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ANALYSES OF LIGHT HYDROCARBONS FROM THE FLORIDA-HATTERAS SLOPE AND BLAKE PLATEAU

by

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ABSTRACT

Residual-light-hydrocarbon (C_1 - C_4) concentrations and molecular distributions have been measured in shallow core sediments from the Florida-Hatteras Slope and Blake Plateau. Specific geologic features and geophysical anomalies chosen as core sites included slump masses, an accretionary wedge, an area of water-column acoustic anomalies, a suspected Paleocene bottom exposure, erosional channel features, an area of subsurface faults, and sediments overlying a diapir. Total light hydrocarbon concentrations were less than 10 ppm in most surface sediments, and the major hydrocarbon components present were methane, ethane, and ethene. Slightly greater residual C_1 - C_4 concentrations of 25 ppm were found in samples collected from sediment associated with the floor of an erosional channel. Although low levels of ethane and propane were found, samples taken during a transect over a diapir consisted mainly of methane and contained slightly greater residual-light-hydrocarbon concentrations, less than 39 ppm. Extremely low residual concentrations of the hydrocarbons reported for the slope and plateau surface sediments are believed to be gases dissolved in the pore water and may represent background concentration levels for this study area. The occurrence of ethene in most samples and the extremely low C_1 - C_4 concentrations suggest that the gas is biogenic and did not originate by diffusion from underlying petroleum and natural-gas reservoirs.

INTRODUCTION

The U.S. Atlantic Continental Shelf and Slope regions are presently the focus of considerable exploration aimed at evaluating the petroleum and natural-gas resource potential of the Atlantic Outer Continental Shelf. Earlier exploration included environmental studies to determine the background concentrations of hydrocarbons in sediment prior to actual drilling (Miller and others, 1977, 1979); the Continental Offshore Stratigraphic Test (COST) wells and Atlantic Slope Project (ASP) cores drilled by consortiums of petroleum companies to assess the petroleum potential (Poag, 1978; Scholle, 1977, 1979, 1980); and the Joint Oceanographic Institutions for Deep Earth Sampling (JOIDES) and U.S. Geological Survey Atlantic Margin Coring Project (AMCOR) designed to increase the knowledge of the stratigraphic record along the U.S. Continental Shelf and Slope (Bunce and others, 1965; Charm and others, 1970; Hathaway and others, 1976, 1979; Miller and Schultz, 1977).

In this present study, light-hydrocarbon analyses were performed on surface sediments collected by piston coring during a cruise conducted chiefly for stratigraphic confirmation purposes onboard the R/V EASTWARD. The purpose and scope of this light-hydrocarbon geochemical study were: (1) to measure the C_1 - C_4 hydrocarbon distribution and relative concentrations in shallow cored sediments from the Florida-Hatteras Slope and Blake Plateau; (2) to determine background concentration levels of light hydrocarbons in surface sediment from the Florida-Hatteras Slope and Blake Plateau; (3) to correlate the hydrocarbon-gas distribution and concentrations found in the surface sediments with geophysical data;

and (4) to differentiate biogenic gas from petrogenic light hydrocarbons that may have diffused from deeper petroleum and natural-gas reservoirs.

Previous Studies

Concentrations and distributions of dissolved, low-molecular-weight gaseous hydrocarbons, methane through pentane, have been measured in open ocean and nearshore water columns (Atkinson and Richards, 1978; Swinnerton and Linnenbom, 1967; Swinnerton and others, 1969; Lamontagne and others, 1971, 1973, 1974; Linnenbom and Swinnerton, 1970; Brooks and Sackett, 1973, 1977; Brooks and others, 1973; Swinnerton and Lamontagne, 1974; and Sackett and Brooks, 1975). Generally, average surface concentrations in uncontaminated, open ocean water are 40-60 ppb (nl/l) for methane, 0.25-3 ppb for ethane and propane, 4-6 ppb for ethene, and less than 2 ppb for propene. Gas concentrations in coastal samples are usually higher than those in the open ocean, and in some areas, the higher concentrations appear to be largely a result of anthropogenic activities (Brooks and Sackett, 1977).

During the Deep Sea Drilling Project, gas from gas expansion pockets and from the head space in canned core samples was analyzed for the hydrocarbon gas content. The hydrocarbon gas found was mainly methane and ranged in concentration from about 200 to 256,000 ppm, as determined by a ratio of a fixed volume of unknown gas to an equivalent volume of calibration gas of known concentration (Erdman and others, 1969; Claypool and others, 1973; Hammond and others, 1973; McIver, 1973a, b, c, 1974a, b, c, 1975; Claypool and Kaplan, 1974; Hunt, 1974; Morris, 1974; Doose and others, 1975; and Hunt and Whelan, 1975; among others). Gas expansion pockets that formed in the core liners and the head space in canned, sonicated sediment were sampled during the AMCOR program. The light-hydrocarbon concentrations in these samples were as great as 417,000 ppm (Hathaway and others, 1976, 1979; Miller and Schultz, 1977).

The distribution and concentration of light hydrocarbons in surface sediments have been measured by many investigators. Reeburgh (1969) reported methane in the upper 100cm of the Chesapeake Bay sediments and found that the concentration approached the methane saturation limit in several of the cores. Reeburgh and Heggie (1974) extended the methane study to include anoxic sediments from a freshwater lake in Alaska and reported that the methane concentrations were well below the saturation limit. Methane present in anoxic marine sediments from Long Island Sound was also found to approach the saturation limit (Martens and Berner, 1974, 1977).

Barnes and Goldberg (1976) used an in situ sampler to collect pore water from the upper 500 cm of anoxic Santa Barbara Basin sediments. Methane concentrations as great as 300,000 ppm were measured in the pore water from these sediments. Concentration ranges of gaseous hydrocarbons in cored sediments from several anoxic basins off the coast of Southern California were 10-241,000 ppm for methane, 0.3-30 ppm ethane, 0.4-0.7 ppm ethene, 0.4-1 ppm propane, 0.05-0.3 ppm butane, and 0.08-0.5 ppm isobutane (Emery and Hoggan, 1958). The methane present in the sediment pore water approached the saturation limit.

Rashid and Vilks (1977) reported methane concentrations of as great as 16,000 ppm in basin sediment from offshore eastern Canada. Gaseous hydrocarbons above methane were not detected in the samples.

Vibracore samples from the upper 410 cm of sediment from Norton Sound, Alaska, contained methane concentrations of 1.5 to 38,000 ppm in the interstitial pore water, and ethane through butane were reported (Kvenvolden and others, 1979). The upper 200 cm of sediments in the Bering Sea was cored and methane concentrations were determined to range from 0.9 to 21.0 ppm, with hydrocarbons through butane also reported (Kvenvolden and Redden, 1980). Methane concentrations in the upper 200 cm of sediments from the Texas Continental Shelf and Slope ranged from 0.4 to 25 ppm, and ethane, propane and the unsaturated hydrocarbons ethene and propene were also detected (Bernard and others, 1978). Gaseous hydrocarbons were measured in the upper 10 m of sediments from the North and Mid-Atlantic Outer Continental Slope (R.E. Miller and others, unpublished data, 1981). Ethane, ethene, propane, and butane were found in many of the cores, and methane concentrations ranged from 0.4 to 16,500 ppm.

Methane can be formed by anaerobic microorganisms, and it has also been suggested that very small quantities of the saturated higher permanent homologs ethane and propane may be produced biogenically (Davis and Squires, 1954; Rheinheimer, 1974, p. 126). Further, Hunt and others (1980) have suggested that homologs through pentane may originate from biological precursors at temperatures below 20°C. Wilson and others (1970), Brooks and Sackett (1973), and Swinnerton and Lamontagne (1974) noted that a possible correlation exists between plankton and light-hydrocarbon olefins in the water column and that production of ethene and propene in the euphotic zone may be related to biological processes. Profiles of the distribution of ethane, propane, ethene, and propene with burial depth in Gulf of Mexico sediments suggest that these gases may be biogenically controlled in the sediment (Bernard and others, 1978).

The distribution of light hydrocarbons in the water column and sediments has been interpreted to identify possible natural-gas seeps and to aid in offshore exploration for possible reservoirs of petroleum and natural gas (Horvitz, 1954, 1969; Brooks and others, 1974; Carlisle and others, 1975; Bernard and others, 1976; and Sackett, 1977). Bernard and others (1977, p.435) stated that "in theory, petroleum-related (petrogenic) gas can be distinguished from microbially-produced (biogenic) gas by the presence of significant quantities of ethane-and-higher hydrocarbons." Geochemical degradation and alteration processes can, however, change the character of both biogenic and petrogenic gas, and where possible, carbon isotopic ratios should also be determined to distinguish between biogenic and petrogenic gas (Bernard and others, 1977). To pinpoint anomalous gas concentrations possibly due to gas seeps, it is essential to determine the normal background concentration levels of the light-hydrocarbon gases in the area under study.

Methods

Sediment samples were obtained with a 2 inch (5 cm) i.d. (internal diameter) piston corer onboard the R/V EASTWARD during cruise number E-2E-78, April 2-13, 1978. Location, water depth, and sampling intervals of the 23 piston cores are listed in table 1. Cores were collected with either plastic-lined or unlined core barrels.

Immediately upon retrieval, core samples of approximately 20 cm length were removed at preselected depth intervals for light-hydrocarbon analyses onboard ship and further organic geochemical studies in the laboratory. The surface of each core sediment sample was trimmed with a solvent-rinsed spatula to remove the potential problem of surface contamination of the C_{15}+ hydrocarbons from lubricating grease. Two methods, blending and high-speed shaking, were used to extract the light hydrocarbons from the sediment in this study. After sampling, a 5 cm portion of the cored sediment, 100 cm^3 in volume, was placed in a blender container. Hydrocarbon-free, degassed, distilled water was added to bring the head-space volume to 50 ml in the blender. The blender system was flushed with helium; the lid, which was equipped with a septum, was fitted into place; and the sample was blended for 5 minutes. The remaining 15 cm portion of sediment, 300 cm^3 in volume, was placed in a solvent-rinsed quart can equipped with a septum in the bottom. Hydrocarbon-free, degassed, distilled water was added to the can to bring the head-space volume to 125 ml. The can was flushed with helium, sealed with the lid, and inverted; the resultant water seal prevented loss of the gas from around the lid. The sediment gases were extracted by shaking for 10 minutes on a high-speed shaker. After the gas analyses were completed, the blended samples were also placed in cans, and all samples were stored frozen.

