AA Technical Memorandum OMPA-7

INVESTIGATION OF PETROLEUM IN THE MARINE

ENVIRONS OF THE STRAIT OF JUAN DE FUCA

AND NORTHERN PUGET SOUND

PART II (Second-Year Continuation)

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Boulder, Colorado
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This paper presents results of research sponsored by the Environmental Protection Agency and administered by the National Oceanic and Atmospheric Administration (NOAA).

FOREWORD

Substantially increased petroleum transfer and refining activities taking place in the Strait of Juan de Fuca and Northern Puget Sound areas increase the chances of chronic and/or acute oil inputs into the marine environment. To provide a basis for measuring future changes in environmental levels of petroleum, a study was undertaken to measure the levels of petroleum hydrocarbons in the marine environment of the region. The results reported here are from a second-year continuation of a monitoring study and from a small oil-spill in Port Angeles Harbor in May 1979. The research was conducted by the Northwest and Alaska Fisheries Center in Seattle. It was part of an environmental assessment of the region, and was supported by the U.S. Environmental Protection Agency and administered by the NOAA Marine Ecosystem Analysis Puget Sound Project.

ABSTRACT

The Strait of Juan de Fuca and Northern Puget Sound regions have accommodated the transportation and refining of petroleum without serious problems from spilled oil. However, proposed activities related to the transport and use of petroleum in these regions increase the chances of significant additions of petroleum to this marine environment. This project was designed by the National Analytical Facility, Environmental Conservation Division, Northwest and Alaska Fisheries Center, to provide an accurate and reliable set of data against which future levels of petroleum compounds can be compared.

An investigation was initiated in February 1977 to measure existing levels of petroleum hydrocarbons in the Strait of Juan de Fuca and Northern Puget Sound, and to investigate spatial and temporal trends in occurrence, concentration, and composition of petroleum-related hydrocarbons. Sediment and mussel samples from 19 stations located along shipping lanes in the Strait of Juan de Fuca and Northern Puget Sound were analyzed for alkanes and aromatic hydrocarbons. Although the sites were relatively free from petroleum contamination, hydrocarbons indicative of petroleum were found at Cherry Point, Sandy Point, March Point, False Bay, Dungeness/Three Crabs, Ediz Hook, Kydaka Point, and Baadah Point. As a continuation of these studies, sediment samples were collected at ten of these sites three times (summer and fall 1978 and winter 1979) and analyzed for petroleum hydrocarbons. A small oil spill in Port Angeles Harbor on 13 May 1979 afforded an opportunity to evaluate the premise that hydrocarbon concentrations in intertidal sediment and mussels can be used to monitor for spilled oil in this region.

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ACKNOWLEDGMENTS

The work described in this report was performed for the Marine Ecosystems Analysis (MESA) Puget Sound Project, Dr. Howard S. Harris, Manager, and Edward R. Long, contract supervisor.

We would like to express our appreciation to Neva L. Karrick, who coordinated this project with MESA for the Northwest and Alaska Fisheries Center (NWAFC); to Robert C. Clark, Jr., for technical advice; and to Warren Ames for sample collection; to Marianne R. Tomita for manuscript typing; to James H. Peacock, Nancy A. Williams-Nelson, and Carol E. Hastings for preparation of graphical material; and to National Analytical Facility personnel, Douglas G. Burrows, Karen A. Culler, Teresa I. Federighi, David L. Fisher, Donald D. Gennero, Orlando Maynes, and L. Scott Ramos.

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INTRODUCTION

This investigation was part of a major interdisciplinary study to determine the potential effects of increased petroleum transport and refining activities in the Strait of Juan de Fuca and Northern Puget Sound. The overall effort, funded by the Environmental Protection Agency (EPA) and administered by the Marine Ecosystem Analysis (MESA) Puget Sound Project, was designed to focus the capabilities of NOAA and of Federal, state, and local agencies upon specific environmental problems through intensive research projects. Thus far, the Puget Sound Project has sponsored more than 20 research investigations designed to provide an environmental data base for the region upon which future regulatory, socio-economic, and resource management decisions may be based (1).

Studies are being conducted to define the physical and chemical oceanography of the Greater Puget Sound area, the abundance and distribution of the aquatic biota in the area, and the levels of petroleum hydrocarbons in sediments and aquatic organisms. Models derived from these studies will help in predicting oil spill trajectories, potential trophic pathways, and in the assessment of environmental impacts of an oil spill, if one occurs in these waters.

The Northern Puget Sound and Strait of Juan de Fuca regions have accommodated the transportation and refining of petroleum for years without serious problems from oil spills. However, recent increases in tanker transport of crude oil in this region increases this risk. Knowledge of the presence and the distribution of current levels of petroleum is necessary to assess future changes and to determine whether petroleum compounds are increasing in this environment.

A pilot study (2) established field sampling and laboratory analysis methodologies suitable for investigating baseline levels of petroleum hydrocarbons in the intertidal zone. A baseline study (3) was initiated in February 1977 to measure existing levels of petroleum hydrocarbons in the Strait of Juan de Fuca and Northern Puget Sound, and to determine spatial and temporal trends in their occurrence, concentration, and composition. Sediment and/or mussel samples were collected and analyzed from 19 stations located along shipping lanes in the Strait of Juan de Fuca and Northern Puget Sound (Fig. 1). The principal objective was to provide an accurate and reliable set of data against which future levels of hydrocarbons can be compared.

As a continuation of the baseline study, sediment samples were collected at ten sites, during summer and fall 1978 and winter 1979, and analyzed. On 13 May 1979 a small diesel oil spill (about 400 gallons) occurred in Port Angeles Harbor (Fig. 2), providing a rare opportunity to test the use of sediments and mussels as indicator materials in monitoring petroleum pollution in this region.

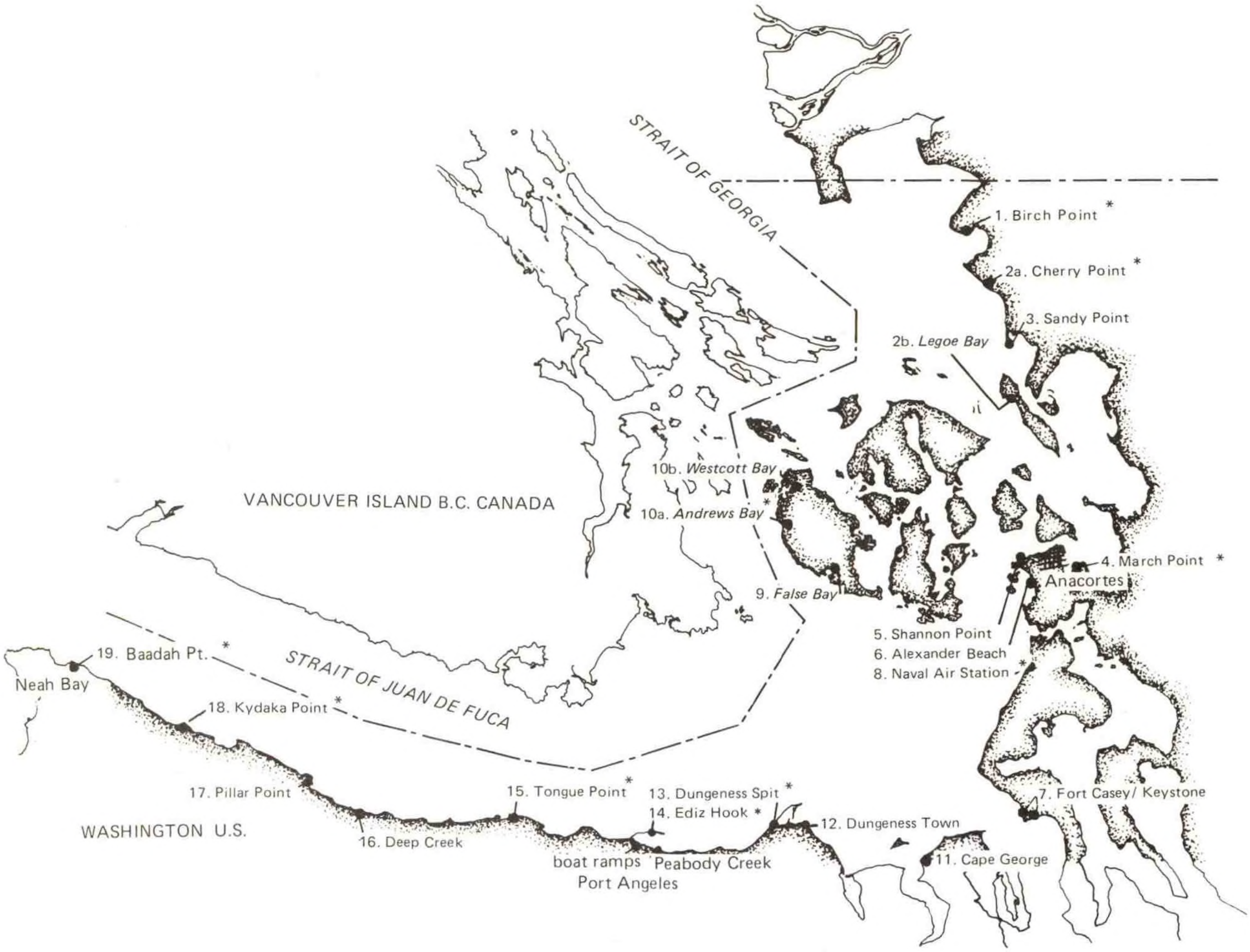

Figure 1. Sampling sites in the Strait of Juan de Fuca and Northern Puget Sound. Sites labeled with an asterisk were used for this (1978-1979) study.

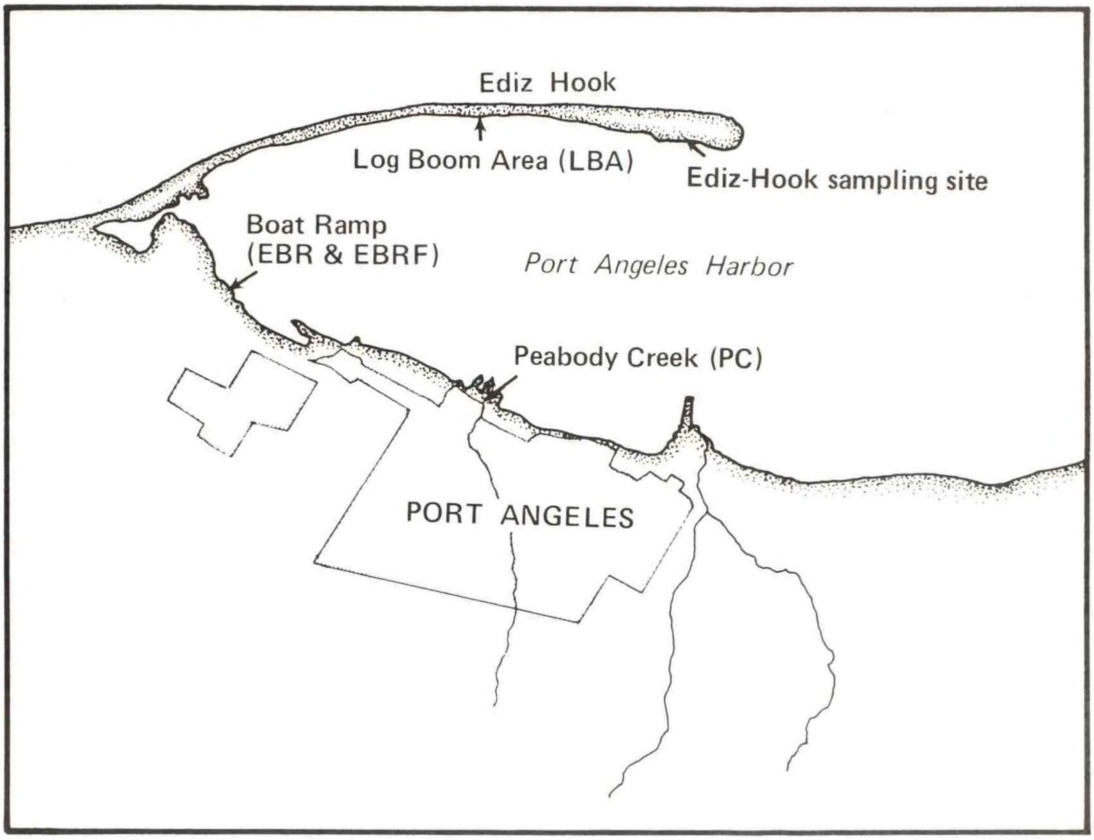

Figure 2. Port Angeles Harbor.

