

Hydrocarbons in Oil Residues on Beaches of Islands of Prince William Sound, Alaska

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Aliphatic and aromatic hydrocarbons were measured on oil residues from beaches on six islands in Prince William Sound, Alaska. In addition to altered products from the *Exxon Valdez* oil spill of 1989, we also found, at two widely separated locations, residues that are similar to each other but chemically distinct from the spilled oil. Terpanes, steranes, monoaromatic steranes, and carbon isotopic compositions of total extracts were most useful in correlating the altered products of the spilled oil. These same parameters revealed that the two non-Valdez samples are likely residues of oil originally produced in California. The results indicate that oil residues currently on the beaches of this estuary have at least two quite different origins.

A major oil spill, the largest in U.S.A. history, occurred when the fully loaded supertanker *Exxon Valdez* struck Bligh Reef on 24 March 1989, in northern Prince William Sound, a complex fjord system on the south coast of Alaska. The spill released about 42 000 m³ (11 million US gal.) of North Slope crude oil into the water, where currents and wind quickly spread the oil through the western part of the sound and southwestward along the coast of the Gulf of Alaska. Along the trajectory of the spill (Fig. 1), crude oil coated the sand, gravel, cobbles, and rocks on the shores of the Sound. Massive efforts were undertaken to clean up the oiled beaches and remove oil from the water (Kelso & Kendziorek, 1991).

About 50 days after the oil spill, the US Geological Survey began a series of investigations to determine the geologic and geochemical fate of the spilled oil (Carlson & Reimnitz, 1990; Carlson, 1991). As a part of these investigations, geochemical analyses were run on eight samples of oil residues (Table 1); these samples were collected about 17 months (August 1990) after the spill from isolated, uninhabited shorelines of six islands—Elrington, Knight, Eleanor, Smith, Naked, and Storey (Fig. 1)—that had previously been affected to various degrees by the spilled oil. At the time of sample collection, the beaches appeared to be remarkably free of obvious oil contamination but closer inspection

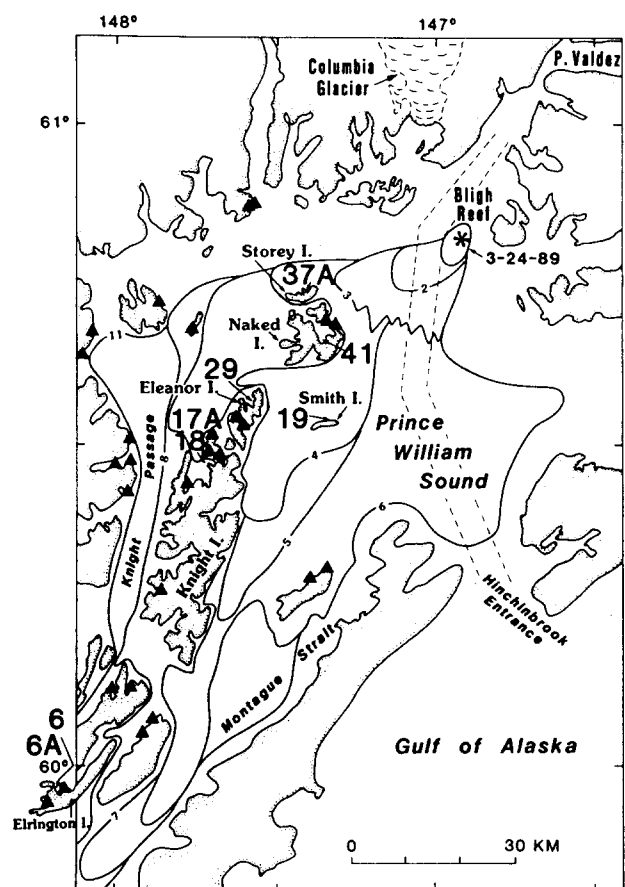


Fig. 1 Prince William Sound, Alaska, showing locations of shoreline sites (bold numbers) where oil residues were collected in August 1990 for geochemical studies of the *Exxon Valdez* oil spill. Triangles (▲) indicate locations on non-*Exxon Valdez* residues observed by the Exxon Co., USA (1990). Numbered lines indicate locations of the spill front and number of days after the oil spill occurred on 24 March 1989, at Bligh Reef.

generally revealed some oil-like coatings on sediment and rock.