The 1 ml volume of gas samples at ambient shipboard temperature and pressure, whether liberated with the 5-minute blender or the 10-minute shaker extraction, was injected on a Hewlett Packard[1] model 5830A gas chromatograph. The instrument was equipped with flame ionization detectors and dual 4 ft. (1.2 m) by 1/8 in. (3.2 mm) i.d. stainless steel columns packed with Chromosorb 102. The columns were operated isothermally at 35°C for 3 minutes, and temperature programmed at 6°C/min

[1]Any trade names in this publication are used for descriptive purposes only and do not constitute endorsement by the U.S. Geological Survey.

to a final temperature of 100°C. A 1 ml volume of light-hydrocarbon standard gas at ambient shipboard temperature and pressure, which contained known concentrations of methane, ethane, and propane, was injected prior to analysis of the unknown, and the instrument was calibrated with this standard gas. Quantitative and qualitative gas analyses were determined by comparing the peak areas of the fixed 1 ml volume of unknown gas at ambient shipboard temperature and pressure to the peak areas of an equivalent volume of the calibrated standard gas. The peak labeled as water vapor in the chromatograms in figures 8, 9, and 10, was a result of the analytical method and was not measured. All concentrations are reported in parts per million, ppm, based on volume of gas at ambient shipboard temperature and pressure per volume of sediment.

GEOLOGICAL FRAMEWORK

The Continental Shelf east of Florida, Georgia, and South Carolina is underlain by the Southeast Georgia Embayment and is bordered on the north by the Cape Fear Arch and on the south by the Peninsular Arch (Dillon and others, 1978; Paull and Dillon, 1979, 1980a). The embayment is a gently southeasterly dipping sedimentary basin under the Coastal Plain and offshore between Cape Fear, North Carolina, and Jacksonville, Florida. The Florida-Hatteras Slope bounds the shelf and is interrupted at a depth of 600 to 1000 meters below sea level by the broad Blake Plateau. Geology of this region has been summarized by Dillon and others (1975, 1978), Edsall (1978), and Paull and Dillon (1979, 1980a).

Seismic surveys of the offshore Southeast Georgia Embayment, Florida-Hatteras Slope, and western Blake Plateau between latitudes 29°30′N and 33°31′N were conducted by the U.S. Geological Survey during the summer and fall of 1976 (Edsall, 1978; Paull and Dillon, 1979, 1980a). A 600-joule minisparker was used to collect the high-resolution siesmic-reflection data presented by Edsall (1978). The single-channel seismic-reflection profiles reported by Paull and Dillon (1979, 1980a) were obtained by the use of four Bolt airguns of 20-, 40-, 80-, and 160-in^3 fired at 2000 p.s.i. The track lines for collection of the minisparker and airgun seismic data are shown in figure 1.

Core sites in this study were selected on the basis of the available seismic data provided by Dillon and others (1975, 1978), Edsall (1978), and M.M. Ball (personal commun., 1978). The sites were chosen to sample surface sediments associated with specific geologic features or geophysical anomolies and included erosional channel features, subsurface fault zones, slump masses, an accretionary wedge, acoustic anomalies in the water column, a suspected Paleocene bottom exposure, and a diapir.

The Gulf Stream skirts the edge of the shelf and has affected the development of the Florida-Hatteras Slope and Blake Plateau (Edsall, 1978). Areas of submarine erosion and scour, probably results of action by the Gulf Stream, were observed on many of the seismic records (Edsall, 1978; Paull and Dillon, 1979, 1980a). Profiles along track lines 9 and 15 (from Edsall, 1978) are shown in figures 2 and 3, respectively. Cores 34973 and 34974 were collected from surface sediment associated with the channel floor shown in figure 3 and sites 34988, 34989, 34991, and 34992 were located in the channel cut just north of line 9 (fig. 2). Cores 34946 and 34954 were collected from surface sediments in the floors of similar channel features.

The activity of the Gulf Stream has either removed sediments younger than Paleocene or prevented deposition of the younger sediments in large areas of the southern and middle portions of the survey area of the Florida-Hatteras Slope and Blake Plateau (Edsall, 1978). Site 34960 was located in an area where Edsall (1978) and Paull and Dillon (1979) suggested that Paleocene sediments may be exposed on the sea floor. Ayers and Pilkey (1981) recently reported that the suspected Paleocene unit was covered by Holocene sediment which was apparently acoustically transparent.

Several seismic records from this study area reported by Edsall (1978) and Paull and Dillon (1979) have been interpreted to show faults which are believed to be related to either compaction or gravity faulting. With the exception of a normal fault observed on line 29, they are small, near vertical features with displacement of 10 meters or less and do not extend to the surface (Paull and Dillon, 1979). The faults generally occur in groups or clusters and are found throughout the inner Blake Plateau (Paull and Dillon, 1979). The features on line 24A shown in figure 4 have been interpreted to represent subsurface faults, although Edsall (1978) also pointed out that the disrupted reflectors may represent the effects of differential compaction and draping on a buried unconformity. Cores 34956, 34957, and 34958 were collected from the sediments overlying the interpreted faults on line 24A, and core 35000 sampled the surface sediments in a similar zone on line 11.

Slumping on the Florida-Hatteras Slope may take place in areas where activity of the Gulf Stream has eroded sediments farther down on the slope, where truncated foreset bedding occurs near the shelf edge, and where fine-grained sediments are accumulating on the slope (Edsall, 1978). A slump about 6 km long and 30 m thick was interpreted to be present at the base of the slope on track line 19 (fig. 5) (Edsall, 1978). Paull and Dillon (1979) point out that the scar associated with this possible slump mass has not been observed on the seismic records. Site 34970 was located near the toe of this feature. Ayers and Pilkey (1981) suggested that the core from the site penetrated a debris flow associated with the slumping. A smaller possible slump mass was interpreted to be present at the base of the slope on line 20 (M.M. Ball, personal commun., 1978). Core 34964 sampled the surface sediments associated with the toe of this smaller possible slump feature.

A progradational accretionary wedge, possibly Oligocene in age, is exposed on the sea floor in the northern portion of the seismic survey area, on track line 32 between line 9 and line 5C (Edsall, 1978). Sediment overlying this wedge was sampled in core 34984.

Acoustic anomalies in the water column were observed on several track lines across the Blake Plateau in water depths greater than 420 m (Edsall, 1978). These anomalies vary in size, shape, and concentration, appear as hyperbolas on the seismic record, and are generally not associated with obvious bottom structures (Edsall, 1978). Edsall (1978) thought that deep-water coral mounds or reefs were the most likely explanations for the water column anomalies although other possible causes for the hyperbolas on the seismic records may include gas seeps, freshwater seeps, and concentrations of fish. Site 35007 was located near line 17 in an area that was interpreted to produce water column anomalies on the seismic record. Core 34959 was also collected from the surface sediments associated with a suspected deep-water coral mound or reef, as interpreted from the hyperbolas observed in the seismic profile track line 24 (fig. 6) (Edsall, 1978).

Single-channel seismic records were collected by Grow and others (1977) on the Blake Plateau, and two perpendicular profiles that cross at the crest of a diapir are shown in figure 7 (Grow and others, 1977; Paull and Dillon, 1980b). Cores 35034, 35035, 35036, 35037, and 35038 were collected on a transect made across the diapir approximately on the line followed in profile B. Site 35036 was located above the crest of the diapir, sites 35034 and 35035 were located over the north and west flank, and sites 35037 and 35038 were over the south and east flank of the diapir.

RESULTS AND DISCUSSION

The R/V EASTWARD core station locations, AMCOR and JOIDES core sites, track lines for collection of the seismic data, and an interpretation of the geologic features sampled are presented in figure 1. The high-resolution seismic data and the interpretation of the geologic features were reported by Edsall (1978) and Paull and Dillon (1979, 1980a, b).

Light-hydrocarbon concentrations present in the upper few meters of surface sediments from the Florida-Hatteras Slope and Blake Plateau are listed in table 2. Concentration values of the light hydrocarbons measured after the blender extraction were statistically greater, an average of 5 times greater, than levels determined after shaking (Schultz and others, unpublished data). The data reported in table 2 were determined after a 5 minute blender extraction, unless otherwise noted.

The gas analyzed in this study was extracted from sediments composed of clay to fine- or medium-grained sands (table 1) (Ayers, written commun., 1978, 1979; Ayers and Pilkey, 1981). Concentration levels of the residual light hydrocarbons were very low, less than 39 ppm (μl gas at STP/liter of sediment) of methane and less than 3 ppm ethane and propane (table 2). Concentrations of the higher homologs, propane, butane, and isobutane, were below the minimum level of detection for the majority of the gas samples analyzed. Ethene was detected in most of the samples, at levels generally less than 1 ppm. Light-hydrocarbon concentrations reported in this study are believed to be residual pore gases retained by the sediment and water after the piston core was raised to ambient shipboard pressure and temperature. These concentrations, therefore, do not represent in situ values.

An estimate of the solubility of methane under in situ temperature and pressure conditions may be made to determine if the residual gas concentrations approach the saturation limit of methane in the pore water. Atkinson and Richards (1967) measured the solubility of methane in seawater having a salinity of 40 ^{o}/oo and reported that the solubility was a linear function of temperature from 0^{o}C to 30^{o}C. The solubility of methane in seawater decreases by about 1% per chlorinity unit, and at a salinity of 35 ^{o}/oo it is about 20% less than the solubility in distilled water (Reeburgh, 1969). Culberson and McKetta (1951) also found that the solubility of methane in distilled water increased by about two orders of magnitude as pressure increased from 1 atm. to 240 atm. at a constant temperature of 25^{o}C.

In this present study, the temperatures of the sediment-water interface for the core sites are estimated to be less than 10^{o}C. This estimate is based on the hydrographic sections off Cape Hatteras reported by Barrett (1965). At a salinity of 35 ^{o}/oo, temperatures of less than 10^{o}C and hydrostatic pressures of 30 atm. or greater, the in situ solubility of methane can be estimated to be greater than 200 ml methane per liter (200,000 ppm) (Claypool and Kaplan, 1974). The solubility of methane in seawater of 35 ^{o}/oo salinity under the ambient shipboard conditions of 1 atm. pressure and 20^{o}C would be approximately 29 ml/l (29,000 ppm) (Atkinson and Richards, 1967).

Molecular distributions and concentrations of the light-hydrocarbons are reported in table 2. Most of the sediment samples contained the permanent gases methane and ethane, and many had the unsaturated homolog ethene present, but few contained the higher homologs propane, butane, or isobutane. Regardless of the geologic feature or geophysical anomaly with which the cores were associated, the total $C_1 - C_4$ hydrocarbon concentrations found in the surface sediments were in all cases less than 40 ppm (μ l gas/l sediment). The total residual-gaseous-hydrocarbon concentrations are believed to be several orders of magnitude below the estimated saturation levels of methane both _in situ_ as well as at ambient shipboard temperatures and pressures. The residual-light-hydrocarbon concentrations measured in the samples from the upper few meters of surficial sediments for the study area probably represent gases dissolved in the interstitial pore water rather than free gas. Loss of gas during recovery of the piston core and subsequent shipboard processing of the cored sediment affects the residual-light-hydrocarbon concentrations. Bernard and others (1978) assumed that extremely low residual-gas concentrations, well below saturation levels, precluded outgassing during sample processing and that loss of gas from sediments that showed low residual concentrations took place only through molecular diffusion. However, _in situ_ measurements of the gas composition, volumes, pore pressures, and temperatures are necessary to evaluate the relationship between the residual-gas concentrations and _in situ_ gas information (R.E. Miller and others, unpublished data, 1981).

