

Oil pollution and the carbon isotope ratio in organisms and recent sediments of coastal lagoons in the Gulf of Mexico

Oil pollution
Carbon isotope ratios
Coastal lagoons
Gas chromatography
Anthropogenic sources

Pollution pétrolière
Proportions de carbone isotopique
Lagunes côtières
Chromatographie gazeuse
Sources anthropogéniques

A. V. Botello ^a, S. A. Macko ^b

^a Instituto de Ciencias del Mar y Limnología, Universidad Nacional Autónoma de México, Apartado Postal 70305, México D.F. 04510, México.

^b Carnegie Geophysical Laboratory, Washington D.C. 20008, USA.

ABSTRACT

Samples of recent sediments and marine organisms from seven coastal lagoons along the Mexican coast of the Gulf of Mexico were analyzed to determine the present levels of fossil hydrocarbons. The presence and identification of these hydrocarbons were determined by gas chromatography and GC-MS coupled system.

The hydrocarbons were extracted from sediments with methanol: benzene and the concentrate was separated by column chromatography into a hexane-benzene eluate on a mixed bed of alumina-silica gel column.

The fractions were analyzed and quantified by gas chromatography in SP1000 or OV-101 Glass Capillary Perkin Elmer Model 910 with FID.

Results of this study show that the highest concentrations of fossil hydrocarbons are present in organisms and sediments located near petrochemical plants and oil refineries, indicating that petroleum hydrocarbons are being released into the coastal lagoons. Stable carbon isotope ratios ($^{12}\text{C}/^{13}\text{C}$) of sedimentary organic carbon in sediment and organism samples were also determined. For most of the lagoons, the $\delta^{13}\text{C}$ ratios for sediments ranged from -19.0 to -23.9 ‰, anomalous ratios ranging from -26.8 to -29.3 ‰ were shown for sediments and organisms (oysters) of two of the studied areas, probably due to the presence of organic carbon from anthropogenic terrestrial sources.

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RÉSUMÉ

Pollution pétrolière et proportions de carbone isotopique dans les organismes et sédiments récents des lagunes côtières du Golfe du Mexique.

Des prélèvements de sédiments récents et d'organismes marins provenant de sept lagunes côtières situées le long de la côte mexicaine du Golfe du Mexique ont été analysés, afin de déterminer le niveau actuel des hydrocarbures fossiles. La présence et l'identification de ces hydrocarbures ont été déterminées par chromatographie en phase gazeuse et par couplage de GC-MS.

Les hydrocarbures ont été extraits des sédiments par le mélange alcool méthylique-benzène. L'extrait a été séparé par chromatographie sur colonne mixte d'alumine et de silice, en éluant par un mélange d'hexane-benzène. Les fractions ont été analysées par chromatographie en phase gazeuse sur colonnes capillaires de SP 1000 ou d'OV-101 à l'aide d'un appareil Perkin-Elmer modèle 910.

Les résultats de cette étude montrent que les concentrations les plus élevées d'hydrocarbures fossiles se trouvent dans des organismes et sédiments situés près des centrales pétrochimiques et raffineries pétrolières, ce qui indique que les hydrocarbures pétroliers sont rejetés dans les lagunes côtières. On a déterminé également les rapports des isotopes stables du carbone $^{13}\text{C}/^{12}\text{C}$ dans les échantillons de sédiments et d'organismes. Dans la plupart des lagunes, les rapports de $\delta^{13}\text{C}$ dans les sédiments varient de -19.0 à -29.3 ‰. Des valeurs anormales variant entre -26.8 et 29.3 ‰ se sont rencontrées dans des sédiments et organismes (huîtres) en deux zones, ce qui est probablement dû à la présence de carbone organique provenant de sources terrestres anthropogéniques.

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INTRODUCTION

Marine pollution by petroleum and its derivatives has aroused great interest in the scientific community, especially after the large oil spills from tankers such as the «Torrey Canyon» in England, the «Tampico Maru» on the coast of Baja California and the «Amoco Cadiz» on the Brittany coast of France, and from the Ixtoc-I oil well in the Gulf of Mexico, where thousands of tons of crude oil were spilled. Recent estimates (National Academy of Sciences, 1975) indicate that a total of 6.2 million tons of crude oil are released into the world's oceans from various sources, 2.2 million tons from the mayor source, maritime transportation. According to Gundlach (1977), 28 per cent of all petroleum released into the ocean winds up in coastal zones. The main sources of fossil hydrocarbons in oceans and coastal zones are the following:

- a) anthropogenic hydrocarbons produced by various human activities;
- b) biogenic hydrocarbons produced naturally by marine organisms;
- c) hydrocarbons from natural seeps.

