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OCCURRENCE AND ORIGIN OF NATIVE SULFUR IN CAPROCK FROM TATUM SALT DOME, MISSISSIPPI

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ABSTRACT

Native sulfur in caprock from Tatum Dome occurs as fine-grained vug linings in the limestone portion of the caprock, and as veinlets and disseminations in the underlying anhydrite zone. Textural relationships indicate that both occurrences formed late in the history of caprock development. In the anhydrite zone, replacement of anhydrite by sulfur occurred simultaneously with gypsification. This observation, along with the recognition that the formation of gypsum "transition" zones here and probably at other domes is a relatively late occurrence, indicates that the gypsum-forming solutions may also be responsible for the oxidation of H_2S to native sulfur. These solutions were probably cool and of relatively low salinity, based on gypsum stability relationships, and were probably shallow circulating seawater or groundwater.

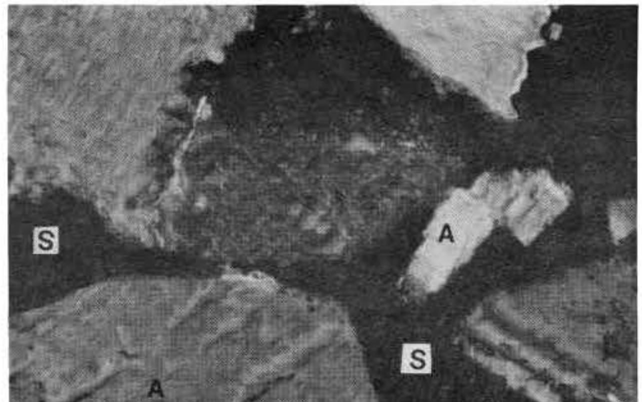

Figure 1. Photomicrograph of sulfur (S) veinlet crosscutting anhydrite (A). Sample WP-1-1075, crossed polars, field of view is 0.64 mm.

INTRODUCTION

Tatum Salt Dome is located in Lamar County, Mississippi, and was the site of two underground nuclear tests in the early 1960's. Caprock at the dome was explored for its sulfur potential by Freeport Sulphur Company in the 1940's, but only minor concentrations were discovered. However, the occurrence of sulfur and its relative timing of deposition at Tatum Dome is significant because it provides insight into the geochemical processes that operate to precipitate native sulfur, particularly when viewed in the context of recent caprock studies.

GEOLOGIC SETTING

Tatum Dome is a shallow piercement-type salt dome, the top of which lies approximately 900 feet below the ground surface. The upper part of the dome is roughly circular in outline and is approximately one mile in diameter. There is an overhang that extends around the entire circumference of the dome and is most pronounced on the southwest margin. Caprock at Tatum Dome exhibits characteristics typical of zoned caprocks from other Gulf Coast salt domes (Halbouty, 1979). The caprock is 500-600 feet thick over the center of the dome and consists of an upper limestone zone 68-202 feet thick, a gypsum-rich zone 0-40 feet thick, and a lower anhydrite zone up to 460 feet thick. Field descriptions (Eargle, 1962a,b) and petrographic studies (Schlocker, 1963; Saunders, in press a,b) of core from two holes drilled by the U.S. Atomic Energy Commission indicate that the limestone caprock is composed of an upper zone of massive equigranular calcite and a lower banded calcite zone. In the banded zone, dark bands of fine-grained calcite alternate with lighter-colored bands of coarse crystalline, euhedral calcite, celestite ($SrSO_4$), and strontianite ($SrCO_3$). The dark calcite bands contain abundant bitumen (very mature hydrocarbons) and minor amounts of pyrite, sphalerite, galena, and doubly terminated quartz crystals. Veinlets of bitumen are present in the upper massive calcite portion of the limestone caprock, and they typically contain sulfide minerals. Strontium minerals and sulfides were apparently deposited during the episodic intrusion of hot and metalliferous brines into the caprock during the development of the limestone portion (Saunders, in press a,b). The degree of activity of sulfate-reducing bacteria, using liquid hydrocarbons as a nutrient source, controlled whether sulfide or sulfate minerals precipitated along with the carbonates.

SULFUR OCCURRENCE

Sulfur has been observed in both the limestone and anhydrite portions of the caprock. Minor amounts of sulfur were reported in the gypsum zone in the logs from the Freeport Sulphur Company exploration wells, but none of the gypsum zone was recovered in the A.E.C. core holes. In the limestone zone, sulfur is fairly rare and occurs as fine crystalline linings along walls of vugs. Textural evidence indicates that sulfur was the last mineral to precipitate in the limestone caprock (Saunders, in press a). In the anhydrite zone, sulfur occurs as veinlets up to 1 mm in width (Figure 1), or as disseminated aggregates up to 1 cm in diameter, both of which appear to crosscut and replace anhydrite. The disseminated sulfur aggregates contain intergrowths of equant, yellow-green sulfur crystals up to 0.5 mm in diameter. Sulfur in the veinlets is finer-grained and commonly contains gypsum and minor amounts of calcite, bitumen, and "islands" of unreplaced anhydrite.