Light-hydrocarbon analyses during the AMCOR program indicated that concentrations of methane and total gas increased with depth of sediment burial (Hathaway and others, 1976, 1979; Miller and Schultz, 1977). The greatest methane concentrations detected in piston cores from the

Mid-Atlantic upper slope sediments were present in the deepest portions of the core, which was generally at a burial depth of 6 to 8 m (R.E. Miller and others, unpublished data, 1981). Results of this present light-hydrocarbon study do not, however, show a similar, consistent trend of increased concentration with increased sediment depth in the shallow cores. Whereas cores from the Mid-Atlantic study were as long as 10 m, the cores collected on the Florida-Hatteras Slope and Blake Plateau were less than 6 m long, and it is possible that greater concentrations may have been observed if more deeply buried sediment had been sampled.

Sediment cores collected from surface sediments associated with channel features (stations 34988, 34989, 34991, and 34992) contained the higher homologs propane, butane, and isobutane and had total residual $C_1 - C_4$ concentrations of about 25 ppm (table 2). A chromatogram of the gases analyzed from core 34991 is shown in figure 8. A thin, 5- to 50-cm-thick layer of coarse sand and lithified manganese-phosphorite nodules overlaid fine-grained, muddy sands in the surface sediments associated with the channel feature. Concentration levels in the surface sediments overlying other channel features, from cores 34946, 34954, 34973, and 34974, were less than 6 ppm total $C_1 - C_4$ residual hydrocarbons, much lower than concentrations in the samples from sites 34988, 34991, and 34992. The surface sediments in these samples were similar in texture to the sediments in the 34988 to 34992 samples, with muddy foraminiferal sand and glauconite in 34946, slightly sandy, silty clay in core 34954, silty clay in core 34973, and silty fine sand with shell fragments and glauconite in core 34974 (table 1). A layer of coarse sand and nodules was not found in the surface sediments associated with the other channel floors, and it is possible that the nodule pavement overlaying the sediments in the channel floor for the site 34988 to 34992 sampling area may have acted as a seal to reduce submarine erosion of the fine-grained sediment, which retained gas. It is also possible that a relatively greater abundance of organic matter was present in the surficial sediment from the channel feature, sites 34988 to 34992, which may have provided an organic source for production of biogenic gas.

Core 34970, collected from surface sediments near the toe of a possible slump mass shown in figure 5, contained very low residual concentrations of methane, ethane, and propane and had a total concentration of 0.94 ppm. The sediment in this core was a very dense silty clay (table 1). Cored sediment from site 34964, a small feature interpreted as a possible slump, consisted of slightly sandy, silty clay and contained only 4.15 ppm total C_1 - C_4 residual hydrocarbons. The residual-gas concentrations detected in these sediments are not believed to represent sufficient gas for further mass movement through bubble coalescence and liquefaction of the sediment because the residual concentration levels are several orders of magnitude below the saturation levels of methane. The mass transport of sediment downslope, which may have been the cause for the geophysical features interpreted as possible slumps, probably resulted in a loss of gas from the sediment during the mass movement, thereby accounting for the very low residual values.

Sediments from surface piston cores collected from areas which had been intepreted to show subsurface faults on seismic profile 24A (fig. 4), cores 34956, 34957, and 34958, were very coarse and ranged from a coarse muddy sand to a sandy gravel (table 1). Because of the very coarse texture of these sediments, the light hydrocarbons from cores 34956 and 34958 were extracted by the shaker method to prevent damage to the blender blade unit by the coarse sand. The surface sediment overlying the fault zone on track line 11 (fig. 1), core 35000, was a slightly muddy sand having a C_1 - C_4 concentration of 0.59 ppm. A chromatogram of the light-hydrocarbons analyzed from core 35000 is shown in figure 9.

The methane-to-ethane-plus-propane ratio decreased from 19.7 to 6.2 with burial depth in the 34957 core, which indicated an increase in the higher homologs relative to methane. Such a decrease in the ratio would be consistent with an interpretation of diffusion of gas from more deeply buried petrogenic reservoirs; however, it is important to note that the total gas concentrations also decreased with depth in core 34957, and this decrease may have affected the ratio. Total light-hydrocarbon residual concentrations in the piston cores, 34956, 34957, 34958, and 35000, taken above the interpreted area of faulting were less than 10 ppm. Although the methane-to-ethane-plus-propane ratio would suggest diffusion of gas from a deeper reservoir in core 34957, the extremely low residual C_1 - C_4 concentration levels determined in the surface sediments overlying the fault zones and the observed decrease in concentration with burial depth do not appear to support an interpretation of diffusion of gas to the surface through the faults deeper in the section.

Core 35007 was collected from the surface sediments associated with an area which had shown surface-water-column acoustic anomalies on the seismic record. The sediment consisted of a muddy silt, and the residual concentration of the total C_1 - C_4 hydrocarbons was 0.77 ppm. Sediments collected from core site 34959, which was located in an acoustically anomalous zone interpreted as showing a possible deep-water coral reef or mound, was a muddy coarse sand and coral gravel. The light-hydrocarbons were extracted by shaking. The concentration of methane was 0.50 ppm and hydrocarbon homologs above methane were not detected. The results of the light-hydrocarbon analyses from these two sites are consistent with the interpretation of Edsall (1978) that the anomalies probably do not result from gas seeps. However, without on-site seismic data, a single core taken from each area may not have sampled the feature causing the water-column anomaly, and a series of cores may be necessary to pinpoint a gas seep, if present.

A muddy fine sand was recovered in the surface sediments from the possible Paleocene bottom exposure, site 34960, and a muddy medium sand containing numerous small phosphorite nodules was present in core 34984 from the surface sediments overlying the accretionary wedge. The residual concentration levels of methane were less than 4 ppm in these sampling areas, well below the saturation limit. These levels are probably gases dissolved in the pore water, rather than free gas.

Cores 35034 to 35038 were collected from surface sediments along a transect across the location of a diapir on the Blake Plateau (fig. 7). Concentration levels of methane in these samples reached the greatest values reported in this study, 38.34 ppm. These concentrations are still several orders of magnitude below the theoretical saturation limit of methane and are probably gases dissolved in the interstitial water. Ethane and propane were also detected, and were generally less than 1 ppm in concentration. A chromatogram of the gases analyzed from core 35037 is presented in figure 10. The total light-hydrocarbon concentrations increased eastward along the transect to the area above the diapir crest and reached the greatest concentration over the southeast flank of the diapir in core 35038 (table 2).

The seismic reflector which appears on the flank of the diapir at a reflection time of about 0.4 sec subbottom in the seismic profile in figure 7 has been interpreted as evidence for the presence of a frozen-gas-hydrate or clathrate layer in the shallow subsurface (Grow and others, 1977; Dillon and others, 1980; Paull and Dillon, 1980b). Such gas hydrates are formed under certain conditions of high pressure and low temperature and are crystalline solids in which the ice lattice framework is expanded to form cages that contain trapped gas molecules, which may include methane, ethane, propane, isobutane, carbon dioxide, and hydrogen sulfide (Hunt, 1979, p. 156). The temperature of the sediment-water interface is estimated to be 3°C at a water depth of 2,000 to 2,360 m (6,562 to 7,743 ft) near the diapir site based on the temperature-depth relationship presented by Tucholke and others (1977). Provided that excess methane is present in the sediments, gas hydrates would form and be stable under the pressure-temperature conditions at the diapir site (see Hunt, 1979, p. 158). It can be estimated from Claypool and Kaplan (1974) that under 2,000 m (6,562 ft) of water and at a 2°C bottom temperature, a methane concentration of about 52 mmol/kg, approximately 1,200,000 ppm, would approach the levels required for the formation of gas hydrates.

The presence of bottom-simulating reflectors (BSR) along the crest of the Blake Outer Ridge and beneath the upper Continental Rise off New Jersey and Delaware has been interpreted as evidence for the presence of gas hydrates in the sediment (Tucholke and others, 1977). The reflectors examined by Tucholke and others (1977) follow the bottom sediment contours very closely, at a reflection time of about 0.6 sec subbottom, whereas the reflecting horizon on the diapir flank in this present study dips more sharply seaward than the bottom contour does at a reflection time of about

0.4-sec subbottom. Grow and others (1977) interpreted the diapir to be a salt dome and observed that the reflectors domed up around diapirs. Tucholke and others (1977) have suggested that salt may be an inhibitor to clathrate formation whereas Hunt (1979) has indicated that gas-hydrate sections may undergo thinning of as much as 50 percent over salt diapirs. Paull and Dillon (1980b) attributed the apparent doming of the reflectors to the fact that salt is a good thermal conductor and that heat flow through the diapir thus may be higher than the heat flow through the surrounding sediments. The increased heat flow would cause the hydrate-to-gas phase boundary to occur at a shallower depth in the sedimentary section and result in a thinning of the gas-hydrate section above the diapir. Tucholke and others (1977) also suggested that the reflector may be caused by minerals, such as ankerite or siderite, in the sediment. Paull and Dillon (1980b) believed, however, that the velocity structures observed cannot be explained by a thin layer of authigenic minerals and concluded that the BSR results primarily from a gas-hydrate layer.

The presence of gas hydrates in sediments may cause a decrease in permeability (Dillon and others, 1980). If the primary source of the gas is below the hydrate-formation zone, gas diffusing upward would become hydrated at the phase boundary, and gas diffusion toward shallower depths would be strongly retarded (Tucholke and others, 1977). The hydrate layer may then act as a seal and trap gases diffusing upward (Dillon and others, 1980; Paull and Dillon, 1980b). Core 35038 was collected over the southeast flank of the diapir and contained the greatest light-hydrocarbon concentrations (39 ppm), where the BSR was most pronounced on the seismic record. The BSR does not appear, however, to be present on the seismic record over the northwest flank of the diapir, and C_1 - C_4 concentrations in the two cores, 35034 and 35035, taken over this flank were the lowest concentrations in this transect. It is possible that the greater concentrations noted in the cores in this transect, particularly in cores 35036 and 35038 taken over the crest of the diapir and over the southeast flank, may be surface manifestations of slightly increased hydrocarbon-gas diffusion, which results from the thinning of the gas-hydrate layer. However, as the BSR is still present, it would appear unlikely that gas would diffuse to the surface through the hydrate seal, unless microfractures are present.