Methods

The procedures used for geochemical analyses were described previously by Kvenvolden *et al.* (1991). Briefly, the oil residues, as well as 30 mg of North Slope crude oil impounded after the oil spill from the supertanker *Exxon Valdez* and referred to here as *Exxon*

TABLE 1

Molecular and carbon-isotopic parameters of oil-like residues collected in August 1990, Prince William Sound, Alaska, and of an oil sample from the supertanker *Exxon Valdez*.

Sample	Description and location	Pr/Ph	m/z 191				m/z 217			BNO	$\delta^{13}\text{C}$ (‰)
			$\text{C}_{23}/\text{C}_{30}$	$\text{C}_{30}/\text{C}_{29}$	$\text{C}_{31} \frac{\text{S}}{\text{S}+\text{R}}$	Triplet	$\text{C}_{29} \frac{\text{S}}{\text{S}+\text{R}}$	$\frac{\text{D}(27)}{\text{St}(29)}$	$\text{C}_{28}/\text{C}_{29}$		
EVO	<i>Exxon Valdez</i> spill oil, North Slope crude oil	1.4	0.71	1.4	0.60	2.0	0.45	2.4	0.63	—	−29.2
EVR 6	Oily beach sediment, Elrington Island	1.2	1.0	1.4	0.60	2.0	0.48	2.6	0.67	—	nd
17A	Solidified oil on cobbles, Knight Island	0.69	0.83	1.4	0.59	2.0	0.53	2.0	0.50	—	nd
18	Oily beach sand, Knight Island	1.0	0.59	1.3	0.59	2.0	0.49	1.7	0.46	—	−29.2
19	Oil coated cobbles, Smith Island	0.68	0.86	1.6	0.60	2.2	0.48	1.6	0.61	—	nd
29	Solidified oil on rock, Eleanor Island	0.26	1.2	1.4	0.61	2.0	0.45	2.2	0.68	—	−29.1
41	Oily cobbles, Naked Island	0.75	1.0	1.4	0.62	2.2	0.48	2.1	0.61	—	nd
NEVR 6A	Tar on rocks, Elrington Island	2.5	0.17	1.9	0.56	4.3	0.30	0.65	1.0	+	−23.8
37A	Tar on rocks, Storey Island	1.2	0.17	1.8	0.57	5.1	0.32	0.57	1.0	+	−23.9