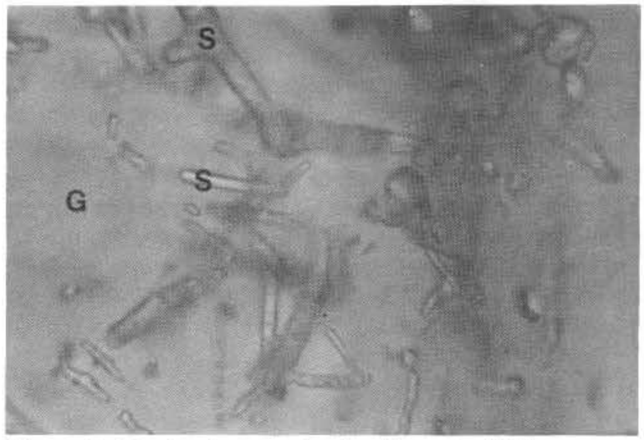

Figure 2. Photomicrograph of sulfur rods (S) in gypsum (G). Sample WP-1-1075, transmitted light, field of view is 0.12 mm.

Sulfur in the veinlets exhibits a variety of textures. Near the edge of the veinlets, sulfur typically occurs as rod-shaped or curved or anastomosing tubules up to 0.1 mm in length (Figures 2 and 3). This sulfur morphology occurs in the rims of anhydrite crystals adjacent to the veinlets and, more commonly, in gypsum within the veinlets. In the latter case, the rods are commonly highly segmented (Figure 3). The sulfur rods appear to coalesce toward the center of the veinlets to form the more massive sulfur intergrowths typical of the bulk of sulfur in veinlets (Figure 4). This sulfur is more equant and may have formed by the recrystallization of the rods.

DISCUSSION

The general aspects of the formation of sulfur at the expense of anhydrite in the caprock were established by Feely and Kulp (1957). The process involves two steps, including: 1) reduction of sulfate sulfur to sulfide (i.e. H_2S) by anaerobic bacteria with simultaneous oxidation of the hydrocarbon nutrient source to produce CO_2, resulting in the precipitation of biogenic calcite,

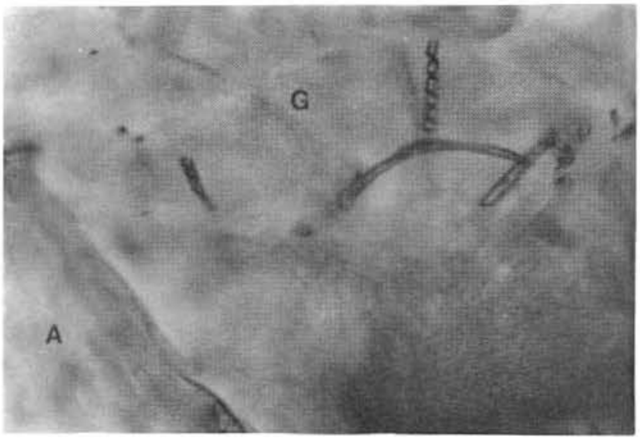

Figure 3. Photomicrograph of a segmented sulfur rod in gypsum (G) adjacent to anhydrite (A). Sample WP-1-1457, transmitted light, field of view is 0.16 mm.

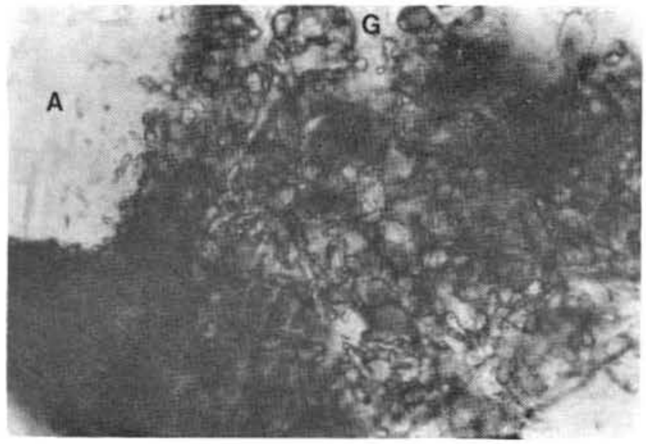

Figure 4. Photomicrograph of massive sulfur intergrowth in gypsum (G) adjacent to anhydrite (A). Sample WP-1-1457, transmitted light, field of view is 0.16 mm.

and 2) oxidation of sulfide to native sulfur. The mechanism by which the oxidation is accomplished is a matter of considerable debate. Feely and Kulp (1957) proposed that aqueous sulfate could oxidize sulfide, but Davis et al. (1970) demonstrated that this reaction is possible only at unreasonably high temperatures. Instead, they proposed that oxygenated groundwater is the likely oxidant, as did Ruckmick et al. (1979).