Gas concentrations from sediments taken during the diapir transect are slightly greater than concentrations measured in the other sampling sites. These concentrations are, however, several orders of magnitude below the critical concentration levels reported by Claypool and Kaplan (1974) as being necessary for methane-hydrate formation. Hydrotroilite was noted in the diapir transect cores associated with numerous small burrows (M. Ayers, written commun., 1979). The hydrotroilite may indicate the presence of anaerobic conditions in the sediment and may be indicative of a relatively higher rate of methanogenic microbial activity which may also explain the presence of the greater methane concentrations in the diapir transect cores. Zobell (1946, p. 94) reported that bacterial populations are closely related to the character of the sediment and that finer grained sediments generally contain a greater abundance of bacteria. In this respect, Ayers (written commun., 1978) pointed out that the finest grained sediments tend to be present in the northern and western extremities of the study area where the diapir transect is located. In addition, the relatively greater gas concentrations may be, in part, due to absorption of gas by the finer grained silty clay in this portion of the study area.

In this study, the methane-to-ethane-plus-propane ratios average 31.5, which are considered to be in the range of ratios characteristic of petrogenic gases (table 2). Bernard and others (1977) suggested that petroleum-related hydrocarbon gases generally have methane-to-ethane-plus-propane ratios of less than 50. The use of this ratio alone is inconclusive and should be compared to the $\delta^{13}C$ isotopic composition of the gas in order to be useful for identifying petrogenic seeps (Bernard and others, 1978). Unfortunately, gas concentrations were too low to collect sufficient gas for stable carbon isotope determinations. Several cores collected from surface sediments associated with channel site locations 34946 and 34954, with the diapir transect, core 35035, and with an area of subsurface faulting, core 34957, show a decrease in the methane-to-ethane-plus-propane ratio with burial depth. The reason for this decrease with very shallow increased burial depth is unknown. The ratio increased, however, with burial depth in the surface sediments associated with the remaining diapir transect sites, 35034, 35036, 35037, 35038, and in the channel core site location 34992. This increase would suggest that the ethane and propane are not diffusing from a more deeply buried reservoir and that these gases may have a microbial rather than a petroleum-related origin. R.E. Miller and others (unpublished data, 1981) also reported that ethane and propane concentrations relative to methane concentrations were greatest in the less deeply buried sediments and attributed this to a biologic origin for ethane, propane and butane. The presence of ethene may also suggest that at least a portion of the light hydrocarbons found in these sediments are biogenic rather than petrogenic.

Low concentrations of the higher permanent homologs ethane and propane may be formed during the microbial production of methane (Davis and Squires, 1954; Rheinheimer, 1974, p. 126). Bernard and others (1978) found that the distribution of ethane, propane, ethene, and propene was relatively constant with burial depth in Texas Continental Shelf and Slope surface sediments and concluded that the background concentrations of these hydrocarbons are controlled by microbial processes. Hunt and others (1980) suggested that pentanes as well as the lower alkane homologs, ethane through the butanes, may be biosynthesized in organisms or formed by decarboxylation of even-numbered carbon chains at temperatures less than 20°C.

The occurrence of low concentrations of ethane through butanes in this present study area would appear to be a result of biogenic production of at least a portion of these gaseous hydrocarbons. Evaluators of the petroleum potential from near-surface light-hydrocarbon data must therefore consider both the background levels in the area under study and the possibility that the occurrence of low concentrations of light hydrocarbons, methane through the higher homologs propane, butane, and pentane, may be dependent upon biological precursors as well as upon petroleum-related sources.

SUMMARY AND CONCLUSIONS

The microbial production of methane has been well established. Bacterial production of the higher homologs ethane, propane, butanes and pentanes has been suggested by several investigators on the basis of field observations but has yet to be demonstrated in the laboratory. Although a great deal of work still needs to be done to explain the presence of low concentrations of these higher homologs in the sediment, a few general statements may be made to summarize the results of this present study.

1) Surface sediments from the Florida-Hatteras Slope and Blake Plateau contained less than 40 ppm total light hydrocarbons; methane, ethene, and ethane were present in most samples, and permanent homologs higher than ethane were detected in several cores.

2) The higher homologs ethane, propane, butane, and isobutane were detected in the gas samples from surface sediments associated with a channel feature. Total C_1 - C_4 concentrations in the cores from this sampling area were less than 26 ppm.

3) Gases found in cores from a transect across the location of a possible salt diapir contained the greatest methane concentrations, 38.34 ppm, and ethane and propane were present.

4) The low residual-light-hydrocarbon concentrations are probably gases dissolved in the sediment pore water and represent background levels for this study area. Although methane-to-ethane-plus-propane ratios were generally in the range of petrogenic gas values, no consistent and direct evidence was found in the surface sediments to suggest diffusion of gas from deeper reservoirs in any of the geologic features and geophysical anomalies

examined. The presence of the unsaturated hydrocarbon ethene in the majority of sediment samples and the extremely low concentration levels of ethane, propane, and the butanes may suggest a microbial origin of the associated saturated gaseous hydrocarbon homologs from organic matter in the sediment, rather than gases diffusing from deeper petroleum or natural gas reservoirs.

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TABLE 1

Station locations and descriptions of samples collected during cruise onboard R/V EASTWARD, April 2-13, 1978

Station Number	Station Location	Water Depth	Sample Interval	Core Sediment Description[1]
34946	29°57.3'N 79°55.9'W	575 m	250-270 cm 530-550 cm	greenish gray, slightly muddy foram sand with green-black glauconite
34954	30°14.2'N 79°44.7'W	620 m	40-70 cm 240-270 cm 450-466 cm	gray, slightly sandy, silty clay
34956	30°48.7'N 79°31.8'W	805 m	0-23 cm	forams and glauconite, sandy gravel, phosphorite nodules
34957	30°50.7'N 79°30.3'W	790 m	0-30 cm 132-147 cm	yellowish gray muddy form-rich sandy coral gravel
34958	30°54.1'N 79°28.1'W	755 m	230-240 cm	yellowish tan coarse sandy mud
34959	30°59.9'N 79°37.0'W	480 m	200-220 cm	pale olive muddy coarse sand, coral gravel
34960	31°04'N 79°30'W	680 m	426-454 cm	greenish gray, muddy fine sand
34964	31°47.3'N 79°16'W	300 m	170-200 cm	grayish olive slightly sandy, silty clay, coral gravel
34970	31°58.4'N 79°00.5'W	400 m	110-140 cm	light olive brown very dense silty clay, phosphorite nodules
34973	32°24.5'N 78°34.2'W	340 m	90-120 cm 160-190 cm	moderate olive brown to grayish olive silty clay
34974	32°23.5'N 78°33'W	360 m	200-230 cm	dusky yellow green, silty fine sand, numerous shell fragments, glauconite

TABLE 1

Station locations and descriptions of samples collected during cruise onboard R/V EASTWARD, April 2-13, 1978

Station Number	Station Location	Water Depth	Sample Interval	Core Sediment Description[1]
34984	32°40.1'N 77°25.4'W	430 m	180-210 cm	grayish olive muddy medium sand, numerous small phosphorite nodules
34988	32°35.6'N 77°32.4'W	450 m	60-90 cm 200-230 cm	dusky yellow green muddy fine sand, glauconite
34989	32°35'N 77°33.9'W	420 m	100-125 cm	grayish olive fine muddy foram-rich sand
34991	32°36.5'N 77°39.5'W	390 m	50-80 cm 130-160 cm 220-250 cm	moderate olive-brown muddy fine sand
34992	32°35.6'N 77°38.7'W	420 m	140-170 cm 230-260 cm	grayish olive muddy sand
35000	32°30.8'N 77°59.9'W	300 m	224-244 cm	yellowish gray slightly muddy sand
35007	32°10.3'N 78°40.8'W	420 m	50-60 cm	yellowish gray muddy silt
35034	32°38'N 76°33.2'W	1050 m	39-55 cm 288-304 cm 460-476 cm 578-594 cm	grayish olive silty clay
35035	32°34.4'N 76°21.4'W	2020 m	110-126 cm 296-312 cm 554-573 cm	olive gray, dense silty clay, hydrotroilite mottling associated with burrowing
35036	32°30.3'N 76°11.7'W	2100 m	124-140 cm 277-293 cm 427-443 cm 560-576 cm	olive gray, dense silty clay, hydrotroilite mottling associated with burrows

TABLE 1

Station locations and descriptions of samples collected during cruise onboard R/V EASTWARD, April 2-13, 1978

35037	32°26.7'N 76°02.5'W	2360 m	10-26 cm 306-322 cm 542-561 cm	grayish olive silty clay hydrotroilite mottling associated with burrows
35038	32°28'N 76°08.8'W	2220 m	10-29 cm 294-310 cm 556-572 cm	grayish olive silty clay hydrotroilite mottling associated with burrows

[1]Ayers, written communication, 1978, 1979; and Ayers and Pilkey, 1981.

TABLE 2

LIGHT-HYDROCARBON CONCENTRATIONS[a] AND RATIOS OF METHANE-TO-ETHANE-PLUS-PROPANE DETERMINED ON CORED SEDIMENT SAMPLES FROM FLORIDA-HATTERAS SHELF AND SLOPE AND BLAKE PLATEAU