In the open sea, concentrations of hydrocarbons are generally low and their origin is not always easily determined. By contrast, in bays, estuaries and coastal areas, hydrocarbons may be present in high concentrations, as a direct effect of oil pollution resulting from spills, wastes from petrochemical plants and refineries, normal loading operations and tropospheric transport.

The effects that large oil spills and the use of dispersants have on marine biota have been widely documented and published (Blumer *et al.*, 1970; Blumer, Sass, 1972; Sanders *et al.*, 1972; Portman, Connor, 1968; Crapp, 1971). Nevertheless, very little is known about the biological and physiological effects of oil pollution in coastal waters or estuarine systems.

Mackin and Hopkins (1964) have postulated that in certain coastal areas the continental release of oil-derived products has little or no effect upon the populations or productivity of the region. However, other researchers suggest that some important biological processes, such as photosynthesis and respiration, are affected by relatively low concentrations of petroleum (Gilfillan, 1973; Jacobson, Boylan, 1973; La Roche, 1973; Parker, Menzel, 1974; Pulich *et al.* 1974). Generally speaking, the biological or biochemical activity of oil components in estuaries and coastal zones is dependent on the following parameters:

- a) the levels at which hydrocarbons are bioaccumulated;
- b) the amount of time hydrocarbons remain in the water column and, subsequently, in organisms and sediments;
- c) the composition of the hydrocarbon mixture in the water and subsequently in organisms.

Furthermore, in marine organisms these parameters are modified by biological factors such as lipid content, efficiency of hydrocarbon consumption and hydrocarbon entry and waste routes.

The use of the carbon isotope ratio ($^{12}\text{C}/^{13}\text{C}$) has increased considerably in recent years. Many scientific disciplines, such as chemistry and geochemistry, use it as a helpful research tool. At present it is effectively employed in the study of trophic relations in coastal and estuarine systems (Haines, 1976 a ; 1976 b ; Haines, Montague, 1979 ; Fry *et al.*, 1977 a ; Fry, 1977) and as an excellent indicator of organic pollution processes (Calder, Parker, 1968 ; Botello *et al.*, 1980 ; Burnett, Schaeffer, 1980 ; Hartman, Hammond, 1981).

Until a few years ago, information about the carbon isotope ratio in coastal systems of the Gulf of Mexico was scanty and limited (Gearing *et al.*, 1978 ; Botello, 1978) and even more so for coastal lagoons (Fry, 1977 ; Botello, 1980). For this reason, the present study was undertaken, with the objective of characterizing the natural and/or artificial source of organic carbon present in coastal areas.

MATERIALS AND METHODS

Hydrocarbons

Sediments and organisms were collected from those lagoons and coastal areas of the Gulf of Mexico considered to be the most important (Fig. 1). Prior to analysis all samples were frozen at -20°C according to the recommendations of Clark (1966).

Gas chromatography.

For the hydrocarbon analysis, a Perkin Elmer gas chromatograph model 910 was used, equipped with flame ionization detector, linear temperature programmer, electronic integrators and capillary columns 30 m long with an internal diameter of 0.25 mm, siliconized with OV-101 and SP-1000. The temperature was programmed from 60° to 255°C with an increase of $3^{\circ}\text{C}/\text{min}$. Nitrogen was used as carrier gas, with an average flow of 25 ml/min. The samples were analyzed before and after the addition of an internal standard containing pristane, phytane and n-paraffins ranging from C_{16} to C_{32} . The amount of each component was determined by its corresponding area in the peaks of each chromatogram.

A Varian Aerograph gas chromatograph model 2700 coupled to a Dupont mass spectrometer model 21-491 was used to analyze the structure of the isomers present in the samples and to confirm the presence of certain n-paraffins, under the same experimental conditions described above.