Pr/Ph, pristane (2,6,10,14-tetramethylpentadecane)/phytane (2,6,10,14-tetramethylhexadecane); $\text{C}_{23}/\text{C}_{30}$, C_{23} -tricyclic terpene/ $17\alpha(\text{H}), 21\beta(\text{H})$ -hopane; $\text{C}_{30}/\text{C}_{29}$, $17\alpha(\text{H}), 21\beta(\text{H})$ -hopane/ $17\alpha(\text{H}), 21\beta(\text{H})$ -30-norhopane; $\text{C}_{31} \text{S}/(\text{S}+\text{R})$, $17\alpha(\text{H}), 21\beta(\text{H})$ -homohopane (22S)/ $17\alpha(\text{H}), 21\beta(\text{H})$ -homohopane (22S)+(22R); Triplet, [C_{26} -tricyclic terpene (S?)+ C_{26} -tricyclic terpene (R?)]/ C_{24} -tetracyclic terpene; $\text{C}_{29} \text{S}/(\text{S}+\text{R})$, 24-ethyl-5 $\alpha(\text{H}), 14\alpha(\text{H}), 17\alpha(\text{H})$ -cholestane (20S)/24-ethyl-5 $\alpha(\text{H}), 14\alpha(\text{H}), 17\alpha(\text{H})$ -cholestane (20S)+(20R); $\text{D}(27)/\text{St}(29)$, 13 $\beta(\text{H}), 17\alpha(\text{H})$ -dia-cholestane (20S)+(20R)/24-ethyl-5 $\alpha(\text{H}), 14\alpha(\text{H}), 17\alpha(\text{H})$ -cholestane (20S)+(20R); $\text{C}_{28}/\text{C}_{29}$, 24-methyl-5 $\alpha(\text{H}), 14\alpha(\text{H}), 17\alpha(\text{H})$ -cholestane (20R)/24-ethyl-5 $\alpha(\text{H}), 14\alpha(\text{H}), 17\alpha(\text{H})$ -cholestane (20R); BNO, $17\alpha(\text{H}), 18\alpha(\text{H}), 21\beta(\text{H})$ -28,30-bisnorhopane, 25-norhopanes, oleananes; $\delta^{13}\text{C}$ (‰), $[^{13}\text{C}/^{12}\text{C}_{\text{sample}}/^{13}\text{C}/^{12}\text{C}_{\text{standard}})-1] \times 10^3$; nd, not determined.

Valdez oil (EVO), were dissolved in dichloromethane. The resulting solutions were concentrated, and elemental sulphur was removed. Portions of these extracts from four sediment samples and from the EVO were taken for determination of stable carbon-isotopic compositions. After solvent removal, the extracts were combusted with oxygen, and the resulting CO_2 was purified under vacuum by differential temperature transfer and measured by isotope-ratio mass spectrometry. The results are reported in delta (δ) notation in parts per thousand (‰) relative to the Pee Dee belemnite (PDB) standard.

The solutions were fractionated by liquid–solid chromatography after the dichloromethane solvent was exchanged with *n*-hexane. Fractions containing aliphatic and aromatic hydrocarbons were analysed by gas chromatography and gas chromatography/mass spectrometry (GC/MS). Polycyclic aliphatic biomarkers were analysed by selected-ion monitoring (SIM) of mass-to-charge (*m/z*) ratios of 177 for 25-norhopanes, 191 for terpanes (tricyclic and tetracyclic terpanes and pentacyclic triterpanes), and 217 for steranes (including diasteranes). Compound identifications of terpanes and steranes were based on a previous study of North Slope crude oil (Kvenvolden *et al.*, 1985). Selected ratios of these compounds were calculated from peak heights on mass chromatograms. Identification of 25-norhopanes was based on work by Curiale *et al.* (1985). Aromatic hydrocarbons were identified by monitoring molecular ions (*m/z*) of naphthalenes (128, 142, 156, 170), phenanthrenes (178, 192, 206, 220), dibenzothio-

phenes (184, 198, 212, 226), and the major-fragment ion of triaromatic steranes (231).

Results and Discussion

When we initially sampled the shorelines of the six islands (Fig. 1), we assumed that all of the oil residues which we collected were related to the *Exxon Valdez* oil spill. Thus, we searched for geochemical criteria that could relate these residues on the shores to the North Slope crude oil that was spilled in March 1989 (Rapp *et al.*, 1990; Kvenvolden *et al.*, 1991). Previous published geochemical studies of hydrocarbons in Prince William Sound had been completed before the oil spill and focused on Port Valdez (Fig. 1) at the north end of the Sound (Shaw & Baker, 1978; Shaw *et al.*, 1985). Our report presents new information on the hydrocarbon geochemistry of oil residues collected since the oil spill at locations throughout the Sound.