Geochemical considerations indicate that Step 2 must have occurred some time after Step 1 in most cases, for it is difficult to envision an environment that could both reduce and oxidize sulfur simultaneously on a large scale. Alternatively, the migration of H_2S gas out of the reducing environment to an oxidizing environment could result in approximately simultaneous redox reactions. In this scenario, sulfur is deposited stratigraphically higher in the typically porous and highly permeable limestone caprock rather than its point of "release". For commercial sulfur deposits to form, both a trap for H_2S and an oxidant are required (Davis and Kirkland, 1979). The redox processes described above cause sulfur to form at a volumetric ratio of 1:4 with respect to the cogenerated biogenic limestone (Davis and Kirkland, 1979). Many domes have large volumes of limestone and little sulfur (as at Tatum), indicating that much of the sulfur has migrated out of the caprock.

Sulfur veinlets in the anhydrite portion of Tatum Dome caprock apparently formed by replacement of anhydrite at the site. Textural relationships indicate how this was accomplished. The association of minor amounts of bitumen and calcite with sulfur indicates that sulfate reduction did take place along the veinlets, although some H_2S probably also migrated into the site along the fractures. One mechanism by which the sulfur rods and tubules may have formed involves the replacement of anhydrite by gypsum initially. Gypsum forms by the hydration of anhydrite below temperatures of 57°C (Holland and Malinin, 1979) and proceeds from the grain boundary inward. If a colony of sulfate-reducing bacteria were active at the advancing anhydrite/gypsum boundary, then it would leave a "trail" behind it where H_2S could be oxidized to produce the rods and tubules. In this situation, the only place where reduction is taking place is in the immediate vicinity of the bacteria colony. The solutions moving through the veinlets that caused anhydrite replacement were probably somewhat oxidizing, low temperature, probably dilute, and may have provided the hydrocarbons necessary for sulfate reduction as well. Volume expansion attendant with the replacement of anhydrite by gypsum may have caused the segmentation of the sulfur rods due to dislocations along cleavage planes.

The source of the sulfur-forming solutions is problematic due to the lack of data on the timing of this mineralization in relation to the salt dome growth history. Two possibilities exist. If the sulfur is of very recent origin, oxidized groundwater could be responsible for veinlet formation, because of the shallow depth of the caprock. For example, groundwater has formed a 400 foot-thick gypsum zone overlying anhydrite at Gyp Hill Dome, Texas (Kreitler and Dutton, 1983). Alternatively, many caprocks appear to have formed at or just below sea level throughout most of their formational history (Posey et al., 1987). Isotopic studies of caprock minerals by Posey (1986) and Posey et al. (1987) suggest some contribution by seawater. In addition, Werner (1986) concluded that the formation of gypsum veins in the anhydrite portion of the caprock at Richton Dome, Mississippi, formed while the top of the caprock was exposed at the sea floor. These gypsum veins commonly contain sulfur and organic matter that apparently replaced gypsum (Drumheller et al., 1982).

CONCLUSIONS

The formation of sulfur at Tatum Dome occurred late in the formational history of the caprock. Although no commercial concentrations of sulfur have been discovered at Tatum Dome, the timing of the oxidation event at Tatum may be similar to that at other domes where large concentrations of sulfur were deposited. The typical stratigraphy of zoned caprock implies a late incursion of low-temperature and probably low-salinity solutions. The gypsum or transition zone that typically separates the limestone zone from the underlying anhydrite probably formed late in the caprock history. Recent studies (cf. Kyle and Price, 1986; Posey et al., 1987; and Saunders, in press a) have indicated that banded limestone caprock forms by the dissolution of the underlying anhydrite by the episodic intrusion of hot brines. During this process, no gypsum was deposited. The formation of the gypsum "transition" zone in most domes probably occurred later, as lower temperature solutions flowed through the typically cavernous and permeable limestone caprock and encountered the underlying anhydrite. Thus, the gypsification process that resulted in the oxidation of sulfide to sulfur at Tatum Dome on a local scale may be equivalent to the event that formed the gypsum transition zone in other domes, perhaps simultaneously causing the oxidation that produced commercial sulfur deposits.

ACKNOWLEDGMENTS

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LATE HOLOCENE BARRIER AND MARSHLAND EVOLUTION, SOUTHWEST MISSISSIPPI

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INTRODUCTION

The south Hancock County barrier-salt marsh complex represents the largest Holocene area of mainland coastal accretion in the state (Figures 1, 2). Other such areas include the barrier spit-marsh complex in Jackson County, attached to the Late Pleistocene Belle Fontaine Beach barrier headland, the Pascagoula and the (now relict) Escatawpa deltas. This is a brief, but thus far the most detailed, stratigraphic discussion of the south Hancock Quaternary.

METHODS

Samples from about 30 rotary drillholes were utilized. In the northern, Pleistocene area drilling was on land; in the southern marshlands barges and specialized marsh equipment were used.