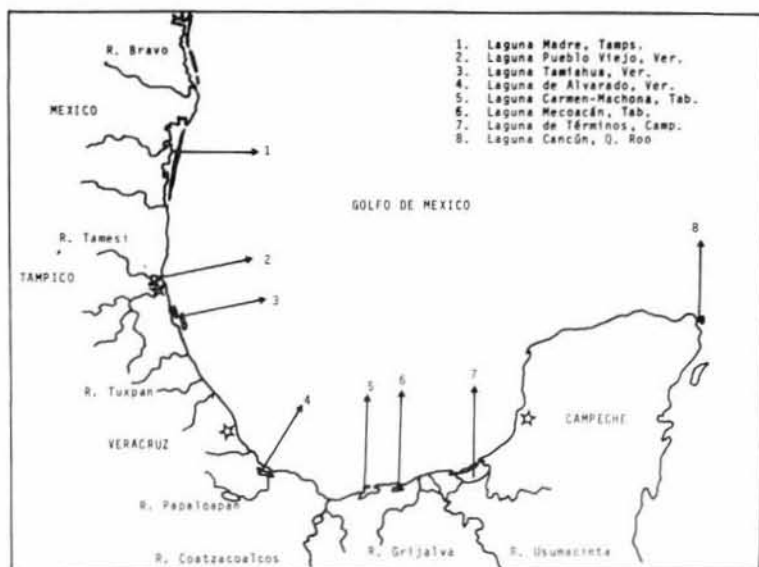


Figure 1
Location of sampling sites for recent sediments and oysters.

Carbon isotope ratios.

To determine $\delta^{13}\text{C}$ (carbon isotope) ratios, the sediment and organism samples were treated with HCl 0.2N to eliminate carbonates, according to the method described by Craig (1953). A small aliquot of the samples was mixed with OCu and burned in a LECO radio frequency furnace at 750 °C in the presence of pure oxygen at a pressure of 800 mm of mercury. The products of the combustion were kept in a glass tube submerged in liquid nitrogen which was later recycled over MnO at 500 °C.

The purified CO_2 was analyzed in a mass spectrometer for isotope ratio (THN 204 Nucleide Inc.), using as standard graphite NBS = - 21 ‰. Based on the analysis of 40 duplicated samples, the total average of error in the combustion and measurement was ± 0.2 ‰.

RESULTS AND DISCUSSION

Hydrocarbons in recent sediments.

Sediments serve as the receptacle for dispersed substances and matter in the water column, which means that their chemical analysis is very useful for the detection of certain pollutants in aquatic ecosystems.

Hydrocarbons of biological origin are present in a large number of marine organisms and in all recent sediments. Fortunately, there are differences in composition and structure between these indigenous hydrocarbons and those derived from petroleum, thus making it possible to distinguish clearly between them (Clark, Blumer, 1967).

Nonpolluted recent sediments reflect the composition of organisms which have contributed organic matter to them. Among the hydrocarbons there is a marked predominance of n-alkanes with odd-numbered carbon atoms (Stevens *et al.*, 1965; Bray, Evans, 1961; Clark, Blumer, 1967; Anderson *et al.*, 1974; Farrington, Quinn, 1973).

In nonpolluted coastal zones the concentration of biogenic hydrocarbons in recent sediments is less than 70 ppm (Blumer, Sass, 1972; National Academy of Sciences, 1975).

In sediments of polluted zones, concentrations of fossil hydrocarbons, which may vary widely depending on the source of the pollution and the characteristics of the ecosystem, are much greater than concentrations of biogenic hydrocarbons; similarly, the composition of fossil hydrocarbons in sediments may be quite varied, depending on the amount of time they have remained in the sediments and, obviously, the original composition of the petroleum from which they come.

Fossil hydrocarbons may remain in the sediments from 3 to 10 years, depending on the rate of degradation of the sediment (Moore *et al.*, 1973). In coastal zones the rate of

Table 1

Total hydrocarbons concentration in recent sediments from coastal lagoons of the Gulf of Mexico.

Lagoon	Total hydrocarbons ($\mu\text{g/g}$ dry weight)		
	Range	Average	OEP Value*
Madre, Tams.	10-40	26	3.5
Pueblo Viejo, Ver.	17-80	53	2.6
Tamiahua, Ver.	15-62	31	3.7
Alvarado, Ver.	10-30	18	4.2
Carmen-Machona, Tab.	6-150	45	3.0
Mecoacán, Tab.	10-1 060	88	2.7
Términos, Camp.	10-50	37	4.1
Cancún, Q. Roo	8-18	12	4.4

* Scalap and Smith, 1970.

degradation is much higher because the presence of nutrients, light, temperature, oxygen and substrate permits high rates of bacterial biodegradation and photochemical oxydation.

To date, very little research on this aspect has been carried out in the coastal area of the Gulf of Mexico (Parker, Calder, 1972; Gearing *et al.*, 1977; Botello *et al.*, 1976; Botello, Mandelli, 1978 and Botello, 1980).