METHODS

The methods used were described in detail by Brown et al. 1979 (3), and are summarized below.

Sampling

Sediment samples were collected at 10 stations in Northern Puget Sound and the Strait of Juan de Fuca during summer and fall 1978 and in winter 1979 (Fig. 1). On 18 May and 12 June 1979 (after the diesel oil spill of 13 May), samples of sediment were collected at several locations in Port Angeles Harbor (Fig. 2), including the baseline sampling site near the Coast Guard station; mussels were collected at the baseline sampling site on both dates. For comparative purposes, a sample of diesel fuel was obtained from a local supplier.

Sediment was collected at ten random positions along a 30-m length of beach at the 0.6-m tidal elevation and combined. Surface sediment cores were taken with a 5.5-cm-diameter by 2-cm-deep aluminum coring device (total volume = 47 cm^3). A mussel sample consisted of 10 to 15 mussels 3- to 5-cm in length collected randomly within the 0.6-m to 0.9-m tidal height along 30 m of beach.

Hydrocarbon Extraction and Separation

The analysis schemes for sediment and mussel tissue are diagrammed in Fig. 3. Subsamples of thawed sediment were extracted two times with methanol, and then three times with a 2:1 mixture of dichloromethane and methanol using a ball-mill tumbler. The combined extracts (dichloromethane and methanol) were washed with water to remove the methanol. The dichloromethane extract was treated with activated copper to remove elemental sulfur and filtered through silica gel to remove some of the polar compounds. The concentrated extract in *n*-hexane was separated into two fractions using silica-gel chromatography and eluting with pentane and dichloromethane/pentane; the first fraction contained the saturated hydrocarbons, and the second contained the unsaturated hydrocarbons including the aromatic compounds.

Subsamples of the blended mussel tissue were saponified with 4N sodium hydroxide by allowing the mixture to stand overnight (18 h) at 30°C. The digested mixture was extracted twice with peroxide-free diethyl ether. The ether extract was filtered through a silica-gel column. This extract was exchanged into *n*-hexane and fractionated using silica gel. The resulting eluates were concentrated to 1 ml in *n*-hexane for analysis.

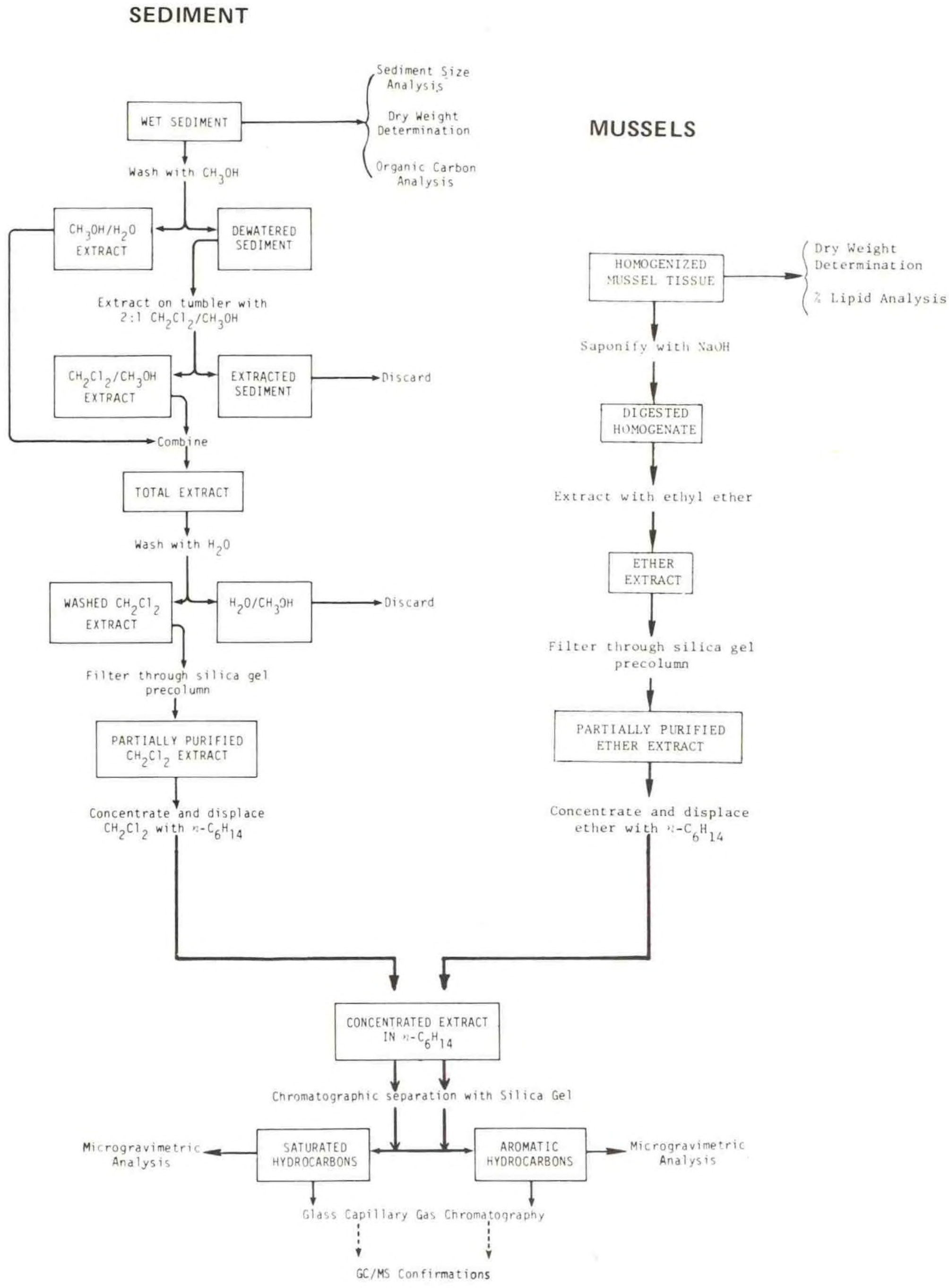

Figure 3. Analytical scheme for hydrocarbons in sediment and mussel tissue.

Gas Chromatography/Mass Spectrometry

The extracts were analyzed for the hydrocarbons using a Hewlett-Packard (model 5840A) microprocessor-controlled gas chromatograph (GC) equipped with an automatic sample injector, model 7671A, glass capillary column (20- to 30-m long and 0.25-mm i.d.) coated with either SE-30 or SE-54, and a hydrogen flame ionization detector. The GC injection port was modified for splitless injections using capillary columns, as described by Ramos et al. (4). Column temperature was programmed at $4^{o}C$/min. from 50^{o} to 280^{o} and held at $280^{o}C$ for 20 minutes. Peak areas were automatically integrated and the concentrations calculated using internal standards. The identity of compounds detected and quantitated by GC were confirmed by gas chromatography-mass spectrometry (GC-MS) as necessary, using an identical GC system interfaced with a Finnigan 3200 mass spectrometer equipped with an Incos 2300 data system.

Gravimetric Determinations

The dry weight of sediment was determined by heating 10-20 g of sediment in a drying oven for 24 h at $120^{o}C$. Dry weight of mussel tissue was determined by drying ca. 0.5 g of tissue mixed with sand.

Total saturated and unsaturated hydrocarbons was determined by transferring 25 μl of the respective extracts from the silica-gel chromatographic separation (Fig. 3) onto the balance pan of a microbalance and allowing the solvent to evaporate until a constant weight was reached.

RESULTS

Site locations, field conditions at the time of sampling, and all data resulting from chemical analyses were recorded on computer punch cards and archived at the National Oceanographic Data Center (3).

Field Observations

Oil slicks or other overt evidence of oil contamination were not observed at any of the sites during the one-year study except at Port Angeles after the oil spill in May 1979. No physical changes in the beaches were noticed.

Total Saturated and Total Unsaturated Hydrocarbons

Total saturated hydrocarbons were determined as the weight of extractable material present in the fraction that is eluted from the silica column with pentane. The fraction eluted from the column with 40% dichloromethane in pentane was determined similarly as the total unsaturated hydrocarbons. In both cases the results included contributions from all materials present in that silica-gel eluate, not just from the individual hydrocarbons selected for GC quantitation. Total gravimetric data are presented in Tables 1 and 2.

The concentrations of selected alkanes and selected aromatic hydrocarbons in sediments are reported in Tables 3, 4 and 5. The data were summarized by (1) adding the concentrations of the alkanes (ΣSelected Alkanes, Table 6), and (2) adding the concentrations of the selected aromatic compounds (ΣSelected Aromatics, Table 7). The Σ Selected Alkanes are also graphed in Figures 4 and 5.

As in the first year's study (3), the mean Σ Selected Alkanes in sediment during the second year continuation was highest at Ediz Hook (680 ng/g dry weight). The highest mean ΣSelected Aromatics in sediments was found at March Point (250 ng/g dry weight).

As reported for the previous study (3), when benzpyrenes were detected, both benzo(e)pyrene and benzo(a)pyrene were found, and at similar concentrations.

Sediment Analysis

Total organic carbon (TOC) and sand-to-mud ratios were used to characterize the sediment and these results are reported in Table 8.

Port Angeles Oil Spill

The concentrations of the alkanes and aromatic compounds analyzed in the samples collected following the Port Angeles oil spill on 13 May 1979 are given in Table 9. Chromatograms pertaining to these samples are included in Figures 6 through 11. The Σ Selected Alkanes and the Σ Selected Aromatics are included in Table 10.

Sediment and mussel samples were collected in Port Angeles Harbor (Fig. 2) on 18 May and 12 June 1979, following the spill of about 400 gallons of diesel fuel oil on 13 May. GC patterns and relative hydrocarbon concentrations found in sediment samples from the boat ramp site (Fig. 7A,B) on both sampling dates and in the mussel sample from the Ediz Hook site on 18 May (Figs. 8B and 9B) were similar to No. 2 fuel oil (Fig. 6). For both dates, the sediment samples from the Ediz Hook site had GC patterns and relative concentrations for the alkanes (Fig. 10B,10C) similar to that of the 18 May boat ramp sediment sample (Fig. 7A,B), but did not contain measureable quantities of aromatic hydrocarbons (Fig. 11B,11C). The concentrations of the aromatic hydrocarbons in the Ediz Hook site sediments were too low to be measured (about ten times lower than the 18 May boat ramp sediment). The amplified mass spectral data (Fig. 12) shows that aromatic hydrocarbons were present in the 18 May Ediz Hook sediment sample, though at concentrations below normal measureable limits.

