Spring 1983

Input and Distribution of Sewage Derived Sedimentary Material Adjacent to Chesapeake-Elizabeth Sewage Outfall, Virginia Beach, Virginia

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INPUT AND DISTRIBUTION OF SEWAGE DERIVED SEDIMENTARY MATERIAL ADJACENT TO CHESAPEAKE-ELIZABETH SEWAGE OUTFALL, VIRGINIA BEACH, VIRGINIA

by

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B.S., May 1978, Virginia Commonwealth University

A Thesis Submitted to the Faculty of
Old Dominion University in Partial Fulfillment of the
Requirements for the Degree of

MASTER OF SCIENCE

OCEANOGRAPHY

OLD DOMINION UNIVERSITY
May 1983

Approved by:

Terry L. Wade (Director)

George F. Oertel

George T.F. Wong

ABSTRACT

INPUT AND DISTRIBUTION OF SEWAGE DERIVED SEDIMENTARY MATERIAL ADJACENT TO CHESAPEAKE-ELIZABETH SEWAGE OUTFALL, VIRGINIA BEACH, VIRGINIA

Robert Carroll Brown
Old Dominion University, 1983
Director: Dr. Terry L. Wade

The concentrations of coprostanol and hydrocarbons were measured in the effluent from the Chesapeake-Elizabeth sewage treatment plant and surface sediments from the area surrounding the effluent discharge site. Most of the coprostanol (>84%) and hydrocarbons (>91%) were associated with particulates in the effluent. Some of these particles were incorporated into the sedimentary column within the study area, while some may have escaped from the area.

The study area is found to be a dynamic area where changes in the percentage and distribution of fine-grained sediments occur over periods of months. The movement of fine-grained sediments is an important determinant of the distribution of sewage derived contaminants.

The Chesapeake-Elizabeth STP was responsible for, at most, 7% of the hydrocarbon contamination of the sediments in the study area, however, the STP is not a major source (<1%) of the fine-grained sediments in the study area. The distribution of hydrocarbons suggest that the Bay

Bridge Tunnel may be a unique source of hydrocarbons to the lower Chesapeake Bay. This study shows the usefulness of coprostanol in providing a better understanding of the fate and importance of sewage derived contaminants in the area around sewage outfalls.

ACKNOWLEDGEMENTS

I would like to express my deepest appreciation to Dr. Terry L. Wade, my thesis advisor, whose intellectual guidance and sincerest friendship aided in the completion of this investigation. A special thanks is due Drs. George T.F. Wong and George F. Oertel for their expert guidance and critical review of this manuscript.

I would like to thank the crew of the R/V Linwood Holton, David Velinsky, and David Webber for their assistance in sample collection; the staff of Hampton Roads Sanitation District for collecting effluent samples and providing information about Chesapeake-Elizabeth STP; and a special thanks to Mark Byrnes for his assistance with the grain size analysis.

A very special thanks go to my mother, father, and sister whose moral encouragement and financial support was invaluable throughout my studies.

Finally, I would like to dedicate this work to my wife, Nancee, without her steady encouragement and loving support none of this would have been possible.

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

Chapter 1

INTRODUCTION

In the United States, the current trend is toward collection and centralized treatment of municipal waste. In most large urban areas, this waste receives some treatment and is then discharged into rivers, estuaries, and coastal waters via sewage outfalls. Most sewer systems service various industries and storm drainage systems besides servicing individual homes. As a result, the influent to sewage treatment plants may contain many materials including heavy metals, pesticides, and petroleum hydrocarbons, as well as, pathogenic bacteria and viruses associated with human wastes (Geldrich, 1972; Metcalf and Eddy, 1979; Van Vleet and Quinn, 1977; Burlingame et al., 1972). Even secondary sewage treatment does not remove all of these contaminants (Metcalf and Eddy, 1979; Van Vleet and Quinn, 1977; Burlingame et al., 1972; Dutka et al., 1974). Therefore, it is important to delineate areas within aquatic systems that may be adversely influenced by sewage discharges.

Sewage contamination is routinely determined by enumeration of fecal coliform bacteria (Metcalf and Eddy, 1979; Tabak et al., 1972; Smith and Gouron, 1969),

because coliform bacteria are thought to be specific to sewage, present in large quantities, and easy and inexpensive to quantify. Recent studies (Dutka et al., 1974; Goodfellow et al., 1977; Loh et al., 1979) describe the limitations of the coliform test as an adequate indicator of sewage contamination in aquatic environments (i.e. rapid bacterial death due to environmental stress).

The inadequacy of the method has led researchers to investigate other parameters that may be more accurate indicators of the fate of sewage associated contaminants. One of the promising alternatives is the fecal sterol, coprostanol. Coprostanol (5B-cholestan-3B-ol) is thought to be formed exclusively by stereospecific bacterial reduction of cholesterol in man and higher animals and is one of the principle sterols found in their feces (Rosenfield et al., 1954; Rosenfield and Gallagher, 1964; Teshima and Kanazawa, 1978; Murtaugh and Bunch, 1967). Unlike its precursor cholesterol, coprostanol is not a naturally occurring sterol in aquatic systems, and its detection indicates contamination form sewage or runoff from pastures and barnyards (Murtaugh and Bunch, 1967). Therefore, coprostanol isolation and identification may be used to compliment the routine coliform test.

Coprostanol has been shown to be a reliable marker of fecal pollution, even when coliform bacteria may have been destroyed through various processes (i.e. chlorination, addition of heat, etc.; Tabak et al., 1972;

Goodfellow et al., 1977). These processes seem to have no effect on the structural configuration of coprostanol and do not interfere with the analysis of the fecal sterol. This is critical, because various contaminants, including viruses, are also unaffected by these processes and may go undetected by the conventional fecal coliform method. It has also been shown that once the effluent has reached the marine environment, the water quality analysis, by coliform enumeration, may be tenuous because of rapid bacterial death. Loh et al. (1979) have shown that 90% of coliform bacteria entering Mamala Bay, Hawaii were destroyed in less than one hour. The destruction of 90% of various enteric viruses required approximately 48 hours. This suggests that the absence of coliform bacteria would not indicate the absence of human enteric viruses in the ocean environment. Coprostanol, however, has been found to be fairly resistant to microbial degradation in the marine environment and, therefore, can provide information on cumulative loading, the historical influx to aquatic systems, and the fate of sewage associated contaminants (Dutka et al., 1974; Hatcher and McGillivary, 1977, 1979).

Measurements to determine water quality of aquatic systems are traditionally made in the water column. Many contaminants associated with sewage effluents are known to readily adsorb to particulates once they enter the marine environment. They may settle out and concentrate

in the sediments. Van Vleet et al. (1977) noticed this trend for petroleum hydrocarbons being discharged from a sewage treatment plant. They reported that half of the hydrocarbons was deposited near the outfall and the other half was removed from the discharge area. Pathogenic microorganisms and viruses have also been shown to adsorb to particulates and concentrate in the sediments. In the sediment these organisms remain viable and, in some cases, reproduce in predator-free marine sediments (LaBelle and Gerba, 1979). Matson et al. (1978) reported a consistently higher concentration of microorganisms in the sediments than in overlying waters. In some instances they reported that the amount of bacterial indicators in the sediment was sufficient to create a potential health hazard. Therefore, in addition to water analysis, sediment analysis must be performed when trying to determine if a health hazard exists. Coprostanol has been used as an indicator of fecal contamination in both water and sediment samples (Dutka et al., 1974; Tabak et al., 1972; Murtaugh and Bunch, 1967; Kanazawa and Teshima, 1978; Hatcher and McGillivary, 1979). McCally et al. (1980) have shown that more than 95% of the coprostanol found in sewage effluent and in the marine environment is associated with particulates. Coprostanol associated with particulates may rapidly settle out near the point of discharge or may be transported away from the discharge site, depending upon localized conditions (i.e. currents,

tides, particle size, etc.). Therefore, coprostanol is a valuable tracer of sewage contaminated materials in aquatic systems and is particularly important where the discharge of sewage contaminated materials may impact living marine resources or cause a public health hazard. Studies utilizing coprostanol may aid in gaining information about sediment transport and sedimentation patterns in the area of sewage discharges. This information is valuable for making managerial decisions regarding the siting of sewage outfalls and modifications of existing discharges.

With the increased discharge of treated wastes from our environment into aquatic systems, it is extremely important to determine where the contaminants are going. Many of these contaminants are insoluble in water and are found to readily associate with suspended particulate matter. Once discharged into the marine environment, these particles may flocculate and settle out or be carried away by currents. The distribution of this material is difficult to determine using traditional techniques. Coprostanol, on the other hand, has been known to serve as an adequate indicator in such cases. Therefore, the sewage specific indicator, coprostanol, was employed to determine the distribution of sewage associated material in sediment adjacent to Chesapeake-Elizabeth sewage outfall, Virginia Beach, Virginia. The importance of this

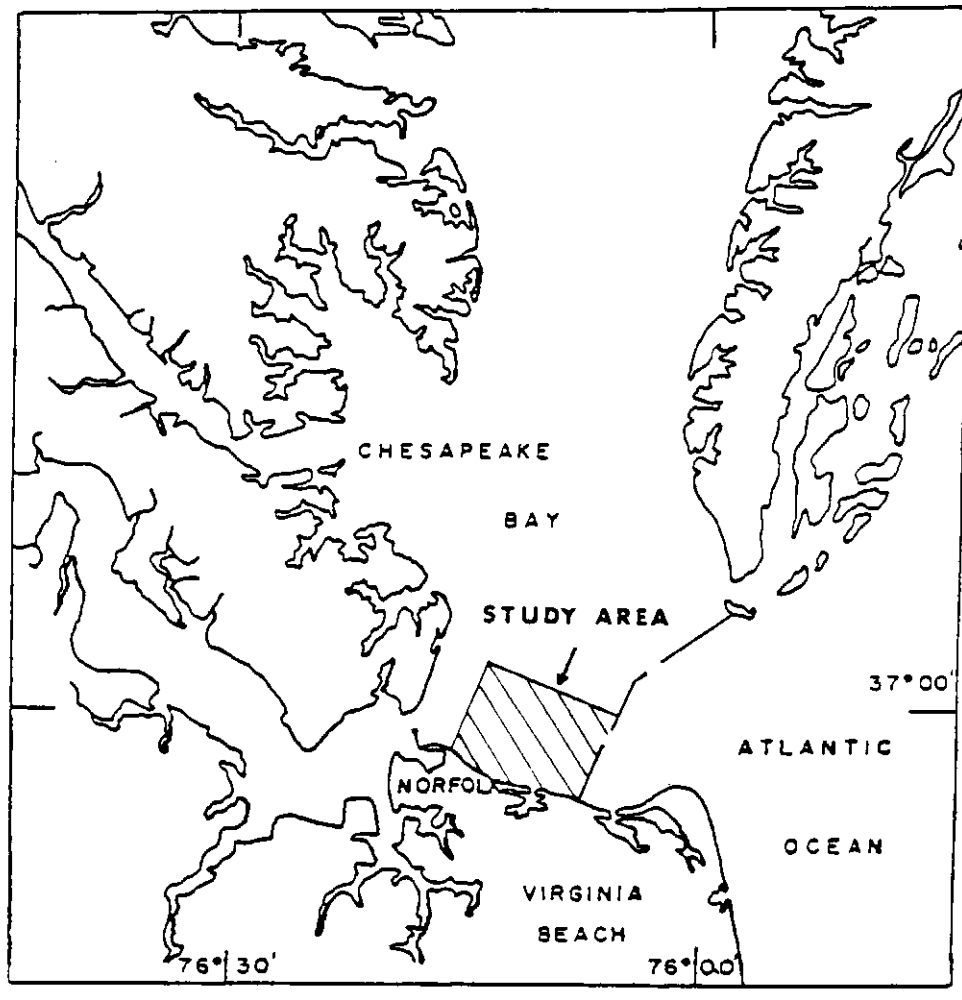
sewage outfall as a source of anthropogenic hydrocarbons to the study area could then be assessed.

Chapter 2

STUDY AREA

The area chosen for this study is approximately 125 km^2 (Figure 1). It is adjacent to the Chesapeake-Elizabeth sewage outfall (Hampton Roads Sanitation District) which is located north of Little Creek Harbor, Virginia Beach, Virginia at 36°56'10" north latitude and 76°10'35" west longitude. This area was chosen to minimize possible influences from other sources of input. Other sources to this area include the influence from James River water entering from the west, the discharge of sewage from the large number of coal colliers moored in the bay east of the Chesapeake Bay Bridge Tunnel, small spills in the channel, and atmospheric deposition.

The northern portion of the study area is defined by Thimble Shoal Channel which is a major navigational waterway in lower Chesapeake Bay. This waterway provides access for large ships from the Atlantic Ocean to upper Chesapeake Bay and the ports of Norfolk and Hampton Roads, Virginia. Water depth within the study area averages approximately 7.6 m (25 ft.) with a maximum depth of approximately 13.7 m (45 ft.) in Thimble Shoal Channel (Ludwick, 1981).