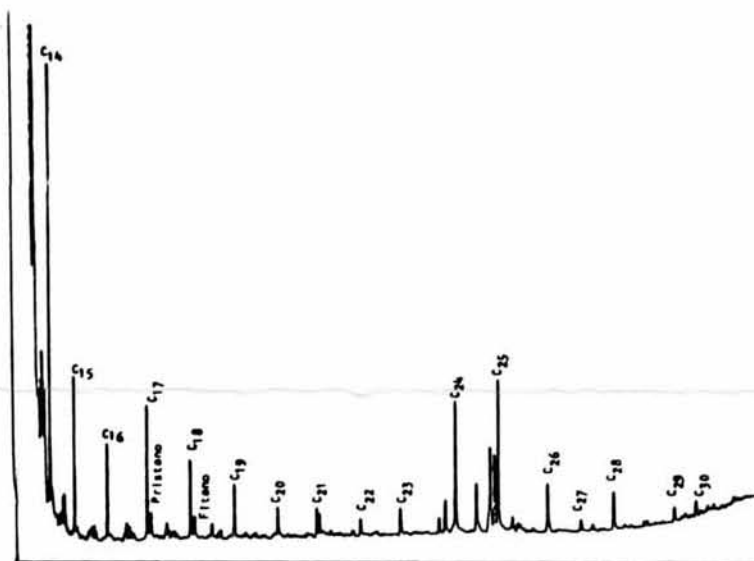
Analysis of hydrocarbons in sediments of coastal lagoons in the Gulf of Mexico confirms the predominance of n-paraffins with odd-numbered carbon atoms in an area for which no previous information existed. This predominance of odd-numbered paraffins falls within the range of values reported by Cooper and Bray (1963) for sediments of other coastal regions and indicates that the natural source of a large percentage of hydrocarbons may be organisms indigenous to the sediments or detritus of terrestrial plants, rather than pollution caused by the various human activities carried out in these areas.

Table 1 presents the total hydrocarbon concentration ratios obtained in the analysis of recent sediments. Clearly evident is the great uniformity of low hydrocarbon concentrations, with the exception of the mouth of the Mecoacan lagoon in Tabasco, which recorded ratios up to 1060 ppm dry weight, due to the arrival of crude oil from the Ixtoc-I spill. In 90 % of the samples the total concentration of hydrocarbons in the sediments studied was less than 70 ppm, tallying with ratios reported for nonpolluted coastal zones, deep marginal seas and certain ocean basins (Blumer, Sass, 1972; 1975).

The chromatograms for the analyzed sediments provide

Figure 2

Chromatogram showing the distribution of n-paraffins in recent sediments from Términos Lagoon, Tabasco, Mexico.



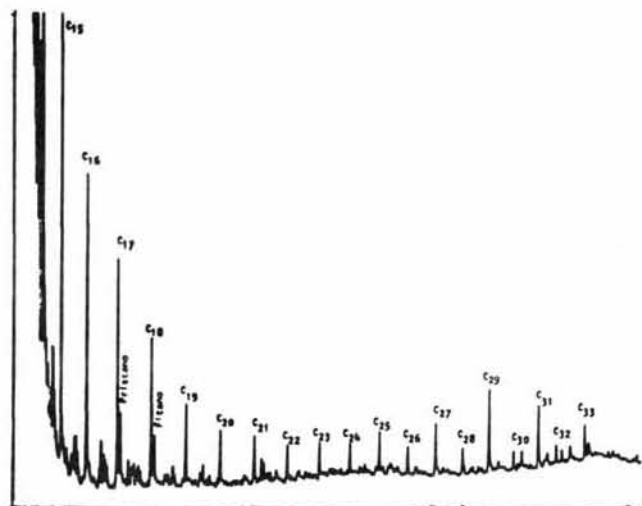


Figure 3
Chromatogram showing the distribution of n-paraffins in recent sediments from Carmen-Machona Lagoon, Tabasco, Mexico.

evidence of the nature of the hydrocarbons present and are of great help in the interpretation of observed differences among the samples.

Figures 2 and 3, which present chromatograms of samples from sediments with low hydrocarbon concentrations, consist principally of a series of peaks representing the n-paraffin distribution in a range from n-C₁₄ (tetradecane) to n-C₃₂ (dotriacontane).