Microfossil and sediment analysis results were used in the construction of cross sections (Otvos, 1978, 1985, p. 26-27). Correlation of Recent foraminifer assemblages with salinity ranges measured over the sample locations resulted in four empirically tabulated biotope groups (Table 1) that define brackish and marine stratigraphic intervals. Much less successful was the attempt to utilize the 13 most common ostracode species, associated with well-defined foram faunas in a number of south Hancock, barrier island and Mississippi Sound coreholes, in order to further refine the facies designations. In the absence of percentage values of faunal composition, thickness values of the sample intervals that contained a given species were used in describing frequency of occurrence. Several factors (e.g., edaphic conditions) other than salinity were found to be equally important in ostracode distribution. Only two broad ostracode bio-

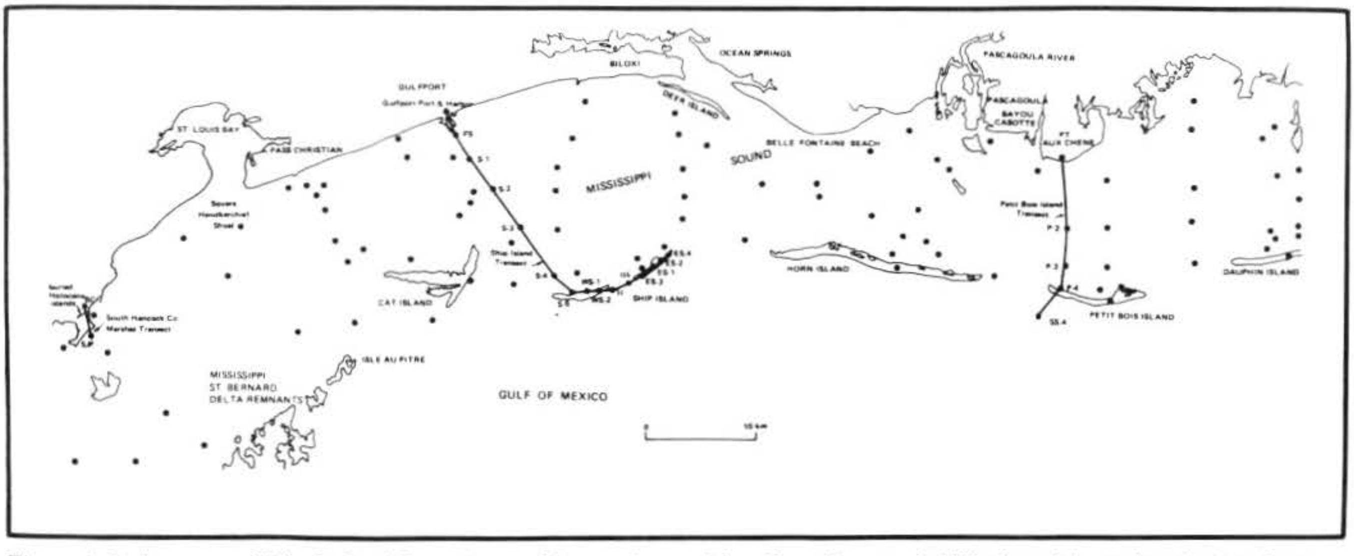

Figure 1. Index map of Mississippi Sound area, Hancock marshland location, and drillholes with analyzed microfauna.

TABLE 1. SALINITY RANGE CATEGORIES, BASED ON FORAMINIFER ASSEMBLAGES

(1) Oligohaline – lower mesohaline (c. 2–16 ppt)

Dominant

Ammotium salsum (0–100%)
Ammonia beccarii parkinsoniana (0–100%)

Secondary and minor (Each species usually less than 10–50%).
(Several are common in salt marshes).

Ammobaculites exiguus
A. exilis
Ammonia beccarii tepida
Trochammina sp.
Miliammina fusca
Jadammina polystoma
Arenoparella mexicana
Haplophragmoides subinvolutum
H. canariense
Ammoastuta inepta
thecamoebians

Very few (1–3) species dominate each sample.

(2) Mesohaline – lower polyhaline (c. 10–26 ppt)

Dominant

Ammonia beccarii parkinsoniana (10–60%)
Ammonia beccarii tepida (10–80%)
Elphidium galvestonense (10–50%)

Secondary

Ammotium salsum (0–20%)
Nonion depressulum matagordanum (0–15%)

Minor (usually < 5% of total)

Cribroelphidium poeyanum
Palmerinella gardenislandensis
Elphidium latispatium pontium
E. incertum mexicanum

Altogether 15 or less species

(3) Polyhaline – lower euhaline (c. 20–30 ppt)

Dominant (40–60%)

Ammonia beccarii tepida (10–35%)
Nonion depressulum matagordanum (5–30%)
Elphidium galvestonense (10–30%)

Secondary (30–40%)

(a) Lower salinity subgroup
Cribroelphidium poeyanum (0–10%)
Ammonia beccarii parkinsoniana (0–10%)
Buliminella elegantissima (0–10%) (significant organic content in sediments also favors this species)
Hanzawaia strattoni (0–5%)

(b) Higher salinity subgroup
Hanzawaia strattoni (0–10%)
Nonionella opima (0–15%)
Elphidium incertum mexicanum (0–5%)

Minor

Sum: c. 5–10% of total. Each species less than 1%.
Fursenkoina sp.
Elphidium latispatium pontium
E. advenum
E. sp.
Brizalina lowmani
Quinqueloculina sp.
Triloculina sp.
Guttulina sp.
Cibicides sp.
Nonionella atlantica
Globigerinoides sp.
Globigerina sp.