Distributions of certain aliphatic and aromatic hydrocarbons (Fig. 2, Table 1) and carbon isotopic compositions of total extracts (Table 1) provided evidence that our sample set contained oil residues from two distinct sources, referred to here as '*Exxon Valdez* residues (EVR)' and '*Non-Exxon Valdez* residues (NEVR)'. Six samples 6, 17A, 18, 19, 29, and 41 comprise the EVR, because these samples have chemical and isotopic compositions that match those of the *Exxon Valdez* oil (EVO). These samples ranged from oil with odour and sheen, through sticky oil with no odour, to solid oil which could be scraped from the

rocks (Table 1). Two samples, 6A and 37A, comprise the NEVR, because their compositions are different from EVR and EVO. These two samples, which were tar-like in appearance (Table 1), came from localities about 100 km apart, on Elrington Island and Storey Island (Fig. 1).

The following discussion presents the arguments that are used in the correlation of the EVR samples with the EVO and in the differentiation of EVR samples and NEVR samples. In this discussion, much of the molecular data is condensed in Fig. 2. Gas chromatograms of aliphatic hydrocarbons and mass chromatograms of polycyclic aliphatic and aromatic hydrocarbons of the EVO are compared with sample 41 chromatograms, which are typical of all EVR samples, and with sample 37A chromatograms, which are similar to those of the other NEVR sample.

Exxon Valdez Residues (EVR)

Gas chromatograms of the aliphatic hydrocarbons in the EVO and EVR (Fig. 2A, B) illustrate the effects of

oil degradation. The gas chromatogram of the aliphatic hydrocarbons in the EVO shows a mixture dominated by an homologous series of *n*-alkanes that decrease monotonically in relative concentrations with increasing molecular weight and range in carbon number from about *n*-C₁₁ to at least *n*-C₄₀ (Fig. 2A). Isoprenoid hydrocarbons which are commonly considered in geochemical studies (Didyk *et al.*, 1978; ten Haven *et al.*, 1987; Powell, 1988), are also present, and pristane and phytane are the most abundant. In contrast, the gas chromatogram of the aliphatic hydrocarbons in the representative EVR sample shows that the residues have undergone significant alteration (Fig. 2B). The *n*-alkanes have essentially disappeared, and pristane and phytane are the only aliphatic compounds that can still be confidently identified by gas chromatographic retention times. Pristane/phytane ratios vary (Table 1), probably as a result of alteration processes. A further example of alteration is indicated by the presence of an unresolved complex mixture (UCM) of hydrocarbons (Farrington & Meyers, 1975) that manifests itself as a