In these chromatograms there is an obvious predominance of paraffins with odd-numbered carbon atoms. The major peaks represent n-C₁₅, n-C₁₇, n-C₂₁, n-C₂₅, n-C₂₉ and n-C₃₁, respectively. This predominance of odd-numbered carbon atoms in the n-C₂₀ range is common for recent non-oil-polluted freshwater and marine sediments (Clark, Blumer, 1967; Blumer, Sass, 1972; Giger, Blumer, 1974).

The most important source of hydrocarbons in recent sediments of the areas studied is thought to be detritus of terrestrial plants, especially mangroves, which is carried to lagoons by rivers and land run-off.

Figure 4 shows the chromatogram of n-paraffins from the sampling station located at the mouth of the Mecoacan lagoon, which receives considerable amounts of crude oil from Ixtoc-I.

It may be observed in this figure that the distribution of n-paraffins differs considerably from the previous chromatograms, as there is no predominance of odd-numbered paraffins, a condition which is characteristic of crude oil. In addition, the figure shows the presence of the isoprenoid

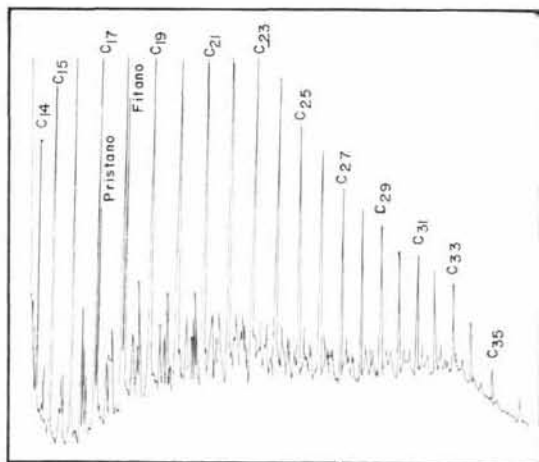


Figure 4
Chromatogram showing the distribution of n-paraffins in recent sediments from the sampling station located at the mouth of Mecoacan Lagoon.

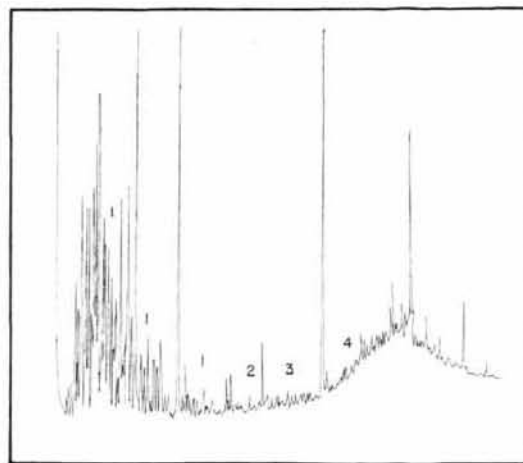


Figure 5
Chromatogram showing the aromatic compounds in recent sediments from Mecoacan Lagoon, Tabasco, Mexico: 1: naphthalene and methyl naphthalenes; 2: dibenzothiophene; 3: phenanthrene; 4: fluorene.

compounds pristane and phytane and components ranging from n-C₁₄ to n-C₃₅. The Carbon Preference Index (Cooper, Bray, 1963), 1.0, is typical of crude oils, thus confirming the conclusion suggested by the absence of odd-numbered paraffins that the sediment sample contained crude oil from the spill in Campeche Bank.

Figure 5 shows the presence of polynuclear aromatic hydrocarbons such as the dibenzothiophenes, phenanthrene, anthracene, benzoanthracene and fluorene as well as the simplest aromatics such as the naphthalenes and their methyl derivatives.

These results are of singular importance from an ecological point of view, since the majority of these organic compounds are not produced or synthesized naturally and thus their presence in the environment implies that they have been released by anthropogenic actions (Farrington, Quinn, 1973; Bravo *et al.*, 1978).

Many of these compounds, having been released into the environment, wind up in the sediment column, where, due to the great chemical stability afforded them by their complex structure, they are stored for a long period of time before being transformed or biodegraded.

Hydrocarbons in organisms (*Crassostrea virginica*)

The sedentary life of most bivalves makes them extremely vulnerable to acute and chronic exposure to environmental pollutants such as fossil hydrocarbons from petroleum (Blumer *et al.*, 1970; Lee *et al.*, 1972; Clark, Finley, 1973; Di Salvo *et al.*, 1975)

Analysis of marine organisms exposed to oil spills have demonstrated their capacity to assimilate and store hydrocarbons (Blumer, Sass, 1972; Burns, Teal, 1971; Clark, Finley, 1973). Benthic organisms from coastal areas with a high degree of hydrocarbon pollution generally show much higher hydrocarbon concentrations than the water in which they live.