Great species diversity

(4) Euhaline (c. 25–32 ppt)

Dominant

Hanzawaia strattoni (15.0–50.0%)

Secondary

Elphidium galvestonense (5–20%)
Ammonia beccarii tepida (10–15%)
Nonion depressulum matagordanum (10–15%)

Minor

Each species c. 5–10%
Quinqueloculina lamarckiana
Q. seminulum
Buliminella sp.
Rosalina columbiensis
Nonionella opima
Elphidium incertum mexicanum
Cribroelphidium poeyanum
miliolids

Each species c. 0–5%
Bigenerina irregularis
Textularia mayori
T. agglutinans
T. candeiana
Cibicides floridanus
Cassidulina subglobosa
C. crassa
Reussella atlantica
Elphidium discoidale
Buccella hannai
Trifarina bella
Sagrina pulchella primitiva
Globigerina sp.
Globigerinoides sp.

Highest species diversity

Table 2. Ostracode species abundance in foraminifer-based salinity zones, using thickness values of vertical drill sample intervals.

	Cumulative vertical sample interval footage of each ostracode species (number of analyzed samples in parentheses)	Percentage of cumulative sample interval footage per species in foram-based Salinity Range Categories 1 (c. 2-16°/oo)	2 (c. 10-26°/oo)	3 (c. 20-30°/oo)	4 (c. 25-32°/oo)
(a) Relatively low salinity (mostly "upper"-"mid bay") affinities					
Paracytheroma stephensoni	25.5' (55)	29.0	11.0	44.3	15.7
Megacythere repexa	37.2' (65)	11.5	19.1	57.4	12.0
Perissocytheridea brachyforma	29.3' (55)	8.2	8.9	70.6	12.3
Cytherura sandbergi	35.2' (66)	5.4	17.0	53.4	24.2
(b) Relatively high salinity (mostly ("lower bay"-"inlet") affinities					
Peratocytheridea bradyi	17.2' (31)	2.3	16.3	53.5	25.6
Hulingsina sulcata (*Pontocythere s.*)	32.5' (57)	4.3	12.0	55.7	28.0
Cushmanidea seminuda	34.7' (64)	4.0	8.6	63.2	24.2
Loxoconcha moralesi	37.2' (65)	3.8	8.6	66.6	21.3
Puriana krutaki	31.3' (58)	4.4	6.3	63.4	25.9
Cytherura radialirata	42.8' (72)	1.4	11.2	76.9	10.5
C. valentinei	21.2' (40)	-	8.5	62.3	29.2
Pellucistoma magniventra	13.7' (25)	-	17.5	49.6	32.9
Puriana rugipunctata	9.5' (13)	-	-	62.1	37.9

facies categories were found in the drill samples: one of generally higher salinities and another, much smaller group, associated to a greater extent than the first with highly-to-moderately brackish biosomes (Table 2).

W.D. Bock identified the foraminifer faunas, P.R. Krutak and T.R. McFadden the ostracodes. Radiocarbon age dating was performed at the University of Georgia Center for Applied Isotope Studies, Athens, Georgia. Wade Howat supervised drilling and sediment analysis. He also drafted the maps and cross sections.

QUATERNARY HISTORY

Pleistocene

Muddy-sandy, fossiliferous beds of the Biloxi Formation, deposited during the Sangamonian Interglacial in a transgressive-regressive eustatic cycle are detected at shallow depths along most of the northern Gulf Coast (Otvos, 1985). In the Hancock area, the Biloxi is encountered at decreasing depths as far as 20 km north of the Holocene shoreline. In the Magnolia Ridge hole (Figure 4) brackish and open marine units occur between 16-27 m below sea level. The Biloxi's thin, landward "feather edge" that interfingers with alluvial Prairie Formation strata contains highly brackish units. The fossil-free, silty-sandy Prairie overlies the Biloxi and forms the Pleistocene land surface north of the south Hancock Holocene area. Extensive late Pleistocene-to-late-Holocene fluvial-subaerial erosion shaped the Holocene landscape during the low sea level stage. Despite tentative claims (Lins and Rogers, 1986), these erosion features do not suggest a high ("Silver Bluff") mid-Holocene sea level here.

Holocene

The Holocene transgression reached the present Hancock mainland area before 5,000 years B.P. (before the present). The new intertidal-nearshore zone received littoral drift from the recently formed Alabama-Mississippi barrier island chain, via Square Handkerchief Shoal (Otvos, 1978, 1981). Sand was transmit-

