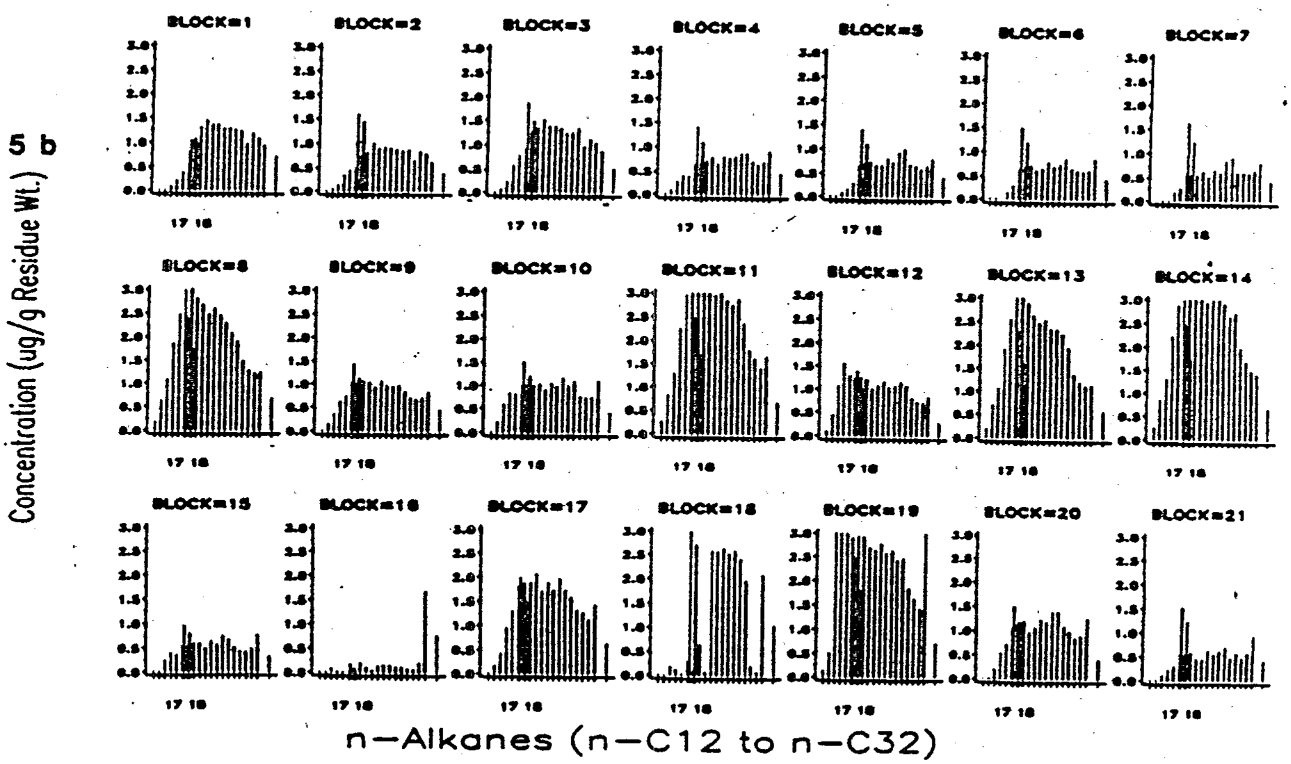
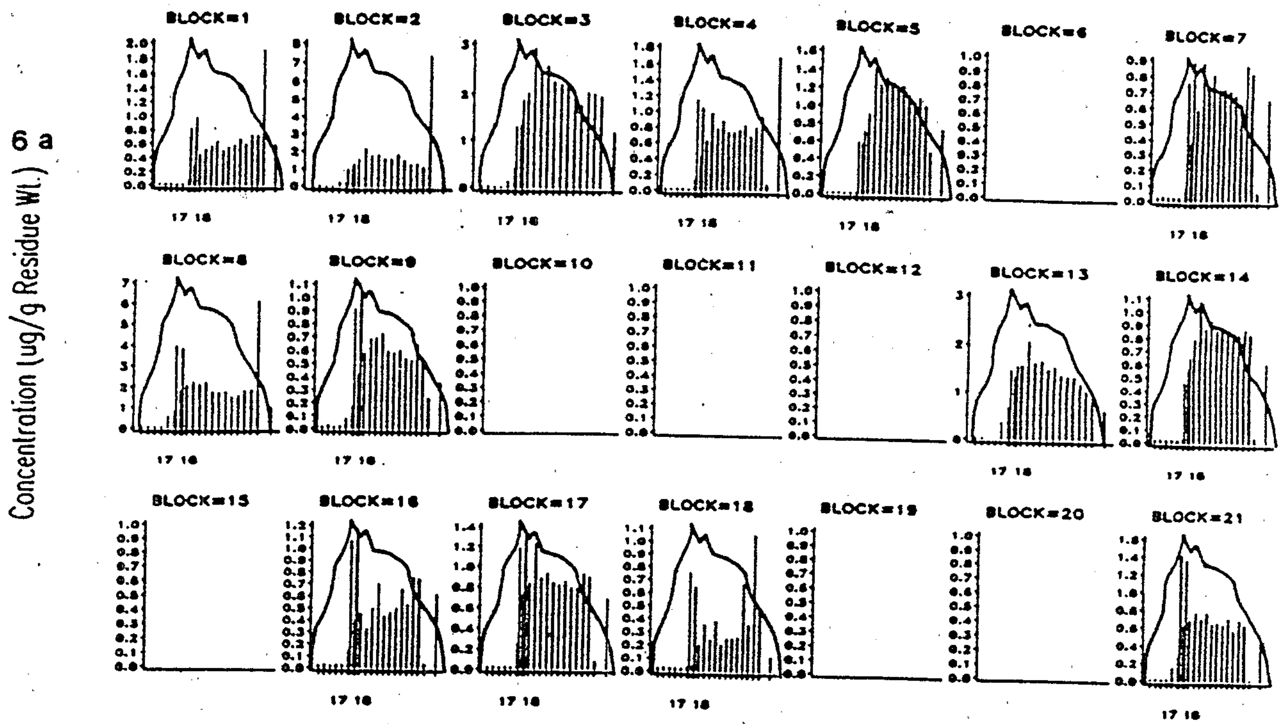