Station	Interval	Core[b] Type	Methane (CH_4)	Ethene (C_2H_4)	Ethane (C_2H_6)	Propane (C_3H_8)	Isobutane (iC_4H_{10})	Butane (C_4H_{10})	Total $C_1 - C_4$	$\frac{C_1}{C_2 + C_3}$
					CHANNEL					
34946	250-270	lined	2.81	0.10	0.09	ND[c]	ND	ND	3.00	31.2
	530-550		1.89	0.01	0.23	ND	ND	ND	2.13	8.2
34954	40-70	unlined	5.83	0.04	0.11	ND	ND	ND	5.98	53.0
	240-270		3.84	0.09	0.14	0.15	ND	ND	4.22	13.2
	450-466		1.59	0.05	0.11	0.31	ND	0.08	2.14	3.8
34973	90-120	unlined	2.70	0.09	0.08	0.02	ND	ND	2.89	27.0
	160-190		2.54	0.10	0.08	ND	ND	ND	2.72	31.8
34976	200-230	unlined	1.94	0.10	0.07	ND	ND	ND	2.11	27.7
34988	60-90	unlined	12.92	0.22	2.03	1.82	1.60	0.82	19.41	3.4
	200-230		9.30	0.11	1.42	1.15	1.50	0.49	13.97	5.9
34989	100-125	lined	0.51	0.01	0.01	ND	ND	ND	0.53	51.0
34991	50-80	unlined	16.53	0.25	2.65	2.19	1.90	1.10	24.62	3.4
	130-160		11.43	0.13	1.97	1.67	1.48	0.87	17.55	3.1
	220-250		17.06	0.24	2.75	2.30	1.78	1.27	25.40	3.4
34992	140-170	unlined	13.55	0.18	1.49	1.05	0.74	0.45	17.47	5.3
	230-260		0.40	ND	0.01	ND	ND	ND	0.42	40.0
					SLUMP MASS					
34964	170-200	unlined	3.68	0.17	0.12	0.18	ND	ND	4.15	12.3
34970	110-140	unlined	0.86	ND	0.02	0.06	ND	ND	0.94	10.8
					ACCRETIONARY WEDGE					
34984	180-210	unlined	1.86	0.06	0.11	0.03	ND	ND	2.06	13.3
					DIAPIR TRANSECT					
35034	39-55	lined	15.10	0.71	0.57	1.71	ND	ND	18.09	6.6
	288-304		4.95	0.15	0.16	0.23	ND	ND	5.49	12.7
	460-476		0.94	0.08	0.07	0.05	ND	ND	1.14	7.8
	578-594		ND	ND	ND	ND	ND	ND	ND	--
35035	110-126	lined	2.88	0.02	0.03	ND	ND	ND	2.93	96.0
	296-312		5.22	0.01	0.06	0.04	ND	ND	5.33	52.2
	554-573		6.67	0.06	0.09	0.55	ND	ND	7.37	10.4
35036	124-140	lined	7.59	0.17	0.11	ND	ND	ND	7.87	69.0
	277-293		23.02	0.33	0.27	0.15	ND	ND	23.77	54.8
	427-443		23.78	0.17	0.28	ND	ND	ND	24.23	84.9
	560-576		36.61	0.08	0.22	ND	ND	ND	36.91	166.4
35037	10-26	lined	26.67	0.77	1.03	0.54	ND	ND	29.09	17.0
	306-322		6.71	0.10	0.12	0.30	ND	ND	7.23	15.9
	542-561		11.76	0.58	0.47	0.13	ND	ND	12.94	19.6
35038	10-29	lined	20.33	0.36	0.59	0.26	ND	ND	21.54	23.9
	294-310		11.25	0.15	0.17	0.14	ND	ND	11.71	36.3
	556-572		38.34	0.18	0.40	0.05	ND	ND	38.97	85.2
					FAULTING					
34956[d]	0-23	lined	0.85	0.01	0.05	ND	ND	ND	0.91	17.0
34957	0-30	lined	7.87	1.10	0.40	ND	ND	ND	9.37	19.7
	132-147		1.24	ND	0.07	0.13	ND	ND	1.44	6.2
34958[d]	230-240	lined	0.34	ND	ND	ND	ND	ND	0.34	--
35000	224-244	lined	0.57	0.01	0.01	ND	ND	ND	0.59	57.0
					WATER COLUMN ACOUSTIC ANOMALY					
35007	50-60	lined	0.76	ND	0.01	ND	ND	ND	0.77	76.0
34959[d]	200-220	unlined	0.50	ND	ND	ND	ND	ND	0.50	--
					PALEOCENE BOTTOM EXPOSURE					
34960	426-454	unlined	3.71	0.24	0.19	0.16	ND	ND	4.30	10.6

[a]Concentrations reported in ppm based on volume of gas/volume of sediment, determined following a 5-minute blender extraction, except where noted.

[b]Cores were collected either using a plastic core liner in the core barrel or without the liner.

[c]ND = Below detection limits.

[d]Concentrations determined following a 10-minute shaker extraction.

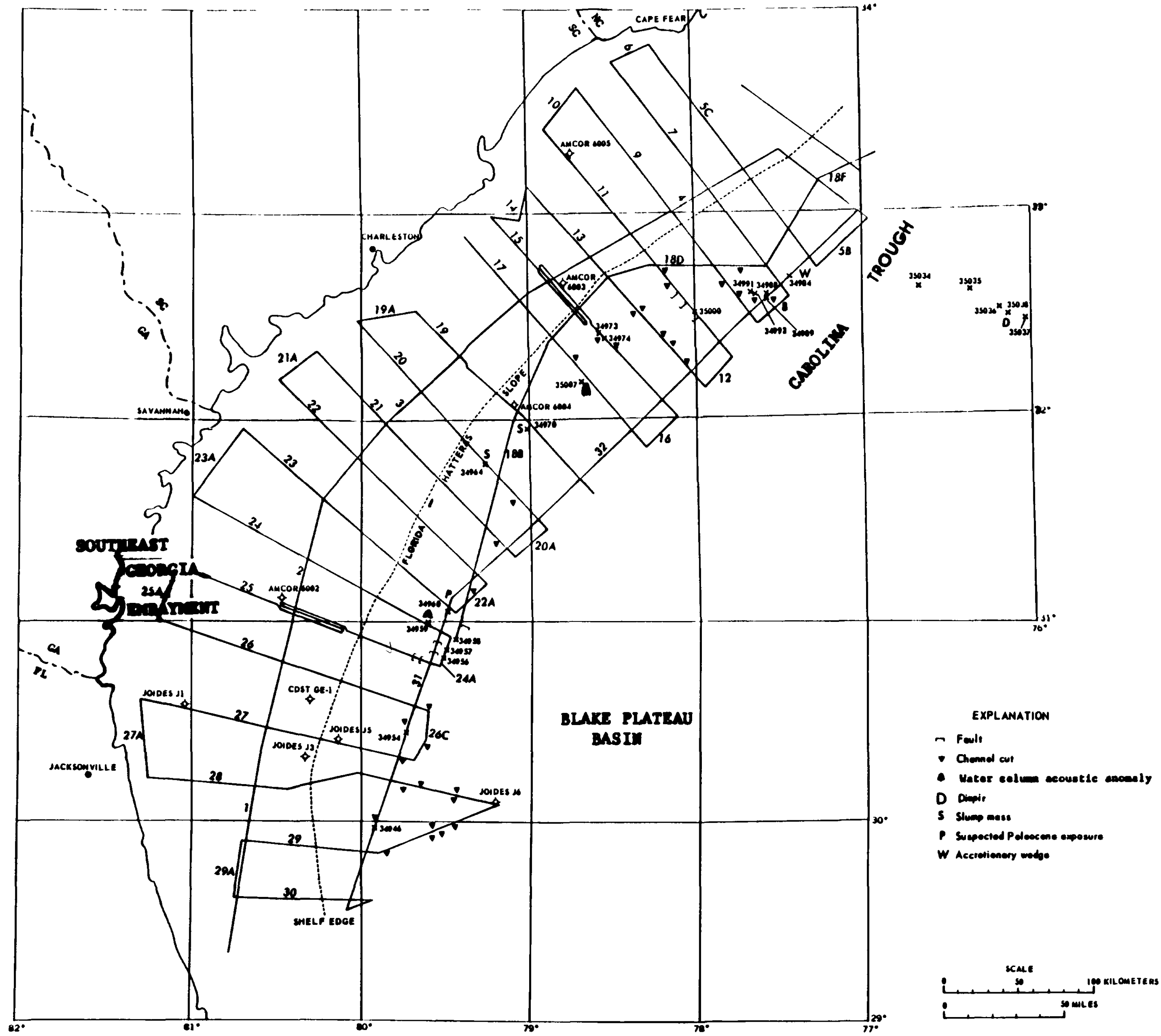

Figure 1. Track lines of seismic profiles and station locations of piston cores on the Florida-Hatteras Slope and Blake Plateau. Figure also shows major depositional basins.

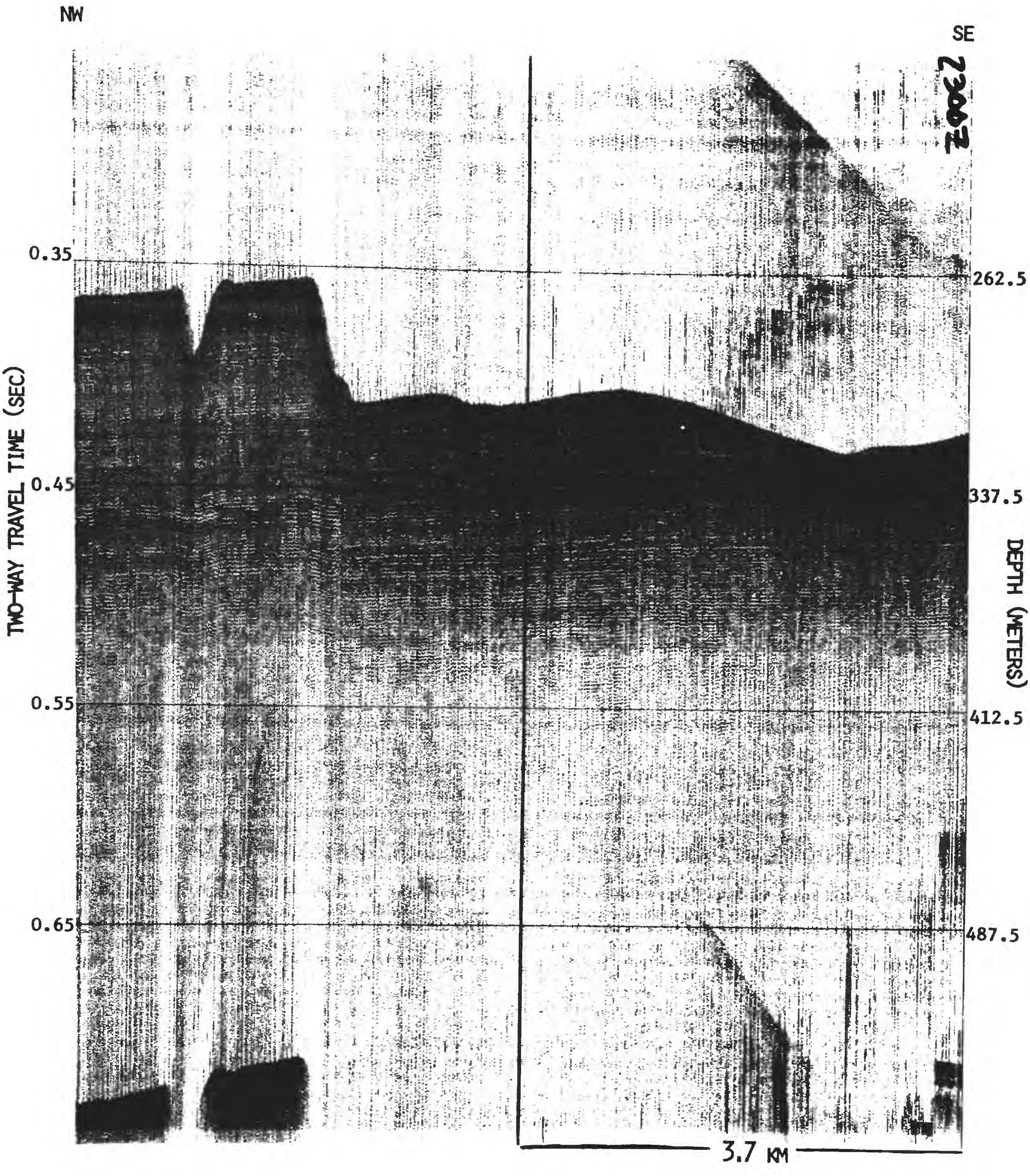

Figure 2. Typical erosional nature of channel cutting on the inner Blake Plateau, seismic line 9, near core 34988, 34989, 34991, and 34992. Profile from Edsall (1978). Location of seismic line 9 is shown in figure 1.