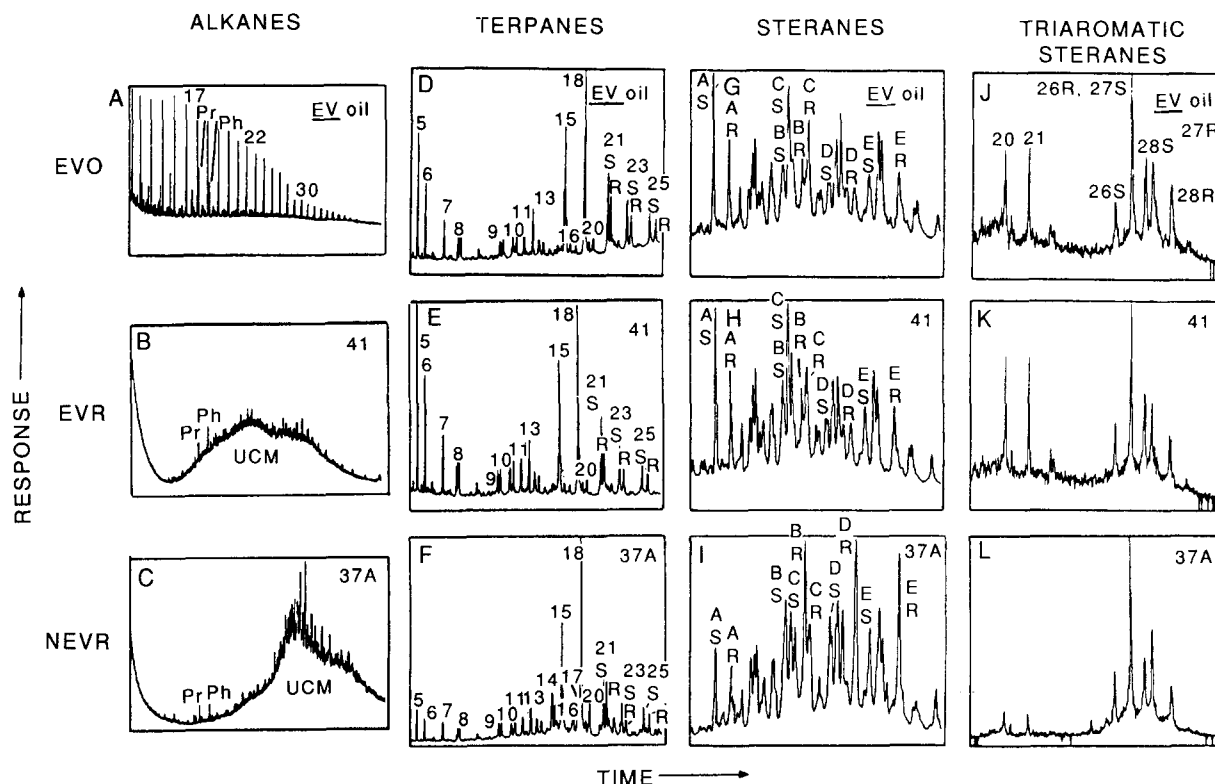


Fig. 2 Chromatograms of alkanes (A, B, C), terpanes (D, E, F), steranes (G, H, I), and triaromatic steranes (J, K, L) in oil and oil residues from Prince William Sound, Alaska: EVO, *Exxon Valdez* Oil [North Slope crude oil impounded from supertanker *Exxon Valdez* (A, D, G, J)]; EVR, a typical *Exxon-Valdez* residue [sample 41, Naked Island (B, E, H, K)]; NEVR, a typical non-*Exxon Valdez* residue [sample 37A, Storey Island (C, F, I, L)].

Fig. 2A, B, C Numbers on Fig. 2A, provide guide to the carbon numbers of the homologous series of *n*-alkanes; Pr, pristane; Ph, phytane.

Fig. 2D, E, F Mass chromatograms of terpanes ($m/z=191$): 5, C₂₃ tricyclic terpane; 6, C₂₄ tricyclic terpane; 7, C₂₅ tricyclic terpane; 8, Triplet: C₂₄ tetracyclic terpane, C₂₆ tricyclic terpane (S?), and C₂₆ tricyclic terpane (R?); 9, C₂₈ tricyclic terpanes (S? and R?); 10, C₂₉ tricyclic terpanes (S? and R?); 11, 18 α (H)-22, 29, 30-trisnorhopane (Ts) (C₂₇); 13, 17 α (H)-22,29,30-trisnorhopane (Tm) (C₂₇); 14, 17 α (H),18 α (H),21 β (H)-28,30-bisnorhopane; 15, 17 α (H),21 β (H)-30-norhopane (C₂₉); 16, 17 β (H),21 α (H)-30-normoretane (C₂₉); 17, 18 α (H) and (or)

β (H)-oleanane (C₃₀); 18, 17 α (H),21 β (H)-hopane (C₃₀); 20, 17 β (H),21 α (H)-moretane (C₃₀); 21, 17 α (H),21 β (H)-homohopane (22S and 22R) (C₃₁); 23, 17 α (H),21 β (H)-bishomohopane (22S and 22R) (C₃₂); 25, 17 α (H),21 β (H)-trishomohopane (22S and 22R) (C₃₃).