CHESAPEAKE
BAY
STUDY AREA
37°00'
ATLANTIC
OCEAN
NORFOLK
VIRGINIA
BEACH
76°30'
76°00'

Chesapeake-Elizabeth Sewage Treatment Plant

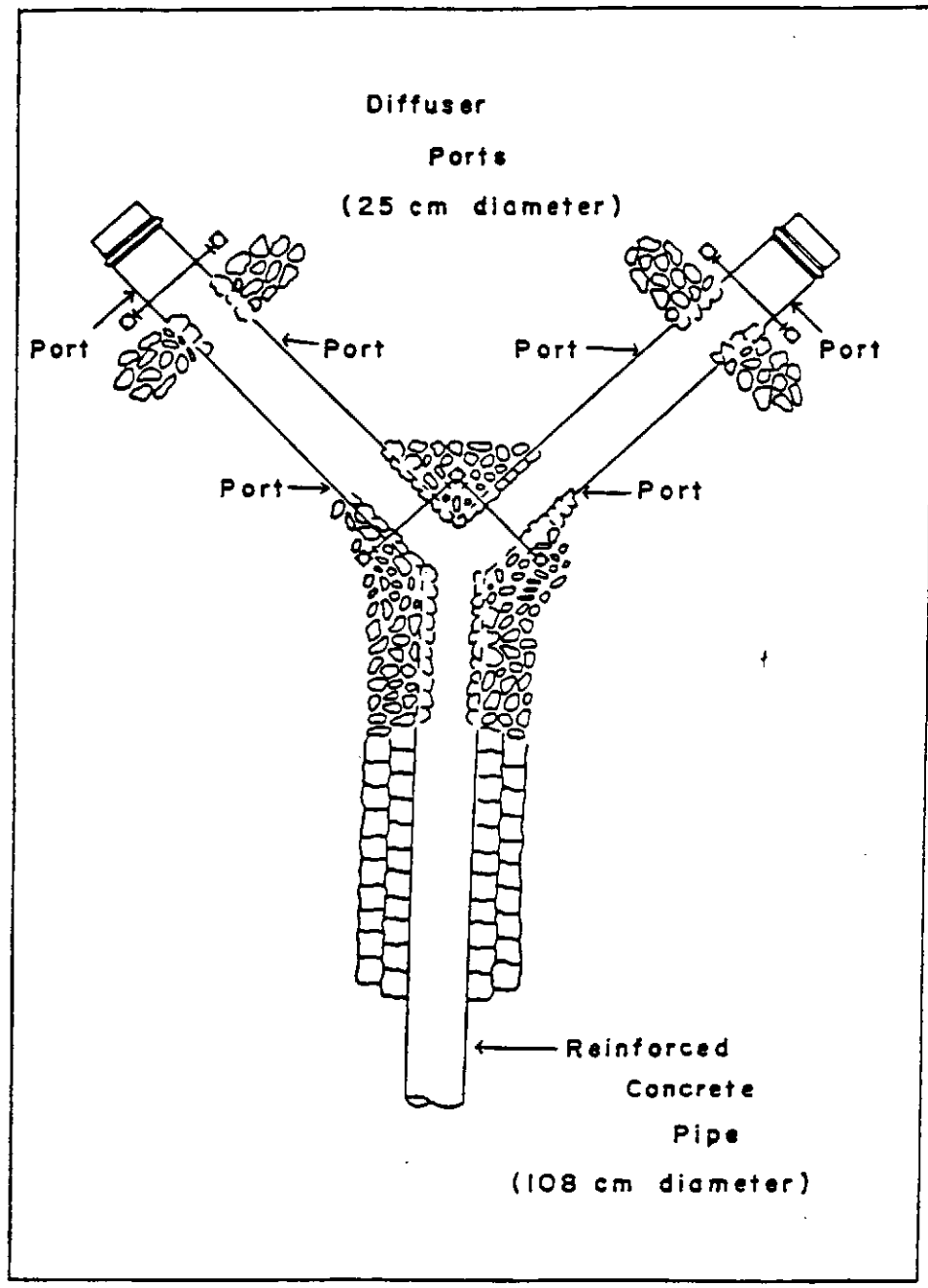
In 1965 Chesapeake-Elizabeth sewage treatment plant began providing primary and secondary treatment (activated sludge) from raw sewage. During the last few years the average discharge has increased (Table 1). During this investigation the daily discharge was approximately 24 million gallons per day.

The effluent is discharged into Chesapeake Bay through a 108 cm diameter reinforced concrete pipe that extends 1,128 m from the shore and terminates at a depth of 10 m. A multiport diffuser (Figure 2) is used to provide better dispersion of the effluent than a straight pipe system. The effluent from this outfall discharges into lower Chesapeake Bay where tidal currents range from 0.5 to 1.0 knot.

Table 1. Chesapeake-Elizabeth STP Annual Effluent Discharge Data

Year	Discharge (MGD)	Total Suspended Solids (Kg/day)
1975	15.6	2675
1976	16.3	4052
1977	19.1	2273
1978	23.5	3395
1979	26.2	4568
1980	23.1	2031

[1]MGD = Million gallons per day.


Diffuser
Ports
(25 cm diameter)
Port
Port
Port
Port
Port
Port
Reinforced
Concrete
Pipe
(108 cm diameter)

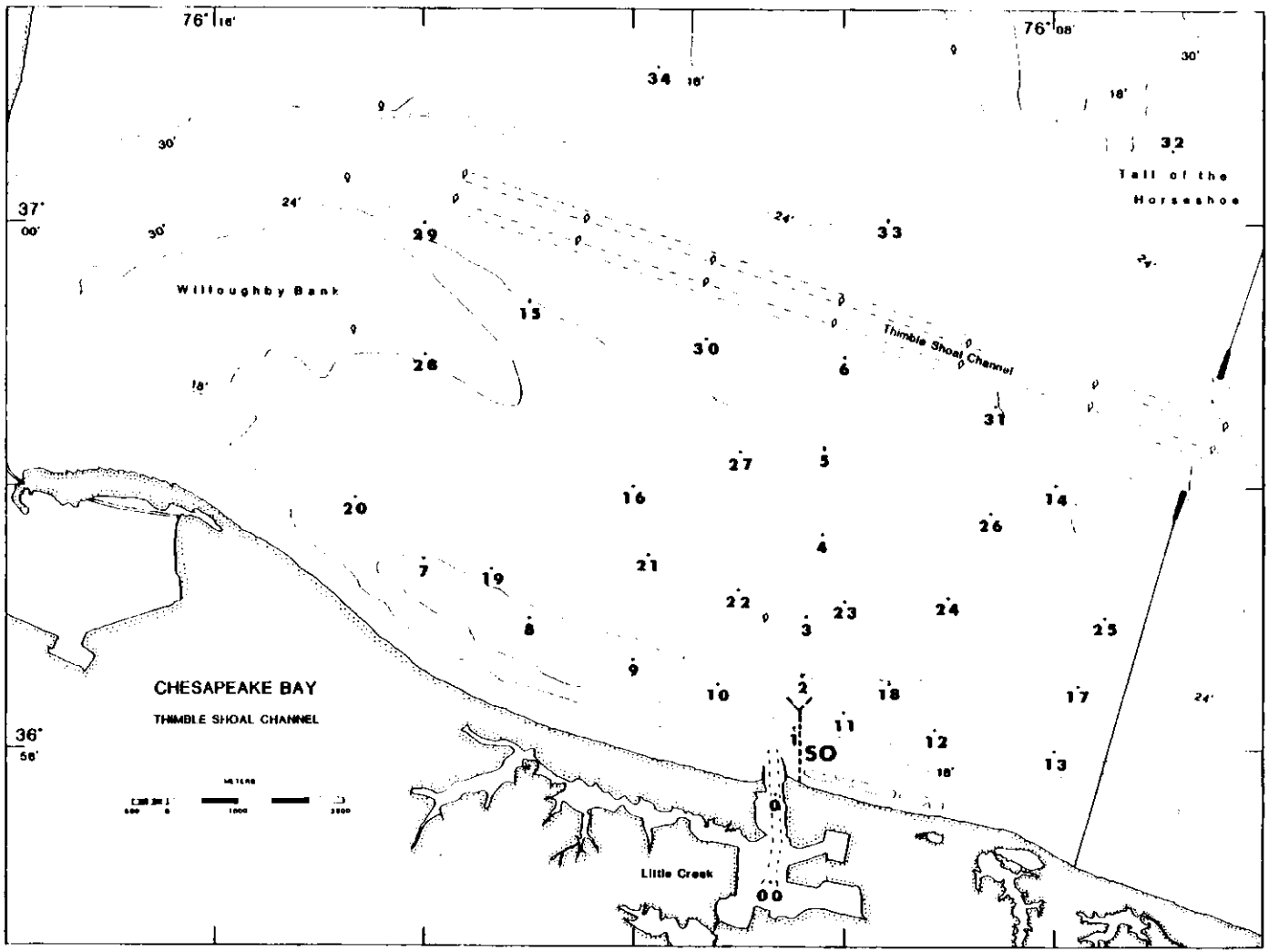
Chapter 3

METHODS AND MATERIALS

Sample Collection

Effluent. Effluent from the Chesapeake-Elizabeth treatment plant (C-E STP) was collected daily just prior to discharge between 23 April and 22 May 1981. Approximately 100 ml of effluent was collected every day at the same time, and the daily samples were combined to form weekly composite samples. Subsamples of these weekly composites were subsequently analyzed for coprostanol and petroleum hydrocarbon concentrations. All effluent samples were collected in pre-washed, solvent rinsed, glass bottles and were refrigerated until they were analyzed.

Sediment. Sediment samples were collected at 36 sites adjacent to the C-E STP outfall in May and July 1981 (Figure 3). Surficial sediment samples, ca. 10 cm in depth, were collected with a Shipek grab sampler aboard Old Dominion University's research vessel, R/V Linwood Holton. The sediment samples were brought aboard ship where they were transferred with a clean stainless steel spatula to pre-washed, solvent rinsed glass jars.


76° 16'
76° 08'
37° 00'
36° 56'
30'
24'
18'
34
32
33
29
15
30
6
31
28
27
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17
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13
0
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Willoughby Bank
Thimble Shoal Channel
Tail of the Horseshoe
CHESAPEAKE BAY
THIMBLE SHOAL CHANNEL
METERS
1000
2000
Little Creek

Precautions were taken to guard against the contamination of the samples from the ship and collection apparatus. The jars were sealed and placed on ice for transport to the laboratory where they were frozen and stored until they could be analyzed.

Sediment Analysis

Sediment samples were thawed and then mixed to ensure homogeneity. The moisture content of the sediment was determined by drying a small sample (ca. 1 g) of the sediment at 105-110°C to a constant weight. Approximately 50 g of wet sediment was placed in a 250 ml round bottom boiling flask. Internal standards, nonadecanol and docosane, were then added to the flask along with 100 ml of 0.5 N methanolic-potassium hydroxide (MeOH-KOH) and 10 ml of toluene. The samples were placed in a fume hood and were saponified/extracted under reflux for 2 hours. Saponification converts bound sterols to free sterols. After the samples cooled, they were filtered through a Whatman #4 filter. The flask and filter were rinsed with 20 ml methanol (MeOH) and 50 ml dichloramethane (CH_2Cl_2) and these solvents combined with the filtrate. The filter and sediment were discarded. The filtrate was transferred to a 500 ml separatory funnel containing 100 ml of 10% sodium chloride solution, with the pH adjusted to 1.0 with hydrochloric acid. The CH_2Cl_2 fraction was isolated, and the solution was then extracted twice more with 50 ml

CH_2Cl_2 each time. The CH_2Cl_2 extracts were combined and then taken to dryness on a rotary flash evaporator under vacuum at a temperature not exceeding 35°C. Coprostanol, along with other sterols and alcohols, were separated from petroleum hydrocarbons and other organics using column chromatography.

Effluent Analysis

Coprostanol and petroleum hydrocarbon concentrations were determined in effluent samples by taking a 250 ml aliquot, adding internal standards, 20 ml of 10% sodium chloride solution (pH =1), and extracting the solution three times with 50 ml CH_2Cl_2 each time. Centrifugation was used to break any emulsions. The combined extracts were taken to dryness on a rotary flash evaporator. The residue was saponified by adding 50 ml of 0.5 N MeOH-KOH, 5 ml toluene, 10 ml H_2O and refluxing this mixture for 2 hours. The saponification mixture was then extracted and further purified using column chromatography.

To determine the coprostanol and petroleum hydrocarbons that were associated with particulates in the final effluent, 250 ml aliquots of effluent were filtered through a pre-ignited Gelman A/E glass fiber filter (nominal pore size 1.0 µm). The organics that were retained on the filter (particulate) were saponified/extracted and concentrated using the techniques previously described for sediment samples.

Column Chromatography

The column consisted of a large volume (4 ml) pasteur pipet filled with 1 g of silica gel over 1 g of alumina both deactivated with 5% water by weight. The column was pre-rinsed with 3 ml MeOH followed by 6 ml CH_2Cl_2 and finally with 6 ml of 95:5 (v/v) hexane-toluene mixture. The concentrated organics, as described in the previous section, were charged to the column in 2 ml 95:5 hexane-toluene mixture. The hydrocarbons contained in the sample were eluted from the column with 6 ml 95:5 hexane: toluene mixture and were collected in a 50 ml pear-shaped boiling flask. The remaining concentrated organics were charged to the column in 2 ml of MeOH in order to ensure complete recovery of the sterols and alcohols. These compounds were then eluted from the column with 8 ml MeOH, and each fraction was taken to dryness on the rotary flash evaporator.