Table 1. Total saturated and total unsaturated hydrocarbons in samples of sediment from Port Angeles Harbor after oil spill (μg/g dry weight; ppm).

Sediment collection site	Total saturated hydrocarbons		Total unsaturated hydrocarbons	
	18 May 1979	12 June 1979	18 May 1979	12 June 1979
Peabody Creek, sediment	210	100	6	37
Boat Ramp, sediment	110	1,500	12	160
Boat Ramp, fine sediment		28		0.5
Log Dump, sediment	7	7	0.2	0.2
Ediz Hook site, sediment	27	33	1	ND[1]
Ediz Hook site, mussel	1,700	450	930	310

1 ND means there were no measurable quantities

NOTE: Blank space indicates no samples taken

Table 2. Total saturated and total unsaturated hydrocarbons in sediment samples collected during 1978-1979 (μg/g dry weight; ppm).

Sediment collection site	Total saturated hydrocarbons			Total unsaturated hydrocarbons		
	6/78	9-11/78	1-3/79	6/78	9-11/78	1-3/79
Birch Point	0.73	1.0	2.0	0.24	ND[1]	ND[1]
Cherry Point	2.3	1.3	1.5	ND[1]	0.97	ND[1]
March Point	4.9	11	4.4	0.44	3.2	0.18
Naval Air Station	0.23	ND[1]	0.39	0.01	ND[1]	0.76
Andrews Bay	2.6	1.4	2.3	ND[1]	ND[1]	0.17
Dungeness Spit	0.33	ND[1]	0.39	1.4	ND[1]	0.09
Ediz Hook	19	19	18	0.86	ND[1]	0.15
Tongue Point	1.5	ND[1]	0.85	0.08	0.63	ND[1]
Kydaka Point	1.2	0.95	1.7	0.19	0.08	ND[1]
Baadah Point	3.2	3.5	2.5	0.50	ND[1]	0.29

[1] ND means there were no measurable quantities

Table 3. Hydrocarbons in sediment samples from ten sites in the Strait of Juan de Fuca and Northern Puget Sound collected during June 1978 (ng/g dry weight; ppb).

Alkanes	Birch Point	Cherry Point	March Point	NAS Whidbey Island	Andrews Bay	Dungeness Spit	Ediz Hook	Tongue Point	Kydaka Point	Baadah Point
n-C_{10}	0.7	1	3	1	5	2	2	1	4	5
n-C_{11}	0.7	1	2	1	6	3	5	1	6	6
n-C_{12}	0.8	1	4	1	7	3	10	1	7	7
n-C_{13}	1	2	6	1	8	3	20	2	9	9
n-C_{14}	1	2	8	2	8	3	20	2	9	9
n-C_{15}	20	5	30	2	10	3	30	3	9	10
n-C_{16}	2	4	20	1	8	3	30	2	10	10
n-C_{17}	40	20	30	7	300	3	40	5	20	90
Pristane	2	8	20	1	10	2	60	5	20	20
n-C_{18}	4	5	20	1	8	2	40	3	10	10
Phytane	3	3	10	0.5	3	0.6	50	2	4	7
n-C_{19}	3	6	20	1	8	2	40	4	10	10
n-C_{20}	2	5	20	1	7	2	40	4	10	10
n-C_{21}	2	6	20	3	9	3	40	7	20	20
n-C_{22}	2	5	10	3	7	3	20	5	20	10
n-C_{23}	4	8	10	5	9	4	20	9	20	20
n-C_{24}	2	6	8	4	6	3	10	6	20	20
n-C_{25}	6	10	10	6	9	5	20	10	20	20
n-C_{26}	2	6	6	5	5	4	7	6	20	20
n-C_{27}	5	20	10	8	10	7	20	20	30	30
n-C_{28}	2	6	5	3	4	3	20	5	20	20
n-C_{29}	4	9	9	7	10	5	20	10	20	20
n-C_{30}	<0.3	5	4	3	5	3	7	5	10	10
n-C_{31}	4	9	10	8	10	7	10	20	20	20

Table 3. Cont.

Aromatic Hydrocarbons	Birch Point	Cherry Point	March Point	NAS Whidbey Island	Andrews Bay	Dungeness Spit	Ediz Hook	Tongue Point	Kydaka Point	Baadah Point
Isopropylbenzene	<0.1	5	0.4	<0.1	0.4	<0.3	0.3	<0.1	1	1
n-Propylbenzene	<0.2	1	1	<0.1	0.7	<0.3	0.3	<0.1	<0.1	0
Indan	<0.1	0.2	0.4	<0.1	0.4	<0.3	1	<0.1	<0.1	0
1,2,3,4-Tetramethylbenzene	<0.1	<0.1	0.9	<0.1	<0.1	<0.3	<0.1	<0.1	<0.1	0
Naphthalene	<0.1	<0.1	1	<0.1	2	2	2	<0.1	1	3
Benzothiophene	<0.2	<0.1	<0.1	<0.2	<0.1	<0.4	<0.2	<0.1	<0.2	<0
2-Methylnaphthalene	0.7	0.7	3	0.6	2	<0.3	4	<0.3	4	6
1-Methylnaphthalene	<0.1	0.2	0.9	<0.1	0.8	<0.2	0.9	<0.1	2	3
Biphenyl	<0.1	<0.1	0.7	<0.1	0.6	<0.3	0.6	<0.1	1	2
2,6-Dimethylnaphthalene	<0.1	<0.1	2	<0.1	1	<0.3	2	<0.1	2	3
2,3,5-Trimethylnaphthalene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.3	<0.1	<0.1	<0.1	2
Fluorene	<0.1	0.6	1	<0.1	2	<0.3	0.8	<0.1	0.4	2
Dibenzothiophene	<0.3	<0.2	1	0.2	<0.2	<0.6	<0.2	<0.2	<0.2	2
Phenanthrene	0.6	4	10	0.9	5	<0.3	6	<0.1	8	8
Anthracene	2	0.5	3	0.8	2	<0.3	0.5	<0.1	<0.1	0
1-Methylphenenthrene	<0.1	<0.1	2	<0.1	0.7	<0.3	0.4	<0.1	2	2
Fluoranthene	0.8	8	12	2	7	<0.3	7	1	1	8
Pyrene	1	6	13	2	4	<0.3	5	2	2	5
Benz[a]anthracene	<0.3	4	9	1	3	<0.7	3	<0.3	<0.3	2
Chrysene	<0.2	4	6	0.8	2	<0.4	2	<0.1	2	2
Benzo[e]pyrene	<0.2	1	3	0.8	0.7	<0.4	1	<0.2	0.8	1
Benzo[a]pyrene	<0.2	0.9	4	1	0.5	<0.4	1	<0.1	2	1
Perylene	<0.2	0.9	1	2	0.8	<0.5	0.6	<0.2	3	4

Table 4. Hydrocarbons in sediment samples from ten sites in the Strait of Juan de Fuca and Northern Puget Sound collected during September-November 1978 (ng/g dry weight; ppb).

Alkanes	Birch Point	Cherry Point	March Point	NAS Whidbey Island	Andrews Bay	Dungeness Spit	Ediz Hook	Tongue Point	Kydaka Point	Baadah Point
n-C_{10}	0.9	0.7	3	0.3	5	2	0.9	0.8	6	4
n-C_{11}	0.7	0.9	4	0.6	6	2	2	0.7	8	4
n-C_{12}	0.8	1	4	0.9	6	2	5	0.8	8	5
n-C_{13}	1	2	10	1	7	3	10	1	10	6
n-C_{14}	2	2	10	1	7	3	20	2	10	7
n-C_{15}	20	20	40	2	10	3	30	4	10	10
n-C_{16}	3	4	40	2	8	4	30	3	10	9
n-C_{17}	20	20	70	5	70	4	40	6	20	60
Pristane	3	4	100	0.9	6	2	60	6	20	10
n-C_{18}	4	4	40	1	8	4	40	3	10	9
Phytane	1	2	10	0.6	3	1	40	2	5	5
n-C_{19}	5	5	50	2	8	3	40	4	20	10
n-C_{20}	2	4	30	1	7	3	40	4	20	9
n-C_{21}	4	6	40	1	8	3	40	7	20	10
n-C_{22}	3	5	20	0.2	7	2	30	4	20	8
n-C_{23}	6	7	30	<0.2	7	2	20	9	20	9
n-C_{24}	0.6	2	10	<0.2	2	0.7	8	5	20	6
n-C_{25}	5	6	30	<0.2	3	0.8	7	20	20	7
n-C_{26}	<0.2	<0.2	10	<0.2	4	<0.1	3	4	20	4
n-C_{27}	8	7	70	<0.2	4	2	6	20	30	10
n-C_{28}	<0.2	<0.2	10	<0.2	<0.2	<0.1	10	3	20	4
n-C_{29}	8	8	50	<0.2	5	2	4	10	20	8
n-C_{30}	<0.3	0.8	9	<0.2	1	2	1	5	10	4
n-C_{31}	5	4	50	<0.2	5	0.4	20	10	40	10

Table 4 Cont.

Aromatic Hydrocarbons	Birch Point	Cherry Point	March Point	NAS Whidbey Island	Andrews Bay	Dungeness Spit	Ediz Hook	Tongue Point	Kydaka Point	Baadah Point
Isopropylbenzene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	1	<0.1	0.3	<0.1
n-Propylbenzene	0.2	1	1	<0.1	0.3	<0.1	0.7	1	0.6	<0.1
Indan	<0.1	<0.1	1	<0.1	<0.1	<0.1	1	<0.1	0.3	<0.1
1,2,3,4-Tetramethylbenzene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Naphthalene	<0.1	<0.1	7	<0.1	<0.1	<0.1	1	<0.1	1	2
Benzothiophene	<0.1	<0.2	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1	<0.2	<0.2
2-Methylnaphthalene	0.5	0.9	4	0.4	1	0.4	3	0.8	3	3
1-Methylnaphthalene	<0.1	0.3	3	<0.1	0.3	<0.1	0.7	0.3	2	2
Biphenyl	<0.1	<0.1	1	<0.1	0.3	<0.1	0.5	<0.1	1	1
2,6-Dimethylnaphthalene	<0.1	<0.1	3	<0.1	0.5	<0.1	1	<0.1	3	1
2,3,5-Trimethylnaphthalene	<0.1	<0.1	3	<0.1	<0.1	<0.1	<0.1	<0.1	2	<0.1
Fluorene	<0.1	0.7	5	<0.1	0.7	<0.1	0.5	<0.1	0.5	1
Dibenzothiophene	<0.2	<0.2	5	<0.2	<0.2	<0.2	<0.1	<0.2	<0.2	<0.2
Phenanthrene	1	7	49	2	2	2	3	1	8	7
Anthracene	<0.1	0.7	9	<0.1	1	<0.1	0.2	<0.1	<0.1	0.3
1-Methylphenenthrene	<0.1	0.4	9	<0.1	<0.1	<0.1	0.3	<0.1	2	1
Fluoranthene	1	14	121	2	4	1	3	3	3	9
Pyrene	1	11	98	1	2	1	3	2	2	5
Benz[a]anthracene	<0.3	9	97	<0.3	1	<0.3	0.9	1	<0.3	2
Chrysene	0.7	6	62	<0.1	0.8	<0.1	1	0.8	2	3
Benzo[e]pyrene	0.5	3	45	<0.2	<0.2	<0.2	0.7	1	<0.2	2
Benzo[a]pyrene	0.8	3	44	<0.1	<0.1	<0.1	<0.1	0.5	0.2	1
Perylene	1	1	22	<0.1	<0.2	<0.2	<0.1	2	2	3

Table 5. Hydrocarbons in sediment samples from ten sites in the Strait of Juan de Fuca and Northern Puget Sound collected during January-March 1979 (ng/g dry weight; ppb).