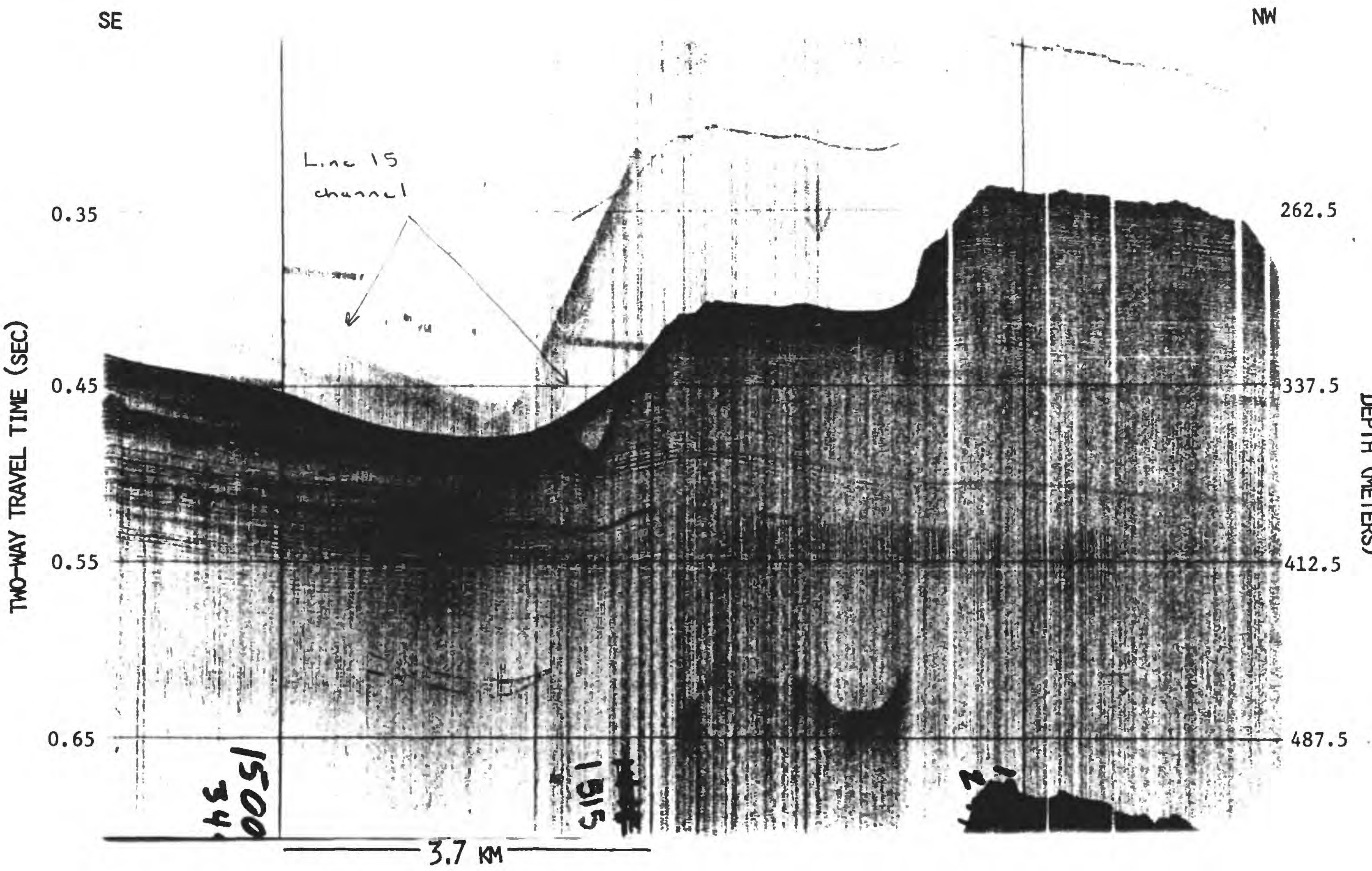

Figure 3. Erosional nature of channel cutting on the inner Blake Plateau, seismic line 15, near core sites 34973 and 34974. Profile from Edsall (1978). Location of seismic line 15 is shown in figure 1.

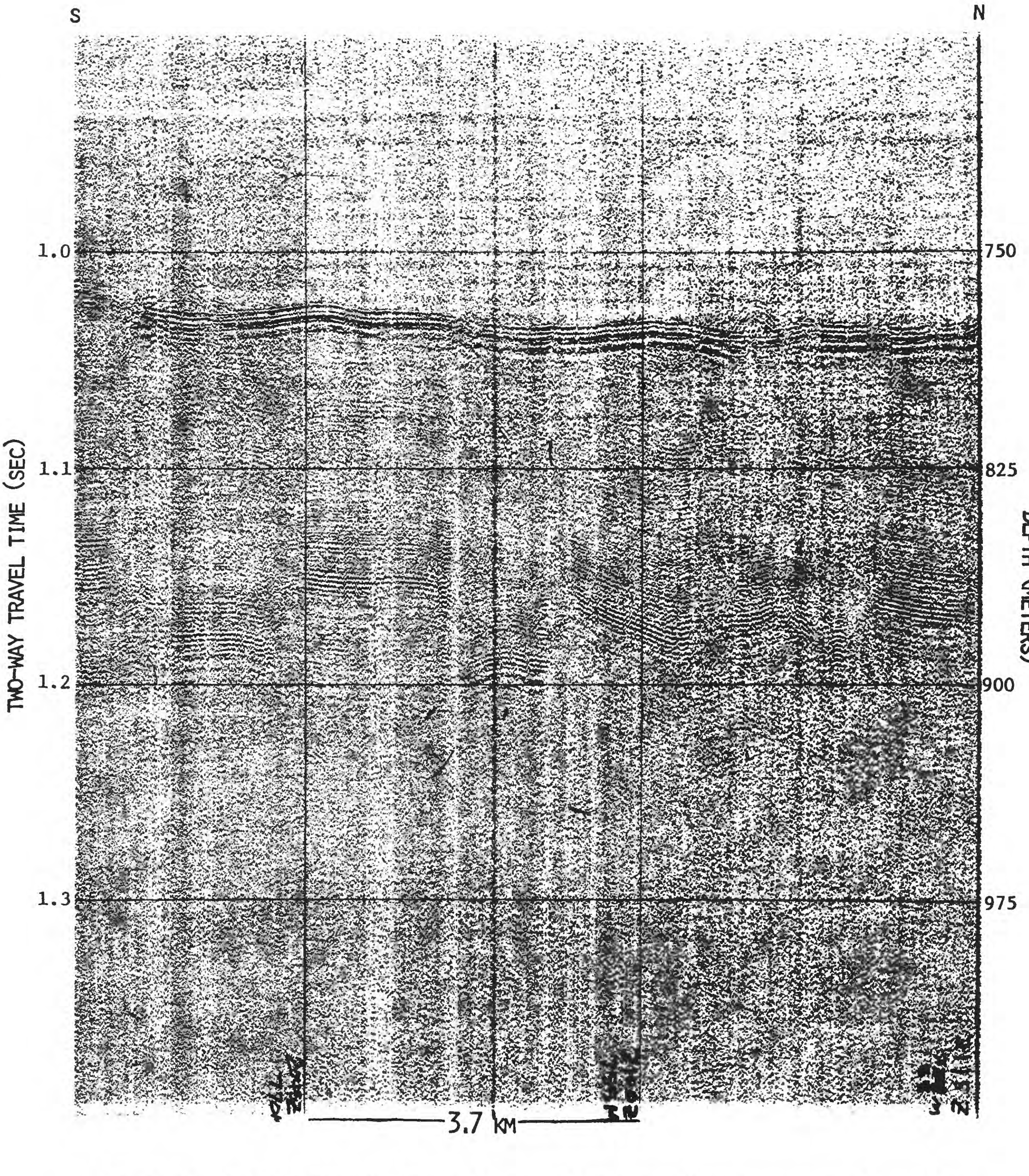

Figure 4. Seismic record showing features interpreted to represent sub-surface faults, line 24A, near core sites 34956, 34957, and 34958. Profile from Edsall (1978). Location of line 24A is shown in figure 1.

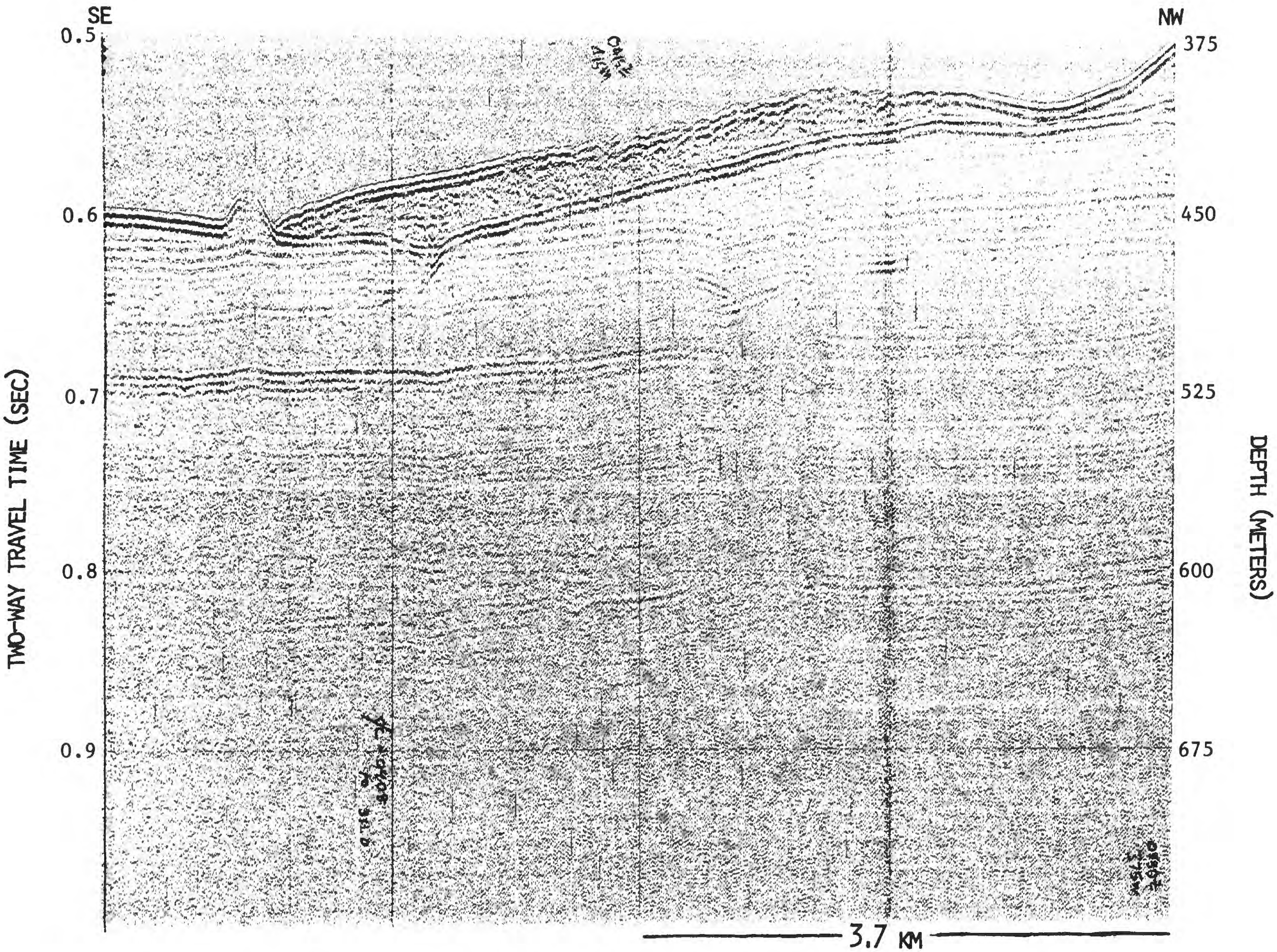

Figure 5. Seismic record showing a feature interpreted to represent a slump mass, seismic line 19, near core site 34970. Profile from Edsall (1978). The feature overlies several shallow faults. The location of seismic line 19 is shown in figure 1.