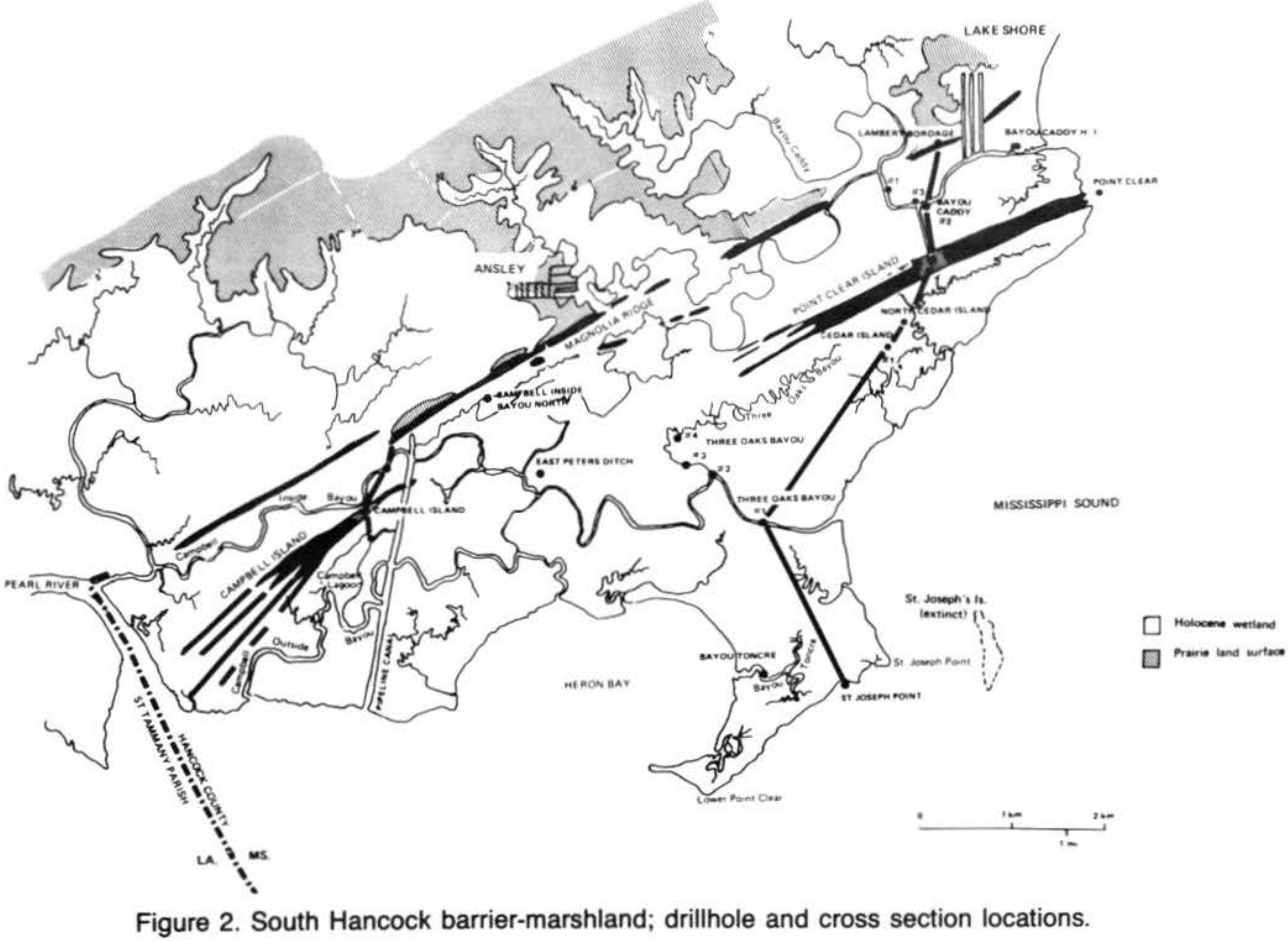

Figure 2. South Hancock barrier-marshland; drillhole and cross section locations.

ted southwestward, toward a line of shoals and islands, then located at the mouth of the Pontchartrain Embayment. This line of mostly buried ridges occurs west of Campbell Island beneath the Pearl River delta and continues south of Lake St. Catherine. It extends well into Orleans Parish, Louisiana (Otvos, 1978).

Brackish waters inundated the seaward sector of the Pearl River valley and incised creek valleys, creating indented, estuarine embayments, flanked by marshes. A straight barrier shore (Magnolia Ridge) developed on the seaward rim of the Prairie surface between the embayments. Barrier progradation also took place from these Prairie headland sectors into Holocene deposits, as shown by sediments in an auger hole (Figures 2, 4). Rectangular creek patterns and several short, largely drowned, relict sand ridge segments in the marsh south of Ansley suggest a localized, 300-m seaward progradation of Magnolia Ridge. Three of these ridge segments form a trend that closes an acute angle with Magnolia Ridge, with the strike toward eastern Point Clear (Figure 2). This raises the alternate possibility that prograding beach ridges of that island might have approached the contemporary mainland shore and were not part of the prograding mainland strandplain ridge sequence.

Muddy sands and sandy, silty muds were deposited offshore in environments that ranged from oligohaline (highly brackish) to euhaline (fully marine). The earliest most brackish transgressive biotopes often became eroded prior to the deposition of later units (Figures 4, 5). Interestingly, Rogers (1984) reported relatively high salinity foraminifer species *(Nonion affinis, Quinqueloculina compta*, and others) at undetermined depth from the auger hole, while seaward the Campbell Inside Bayou drillhole #2 contained only highly brackish forms in the Holocene interval (Figure 4). An oyster date from the Bayou Caddy #2 drillhole suggests that nearshore brackish sediment deposition started c. 3600 years B.P. An oyster reef under northeastern Campbell Island, excavated by the pipeline canal, provided dates in the 3165-2920 years B.P. range (Otvos, 1978). Other oyster dates near the base of the Holocene suggest that Point Clear and Campbell Islands postdate 2650-2325 years B.P.