Some of the hydrocarbons ingested with food or taken from the water area stored in different tissues of marine animals. Perhaps because of their high lipid content, the liver of fish and the hepatopancreas of invertebrates are sites where hydrocarbons are stored (Lee *et al.*, 1972). There is some evidence to indicate that different hydrocarbons have different retention time in organisms; for example, oysters exposed to oil polluted waters accumulate more aromatic hydrocarbons than paraffin hydrocarbons (Anderson, 1973; Blumer *et al.*, 1970).

Consumption and degradation of aromatic and paraffin hydrocarbons occur in some fish and crustaceans (Lee *et al.*, 1972; Corner *et al.*, 1973; Lee, 1975; Botello, 1975).

Table 2

Total hydrocarbons concentration in *Crassostrea virginica* from coastal lagoons of the Gulf of Mexico.

Lagoon	Total hydrocarbons ($\mu\text{g/g}$ dry weight)		OEP Value*
	Range	Average	
Pueblo Viejo, Ver.	27-130	85	1.2
Tamiahua, Ver.	20-129	43	1.8
Alvarado, Ver.	40-46	43	1.6
Carmen-Machona, Tab.	30-352	92	1.4
Mecoacán, Tab.	8-552	81	1.2
Términos, Camp.	30-50	37	2.4

* Scalan and Smith, 1970.

However, the degree of hydrocarbon metabolism in organisms belonging to other zoological groups is not known with certainty at present. Oysters, mussels and other benthic mollusks remove hydrocarbons from the water column by filtration, but they seem to lack the capacity to metabolize these compounds.

In this study, examples were analyzed of *Crassostrea virginica* from different oyster's beds located in the coastal lagoons of the Gulf of Mexico, areas where many physical, chemical and biological factors as well as various geological processes interact, making each lagoon a unique environment.

The total hydrocarbon concentration is shown in Table 2. The results obtained are similar to those reported previously by Bravo *et al.* (1978) and Botello (1978), thus confirming that certain concentrations of hydrocarbons generated by diverse oil-industry activities carried out in those zones are released regularly into the above-mentioned lagoon ecosystems. These concentrations agree with those reported by Blumer *et al.* (1970), Erhardt (1972) and Farrington (1973) for *Crassostrea virginica* from sites affected by spills and continual oil discharges, such as Buzzards Bay, Massachusetts, Narragansett Bay, Rhode Island and Galveston Bay, Texas.

As far as the distribution pattern of n-paraffins in the organisms that were analyzed, the corresponding chromatograms (Fig. 6) indicate the presence of saturated compounds which range from n-C14 (tetradecane) to n-C29 (nonacosane), with a slight predominance of odd-numbered

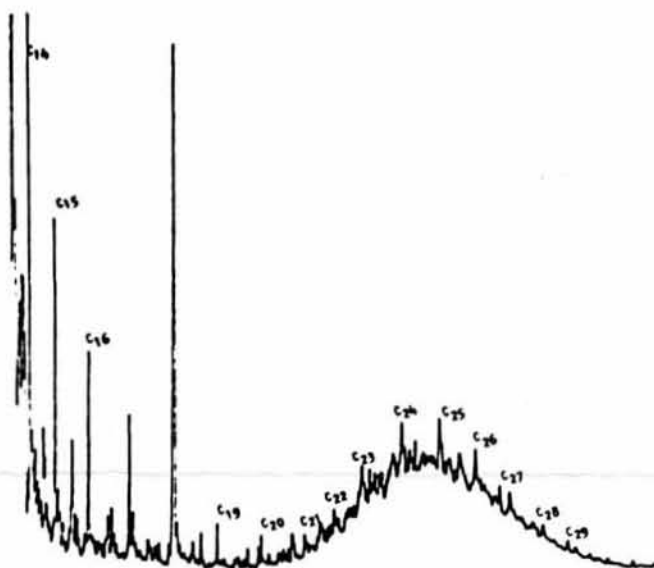


Figure 6
Chromatogram showing the distribution of n-paraffins in *Crassostrea virginica* from Carmen-Machona Lagoon, Tabasco, Mexico.

carbons; the major peaks represent C15, C16, C19, C24 and C25, respectively. The presence, as well, of the isoprenoid hydrocarbons pristane and phytane is significant, since, taking into consideration the findings of Blumer *et al.* (1964) that zooplankton seems to be the principal source of these isoprenes for other marine organisms, it suggests that planktonic organisms make up an essential part of the diet of *Crassostrea virginica*.