Gas Chromatography

Analysis of each column chromatography elution fraction was carried out with a Hewlett-Packard model 5830 gas chromatrograph. The instrument was equipped with a 25 m x 0.25 mm ID fused silica capillary column coated with SP-1000. Some samples were also run on a 30 m x 0.25 mm ID capillary column coated with SE-52 to provide additional support for the positive identification of the compounds being studied. The instrument conditions

are listed in Table 2. The eluting materials were detected with a flame ionization detector (FID), the response of which was recorded and integrated with a Hewlett-Packard model 18850A reporting integrator.

Coprostanol was identified by comparing relative retention times to standard mixtures, co-injection with authentic coprostanol, comparing results on different polarity columns, and by the formation and gas chromatographic analyses of trimethylsilyl (TMS) derivatives (Wells and Makita, 1962; Chambaz and Horning, 1969). Quantitative determinations of coprostanol were provided by establishing the approximate relative response of the FID for sample coprostanol to that of internal standard nonadecanol added to each sample prior to saponification/extraction. The Hewlett-Packard "Internal Standard" program identifies each according to a programmed retention time window and converts the response area to concentration units. Daily checks were made to verify relative response factors and retention times.

Petroleum hydrocarbons consist of numerous resolved peaks and an unresolved fraction that is referred to as an unresolved complex mixture (NAS, 1975). The area of the resolved peaks and the unresolved complex mixture were determined by planimetry. Comparison of these areas with the area of the internal standard, docosane, allows for the concentration of hydrocarbons to be calculated. Procedural blanks and standards were run systematically

Table 2. Gas Chromatographic Conditions

Column	SP-1000	SE-52
Temp 1	150°C	80°C
Temp 2	270°C	270°C
Rate	8°/min	10°/min
Inj. Temp	270°C	270°C
Det. Temp	350°C	350°C
Mode	Splitless	Splitless
Carrier gas/flow	N_2/0.5 ml/min	N_2/0.5 ml/min
Make-up gas/flow	N_2/20 ml/min	N_2/20 ml/min
Detector	FID	FID
Attenuation	8	16
Range	10^{-11}	10^{-11}

in association with all analyses. All concentrations reported have been corrected for the concentrations found in the procedural blanks.

Grain Size Analysis

Grain size analysis was carried out utilizing a modified method described by Folk (1974). Particulates were separated into two size classes: silt and clay fraction (<63 μm) and sand and larger material (>63 μm). A subsample of sediment was dryed at a very low temperature (<40°C) for approximately 2 days or until dry. When dry, the sediment was disaggregated using a mortar and pestle. The sample was then weighed and the weight recorded (total dry weight). After transferring the sample to a 500 ml polyethylene bottle, it was covered with a dispersant (10% sodium hexametaphosphate or Calgon). The bottle is shaken thoroughly and allowed to stand overnight.

The sample was next wet sieved through a 63 μm (230 mesh) screen. The solution is placed onto the screen, making sure all is rinsed from the bottle. The screen is gently rocked back and forth, while spraying gently with water, allowing the silt and clay material to pass through the mesh. Since there is no further analysis to be performed on the fine-grained material, it is discarded. The sediment is continuously washed back and forth over the screen until the water runs through clear (i.e. contains no silts or clays). The sand remaining on

the screen is transferred to a flask and is dryed at 100^{o}C and the weighed. The dry weight of the sieved fraction is subtracted from the dry weight of total before sieved, yielding the amount or percent of material which is <63 μm.

Chapter 4

RESULTS AND DISCUSSION

Effluent Analysis

Chesapeake-Elizabeth STP effluent data for 23 April to 22 May 1981 was obtained from the Hampton Roads Sanitation District monthly operations report (Table 3). The amount of effluent discharged (average flow) and the total suspended solids concentration during the sampling period was fairly constant as evidenced by their low relative standard deviation of ± 7 and 19%, respectively. Fecal coliform bacteria concentrations, on the other hand, were variable with a range of <20 to >800 coliform bacteria per 100 ml of sample and a relative standard deviation of ± 78%.

The concentration of coprostanol and hydrocarbons in the effluent were also measured during this time period. The gas chromatogram of a standard containing nonadecanol, coprostanol, and cholesterol which was analyzed to determine retention times and response factors is shown in Figure 4. A typical chromatogram indicating the presence of coprostanol in sewage effluent is shown in Figure 5. Gas chromatograms of the hydrocarbon standard and hydrocarbons in an effluent sample are shown in

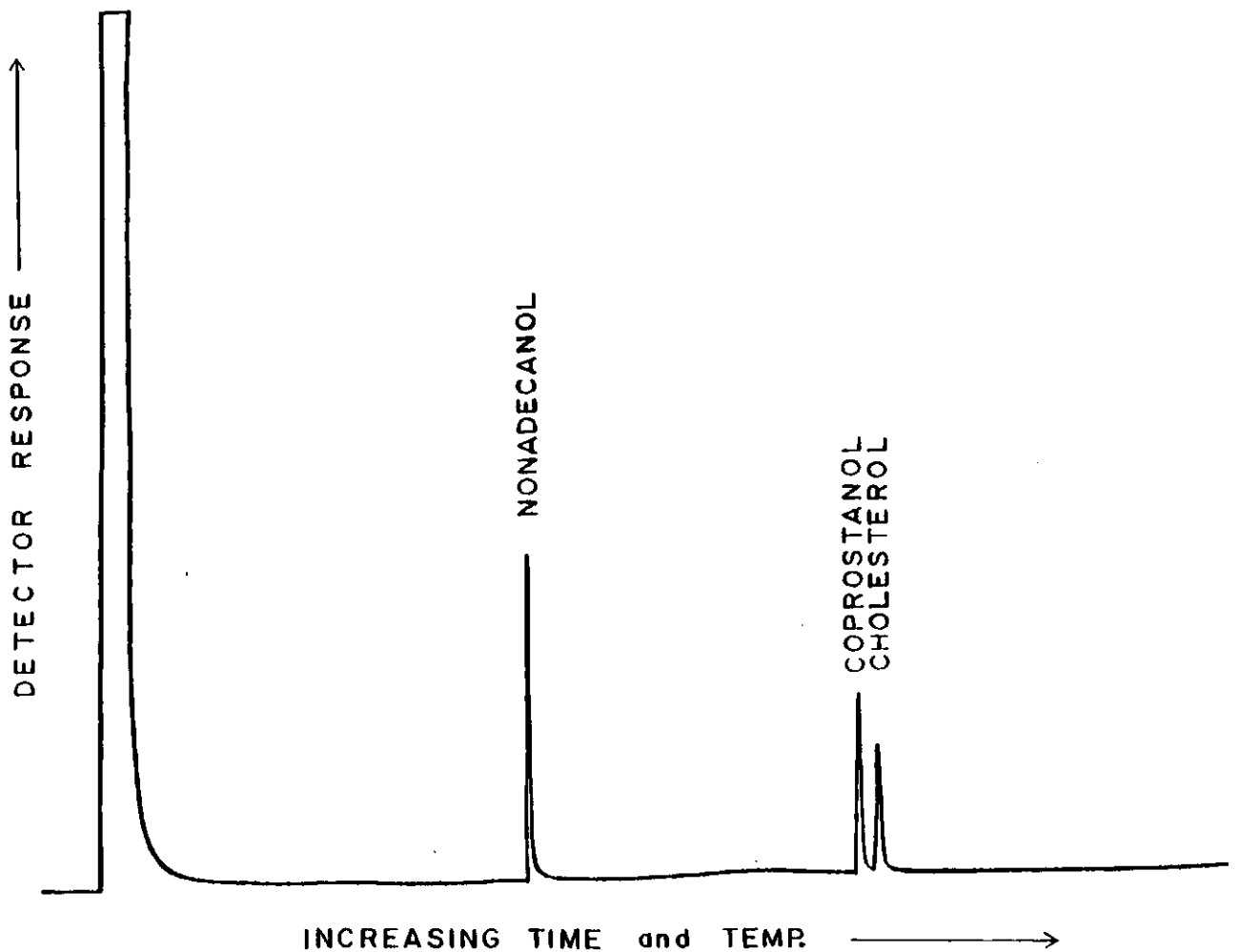
Table 3. Hampton Roads Sanitation District Monthly Operations Report

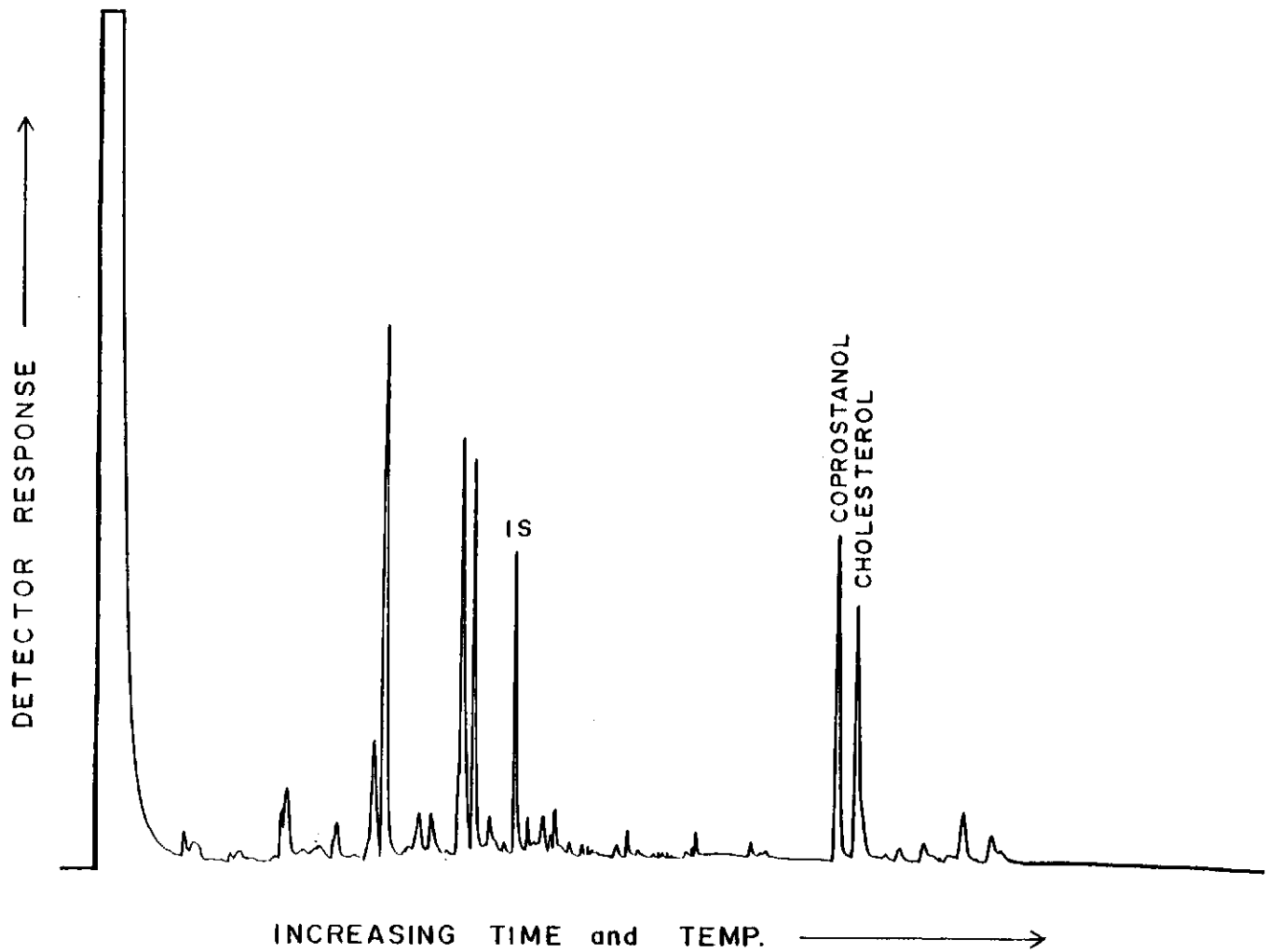
Sample	Date Collected	Average Flow (MGD)[1]	TSS[2] (MG/L)	Fecal Coliform (#/100)[3] Max.	Min.	Avg.
1	23-29 April	19.4	22	340	<20	118
2	30 April -6 May	20.4	14	>8000	80	1607
3	7-13 May	20.0	22	>8000	<20	1351
4	16-22 May	22.9	20	>8000	20	3046

[1]MGD = Million gallons per day.

[2]TSS = Total suspended solids.

[3]#/100 = Number of fecal coliform bacteria in 100 ml of effluent.


DETECTOR RESPONSE
NONADECANOL
COPROSTANOL
CHOLESTEROL
INCREASING TIME and TEMP.


DETECTOR RESPONSE
IS
COPROSTANOL
CHOLESTEROL
INCREASING TIME and TEMP.

Figures 6 and 7, respectively. The standard consists of the even straight chain normal alkanes (n-alkanes) with a carbon range from C_{12}-C_{32}. The hydrocarbons in the effluent sample exhibit an elution range from C_{12}-C_{32} with 80% of the total hydrocarbons consisting of a mixture of cycloalkanes and aromatic hydrocarbons which appear as an unresolved complex mixture (UCM). The presence of the UCM and the lack of odd n-alkane predominance indicates the hydrcarbons found in the effluent are derived from fossil fuels (Farrington and Meyers, 1976).