Figure 5a. Otter beach, sand and gravel, before fertilizer application.

Figure 5b. Otter beach, sand and gravel, four weeks after fertilizer briquet application.



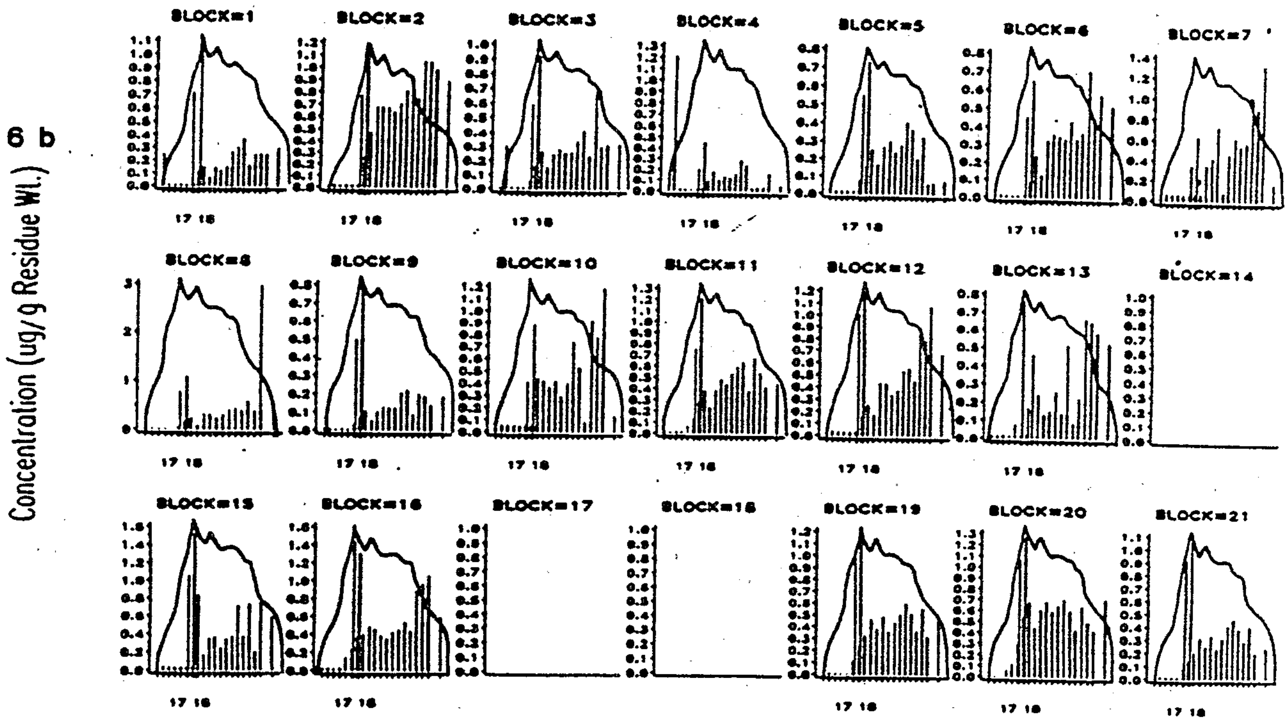


Figure 6a. Raven beach, cobble, before fertilizer application.