Fig. 2G, H, I Mass chromatograms of steranes ($m/z=217$): A(S), 13 β (H),17 α (H)-diacholestane (20S) (C₂₇); A(R), 13 β (H),17 α (H)-diacholestane (20R) (C₂₇); B(S), 5 α (H),14 α (H),17 α (H)-cholestane (20S) (C₂₇); C(S), 24-ethyl-13 β (H),17 α (H)-diacholestane (20S) (C₂₉); B(R), 5 α (H),14 α (H),17 α (H)-cholestane (20R) (C₂₇); C(R), 24-ethyl-13 β (H),14 α (H)-diacholestane (20R) (C₂₉); D(S), 24-methyl-5 α (H),14 α (H),17 α (H)-cholestane (20S) (C₂₈); D(R), 24-methyl-5 α (H),14 α (H),17 α (H)-cholestane (20R) (C₂₈); E(S), 24-ethyl-5 α (H),14 α (H),17 α (H)-cholestane (20S) (C₂₉); E(R), 24-ethyl-5 α (H),14 α (H),17 α (H)-cholestane (20R) (C₂₉).

Fig. 2J, K, L Mass chromatograms of triaromatic steranes (fragment ion $m/z=231$): Peaks of triaromatic steranes are identified in Fig. 2J, showing carbon number and stereochemical configuration (R and S).

chromatographic hump. In all of the EVR samples the UCM is bimodal and broad.

Because polycyclic-aliphatic-hydrocarbons are less susceptible to rapid degradation than are *n*-alkanes and isoprenoid hydrocarbons (Volkman, 1984), the polycyclics are more useful in correlations. The mass chromatograms of terpanes and steranes in the EVR samples are remarkably similar to each other and to those of the EVO. This similarity between EVO and a representative sample of EVR is illustrated in Fig. 2D, E, and 2G, H.

Similarities and differences in these mass-chromatographic patterns can be described by molecular ratios which are defined in Table 1. Although some molecular ratios are controlled more by thermal maturity than by geological source, we have used these ratios as source indicators because the spilled oil is not expected to have undergone any thermal maturation since the time of its release in March 1989. For example, the C_{23}/C_{30} ratio (Waples & Machihara, 1991) for the EVO of 0.71 falls within the range of values (0.59–1.2) observed for the EVR (Table 1); this range may reflect differential weathering of pentacyclic vs. tricyclic terpanes (Reed, 1977; Williams *et al.*, 1986) in the EVO. The similarity of values of other ratios (Table 1), such as C_{30}/C_{29} (Palacas *et al.*, 1984), $[C_{31} S/(S+R)]$ (Ensminger *et al.*, 1974; Mackenzie, 1984), and Triplet (Kvenvolden *et al.*, 1985) from the *m/z* 191 chromatograms, and the $[C_{29} S/(S+R)]$ (Mackenzie *et al.*, 1980), $[D(27)/St(29)]$ (Jiamo *et al.*, 1990), and C_{28}/C_{29} (Grantham & Wakefield, 1988; Waples & Machihara, 1991) from the *m/z* 217 chromatograms, suggests that the EVR samples are related.

The aromatic fraction of the EVO contains primarily polycyclic aromatic hydrocarbons (PAHs), particularly naphthalene and phenanthrene and their alkylated derivatives with naphthalenes being dominant (Kvenvolden *et al.*, 1991). In the methylphenanthrenes, as identified by GC/MS, the 9-methyl isomer is predominant, and alkylated C_1 – C_3 homologs are present in concentrations comparable to those in the parent PAH, as is common in other typical oils (Prah & Carpenter, 1983). Alkylated dibenzothiophenes also are present, but in substantially lower concentrations than phenanthrenes, and a series of triaromatic steranes (Fig. 2J) occur in distributions (C_{20} , C_{21} , C_{26} – C_{28}) similar to those reported in other oils (Killips & Howell, 1988).