Alkanes	Birch Point	Cherry Point	March Point	NAS Whidbey Island	Andrews Bay	Dungeness Spit	Ediz Hook	Tongue Point	Kydaka Point	Baadah Point
n-C_9	0.8	<0.2	<0.2	1.4	0.2	2.4	1.0	<0.2	4.9	2.9
n-C_{10}	0.7	1.3	2.5	1.1	5.3	2.5	1.2	0.7	6.2	3.5
n-C_{11}	0.5	1.7	3.5	1.4	6.7	2.8	4.9	0.8	8.6	4.4
n-C_{12}	0.8	1.8	3.3	1.3	6.3	2.6	18	1.0	8.6	4.4
n-C_{13}	0.8	1.9	4.7	1.4	7.8	2.7	37	1.5	10	5.3
n-C_{14}	0.9	2.1	6.3	1.4	7.5	2.6	56	1.7	11	5.6
n-C_{15}	8.9	3.1	10	1.5	10	2.5	64	2.4	11	6.3
n-C_{16}	1.7	3.1	11	<0.2	<0.2	3.1	80	2.6	12	7.5
n-C_{17}	16	5.4	17	5.3	49	2.9	84	4.2	15	39
Pristane	3.1	3.2	12	<0.2	64	1.5	110	5.4	17	12
n-C_{18}	1.6	3.2	12	1.9	8.1	2.7	79	2.4	13	10
Phytane	6.5	1.3	7	<0.2	3.1	<0.2	72	1.6	3.5	5.3
n-C_{19}	3.1	4.4	14	2.3	9.0	2.8	64	3.7	14	11
n-C-$_{20}$	3.1	3.1	9	1.6	8.0	2.7	53	2.7	14	8.1
n-C-$_{21}$	2.7	4.7	11	2.7	9.3	2.9	50	5.6	17	11
n-C-$_{22}$	2.8	4.4	9.4	2.7	7.7	3.9	32	3.7	18	9.3
n-C-$_{23}$	5.5	8.0	12	4.3	8.0	6.6	25	6.9	23	12
n-C-$_{24}$	5.3	6.6	11	4.9	5.9	8.2	15	4.5	23	13
n-C-$_{25}$	12	14	18	6.8	7.8	11	15	12	28	16
n-C-$_{26}$	6.2	7.6	13	5.6	5.0	11	13	5.1	24	14
n-C-$_{27}$	14	19	24	7.7	9.7	11	24	13	33	17
n-C-$_{28}$	3.8	8.1	13	3.5	5.6	5.8	16	4.4	18	10
n-C-$_{29}$	8.0	13	19	4.9	8.0	6.5	21	12	23	12
n-C-$_{30}$	5.9	6.5	26	2.7	6.2	5.9	23	5.5	19	11
n-C-$_{31}$	7.5	14	17	5.1	9.1	6.7	12	11	19	<0.5
n-C-$_{32}$	<0.4	<0.6	<0.6	<0.5	<0.5	<0.4	<0.3	<0.5	4.8	<0.5

Table 5 Cont.

Aromatic Hydrocarbons	Birch Point	Cherry Point	March Point	NAS Whidbey Island	Andrews Bay	Dungeness Spit	Ediz Hook	Tongue Point	Kydaka Point	Baadah Point
o-Xylene	---[a]	---	---	---	---	---	---	---	---	---
Isopropylbenzene	<0.1	<0.1	<0.1	<0.1	<0.1	1	<0.1	<0.1	<0.1	<0.1
n-Propylbenzene	<0.2	<0.1	<0.2	<0.2	<0.1	0.4	<0.1	<0.1	<0.1	<0.1
Indan	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1	0.3	<0.1	<0.1	<0.1
1,2,3,4-Tetramethylbenzene	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Naphthalene	<0.1	1	1	<0.1	1	<0.1	2	<0.1	<0.1	<0.1
Benzothiophene	<0.2	<0.1	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
2-Methylnaphthalene	<0.1	0.7	2	<0.1	1	0.4	3	0.5	3	1
1-Methylnaphthalene	<0.1	0.2	0.5	<0.1	0.4	<0.1	0.8	<0.1	1	0.8
Biphenyl	<0.1	<0.1	0.5	<0.1	<0.1	<0.1	0.4	<0.1	0.9	0.5
2,6-Dimethylnaphthalene	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	2	<0.1	1	<0.1
2,3,5-Trimethylnaphthalene	<0.2	<0.1	<0.1	<0.2	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Fluorene	<0.1	0.7	0.8	<0.1	1	<0.1	0.7	<0.1	<0.1	0.5
Dibenzothiophene	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Phenanthrene	0.5	7	6	<0.2	4	1	5	1	6	2
Anthracene	<0.2	0.7	2	<0.2	0.6	<0.1	0.4	<0.1	<0.1	<0.1
1-Methylphenenthrene	<0.2	0.3	1	<0.2	<0.1	<0.1	0.9	<0.1	2	<0.1
Fluoranthene	1	10	20	<0.5	5	0.5	6	3	0.5	2
Pyrene	0.6	10	20	<0.2	3	<0.1	4	2	2	1
Benz[a]anthracene	<0.3	8	10	<0.3	2	<0.3	2	<0.3	<0.3	<0.3
Chrysene	<0.2	6	10	<0.2	0.8	<0.2	2	<0.2	2	<0.2
Benzo[e]pyrene	<0.2	3	5	<0.2	<0.2	<0.2	0.7	<0.2	<0.2	<0.2
Benzo[a]pyrene	<0.2	3	6	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Perylene	<0.2	1	2	<0.2	<0.2	0.9	<0.2	1	0.8	1

[a] No data

Table 6. Sum of concentrations of the C_{10} to C_{31} n-alkanes, pristane, and phytane (Σ Selected Alkanes) in sediment samples collected during 1978-1979 (ng/g dry weight; ppb).

	Sampling date				
Site	6/78	9-11/78	1-3/79	5/79[1]	6/79[1]
Birch Point	110	100	120		
Cherry Point	150	120	130		
March Point	300	740	280		
Naval Air Station	80	20	70		
Andrews Bay	470	200	270		
Dungeness Spit	80	50	110		
Ediz Hook	580	510	960	5,700	2,600
Tongue Point	140	130	110		
Kydaka Point	350	400	380		
Baadah Point	410	230	240		

1 Sediments collected at Ediz Hook sampling site as part of the oil spill investigation resulting from spill on 5/13/79.

NOTE: Blank space indicates no samples taken.

Table 7. Sum of concentrations of selected aromatics (ΣSelected Aromatics) in sediment samples collected during 1978-1979 (ng/g dry weight; ppb).

Site	Sampling date				
	6/78	9-11/78	1-3/79	5/79[1]	6/79[1]
Birch Point	4.4	6.7	2.1		
Cherry Point	37	58	52		
March Point	75	589	87		
Naval Air Station	12	5.4	0.5		
Andrews Bay	36	14	19		
Dungeness Spit	2	4.4	4.2		
Ediz Hook	38	22	30	65	ND[2]
Tongue Point	3.3	13	7.5		
Kydaka Point	32	33	19		
Baadah Point	59	43	8.8		

1 Sediments collected at Ediz Hook sampling site as part of the oil spill investigation resulting from spill on 5/13/79.

2 ND means there were no measurable quantities.

NOTE: Blank space indicates no samples taken.

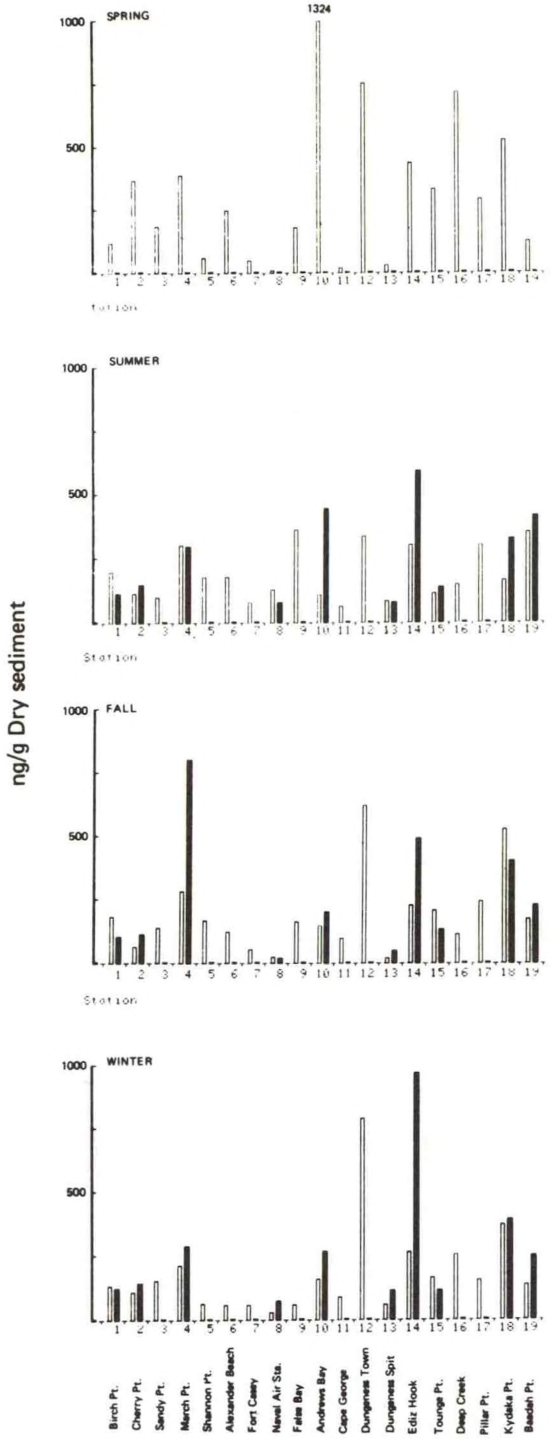

Figure 4. Sums of concentrations of n-alkanes C_{10}-C_{31}, pristane, and phytane from sediment samples. Open bars are for samples collected from spring 1977 through winter 1978 (phase 2) and solid bars are for samples collected from summer 1978 through winter 1979 (phase 3).