The two "marsh islands" prior to our intensive drilling program were termed cheniers by Kolb and Van Lopik (1966). Because of their unusually short distance from Magnolia Ridge, the islands were regarded as the distal members of a partially marsh-covered beach ridge plain, contiguous with that ridge (Otvos, 1973). This view has recently been revived by a cursory study that was not based on subsurface information from the intervening narrow marsh zone (Rogers, 1984; Lins and Rogers, 1986).

The barrier sands in the Point Clear and Campbell Island drillholes rest on Holocene sandy muds and muddy sands, indicating an aggradational origin. Sand sorting generally improves upward, from moderately

Figure 3. Western part of Campbell Island with bifurcating ridges. Magnolia Ridge: in background, on right. Foreground: Campbell Lagoon. View to northwest.

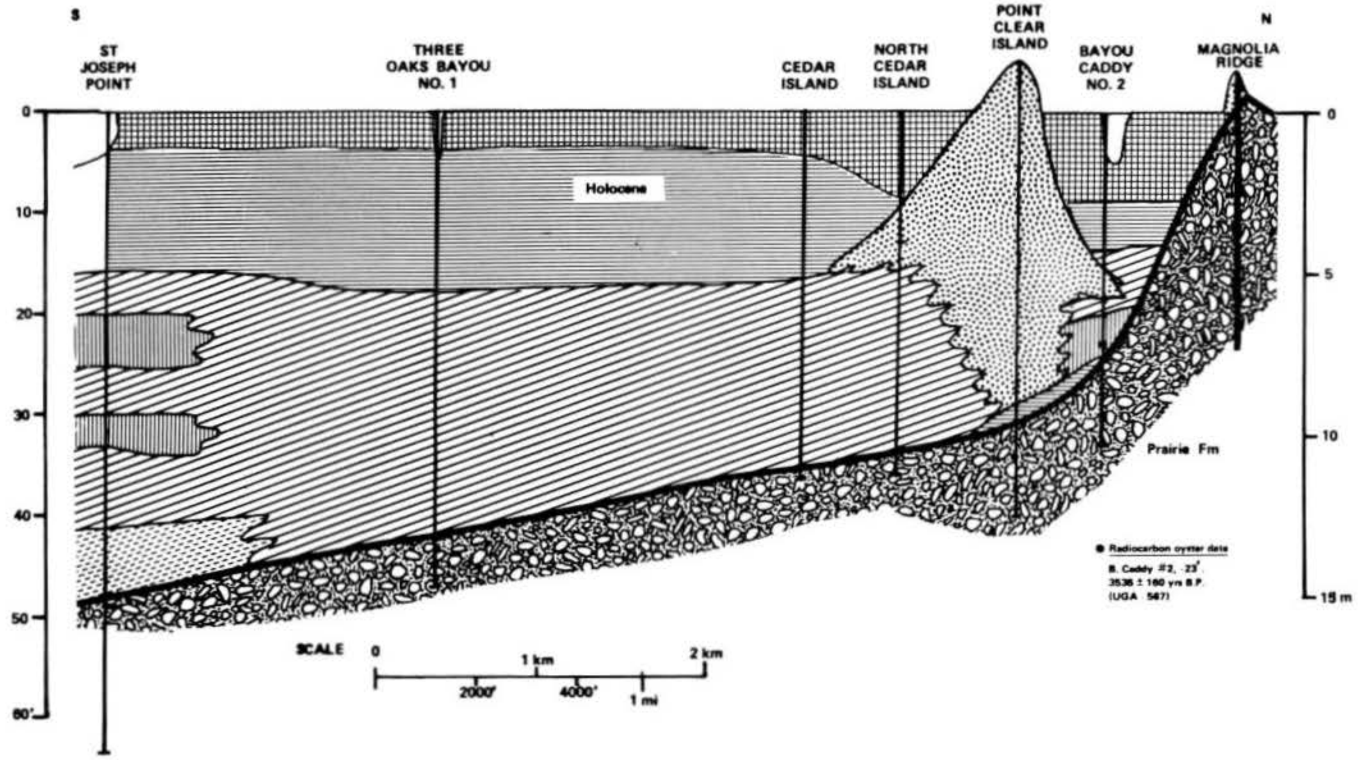

Figure 4. Point Clear Island geologic cross section; see Figure 5 for legend.