Aromatic hydrocarbons are significantly affected by water washing, oxidative processes, and weathering (Gundlach *et al.*, 1983; Brakstad & Grahl-Nielsen, 1988). Weathering causes a progressive loss of lower-molecular-weight PAHs (i.e. naphthalenes before phenanthrenes) and alkylated PAHs with fewer alkyl substituents (Volkman, 1984). Because the aromatic hydrocarbons in the EVR have been altered extensively (Kvenvolden *et al.*, 1991) these compounds are not particularly useful for our present study of EVR and NEVR, with the exception of triaromatic steranes. The distribution of the triaromatic steranes in EVR samples are similar and correlate well with the distribution in the EVO (Fig. 2J, K).

The stable carbon-isotopic compositions of whole

extracts of two EVR samples (18, 29) have $\delta^{13}C$ values of -29.2 and -29.1 ‰, respectively; these values correlate with the $\delta^{13}C$ value of -29.2 ‰ of the EVO. Thus, the carbon-isotopic compositions and the distributions of terpanes, steranes, and triaromatic steranes support the conclusion that the EVR samples are North Slope crude oil residues from the *Exxon Valdez* oil spill.

Non-Exxon Valdez Residues (NEVR)

The gas chromatographic distributions of aliphatic hydrocarbons from the NEVR samples contrast with the distributions from the EVR samples in the pattern of the UCM (compare Fig. 2B and 2C). In NEVR samples the UCM is unimodal and sharp, whereas, the UCM in EVR samples is bimodal and broad. Mass chromatograms of terpanes and steranes of the NEVR samples are similar to each other but different from those of the EVR and EVO (Fig. 2D, E, F and 2G, H, I). All of the molecular ratios in Table 1, except Pr/Ph which apparently varies with the degree of alteration, show that NEVR samples are distinct from EVR samples and EVO. For example, both NEVR samples have C_{23}/C_{30} ratios of 0.17, a value outside of the range of the other samples. Triplet ratios of 4.3 and 5.1 contrast with the average triplet ratio of 2.1 for the EVR and 2.0 of the EVO. The other molecular ratios of Table 1 [C_{30}/C_{29} , $C_{31} S/(S+R)$, $C_{29} S/(S+R)$, $D(27)/St(29)$, and C_{28}/C_{29}] also demonstrate the distinction between NEVR and EVR.

The hopane and sterane epimer ratios [$C_{31} S/(S+R)$ and $C_{29} S/(S+R)$] suggest that the NEVR samples, with average values of 0.56 and 0.31, respectively, have a thermally less mature source than do all of the other samples, with respective average ratios of 0.60 and 0.48. This difference in these maturity parameters (epimer ratios) further distinguishes NEVR from the EVR and EVO. Compelling evidence for this distinction is the presence in only the NEVR samples of $17\alpha(H), 18\alpha(H), 21\beta(H)$ -28,30-bisnorhopane (Fig. 2F), a series of 25-norhopanes (identified in *m/z* 177 mass chromatograms, not shown here), including $17\alpha(H), 18\alpha(H), 21\beta(H)$ -25,28,30-trisnorhopane, and oleananes (Fig. 2F, Table 1, BNO).

Both NEVR samples contain distributions of triaromatic steranes that are similar, but the distributions are different from those in EVR samples and EVO. Figure 2J, K, L compares these distributions in typical samples. NEVR samples have lower relative amounts of C_{20} and C_{21} compounds, a feature suggesting extensive biodegradation (Wardroper *et al.*, 1984).

The stable carbon-isotopic compositions of whole extracts of the two NEVR samples (6A, 37A) have essentially the same $\delta^{13}C$ values, -23.8 and -23.9 ‰, respectively (Table 1). These compositions differ significantly from those obtained for EVR samples (-29.2 and -29.1 ‰) and for EVO (-29.2 ‰). Thus, the carbon-isotopic compositions and the distributions of various hydrocarbons illustrate that our NEVR samples are related and likely come from the same original source, but that they are distinct from EVR and EVO.