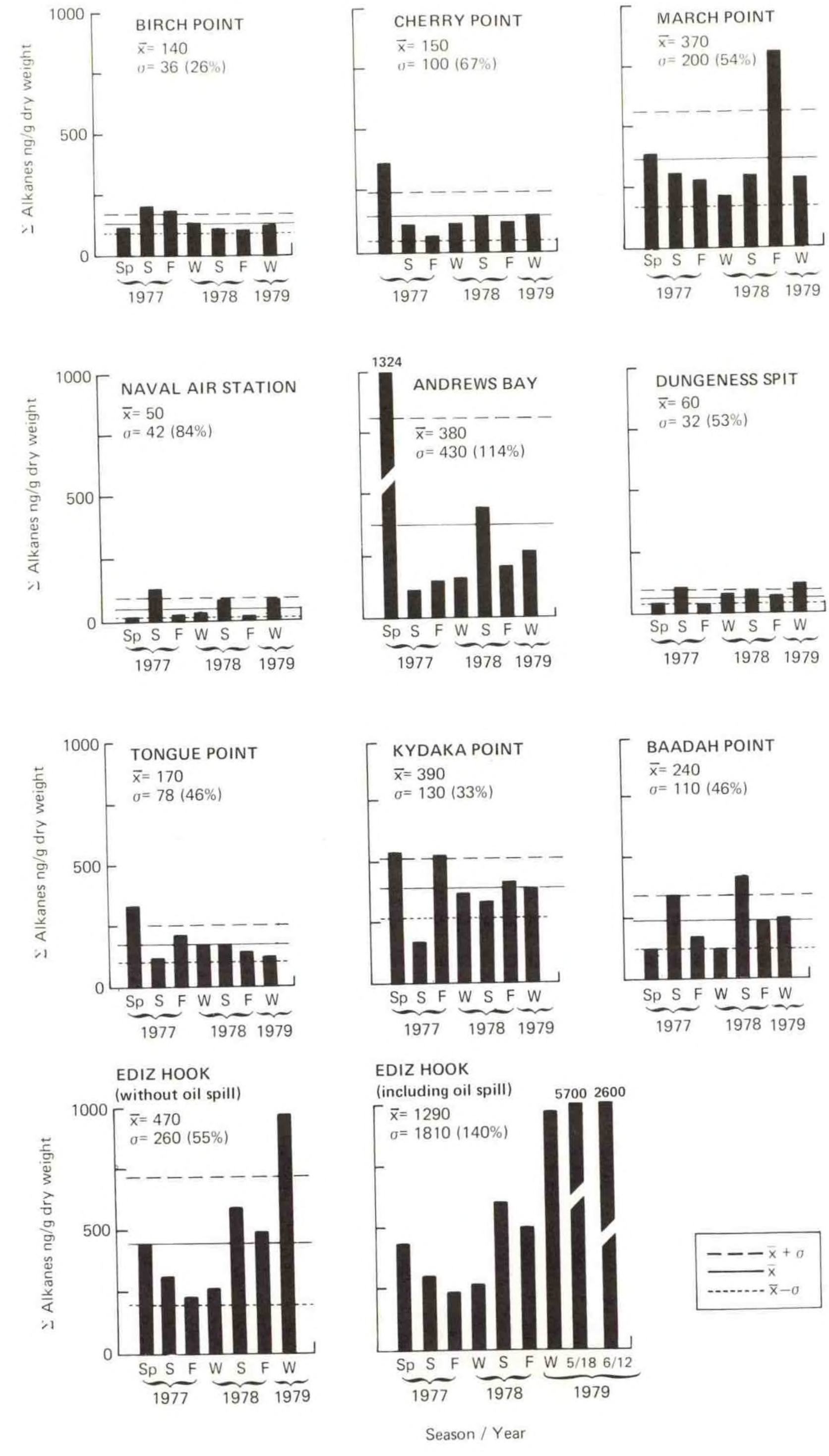

Figure 5. Sums of concentrations of n-alkanes C_{10}-C_{31}, pristane, and phytane from sediment samples (dry weight) collected at ten sites seven times (Sp = spring, S = summer, F = fall, W = winter). A second graph for the alkanes in sediment from the Ediz Hook site includes the data for the two samplings following the May 1979 oil spill.

Table 8. Total organic carbon and sand-to-mud ratio for sediment samples collected during 1978-1979.

Site	Total organic carbon (%)			Sand/Mud ratio		
	6/78	9-11/78	1-3/79	6/78	9-11/78	1-3/79
Birch Point	0.18	0.31	0.09	5.5	6.9	2.3
Cherry Point	0.14	0.10	0.19	19.1	13.4	172
March Point	0.17	0.36	0.26	10.4	8.8	52.6
Naval Air Station	0.35	0.41	0.07	35.1	112	140
Andrews Bay	0.18	0.10	0.08	54.2	40.2	92.6
Dungeness Spit	0.07	0.13	0.08	36.2	146	129
Ediz Hook	0.24	0.13	0.17	25.1	32.5	116
Tongue Point	0.17	0.17	0.26	42.6	7.8	139
Kydaka Point	0.07	0.08	0.10	79.3	31.4	130
Baadah Point	0.12	0.10	0.18	18.4	21.6	169

Table 9. Hydrocarbons in sediment and mussel samples from Port Angeles Harbor collected on 18 May and June 1979 (ng/g dry weight; ppb).

Alkanes	Boat Ramp Sediment 5-18	Boat Ramp Sediment 6-12	Ediz Hook Sediment 5-18 (Baseline site)	Ediz Hook Sediment 6-12 (Baseline site)	Ediz Hook Mussel 5-18 (Baseline site)	Ediz Hook Mussel 6-12 (Baseline site)	Peabody Creek Sediment 5-18	Peabody Creek Sediment 6-12	Log Dump Sediment 5-18	Log Dump Sediment 6-12	Boat Ramp Fine Sediment 6-12
n-C_9	4	<30	4	3	<6	<6	5	8	2	2	4
n-C_{10}	10	30	5	4	<5	70	20	20	2	2	10
n-C_{11}	120	1,400	10	6	390	110	20	20	3	3	10
n-C_{12}	520	6,400	40	20	2,500	150	30	30	2	9	30
n-C_{13}	1,600	20,000	150	50	6,800	340	50	40	3	10	100
n-C_{14}	3,200	40,000	340	110	7,100	50	90	50	5	10	230
n-C_{15}	3,600	60,000	540	190	7,100	560	90	60	8	9	190
n-C_{16}	5,900	71,000	680	270	6,700	1,800	100	80	10	8	250
n-C_{17}	5,200	62,000	610	250	4,600	340	120	100	20	8	210
Pristane	3,200	53,000	360	170	8,400	2,500	140	60	20	20	190
n-C_{18}	4,700	54,000	590	260	4,700	160	100	70	30	10	190
Phytane	2,100	33,000	250	130	4,000	980	110	50	20	7	100
n-C_{19}	3,700	43,000	480	220	4,400	20	80	60	40	9	150
n-C_{20}	3,100	36,000	420	200	3,400	60	110	60	50	10	130
n-C_{21}	2,300	26,000	320	150	3,200	220	110	130	40	10	270
n-C_{22}	1,600	18,000	230	120	2,300	170	30	80	30	10	60
n-C_{23}	1,200	11,000	170	90	1,600	140	140	60	30	20	70
n-C_{24}	690	7,000	110	60	90	180	110	50	20	20	50
n-C_{25}	520	5,000	90	50	760	130	240	110	20	20	50
n-C_{26}	340	3,600	50	30	510	90	170	50	10	10	40
n-C_{27}	350	3,100	70	50	460	120	590	100	20	30	40
n-C_{28}	190	1,900	40	30	260	100	360	110	10	20	40
n-C_{29}	110	1,300	30	20	180	160	460	90	20	50	30
n-C_{30}	360	1,100	70	70	250	170	1,100	490	40	50	140
n-C_{31}	220	1,000	40	20	140	150	440	100	20	20	50
n-C_{32}	190	<55	20	30	70	120	390	120	40	50	5

Table 9. Cont.

Aromatics	Boat Ramp Sediment 5-18	Boat Ramp Sediment 6-12	Ediz Hook Sediment 5-18 (Baseline site)	Ediz Hook Sediment 6-12 (Baseline site)	Ediz Hook Mussel 5-18 (Baseline site)	Ediz Hook Mussel 6-12 (Baseline site)	Peabody Creek Sediment 5-18	Peabody Creek Sediment 6-12	Log Dump Sediment 5-18	Log Dump Sediment 6-12	Boat Ramp Fine Sediment 6-12
o-Xylene	5	<2	<0.3	<0.3	1,700	90	1	6	2	<0.4	<0.3
Isopropylbenzene	1	<2	<0.4	<0.3	70	<10	<0.3	<0.2	<0.2	<0.4	<0.3
n-Propylbenzene	<0.2	<2	<0.4	<0.4	<5	<11	<0.4	<0.3	<0.3	<0.4	<0.4
Indan	0.9	<2	<0.3	<0.3	9	<11	<0.3	<0.3	<0.2	<0.4	<0.3
1,2,3,4-Tetramethylbenzene	1	<2	<0.3	<0.3	<5	<10	<0.3	<0.3	<0.2	<0.4	<0.3
Naphthalene	5	40	<0.3	<0.3	<5	<10	50	<0.3	<0.2	<0.4	710
Benzothiophene	2	<0.2	<0.6	<0.6	<6	<13	<0.5	<0.4	<0.4	<0.6	50
2-Methylnaphthalene	70	200	<0.4	<0.4	70	<10	30	<0.3	<0.3	<0.4	1,500
1-Methylnaphthalene	3-	140	<0.3	<0.3	60	<8	10	<0.2	<0.2	<0.3	470
Biphenyl	2-	70	<0.3	<0.3	340	<10	6	<0.3	<0.2	<0.4	440
2,6-Dimethylnaphthalene	110	850	7	<0.3	3,000	<9	10	<0.3	<0.2	<0.4	360
2,3,5-Trimethylnaphthalene	150	1,100	10	<0.4	8,100	<10	9	<0.3	<0.3	<0.4	70
Fluorene	20	100	2	<0.3	410	<10	90	<0.3	0.1	20	2,700
Dibenzothiophene	40	240	1	<0.7	5,300	<11	50	<0.5	<0.5	<0.8	1,000
Phenanthrene	40	440	20	<0.4	3,200	240	480	<0.3	20	50	4,300
Anthracene	7	60	<0.4	<0.4	1,000	110	50	<0.3	<0.3	<0.5	570
1-Methylphenenthrene	10	100	<0.4	<0.4	1,000	<10	9	<0.3	0.3	<0.4	30
Fluoranthene	20	20	9	<0.4	160	610	550	<0.3	30	<0.5	2,000
Pyrene	10	30	8	<0.4	540	220	270	<0.3	30	20	1,100
Benz[a]anthracene	6	10	4	<1	750	200	130	<0.8	8	30	1,300
Chrysene	10	50	4	<0.6	900	<12	90	<0.4	6	10	740
Benz[e]pyrene	6	30	<0.6	<0.6	310	<13	40	1	5	8	270
Benz[a]pyrene	2	18	<0.6	<0.6	230	140	20	<0.5	<0.6	6	190
Perylene	2	6	<0.8	<0.8	70	<17	30	<0.6	<0.5	<0.8	90