The distribution of n-paraffins in *Crassostrea virginica* is very similar to the pattern reported for recent sediments, which confirms the close interrelation between the water column, sediments and organisms in the distribution and retention of hydrocarbons.

Polynuclear aromatic hydrocarbons are not considered to be normal metabolic products of marine fauna, but they may nevertheless be ingested with food, principally through filtration. The determination of PAH's levels in bivalves has recently aroused much interest, since it helps to determine the present state of oil pollution in coastal regions.

Figure 7 presents aromatic fraction chromatograms for *Crassostrea virginica* from the Mecoacan lagoon. A great variety of PAH's are present, some — such as benzopyrenes, chrysenes, fluorantenes, perylenes, etc. — of biological importance because of their potential carcinogenic activity and thus their implications for public health.

These results agree with those reported earlier by Bravo *et al.* (1978) and Botello (1978), thus confirming that the Carmen, Machona and Mecoacan lagoons receive waste from oil-industry activities carried on in the proximity.

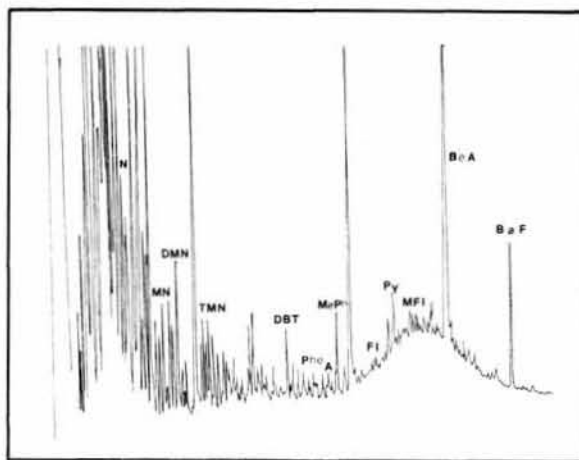


Figure 7

Chromatogram showing the aromatic compounds in *C. virginica* from Mecoacan Lagoon, Tabasco, Mexico. N: naphthalene; MN, DMN, TMN: methyl naphthalenes; DBT: dibenzothiophene; Phe: phenanthrene; A: anthracene; MePh: methylthiophene; FI: fluorene; PY: perylene; MFL: methylfluorene; Be-A: benzoanthracene.

Carbon isotope ratios ($^{13}\text{C}/^{12}\text{C}$)

Because of its biochemical and geochemical importance, carbon has been a useful tool in this research. In turn, this analysis offers valuable information on the sources of carbon in an ecosystem.

For technical reasons, the analysis results are reported in δ (delta) ratios, *i.e.*, the samples's relative deviation per thousand with reference to a standard (Chicago PDB Belemnite, Craig, 1953), based on the following formula:

$$\delta^{13}\text{C}\text{‰} = \frac{(^{13}\text{C}/^{12}\text{C} \text{ sample} - (^{13}\text{C}/^{12}\text{C}) \text{ standard})}{^{13}/^{12} \text{ standard}} \times 1000.$$

The majority of studies on the organic carbon isotope ratios in the marine environment have been carried out in sediments and organisms of regions near the coast of the Gulf of Mexico, specially the region belonging to the USA. Studies carried out by various researchers in this field have reported $\delta^{13}\text{C}$ ratios of -20‰ to -16‰ for sediments and

Table 3
 $\delta^{13}\text{C}$ values and TOC in sediments and oysters (*C. virginica*) from coastal lagoons of the Gulf of Mexico.

Lagoon	Sample No.	Sediments		<i>Crassostrea virginica</i>
		TOC %	$\delta^{13}\text{C}$ ‰	$\delta^{13}\text{C}$ ‰
Madre	1	4.51	-23.9	—
	2	0.67	-20.1	—
	3	2.47	-20.3	—
Pueblo Viejo	4	5.52	-26.8	-24.7
Tamiahua	5	3.90	-20.5	-20.8
Alvarado	6	0.42	-21.8	-21.9
Carmen y Machona	7	4.52	-27.4	-24.9
	8	7.20	-29.2	-24.9
	9	3.84	-23.2	-25.0
	10	4.01	-23.0	-24.4
	11	5.23	-23.1	-23.6
	12	4.82	-23.8	-24.0
	13	6.15	-26.2	-29.7
Mecoacán	14	5.42	-23.3	-27.4
	15	4.73	-25.1	-27.0
	16	3.94	-24.8	-26.7
	17	4.76	-24.3	-26.5
	18	5.24	-25.8	-28.1
Términos	19	2.06	-21.3	—
	20	2.77	-21.2	—
	21	3.69	-22.5	-21.8
	22	2.76	-22.4	-21.6
	23	3.02	-20.9	-21.6
	24	2.44	-19.0	-21.0