The concentrations of coprostanol and hydrocarbons found in the final effluent samples from the Chesapeake-Elizabeth STP are reported in Table 4. Daily samples are combined to obtain four weekly composite samples (Table 3). The mean coprostanol and hydrocarbon concentrations were 33 µg/l (relative standard deviation of ± 6%) and 299 µg/l (relative standard deviation of ± 14%) respectively. Filtration of the effluent indicates that the majority of the coprostanol (>88%) and hydrocarbons (>95%) are associated with particulate material. This is consistent with the findings of other studies (Wun et al., 1978; Van Vleet and Quinn, 1977; Knap and Williams, 1982).

The concentration of particulate coprostanol and hydrocarbons was converted to a weight per weight of suspended solids basis using effluent data from Table 3. The resulting concentrations give mean particulate

Fig. 6. Gas chromatogram of hydrocarbon standard.

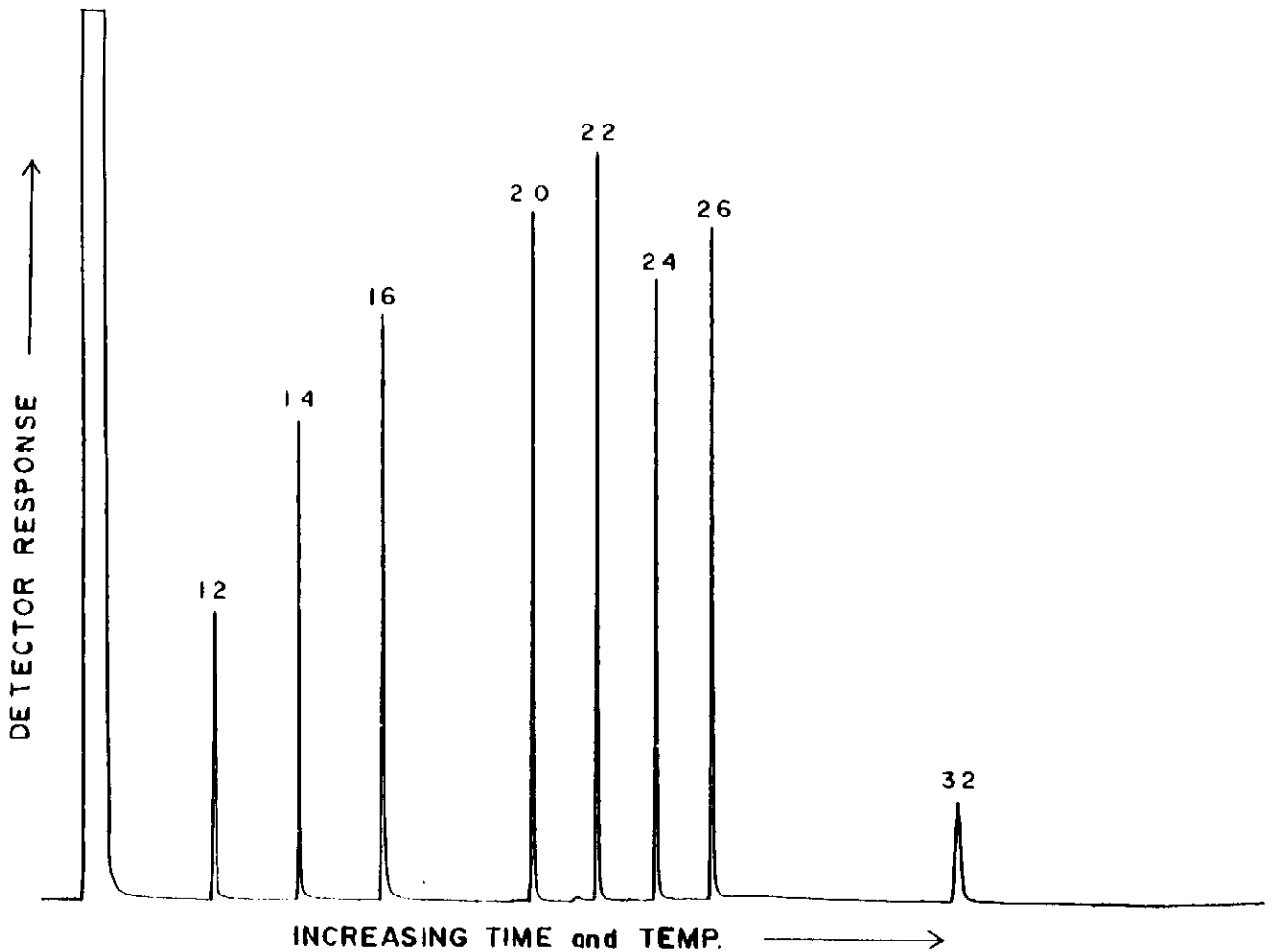
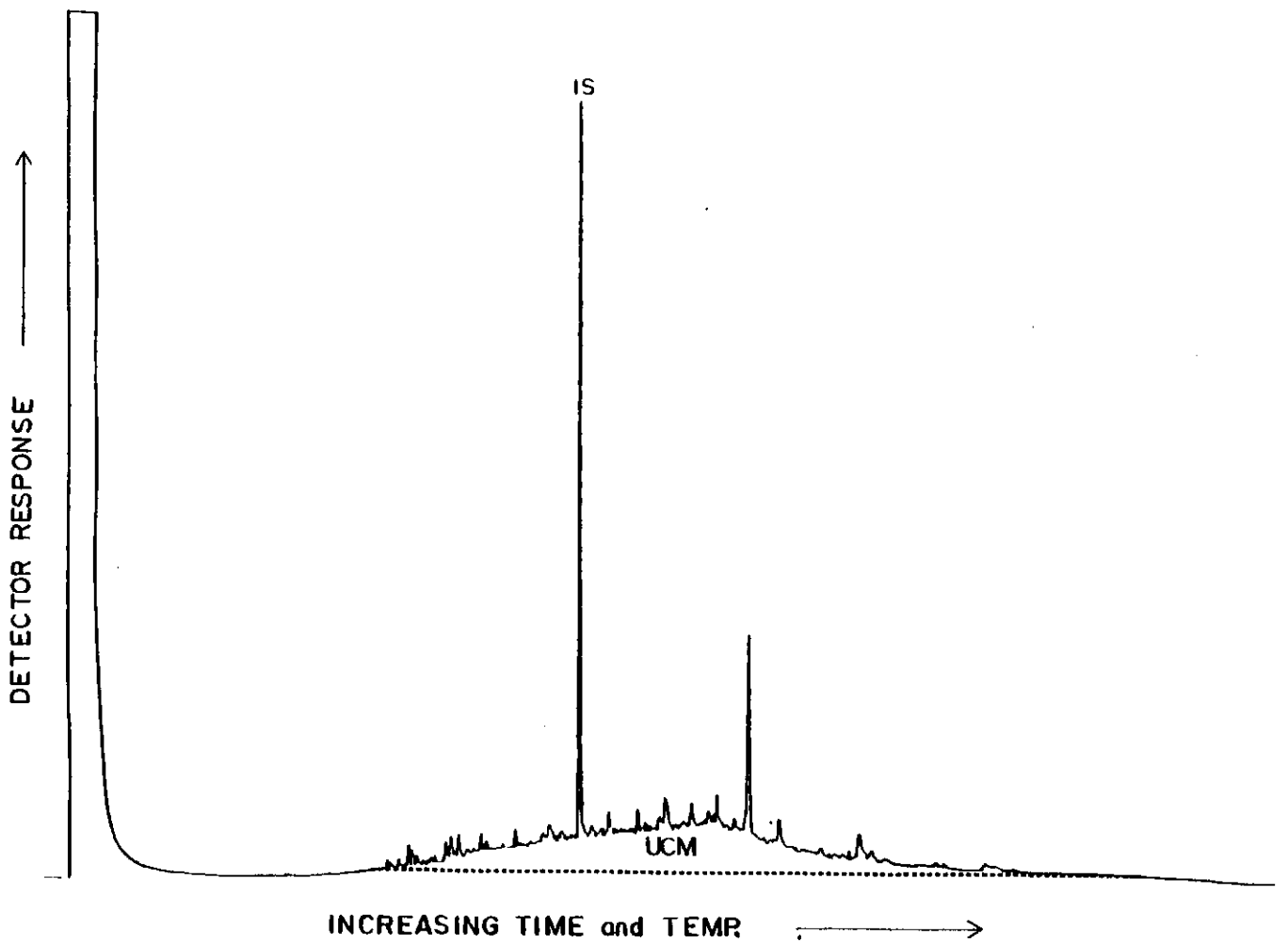

DETECTOR RESPONSE
12
14
16
20
22
24
26
32
INCREASING TIME and TEMP.

Fig. 7. Gas chromatogram of hydrocarbons in STP effluent.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INCREASING TIME and TEMP.

Table 4. Coprostanol and Hydrocarbon Concentrations in Chesapeake-Elizabeth STP Effluent

	COPROSTANOL			HYDROCARBONS			
Sample[1]	µg/l	% part.[2]	mg/g	µg/l	% part.	mg/g	Hydrocarbon/Coprostanol
1	36	88	1.6	247	98	11.2	7
2	32	84	2.3	342	94	24.5	11
3	32	96	1.5	291	91	13.2	9
4	32	85	1.6	317	97	15.9	10
Mean	33	88	1.8	299	95	16.2	9

[1] Composite sample, see Table 3 for dates.

[2] % Particulate = Conc. (filtered)/Conc. (total) x 100%.

coprostanol and hydrocarbon values of 1.6 (± 31%) and 15.4 (± 35%) mg/g (Table 4). Particulate and dissolved coprostanol and hydrocarbon concentrations in the effluent samples analyzed were fairly constant. The ratio of particulate hydrocarbons to particulate coprostanol was also relatively constant with a mean value of 9 and a relative standard deviation of 18%.

Sources of anthropogenic hydrocarbons entering sewage treatment plants are not completely understood; however, possible inputs include oils washed from roads, accidental discharges into sewer systems, atmospheric deposition, industrial discharges, etc. (NAS, 1975). Whatever the means of transport to the plants, the input tends to be fairly constant and the discharge of petroleum hydrocarbons from treatment plants to estuaries and coastal waters is significant (Van Vleet and Quinn, 1977; Eganhouse and Kaplan, 1982).

Sediment Analysis

Grain Size. Lower Chesapeake Bay has been described by Shideler (1975) as a mud-bypass area, because the strength of the tidal currents prevent mud deposition. Brush (1978) did an in-depth sediment study between Thimble Shoal Channel and the south shore and concluded that there was sediment ranging from coarse, gravelly sand to sediment containing 40% silt and clay (<62 μm) material. Ludwick (1981), combining the studies of

Shideler and Brush, defined certain sediment size boundaries (Figure 8). The high silt-clay content located south of Thimble Shoal Channel was attributed to the deposition of fine material from the resuspension of James River sediment on ebb tide. Another high concentration of fine-grained sediment located in the central portion of the study area off Little Creek was postulated to be the result of the settling of fine-grained material discharged from the Chesapeake-Elizabeth STP (Brush, 1978). The specific configuration may be due to advection to the north from ebb currents flowing through Little Creek inlet and the east-west entrainment by tidal currents which may move this fine-grained sediment along the axis of the shore (Ludwick, 1981).

Sediment size analyses were done as part of this investigation. The percentage of the silt-clay fraction (<63 μm) at each station was determined. The results of these analyses are reported in Table 5. The silt-clay fraction ranged from 2.2 to 23.8% of the total sediment on a weight basis. The percentage of fine-grained material at a station in the vicinity of the Chesapeake-Elizabeth STP outfall has been reported to range from 7 to 52% within a one month period (Bates and Spencer, 1979). The percentages of sediments in the fine-grained fraction at the Chesapeake-Elizabeth STP outfall were 7.7 and 10.8%, respectively, for samples collected in June and July 1981. These values are less variable and