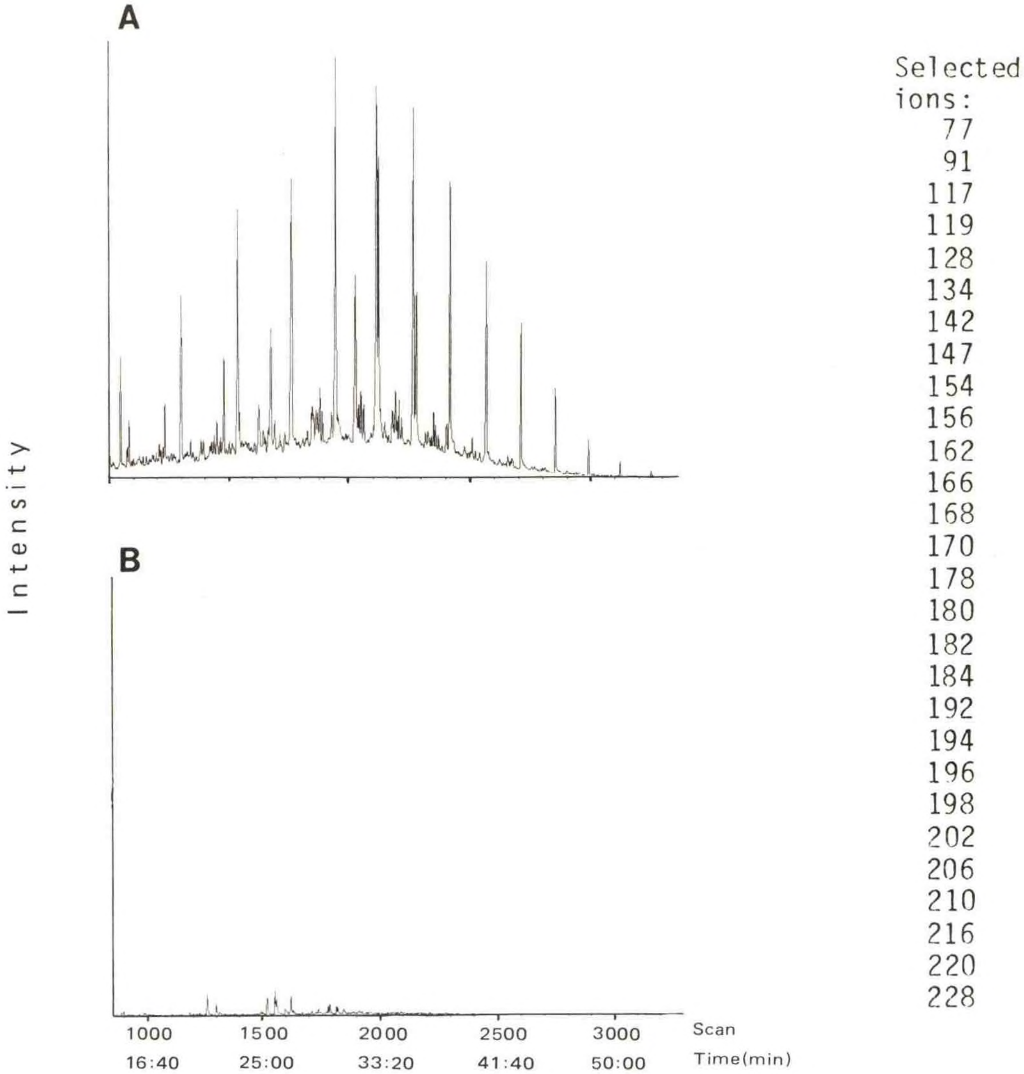

Figure 6. Number 2 fuel oil: (A) total ion current chromatogram of saturated hydrocarbon fraction and (B) selected ion current chromatogram of aromatic fraction. Hewlett-Packard-5840A GC, 30 m x 0.25 i.d. SE-54 glass capillary column, interfaced to Finnigan-3200 mass spectrometer with a NOVA 3 data system. Helium carrier pressure 12 psi, 2 μl splitless injection, split valve opened after 18 sec., $50^{o}C$ initial temp. for 5 min., then $4^{o}C$ per min. to $280^{o}C$. Numbered peaks refer to n-alkanes of given chain length.

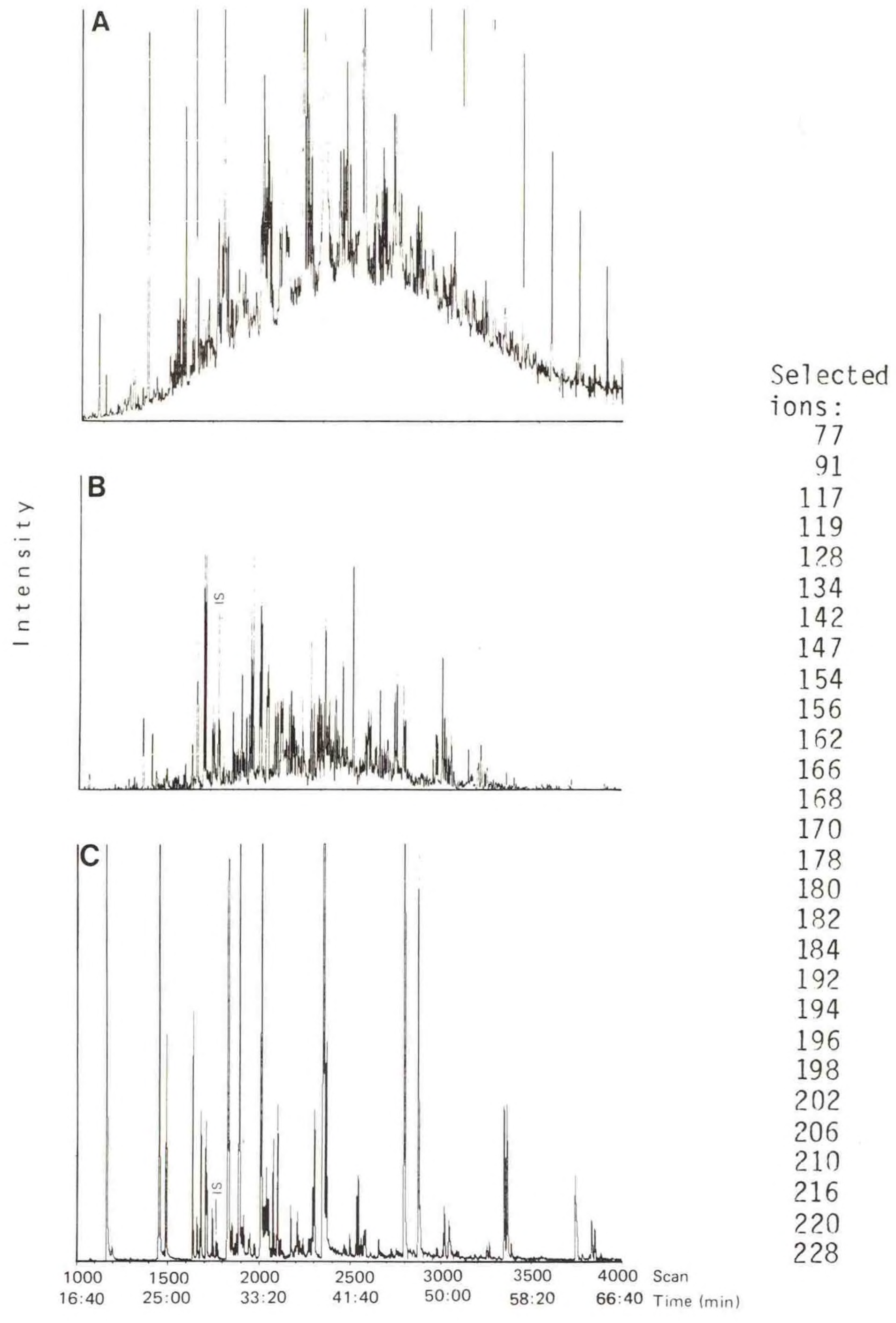

Figure 7. Sediment from the boat ramp in Port Angeles Harbor: (A) and (B) from coarse sediment near ramp and (C) from fine sediment adjacent to the ramp. A: Total ion current chromatogram of the saturated hydrocarbon fraction collected 18 May 1979; B and C: Selected ion current chromatograms of aromatic hydrocarbon fractions collected 18 May 1979 and 12 June 1979, respectively. Conditions same as for Figure 6.

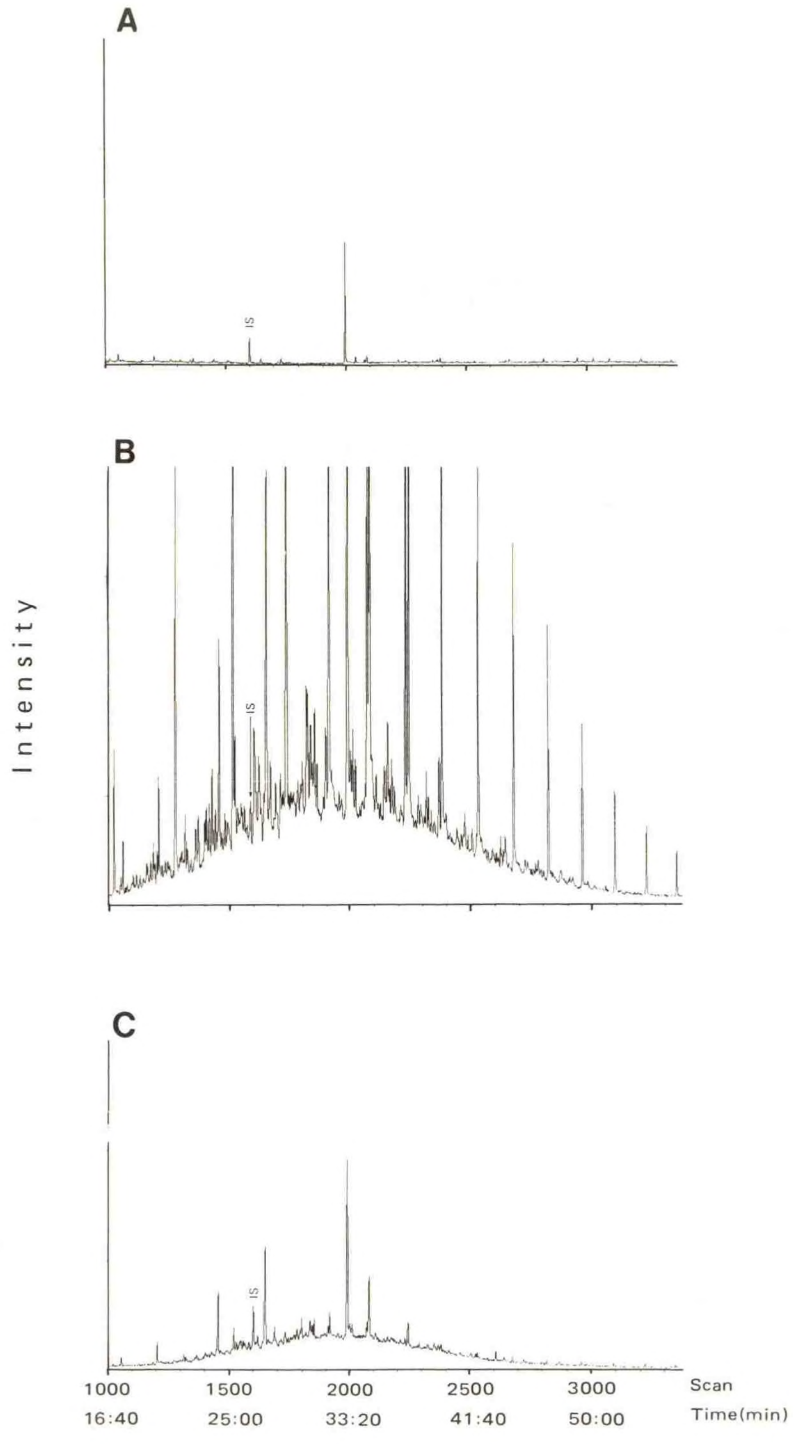

Figure 8. Mussel samples from the Ediz Hook site: Total ion current chromatograms of saturated hydrocarbon fraction collected (A) 28 October 1977, (B) 18 May 1979, and (C) 12 June 1979. Conditions same as for Figure 6.