Figure 6b. Raven beach, cobble, three weeks later, no fertilizer.

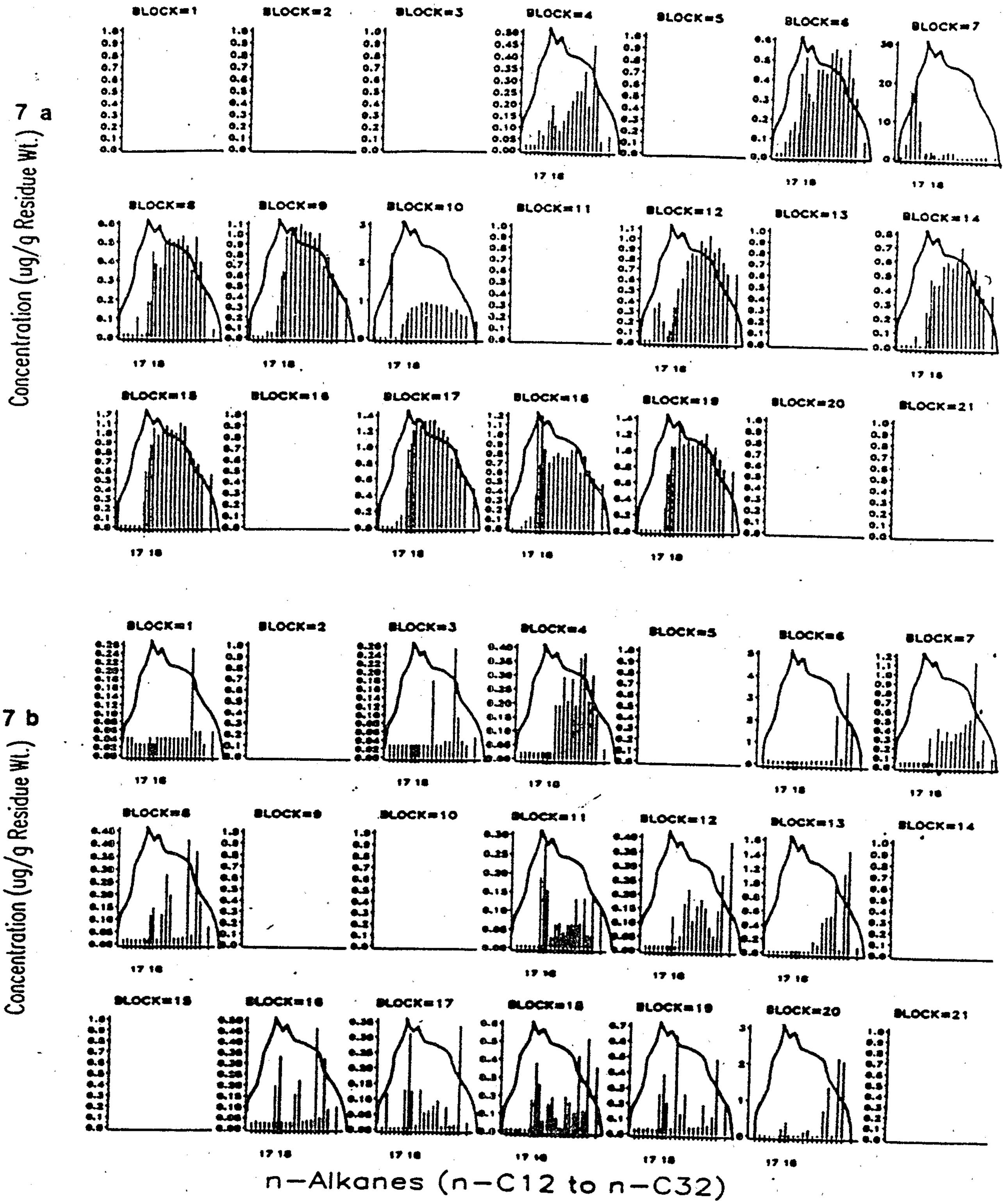


Figure 7a. Kittiwake beach, sand and gravel, before fertilizer application.

Figure 7b. Kittiwake beach, sand and gravel three weeks after application of water soluble fertilizer.