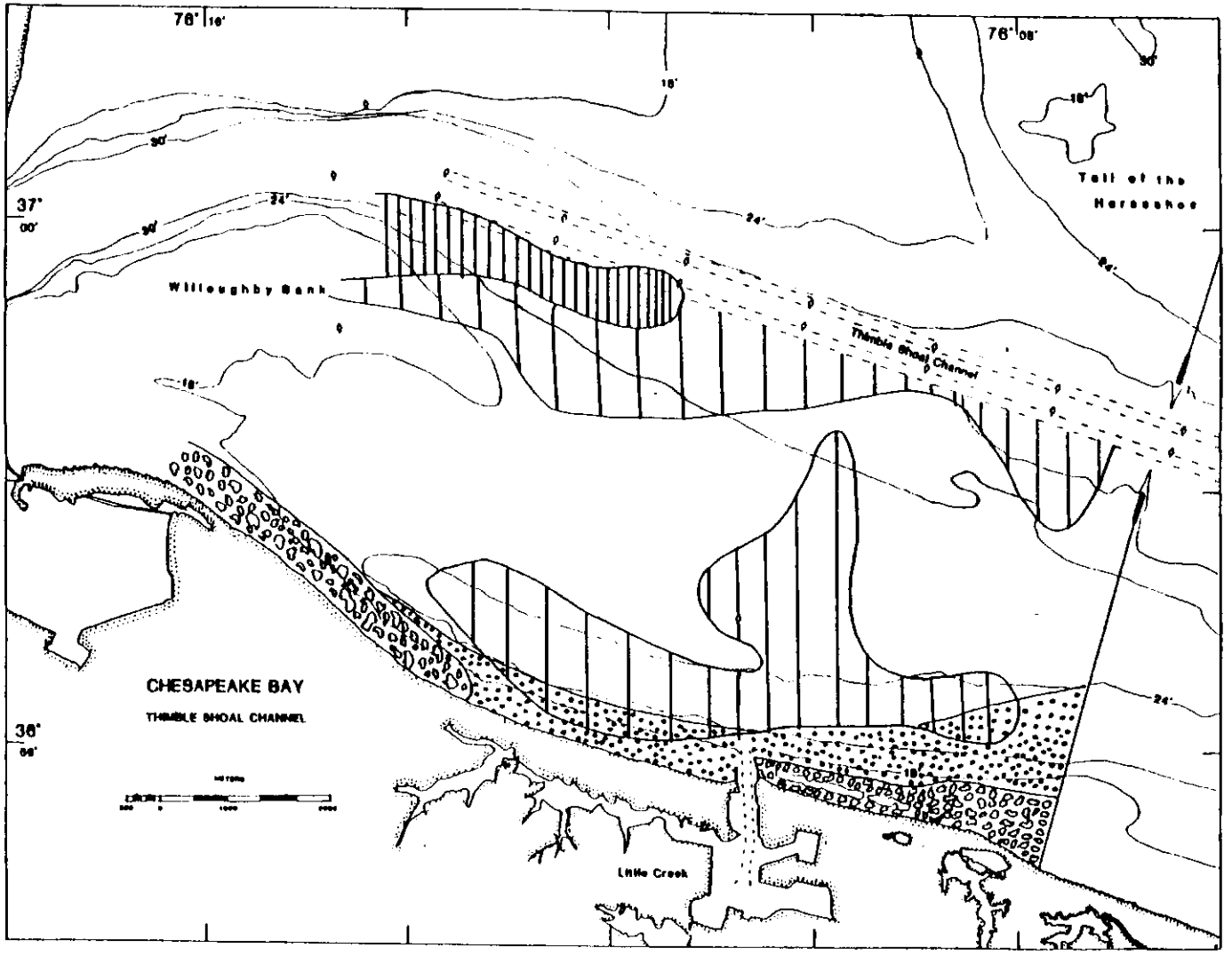

76° 10'
76° 08'
37° 00'
36° 56'
Willoughby Bank
Thimble Shoal Channel
Tail of the Horseshoe
CHESAPEAKE BAY
THIMBLE SHOAL CHANNEL
Little Creek

Table 5. Sediment Grain Size Analyses

Sample #1	% Silt-Clay
00	9.1
0	13.6
1	9.0
2	7.7
2a	10.8
3	5.7
4	14.8
5	10.1
6	19.7
7	9.3
8	16.8
9	18.1
10	18.6
11	21.7
12	21.7
13	2.2
14	10.5
15	20.8
16	10.0
17	6.8
18	12.9
19	9.8
20	5.3
21	11.6
22	9.4
23	9.6
24	5.8
25	16.7
26	23.8
27	12.7
28	4.8
29	16.7
30	16.2
31	15.3
32	7.9
33	7.6
34	7.3
Range = 2.2 - 23.8%	
Mean = 12.2	
Std. dev. = ±5.5	

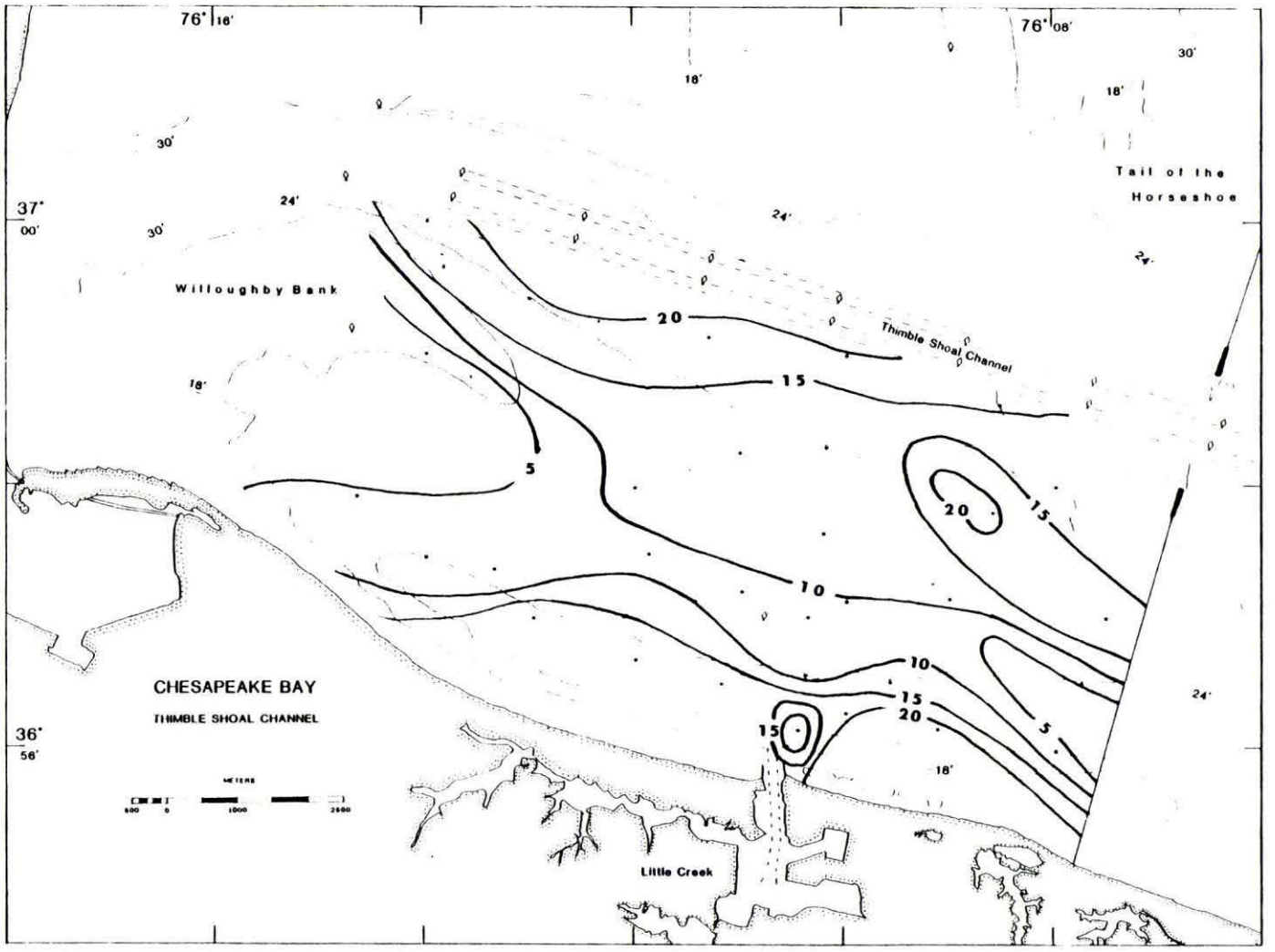
[1]Figure 3 for station location.

fall within the lower range of values reported for this location (Bates and Spencer, 1979).

The distribution of fine-grained materials (FGM) on a percentage basis determined in this study is shown in Figure 9. This distribution is different from that described by Ludwick (1981). This can be seen by comparing Figures 8 and 9. Both distributions show a high percentage of FGM along Thimble Shoal Channel and along the shoreline adjacent to Little Creek Harbor. The percentage of FGM, shown in Figure 8, are generally higher, and the area covered by the 20% contour line is more extensive than that in Figure 9. The high concentration of FGM (20 to 45%) located in the central portion of the study area near the sewage outfall (Ludwick, 1981) in Figure 8 is seen as an area of low FGM (<10%) concentrations in Figure 9. Comparison of the two FGM distributions from this study area suggest it may be a dynamic area, where the distribution and percentage of FGM may change dramatically in periods of a month or more.

Coprostanol. Coprostanol concentrations were determined in the sediments adjacent to the Chesapeake-Elizabeth sewage outfall. A chromatogram of a typical sediment sample (Figure 10) shows that the amount of cholesterol in this sample is greater than the amount of coprostanol. The opposite was seen in the sewage effluent sample chromatogram (Figure 5). This is not

Fig. 9. Percentage silt and clay (<63 μm) distribution.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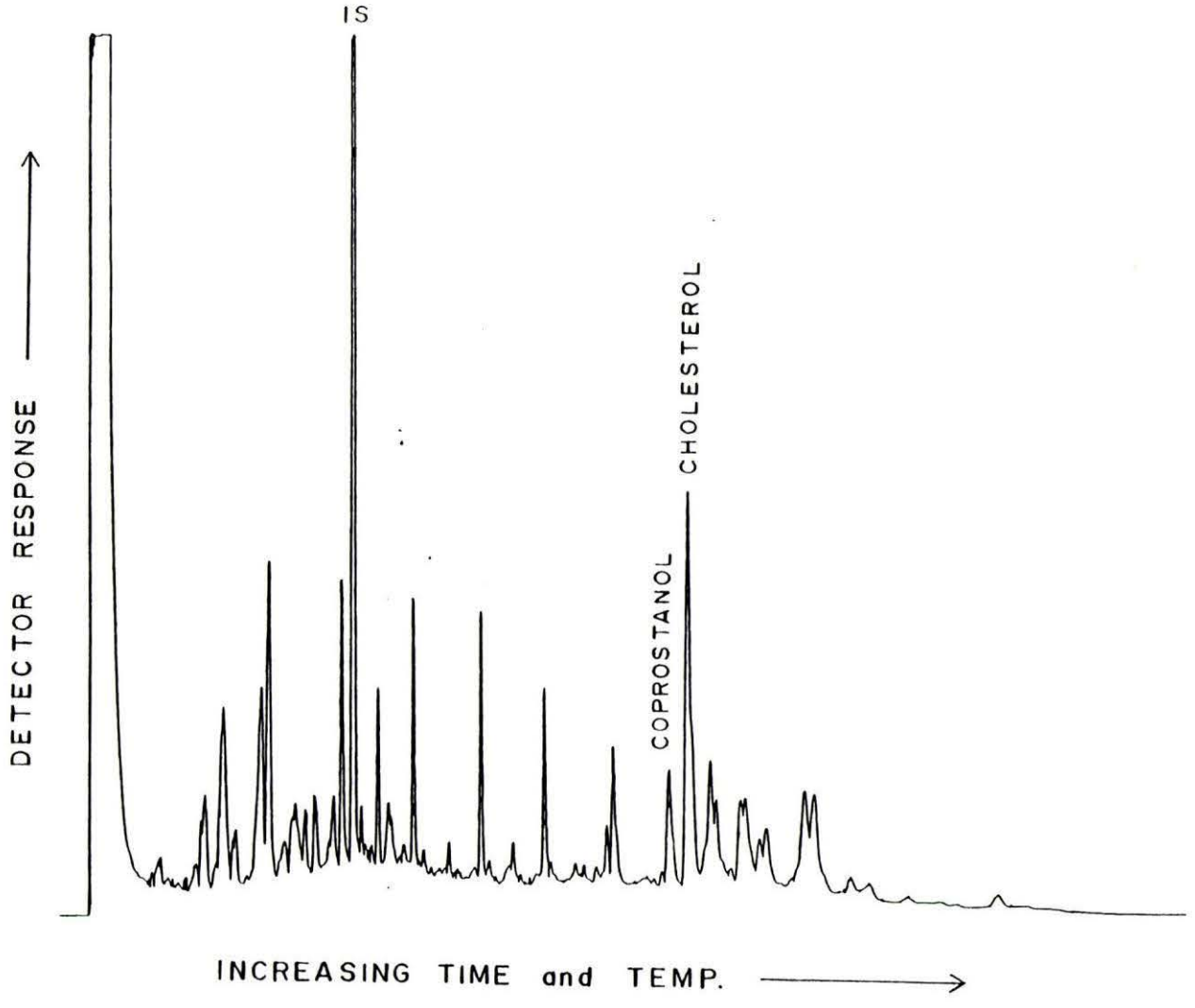
Little Creek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unexpected as cholesterol is produced in the marine environment and also accounts for 9.5% of fecal sterols, while coprostanol is not thought to be produced in the marine environment, but comprises 50 to 80% of fecal sterols (Frerezou et al., 1978; Hatcher et al., 1979). The different relative abundance of cholesterol and coprostanol is due to cholesterol inputs from marine organisms and sewage, while coprostanol comes predominately from sewage.

Sediment coprostanol concentrations (Table 6) ranged from 19 to 450 ng/g with a mean of 142 ng/g. The coprostanol concentrations in this study are on the low end of the ranges reported by Goodfellow et al. (1977) for the Clyde Estuary near Glascow, Scotland (3 to 13,580 ng/g) and Hatcher and McGillivary (1978) for the New York Bight (56 to 5,700 ng/g). It is important to note, however, that their study areas are heavily influenced by the discharge of sewage sludge, while the discharge from the Chesapeake-Elizabeth STP consists of secondarily treated sewage. Studies by Kanazawa and Teshima (1978) in Ariake Sea, Japan and Escalona et al. (1980) in two Mexican harbors report sediment coprostanol concentrations of 2 to 1,770 ng/g and 6 to 44 ng/g, respectively. The sources of input to these areas were reported to be from direct discharge of domestic waste from sewage outfalls, and possibly by discharge from docked ships. These values are in the same range as those reported in this study.