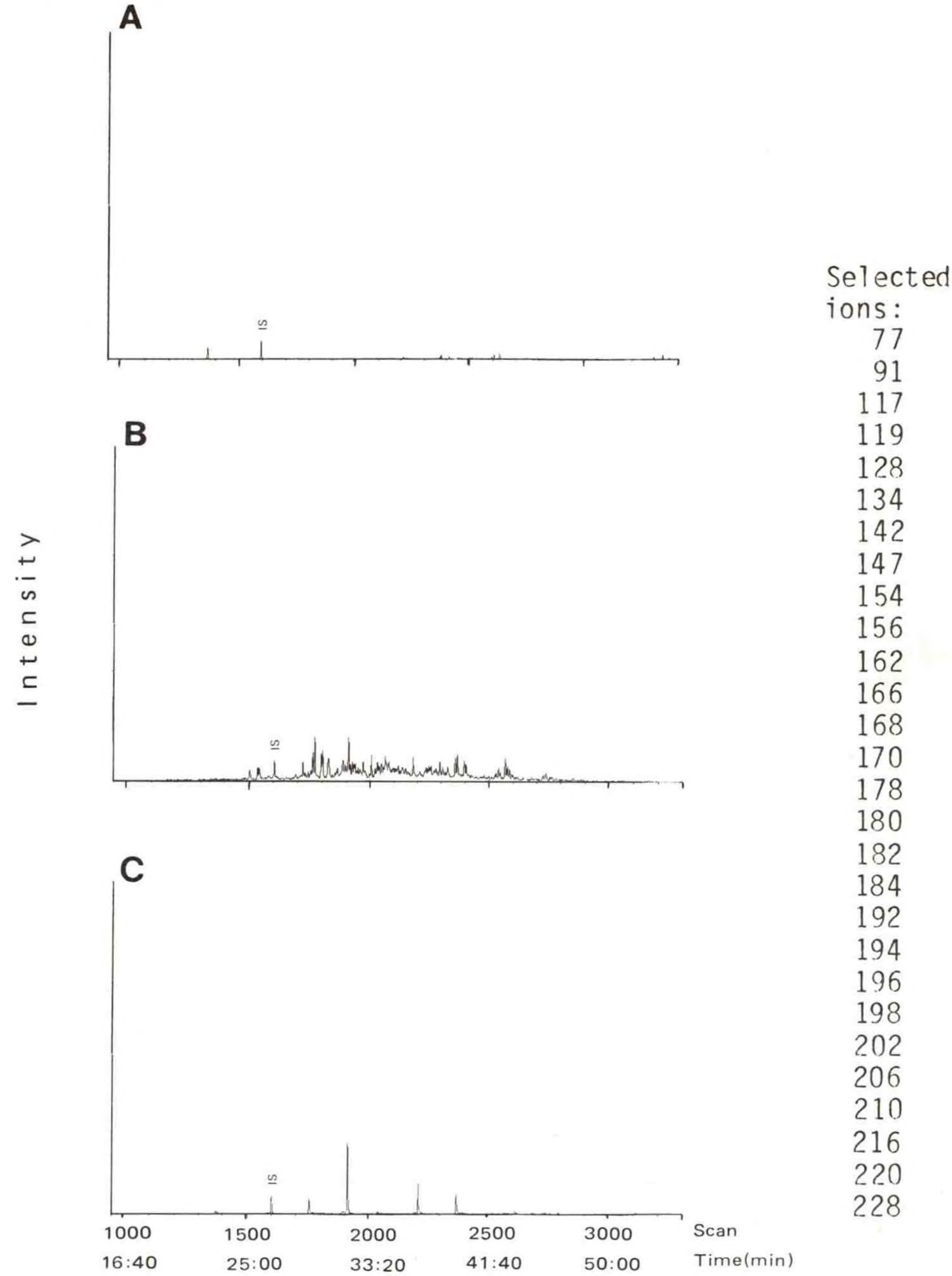

Figure 9. Mussel samples from the Ediz Hook site: Selected ion current chromatograms of aromatic hydrocarbon fraction of mussels collected (A) 28 October 1977, (B) 18 May 1979, and (C) 12 June 1979. Conditions same as for Figure 6.

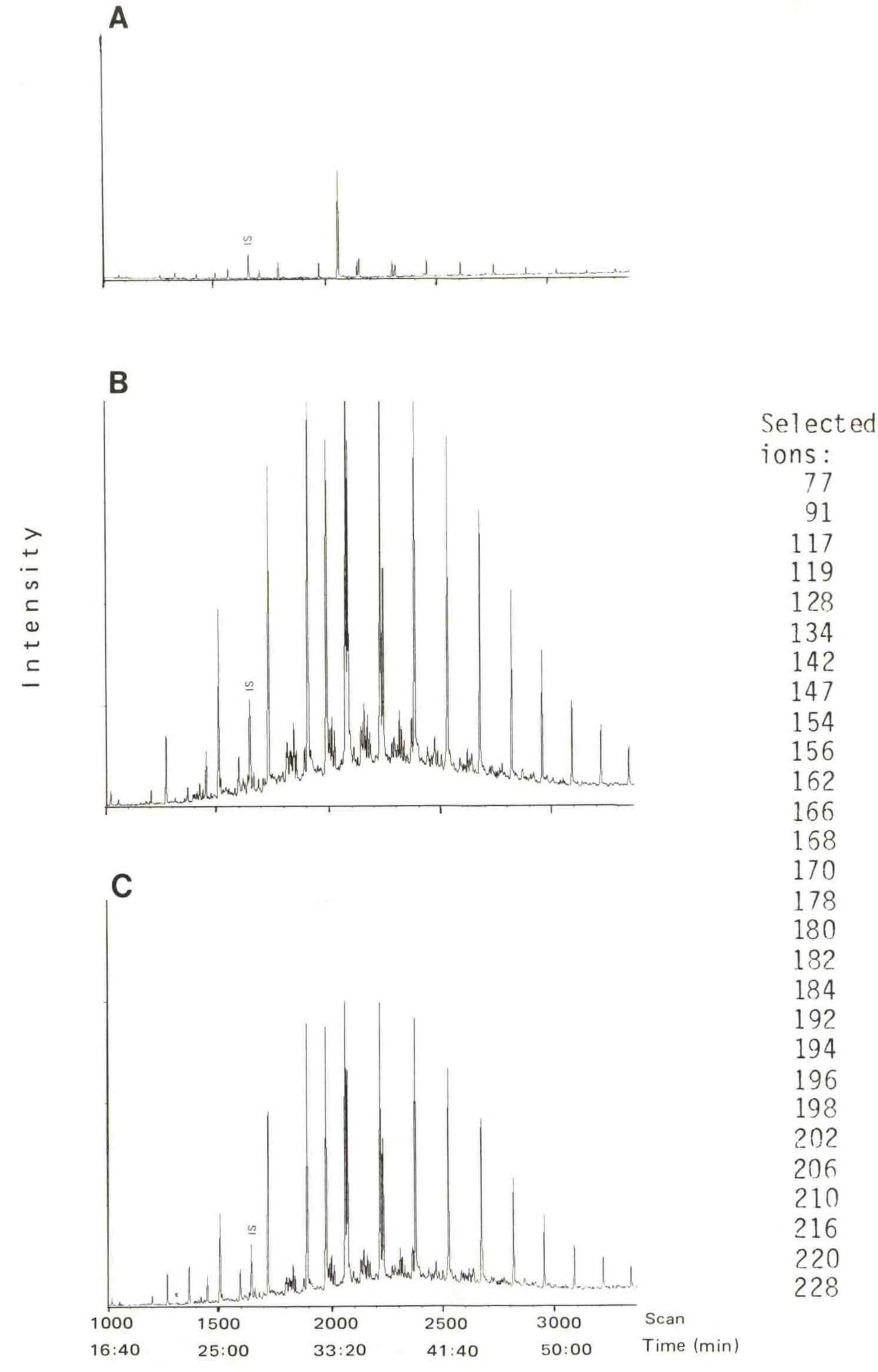

Figure 10. Sediment from the Ediz Hook site: Total ion current chromatogram of the saturated hydrocarbon fraction of sediment collected (A) 28 October 1977, (B) 18 May 1979 - five days after oil spill, and (C) 12 June 1979 - 30 days after spill. Conditions same as for Figure 6.

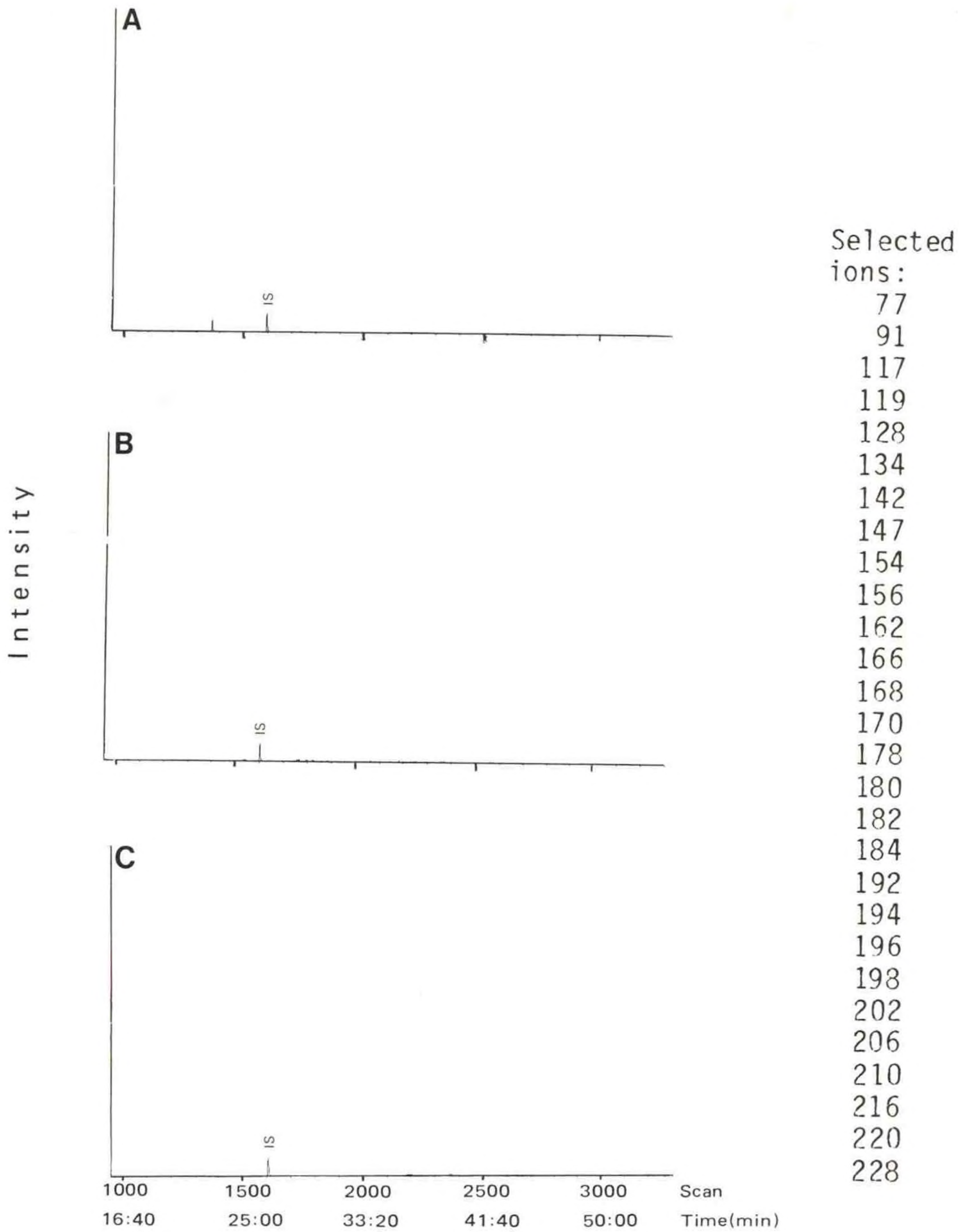

Figure 11. Sediment from Ediz Hook site: Selected ion current chromatogram of aromatic hydrocarbon fraction of sediment collected (A) 28 October 1977, (B) 18 May 1979, and (C) 12 June 1979. Conditions same as for Figure 6.