organic matter from the continental shelf, the ratios becoming more negative as the influence of terrestrial detritus or fossil fuels becomes greater (Parker, 1964; Calder, 1971; Gearing *et al.*, 1977; Haines, Montague, 1979).

The present research was carried out in order to obtain preliminary information on $\delta^{13}\text{C}$ ratios in recent sediments and organisms of the coastal lagoons of the Gulf of Mexico and to complete already available information on this subject for the same region, because of the area's current importance as a site of urban and industrial development. Table 3 shows the distribution of $\delta^{13}\text{C}$ ratios for recent sediments and oysters (*Crassostrea virginica*) from the various sampling stations in the lagoons considered in the present study.

The $\delta^{13}\text{C}$ ratios for recent sediments in these lagoons range from -20.5 to -29.2 ‰. The two most negative ratios are those for the Mecoacan and Carmen-Machona lagoons, which receive waste from the Mecoacan and Sanchez-Magallanes oil fields.

The ratios for the remaining lagoons range from -20.5 to -25.7 ‰ which may reflect the fact that the main source of organic carbon for these lagoons is the detritus contributed by surrounding vegetation, in this case, mangrove, whose $\delta^{13}\text{C}$ ratios range from -23 to -25.5 ‰.

The ratios noted here differ from those reported earlier by Botello (1978) and Botello *et al.* (1980), with more uniform $\delta^{13}\text{C}$ ratios (-23 to -20.1 ‰) recorded for the Madre, Tamiahua, Alvarado, Tampamachoco and Terminos lagoons, results which suggest that a great amount of organic carbon present in recent sediments of these lagoons is basically of marine origin.

Nevertheless, the data reported for recent sediments of the Mecoacan and Carmen-Machona lagoons seems to indicate the existence of a mixture of organic matter of terrestrial and marine origin. In general, the carbon isotope ratio tends to remain constant, so any alteration probably implies a notable change in the composition of the original source of organic carbon.

$\delta^{13}\text{C}$ ratios were also determined for tissues of oysters collected in the lagoons under study. The results are shown

in Table 3, where it may be observed that the most negative ratios (-29.7 and -28.1) are those recorded for the Mecoacan lagoon, which also recorded the most negative $\delta^{13}\text{C}$ ratios for recent sediments. The remaining ratios, although they are not as negative, differ considerably from those reported by Botello (1978) and Botello *et al.* (1980) for the same species from other coastal lagoons of the Gulf of Mexico, where the carbon isotope ratios do not seem to be altered by the presence of non-biogenic organic carbon with very negative $\delta^{13}\text{C}$ ratios, such as fossil hydrocarbons released into these environments by human activities.

CONCLUSIONS

Analysis of samples of recent sediments and organisms from the coastal lagoons under study indicates that both contain fossil hydrocarbons at detectable but not dangerous levels, since the concentrations remained stable throughout the present study.

These hydrocarbons are undoubtedly generated by the various human and industrial activities which are carried on in close proximity to the lagoons. Once discharged, they enter the water column in very low but constant solutions. Of the areas studied, those of the Pueblo Viejo lagoon in Veracruz and the Carmen-Machona and Mecoacan lagoons in Tabasco showed the highest hydrocarbon concentrations. These are considered to be relatively polluted zones because of discharges from refineries, petrochemical plants and oil wells close to these sites.

The presence of polyaromatic hydrocarbons (PAH's) in *Crassostrea virginica* as well as in sediments indicates that these invertebrates retain fossil hydrocarbons in their tissues. These hydrocarbons, which come from petroleum, are released regularly to estuaries, where they interfere with or block important biological processes.

Measurement of the carbon isotope ratio in the different components of the ecosystem studied shows that these lagoons have two natural sources of organic carbon: mangrove and phytoplankton.

In addition to the natural sources mentioned above, there are others of an artificial nature which release into the lagoons anthropogenic organic carbon (fossil hydrocarbons) with isotope ratios similar to those reported for crude oils and/or products derived from the petrochemical industry.

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