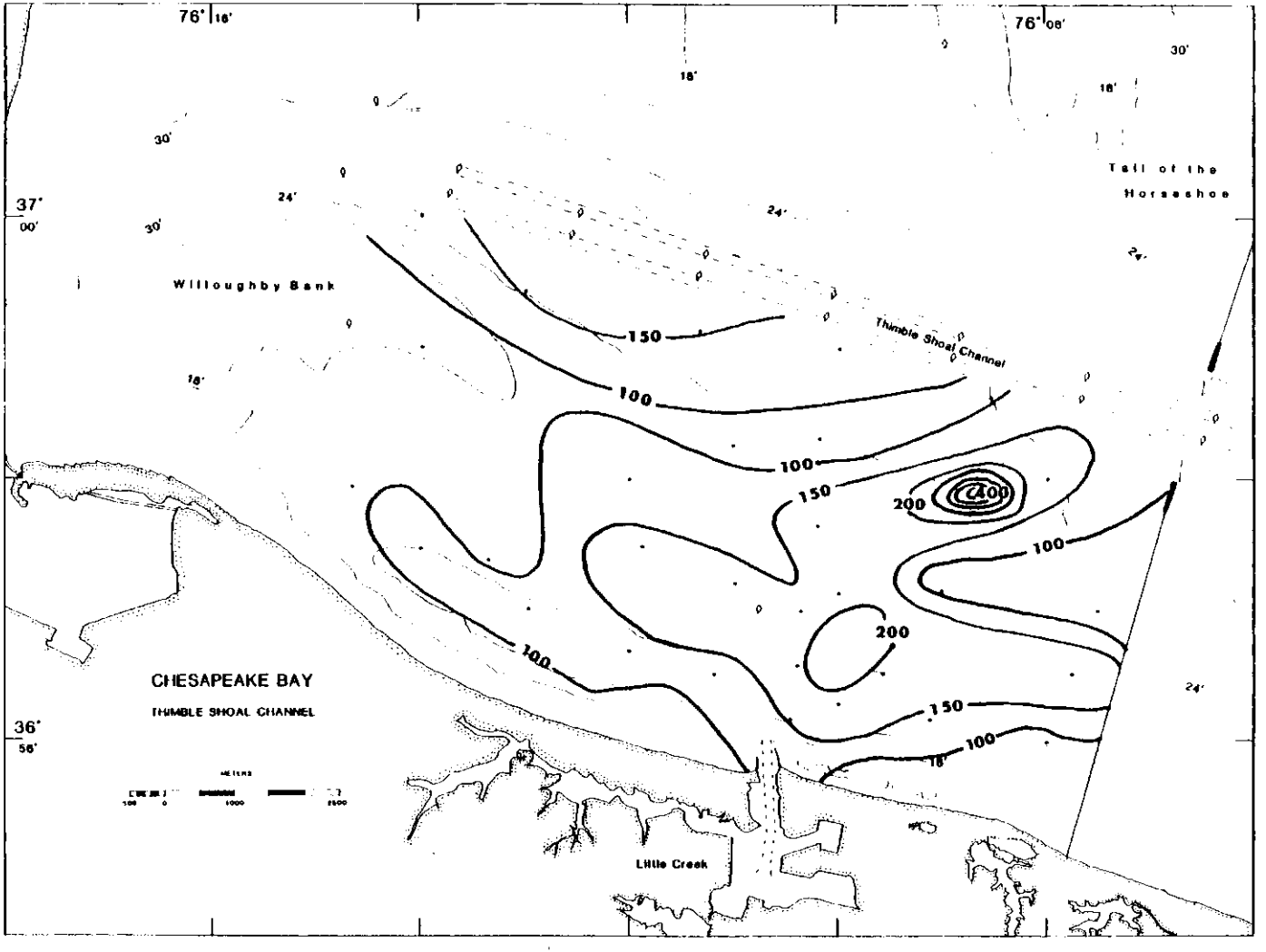
Table 6. Sediment Coprostanol Concentrations

Sample #[1]	ng/g dry sed.	µg/g fine[2]
00	274	3.01
0	185	1.36
1	155	1.74
2	222	2.87
2a[3]	199	1.85
3	166	2.90
4	186	1.26
5	72	0.71
6	139	0.71
7	140	1.50
8	136	0.81
9	145	0.80
10	137	0.74
11	226	1.04
12	120	0.55
13	52	2.42
14	167	1.60
15	166	0.80
16	110	1.10
17	169	2.48
18	199	1.54
19	84	0.86
20	64	1.21
21	151	1.30
22	172	1.83
23	182	1.88
24	91	1.55
25	94	0.56
26	454	1.91
27	80	0.63
28	80	1.68
29	132	0.79
30	152	0.94
31	83	0.54
32	19	0.24
33	28	0.36
34	28	0.38
Range =	19 - 454	0.24 - 3.01
Mean =	142	1.31
Std. dev. =	±79	±.74

[1]Figure 3 for station location.
[2]Sediment size 64 µm.
[3]Duplicate sample.

Pollutants have been found to concentrate on smaller size particles (Meyers and Quinn, 1973; Wade and Quinn, 1979; Moy, 1980). Smaller size particles are also more likely to be affected by currents (Moy, 1980). Therefore, coprostanol concentrations based on the weight of fine-grained (<63 µm, Table 5) sediments was determined. The coprostanol concentrations ranged from 0.24 to 3.01 µg/g fine grained sediment, with a mean of 1.31 µg/g fine-grained sediment. If the stations inside of Little Creek Harbor and on the north side of Thimble Shoal Channel are not included, the range of values is 0.55 to 2.87 µg/g fine-grained sediment. This is not a very large difference if you consider that samples from Station 2 (near the sewage outfall) had concentrations of 2.87 and 1.85 µg/g fine-grained sediment for samples collected in June and July 1981, respectively.

The coprostanol concentrations reported in Table 5 are contoured in Figures 11 and 12. Figure 11 shows an area of higher coprostanol concentration (150 µg/g) adjacent to Thimble Shoal Channel and another area of higher coprostanol concentration surrounding the outfall. The higher concentrations surrounding the outfall probably reflects the deposition of materials from the sewage outfall in that area. The higher concentration of coprostanol adjacent to the channel is separated from the effluent site by areas of lower corpostanol concentration. High percentages of FGM area also found in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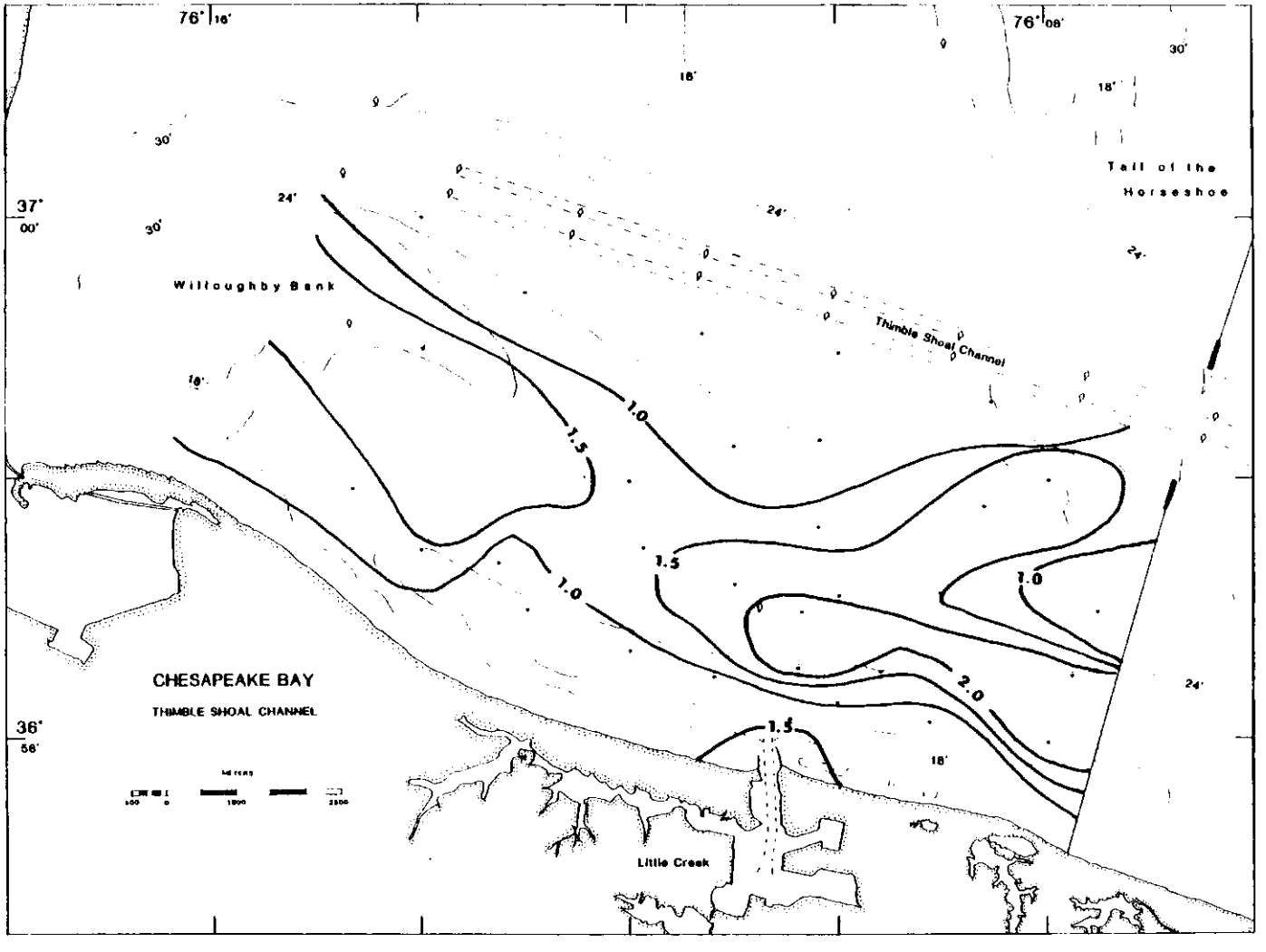
Thimble Shoal Channel
150
100
200
400
CHESAPEAKE BAY
THIMBLE SHOAL CHANNEL
Little Creek

Fig. 12. Coprostanol distribution on fine grain sediments.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CHESAPEAKE BAY
THIMBLE SHOAL CHANNEL
Little Creek

this area (Figure 9) suggesting that some process causes FGM containing coprostanol to concentrate in this area. This observation is also illustrated in the contour of coprostanol concentrations on a µg/g fine-grained sediment bases (Figure 12) which does not show the area adjacent to the channel to be an area of higher concentration, while still showing slightly higher concentrations in the area surrounding the sewage outfall.

Comparison of Figures 9, 11, and 12 indicates that coprostanol sediment concentration is affected by the percent of fine-grained sediments and, to a lesser extent, the distance from the discharge. The coprostanol appears to be spread throughout the study area in association with fine-grained particles. The areas of high concentration seen in Figure 11 are no longer apparent in Figure 12. Coprostanol in association with fine-grained sediments may also be escaping from the study area (Wade et al., 1983). Relatively high concentrations of coprostanol were found inside Little Creek Harbor (Stations 00, 0; Table 6) and lower concentrations on the north side of Thimble Shoal Channel (Stations 32, 33, 34; Table 6). This may result from small particles containing coprostanol being transported into the Little Creek Harbor, with lesser amounts transported across Thimble Shoal Channel. This is only a tentative observation and would require more information to substantiate.

Hydrocarbons. The hydrocarbon concentrations in sediment adjacent to Chesapeake-Elizabeth sewage outfall are presented in Table 7. The concentration ranges from 2.4 to 153.0 µg/g dry sediment. These values are an order of magnitude less than detected by Van Vleet and Quinn (1977) in Providence River sediments near a sewage outfall (570-5410 µg/g). However, the concentration of hydrocarbons discharged by this plant into the Providence River is an order of magnitude higher than that discharged by the Chesapeake-Elizabeth STP. These STP's may also have different magnitudes of hydrocarbon inputs. The hydrocarbon concentrations in lower Narragansett Bay were similar to those found in lower Chesapeake Bay during this study (Wade and Quinn, 1979).

Like coprostanol, hydrocarbon concentrations north of Thimble Shoal Channel were relatively low. The chromatogram for Station 32 (Figure 13) shows litte unresolved complex mixture. The predominant resolved materials are the higher molecular weight n-alkanes, C_{23}-C_{30}, with odd carbon predominance, indicative of material from biogenic sources (Farrington and Meyers, 1976).

The highest concentration of hydrocarbons, like coprostanol, was detected in Little Creek Harbor. The chromatogram for Station 00 is shown in Figure 14. The pattern is unique in that the sample contains few resolved components, but a large UCM. The lack of resolved components and the dominance of the UCM are indicative of

Table 7. Sediment Hydrocarbon Concentrations

Sample #[1]	µg/g dry sed.	µg/g fine[2]	%[4]
00	153.0	1679	2
0	18.3	135	10
1	10.8	121	14
2	12.0	155	19
2a[3]	13.1	122	15
3	6.1	106	27
4	12.1	81	15
5	14.0	139	5
6	19.8	101	7
7	12.7	136	11
8	26.8	159	5
9	17.1	97	8
10	18.8	101	7
11	33.0	152	7
12	13.4	62	8
13	4.7	220	11
14	13.2	126	13
15	25.6	123	6
16	9.4	94	12
17	26.4	387	6
18	14.1	109	14
19	10.8	111	8
20	8.3	157	8
21	15.4	133	10
22	11.1	118	15
23	12.5	130	15
24	8.1	139	11
25	8.4	50	11
26	40.0	168	11
27	18.6	146	4
28	8.6	180	9
29	25.0	150	5
30	23.6	146	6
31	13.3	87	6
32	3.6	46	5
33	3.0	39	9
34	2.4	32	12
Range =	2.4 - 153.0	32 - 1679	
Mean =	18.6	169	
Std. dev. =	±24.2	±262	

[1] Figure 3 for station location.
[2] Sediment size 64 µm.
[3] Duplicate sample.
[4] % of hydrocarbons calculated to come from STP effluent.