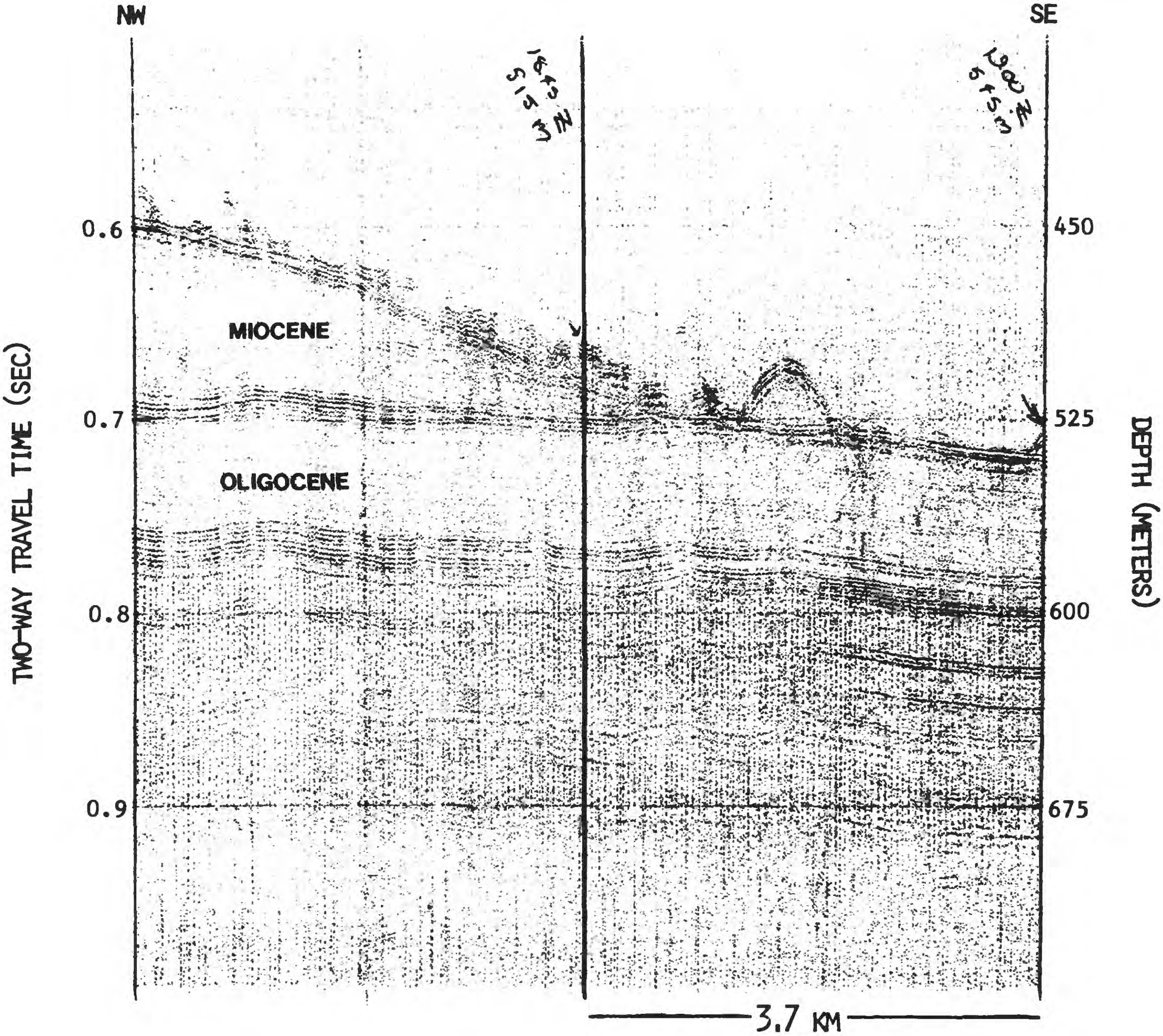

Figure 6. Part of seismic profile 24 showing water-column acoustic anomalies on Blake Plateau interpreted as deep-water coral mounds or reefs, near core site 34959. Profile from Edsall (1978). The location of line 24 is shown in figure 1.

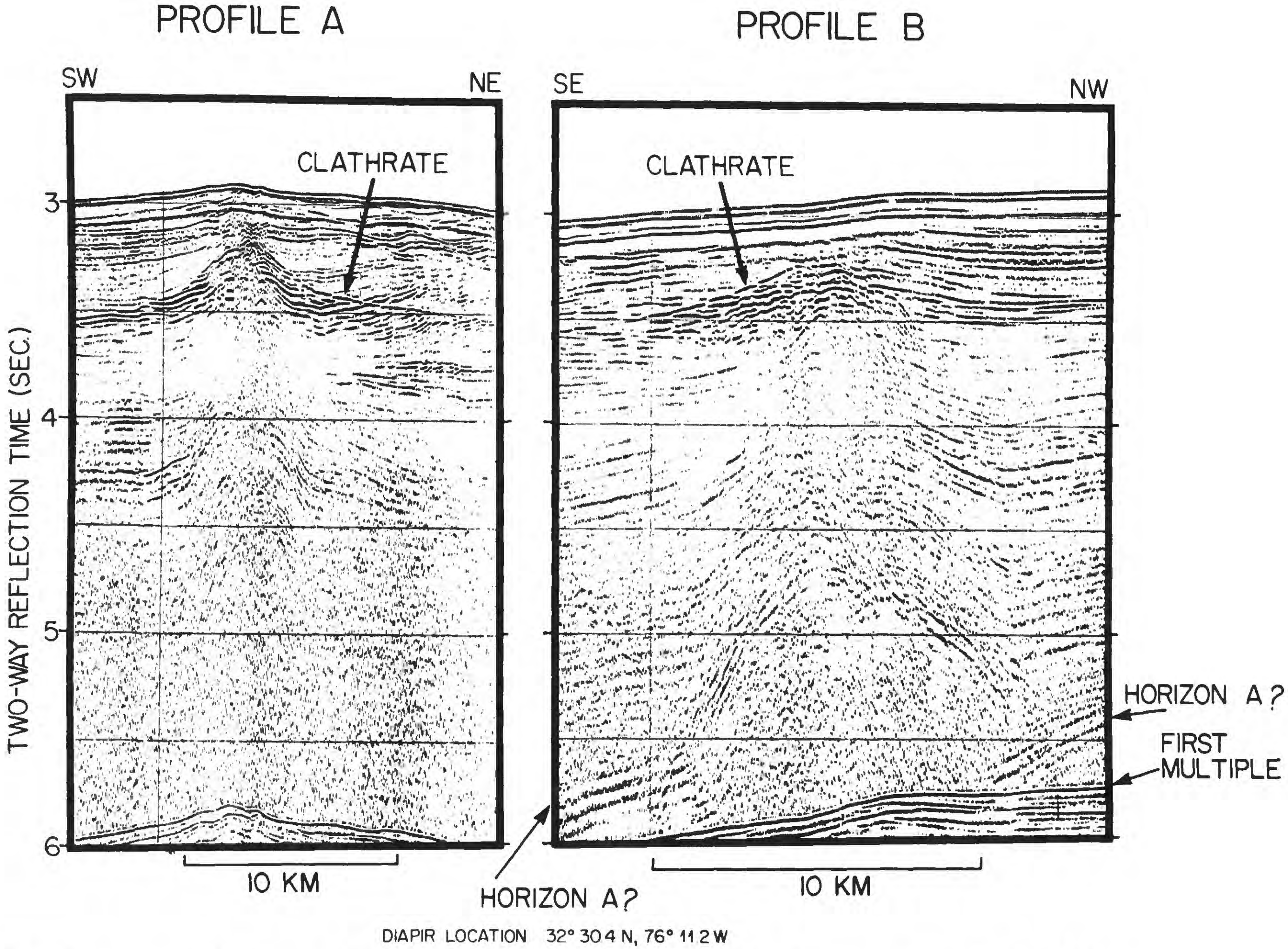

Figure 7. Two perpendicular seismic records of diapir on Blake Plateau, near core sites 35034, 35035, 35036, 35037, and 35038 (fig. 1). Reflector interpreted as the base of a possible clathrate layer is indicated. Profiles from Paull and Dillon (1980b). Strong reflections beneath the reflector have been interpreted as possible trapped free gas or gas-saturated pore water (Paull and Dillon, 1980b).