Apparently non-*Exxon Valdez* oil residues are common in Prince William Sound as observed by Exxon Co., USA (1990), which obtained information on 47 tar samples, unrelated to the *Exxon Valdez* oil spill, that were found during the summer of 1990 at about 27 sites in Prince William Sound (Fig. 1). It is not presently known whether the chemical signatures of these samples match those of our NEVR samples, or whether all of the non-*Exxon Valdez* samples have a common source. Evidently, however, Prince William Sound has other oil contamination besides that from the *Exxon Valdez* oil spill. This contamination could be from any or all of the following: paving and roofing tar, as suggested by Exxon Co., USA (1990), residual spilled fuel oil, and ship discharges.

Source of our Non-Exxon Valdez Residues

Our two NEVR samples have some unusual chemical characteristics that provide a guide to a possible source. First, their carbon-isotopic compositions are heavier ($\delta^{13}\text{C}$ values, -23.8 and -23.9‰) than those of most known crude oils, except for oils from California that are sourced in the Monterey Formation. The $\delta^{13}\text{C}$ values of these California oils fall in the range between about -21 and -23‰ (Magoon & Issacs, 1983; Curiale *et al.*, 1985). Thus, the carbon-isotopic compositions of our NEVR samples closely approach those of California oils. Second, several molecular parameters of our NEVR samples (Table 1) are characteristic of Monterey-sourced California oils studied by Curiale *et al.* (1985). For example, the hopane and sterane epimer ratios of the NEVR samples [$\text{C}_{31} \text{S}/(\text{S}+\text{R})=0.56$ and $\text{C}_{29} \text{S}/(\text{S}+\text{R})=0.31$, respectively] are unusually low and fall within the unusually low ranges of California crude oils, in which hopane epimer ratios are from 0.51 to 0.61 and sterane epimer ratios are from 0.22 to 0.52. Also, the relatively low amounts of diasteranes, compared to steranes, observed in the NEVR samples (Fig. 2I), is another property of California crude oils. Finally, and perhaps most important in assigning a possible source, the NEVR samples contain $17\alpha(\text{H}),18\alpha(\text{H}),21\beta(\text{H})$ -28,30-bisnorhopane, a suite of 25-norhopanes including $17\alpha(\text{H}),18\alpha(\text{H}),21\beta(\text{H})$ -25,28,30-trisnorhopane, and oleananes; these compounds are all molecular marker compounds characteristic of many Monterey-sourced California oils.

In retrospect, finding California oil residues in Prince William Sound is not completely unexpected. Before Alaska had its own internal supply of oil, the State received much of its oil and oil products by barge from California (Standard Oil of California, 1979). Whittier, in the western part of the Sound, and Valdez, at the north end of the Sound, served as a ports for fuel oil and asphalt storage that was greatly disrupted during the 1964 earthquake (National Research Council, 1972, 1973). Thus, there have been many opportunities for California crude oil to reach Prince William Sound.

Conclusions

The geochemical evidence from aliphatic and aro-

matic hydrocarbons and from stable carbon isotopic ratios indicates that oil residues from the shores of Elrington, Knight, Smith, Eleanor, and Naked Islands (Fig. 1) are related to each other and correlate with oil spilled from the supertanker *Exxon Valdez*. The geochemical evidence also indicates that residues from the shores of Elrington and Storey Islands, at localities about 100 km apart at the south and north ends of Prince William Sound, are not derived from North Slope crude oil and are not from the *Exxon Valdez* oil spill but rather are from the same original source, namely, the Monterey Formation in California. The non-*Exxon Valdez* residues may be paving tar, roofing tar, residual fuel oil, or ship discharges. Our results, plus the observations by Exxon Co., USA (1990) suggest that non-*Exxon Valdez* residues may be more common on the shores of Prince William Sound than was previously realized. In addition, our results suggest that oil residues now present in the Sound have at least two very different sources, one being Alaska and the other, California.

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