Fig. 13. Gas chromatogram of hydrocarbons from Station 32.

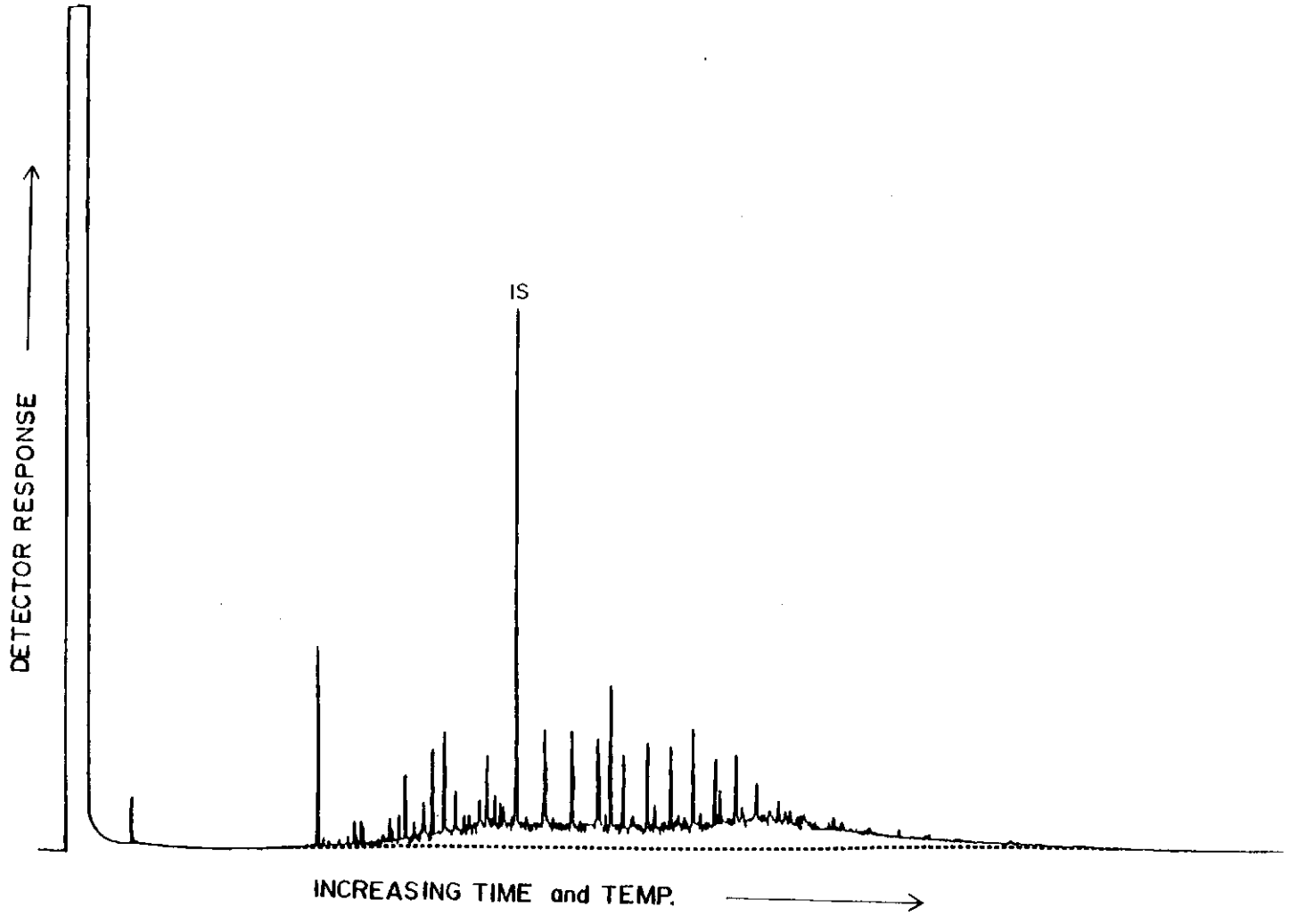
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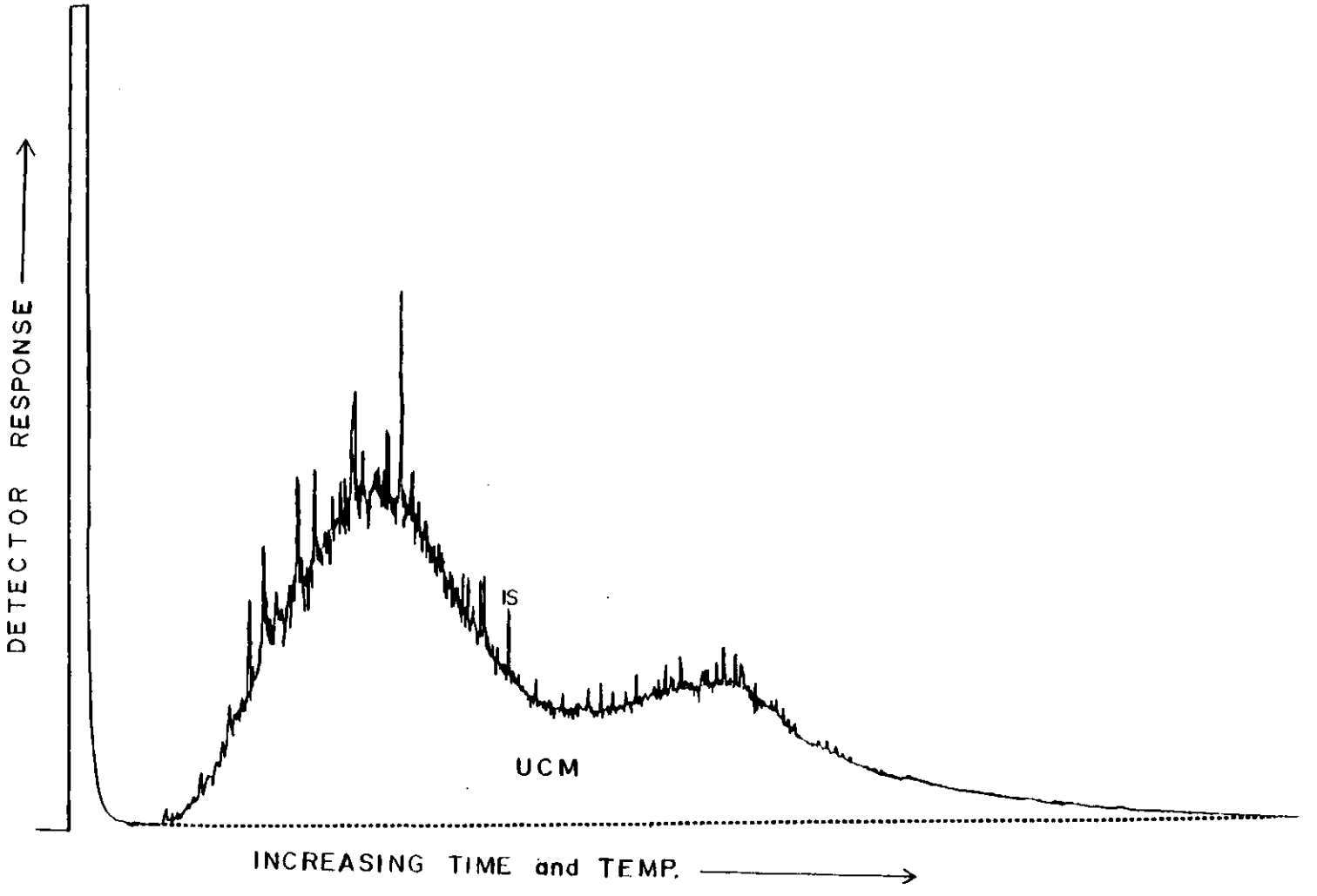
Fig. 14. Gas chromatogram of hydrocarbons from Station 00.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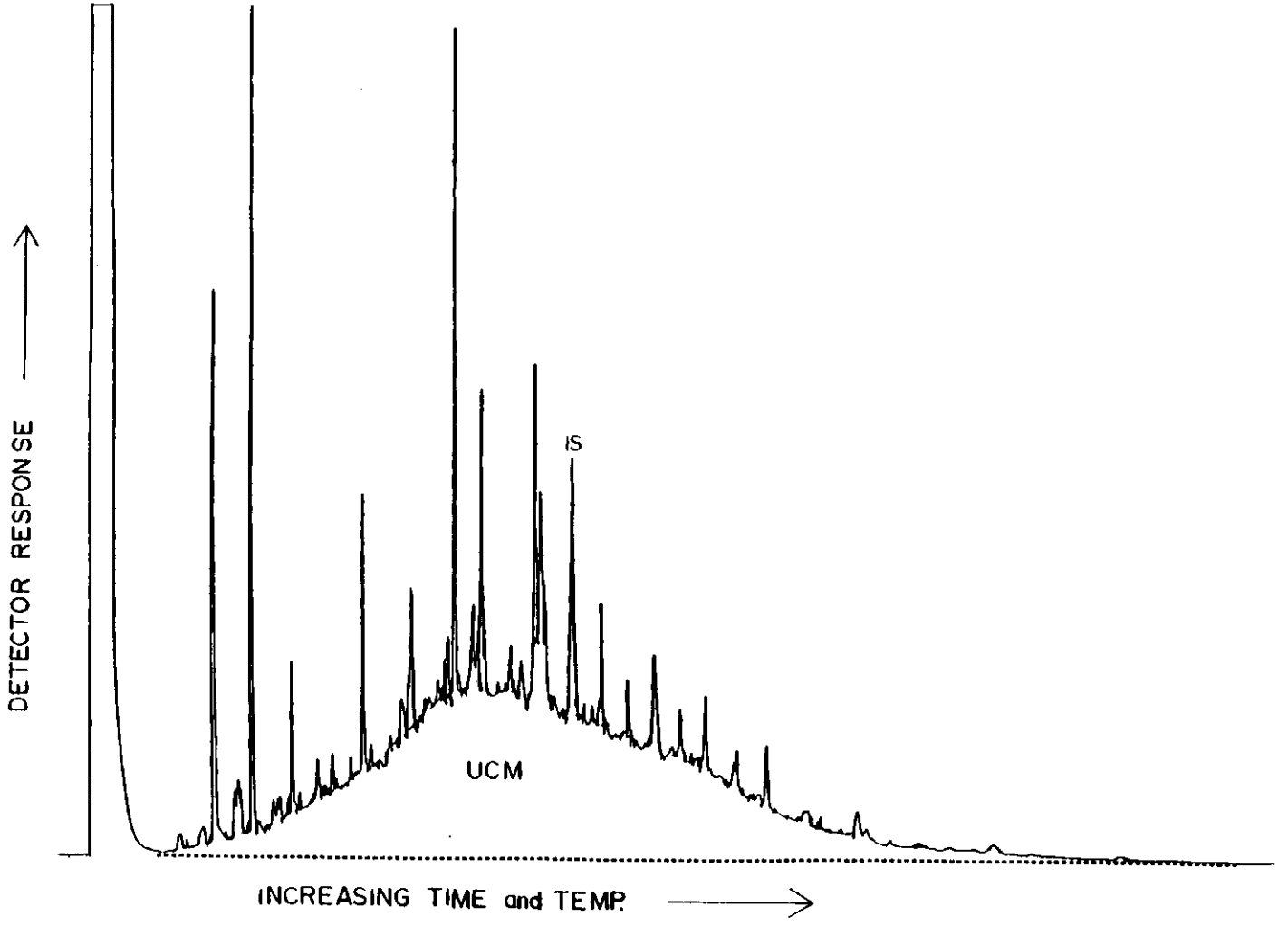
petroleum that has been in the marine environment longer than a few days and has been altered by various "weathering" processes, i.e. evaporation, photochemical oxidation, microbial degradation, etc. (NAS, 1975). An additional feature of this chromatogram is the fact that the UCM has a bimodal distribution (two maxima). This bimodal distribution denotes the possibility of two different petroleum products in the sediments. The first mode consists of low molecular weight or low boiling components. A possible source of this material is diesel or fuel oil which is used by the ships moored in the harbor. Accidental spills and leakages of this fuel may be the source of these hydrocarbons in the sediments. The second mode is typical of a weathered distillate, or crude oil, elution range n-C_{20}-C_{32}. Material in this elution range was detected in Chesapeake Elizabeth STP effluent (Figure 7). A typical chromatogram for sediment samples in the study area is shown in Figure 15. Both resolved and unresolved components are detected with an elution range from C_{12}-C_{32}, 80% of the material is comprised of the UCM. This distribution is similar to that detected in Chesapeake-Elizabeth STP effluent (Figure 7).