Table 10. Sums of the concentrations of C_{10} to C_{31} n-alkanes and pristane and phytane (ΣSelected Alkanes) and sums of concentrations of selected aromatic hydrocarbons (ΣSelected Aromatics) in sediment and mussel samples collected in Port Angeles Harbor, Washington, following a small oil spill on 13 May 1979 (ng/g dry weight).

	ΣSelected Alkanes Sampling date		ΣSelected Aromatics Sampling date	
Site	18 May	12 June	18 May	12 June
Peabody Creek, sediment	5,200	2,200	1,900	<10
Boat Ramp, sediment	45,000	560,000	570	3,500
Boat Ramp, fine sediment		2,600		18,000
Log Dump, sediment	510	430	100	110
Ediz Hook site, sediment	5,700	2,600	60	<10
Ediz Hook site, mussel	71,000	8,900	27,000	1,500

NOTE: Blank space indicates no samples were taken.

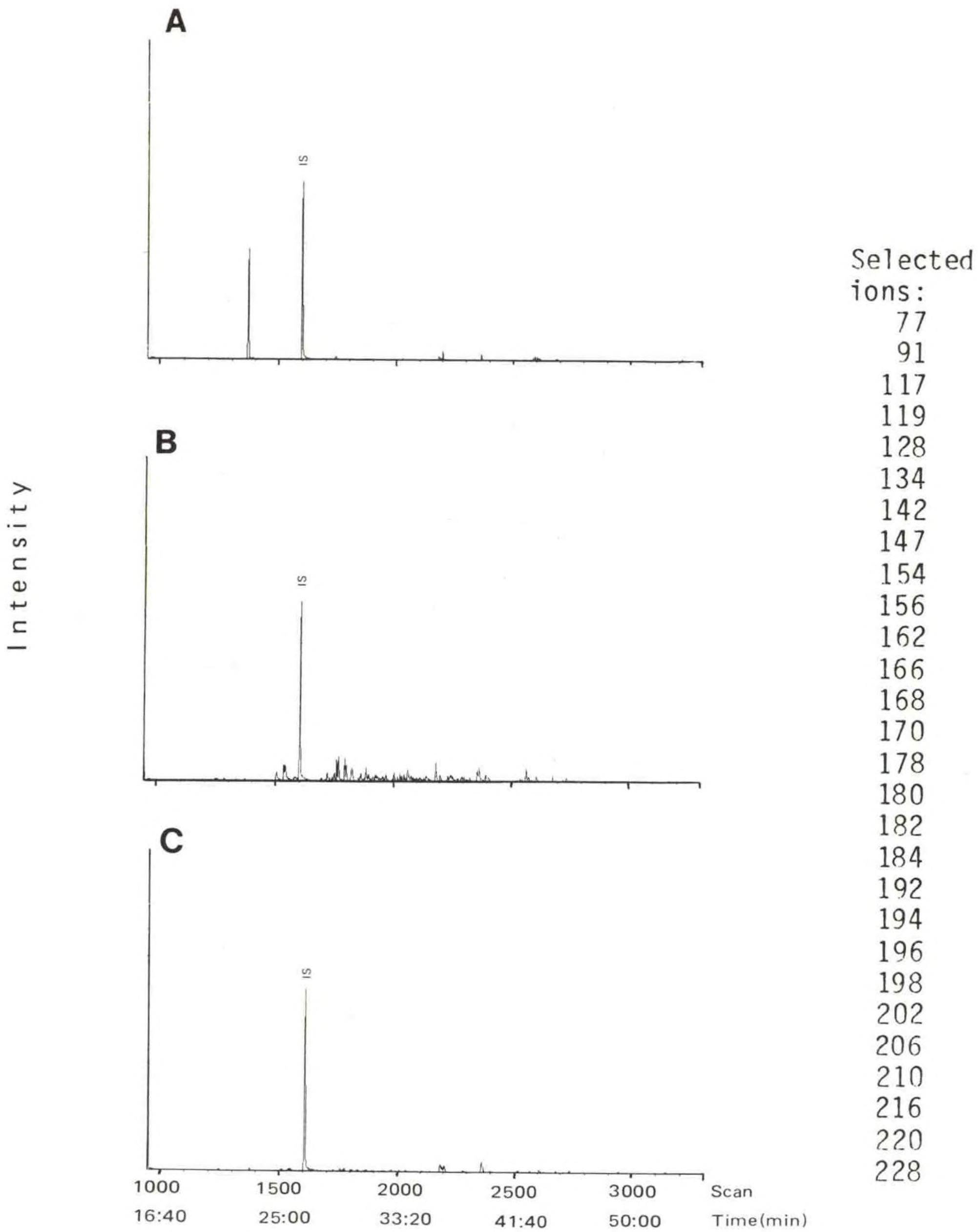

Figure 12. Sediment from Ediz Hook site: Selected ion current chromatograms of the aromatic hydrocarbon fraction from sediment collected (A) 28 October 1977, (B) 18 May 1979, (C) 12 June 1979. The relative intensity was increased, using the computer, as compared to chromatograms in Figure 11. Conditions same as for Figure 6.

DISCUSSION

Hydrocarbon Baseline Study Continuation

As a continuation of the baseline study, sediment samples were collected three times (summer and fall 1978, and winter 1979) from ten stations (Fig. 1) and analyzed for hydrocarbons. Thus, for the two-year period of the entire study, seven sets of data were collected for each station, providing essential information pertaining to temporal variations of hydrocarbons in sediments.

The sums of the concentrations of selected alkanes (ΣSelected Alkanes) at these ten sites (Figs. 4 and 5) show a considerable range of the mean values for the 7 quarterly samplings (50 to 470 ng/g dry weight, Fig. 5). The highest means for ΣSelected Alkanes were measured at Ediz Hook (470 ng/g), Kydaka Point (390 ng/g), Andrews Bay (380 ng/g), and March Point (370 ng/g). The mean of the mean ΣSelected Alkanes for all ten stations was 240 ng/g. The lowest means in sediment were found at the Naval Air Station (50 ng/g) and Dungeness Spit (60 ng/g) sites. The Cherry Point sampling site, although located near two oil refineries, was among the lowest in hydrocarbon concentrations.

The differences among ΣSelected Alkanes at any particular site over the seven samplings were not unusual except for those from March Point (fall 1978), Ediz Hook (winter 1979) and Andrews Bay (spring 1977) (Fig. 5). The ΣSelected Alkanes at these times were considerably greater than the means plus the standard deviations. This suggests that the three sites may have been exposed to non-biogenic sources of hydrocarbons (e.g., oil) prior to the sampling. It is also important to note that by the next quarterly sampling at March Point and Andrews Bay the ΣSelected Alkanes had decreased substantially toward the mean for each site. This suggests that samples may have to be collected more often than quarterly to detect exposures of sediment to small oil inputs, although quarterly sampling may indeed be adequate to monitor for larger or continuous perturbations.

Oil Spill

Generally, the pattern of hydrocarbons in the sediment from the boat ramp site on 18 May and 12 June were the same, though the concentrations differed. The same was true for the sediment samples from the Ediz Hook site. Although the sample collected at the boat ramp 18 May contained elevated levels of alkanes and aromatic hydrocarbons, the sample collected on 12 June contained levels about ten times higher. This may have resulted from the oil having been incorporated into sediment above the sampling elevation when it was boomed and collected there and later having seeped into the lower sediments collected.

Using the ΣSelected Alkanes as an indication of the amount of oil present, the sediment from the boat ramp site on 18 May would appear to have been exposed to roughly ten times the amount of oil than the sediment from the Ediz Hook site.

The alkane patterns from the fine-grain sediment collected beside the boat ramp (Fig. 7C) on 12 June (a few meters from the other boat ramp sampling site) indicate that the sediments probably were exposed to the spilled fuel oil. However, the GC patterns (Fig. 7C) and the relative concentrations for the aromatic hydrocarbons (Table 9) showed that a No. 2 fuel oil was not the only source of the hydrocarbons found.

The sediment extracts from the log boom area and Peabody Creek did not contain hydrocarbons in the same relative ratios as did the fuel oil. The Peabody Creek sediment collected 18 May contained elevated levels of aromatic hydrocarbons, which is consistent with what has previously been reported for this area (2). However, the Peabody Creek sediment collected 12 June did not contain measurable quantities of aromatic hydrocarbons. It was observed before that the stream bed is narrow and shifting, therefore samples collected at apparently the same location may show large differences in hydrocarbon concentrations from time to time (2). This may explain the differences in the above analyses.

Although the mussel sample collected at Ediz Hook 18 May contained a high concentration of hydrocarbons (ΣSelected Alkanes was 71,000 ng/g), the sample collected 12 June contained low concentrations of alkane and aromatic hydrocarbons, similar to those obtained from mussel samples collected before the oil spill (3) (Figs. 8 and 9). Apparently the mussels readily took up the petroleum hydrocarbons (by 18 May), but either rapidly depurated them (by 12 June), and/or converted them to compounds that were not detected by our techniques. The Ediz Hook sediment absorbed hydrocarbons from the spilled oil and retained them longer than did the mussels, in agreement with results of others (5,6,7,8,9).

SUMMARY

Completion of the baseline study, covering a two-year period, has resulted in seven sets of data on the hydrocarbon concentrations in sediment from ten stations in Northern Puget Sound and the Strait of Juan de Fuca. The levels of n-alkanes, pristane, phytane, and aromatic hydrocarbons in the sediment and mussels at each sampling site were generally low, indicating that the area is relatively free from petroleum hydrocarbon contamination. However, our data also suggest that petroleum entered the marine environment at Ediz Hook before the winter of 1978-1979 and again before 18 May 1979, at Andrews Bay before the spring of 1977, and at March Point before the fall of 1978. Otherwise, the hydrocarbon levels at each site maintained a consistent pattern over the two-year period of seven samplings.

The value of baseline data was demonstrated in the investigation of a minor oil spill in Port Angeles Harbor in the area of the Ediz Hook site. The rise and fall in hydrocarbon concentrations in sediment and mussels from baseline levels was consistent with the known intrusion of oil. Sediments took up hydrocarbons more slowly and retained them for more than a month. In contrast, nearby mussels took up hydrocarbons more readily but levels returned to normal in less than a month (through elimination or metabolic conversion).

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