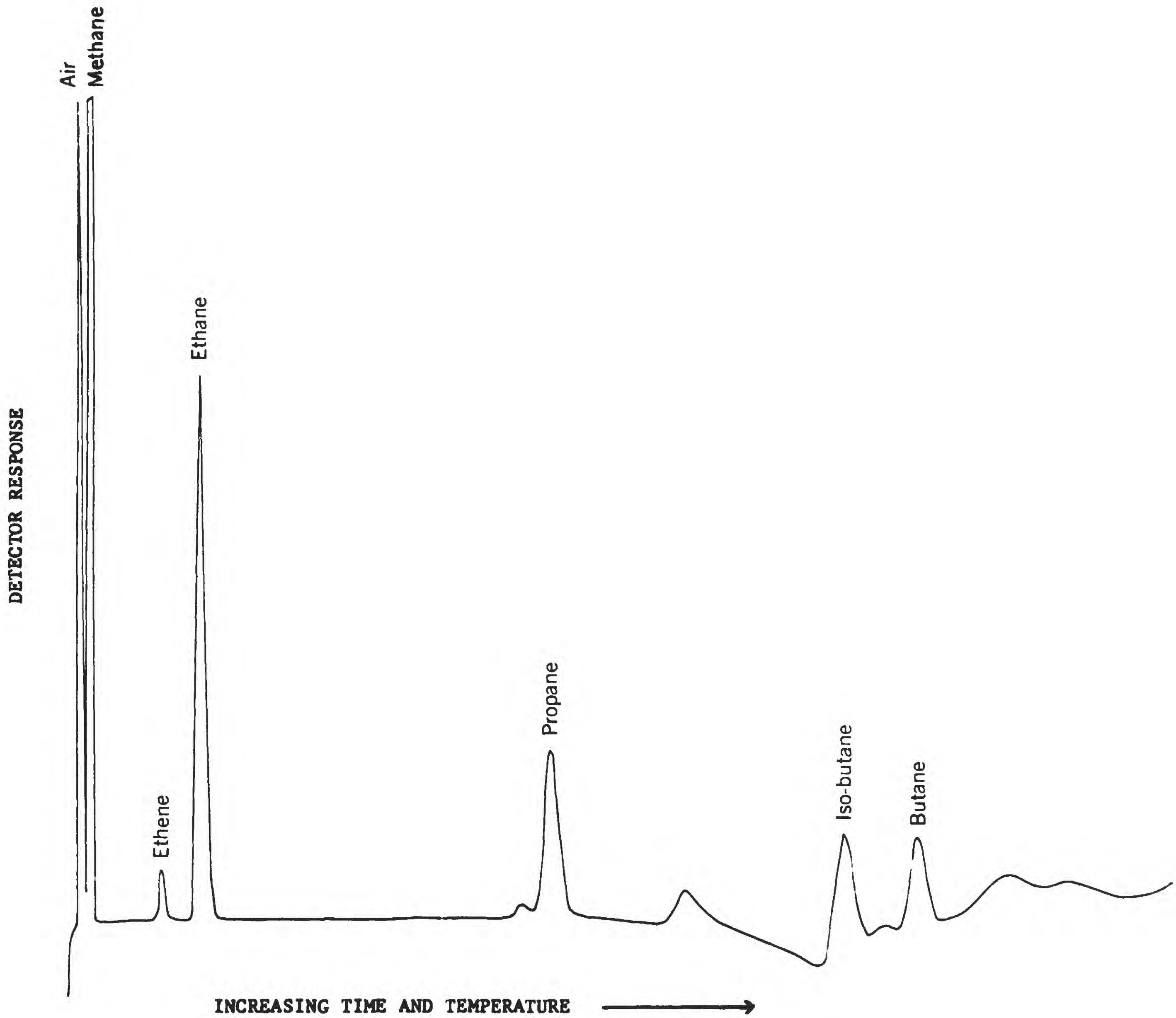

Figure 8. Gas chromatographic analysis of light hydrocarbons in the 130-160 cm depth interval of core 34991. The core was taken from the floor of a channel feature.

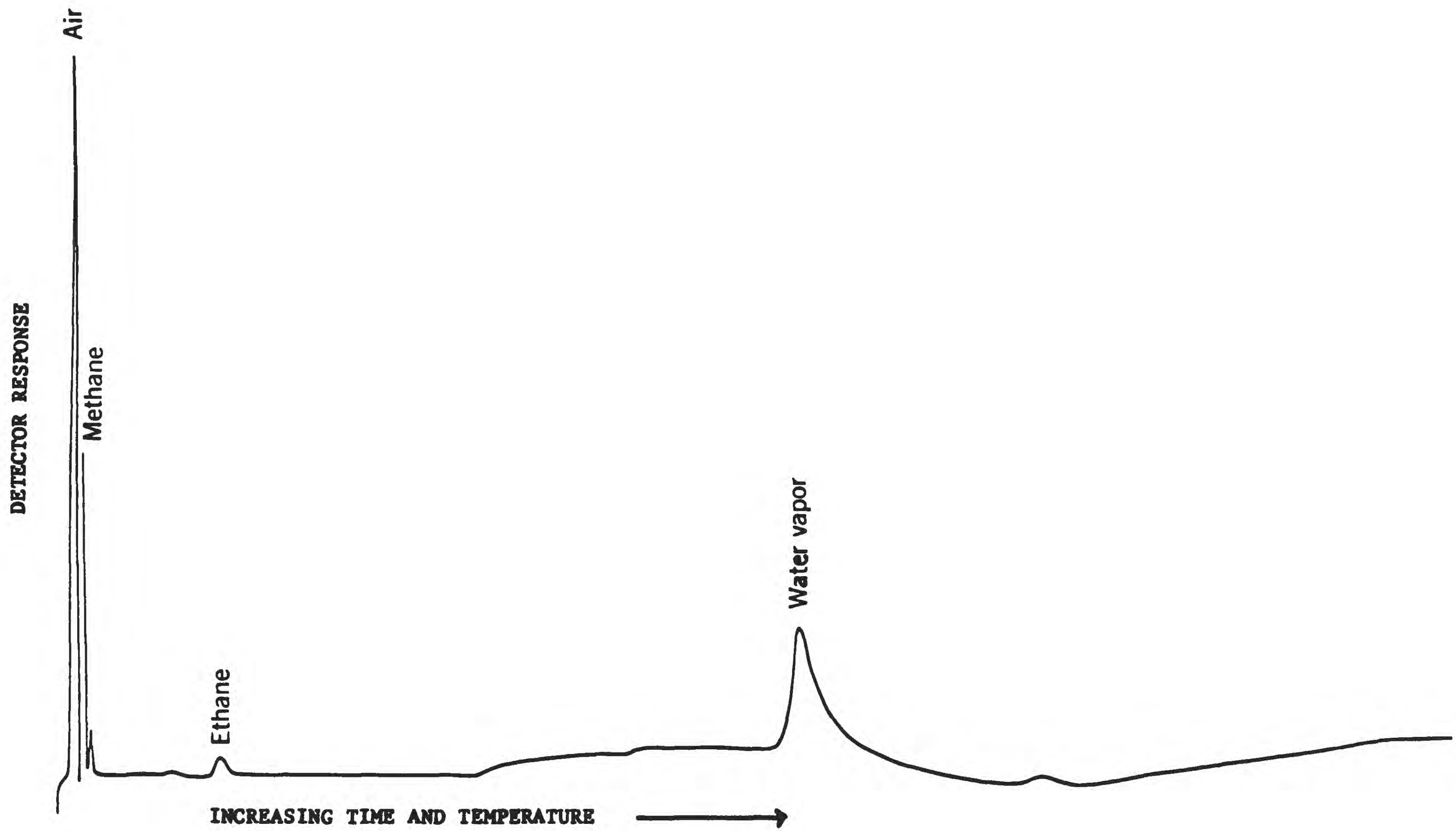

Figure 9. Gas chromatographic analysis of light hydrocarbons in the 224-244 cm depth interval of core 35000 which consisted of sediments overlying a fault zone.

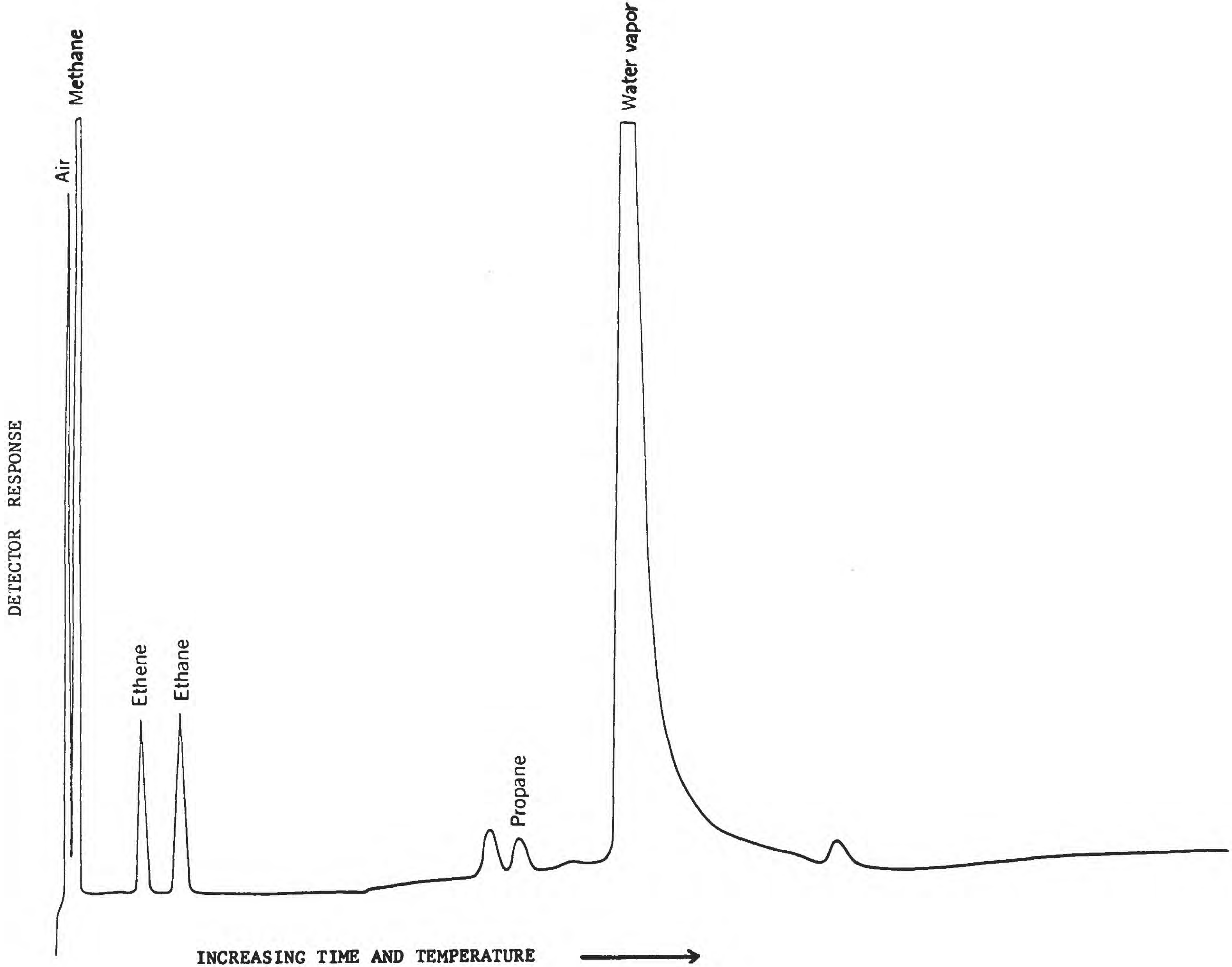

Figure 10. Gas chromatographic analysis of light hydrocarbons in the 10-29 cm depth interval of core 35037 from the diapir transect.