The concentration of hydrocarbons on a µg per gram of fine-grained sediment basis had a range of 32 to 1,679 µg/g fine-grained sediment with a mean of 169 µg/g fine-grained sediment (Table 7). These concentrations are reported in this manner because pollutants are known to

Fig. 15. Gas chromatogram of hydrocarbons from Station 2.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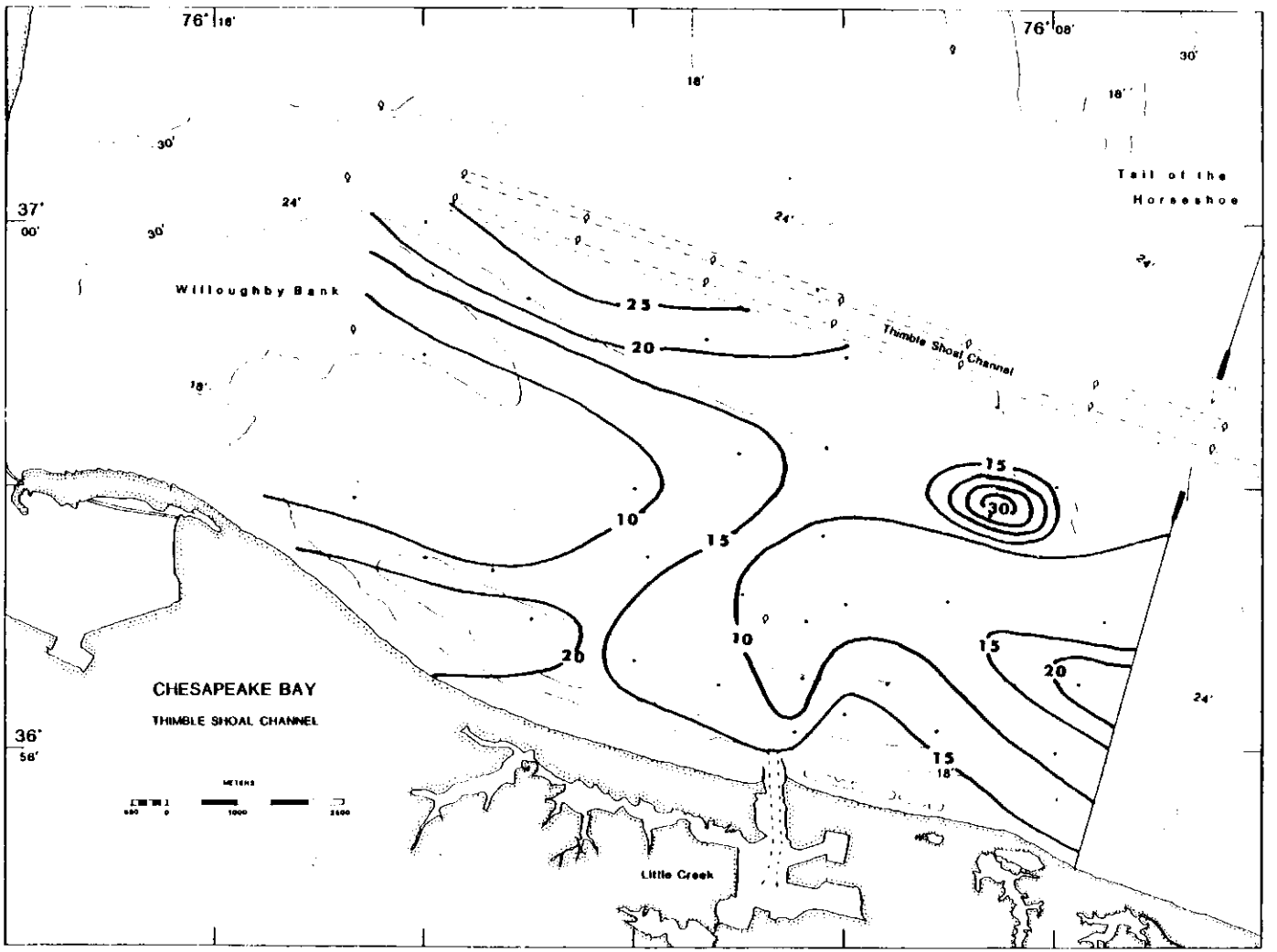
be concentrated on smaller size particles and these particles are more easily transported by currents (Meyers and Quinn, 1973; Wade and Quinn, 1979; Moy, 1980).

The hydrocarbon concentrations reported in Table 7 are contoured in Figure 16 and 17. The contour of hydrocarbon concentrations on a µg/g total sediment basis (Figure 16) shows a higher concentration along Thimble Shoal Channel and also along the shoreline. The concentrations near the STP outfall are lower. This is in contrast to the coprostanol contour (Figure 11) with high concentrations along the channel and the shore, but the highest values nearest the outfall where hydrocarbon concentrations are low (Figure 16). When hydrocarbon concentrations are contoured on a µg/g fine-grained sediment basis, higher concentrations to the east and west of the outfall are seen with much higher concentrations near the Bay Bridge Tunnel. This distribution is much different than the corresponding coprostanol contour (Figure 12). Besides sewage effluents, there are other sources of hydrocarbons entering the study area which may include discharges from ships, accidential spills, refinery activities, atmospheric deposition, etc. (NAS, 1975). Another possible unanticipated source is the Bay Bridge Tunnel. Traffic crossing the bridge may be a source of hydrocarbons to this area in the form of exhaust fumes and oil crank case drippings. These materials may be transported to the Bay by atmospheric deposition or from material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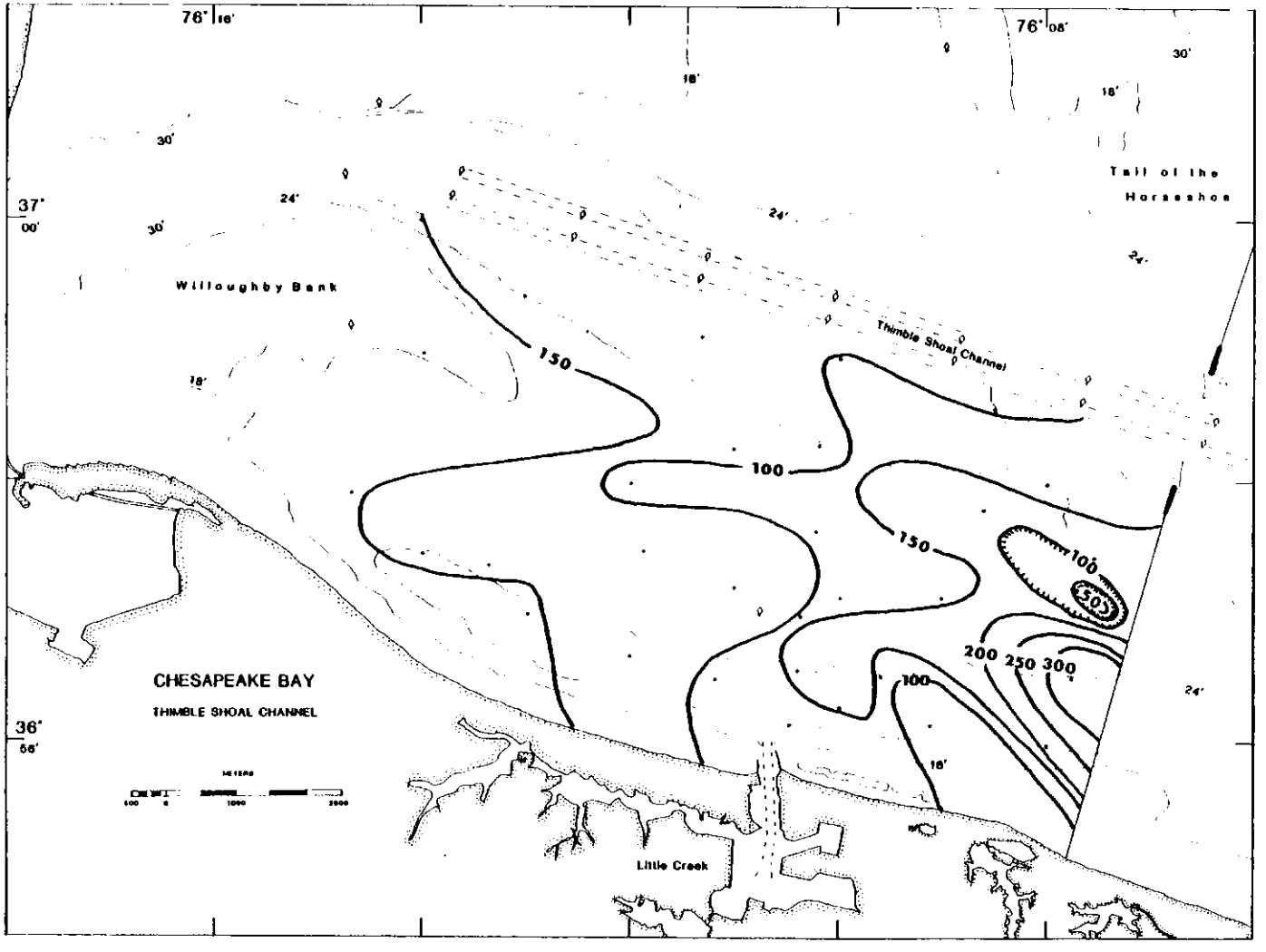
Little Creek

Fig. 17. Hydrocarbon distribution on fine grain sediments (µg/g).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CHESAPEAKE BAY
THIMBLE SHOAL CHANNEL
36°
56'
Little Creek

washed off the road during precipitation events. Additional studies are needed to determine the importance of this rather unique source.

Since the ratio of hydrocarbons to coprostanol associated with particles being discharged by Chesapeake-Elizabeth STP was determined, it is possible to estimate the amount of hydrocarbons in the sediment that originates from the effluent. The mean ratio of hydrocarbon to coprostanol concentrations in the Chesapeake-Elizabeth effluent is 9 (Table 4). The sediment coprostanol concentrations when multiplied by 9, provides an estimate of the hydrocarbons in the sediment originating from the effluent. The percentage of hydrocarbons in the sediment originating from the sewage treatment plant can then be estimated by dividing the value calculated from the coprostanol concentration by the actual sediment hydrocarbon concentrations and multiplying by 100. These percentages are reported in Table 7. The estimate of the percentage of hydrocarbons in the study area coming from the sewage treatment plant ranges from 2 to 27% with a mean of 7%. The mean percentages for 8 stations (1, 2, 2A, 3, 3, 18, 22, and 23; Table 7) close to the outfall is 17%. It appears that a larger percentage of the hydrocarbons near the outfall are derived from the effluent source with lesser percentages as you move away from the source. However, on the average, 93% of the hydrocarbons found in sediments from the study area are not from the

Chesapeake-Elizabeth STP effluent. Van Vleet and Quinn (1977), without the aid of coprostanol analyses, reported that 51% of the hydrocarbons discharged into the Providence River from a sewage treatment plant remained in the vicinity of the discharge and the remaining 49% were transported out of the area. It is likely that some portion of the hydrocarbons and coprostanol input from the Chesapeake-Elizabeth STP effluent do not remain in the study area. Supporting evidence includes detecting hydrocarbons and coprostanol in particulate samples from the Chesapeake Bay mouth and adjacent shelf waters (Wade et al., 1983; Brown and Wade, 1981).

The maximum percent of FGM in the sediments of the study area that originate from the Chesapeake-Elizabeth effluent can also be estimated, if we assume all of the coprostanol in the effluent is deposited in the study area. The concentration of coprostanol in the sediment divided by the concentration of coprostanol in the effluent times 100% gives the percentage of fine-grained sediment coming from the STP. Using mean values from Tables 4 and 6 it is estimated that only ca. 0.07% of the fine-grained sediment in the study area originated from the Chesapeake-Elizabeth STP (range from 0.01 to 0.17%). Therefore, over 99% of the FGM in the study area comes from sources other than the Chesapeake-Elizabeth STP discharge. This is contrary to the suggestion of Bates and Spencer (1979) that the Chesapeake-Elizabeth STP was a major source of

fine-grained sediment to the discharge area. It does, however, agree with Shideler's (1975) description of the lower Chesapeake Bay as being a mud-bypass area.

Conclusions

Both coprostanol and hydrocarbons enter the study area in association with the Chesapeake-Elizabeth STP effluent. Most of the coprostanol (>84%) and hydrocarbons (>91%) are associated with particulates in the effluent. These particles may be transported out of the study area or may become part of the sedimentary column within the study area. Detection of coprostanol within the study area suggest that some of the material from the Chesapeake-Elizabeth STP is deposited in the study area. However, the study area is a dynamic area where changes in fine-grained sediment distribution occur over short time periods (months). The movement of these fine-grained sediments is an important determinant of the distribution of sewage derived materials.

Calculations of the maximum amount of anthropogenic hydrocarbon input to the study area from the Chesapeake-Elizabeth STP indicate that it is only a minor source of these contaminants (~7%). The hydrocarbon sediment distribution suggest that the Bay Bridge Tunnel may be a unique source of hydrocarbons to lower Chesapeake Bay, including the area of this study.

This study shows that sewage derived materials from the Chesapeake Elizabeth STP are spread throughout the study area and perhaps beyond. Dynamics of the sediment bed appear to cause spreading and thus dilution of the sewage derived materials. If this dilution is to an extent that concentrations of potential harmful materials are diluted below their action level it may be considered a positive effect for the discharge area. However, the sewage derived materials may collect and concentrate in other areas outside the area studied, transporting their adverse effect to this new area. Examination of this question would require further research involving studies of sediment dynamics, coupled with coprostanol analyses.

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