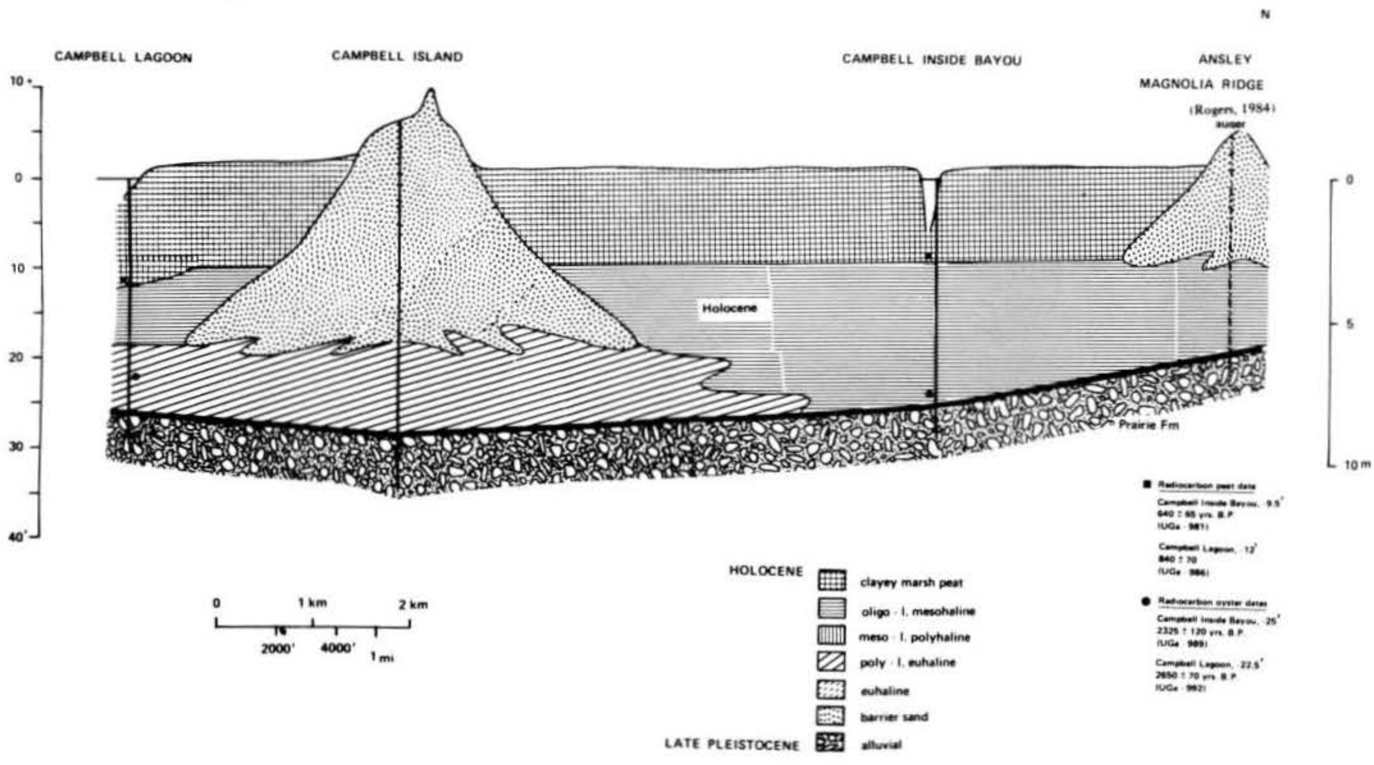

Figure 5. Campbell Island geologic cross section.

and moderately well-sorted, to well- and very well-sorted. Was this aggradation related to the seaward progradation of a continuous mainland strandplain or to progradation of isolated barrier islands? Drillhole data, including subsea depth values of the barrier bases, suggest that upward shoal-to-island growth started prior to mainland beach ridge formation and was completely separate from it. There are no indications yet of buried sand ridges under the marsh between the seaward margin of Magnolia Ridge and the landward margins of the two barriers. Five coreholes in this zone penetrated 5-9 m of a Holocene brackish muddy fine sand, fine sandy mud, clayey fine sand, and marsh peat sequence (Figures 2, 4, 5). The encountered thin, well-sorted sand lenses probably belonged to shoreface facies of the mainland and the two island barriers. No geological process, such as prolonged, high-velocity tidal scouring, may be reasonably invoked as a cause for an assumed wholesale erosion of the hypothetical beach ridges here *after* Point Clear and Campbell Islands came into existence.

After initiation of the Mississippi-St. Bernard subdeltas south-southeast of the Hancock area (c. 2900 years ago), littoral drift and barrier progradation stopped, drastically reducing wave energies, fostering mud deposition, marshland growth and progradation of the Pearl River delta. Peat dates from our drillholes (840-640 years B.P.) indicate the approximate time for the start of intensive marsh buildup in these areas. Bonn and Froelicher (1986) studied these peaty deposits in recent years. Cedar Island is a large Indian shell midden in the marsh south of Point Clear Island, with 4-5 m of *Rangia cuneata* accumulation. A *Rangia* date(2900 $\pm$ 70 years B.P.: UGA-354) from the mound turned out to be highly anomalous, possibly one or two millennia too old. Fanshaped lineaments that trail Campbell and Point Clear Islands on their southwestern ends reflect the presence of shallow buried and semi-submerged beach ridge sets beneath the marsh. Regional tectonic subsidence influencing this Mississippi delta-flank region and compaction of late Holocene sediments resulted in partial to total burial of the ridge sets. Similar subsidence effects are also noted in the partly flooded strandplain swales of nearby Cat Island.

Since sediment supply to the St. Bernard subdeltas stopped c. 2200 years B.P., the erosion of marsh shores has been intensive and the eastern tip of Point Clear Island also disappeared. Extinct St. Joseph's Island (Figure 2), shown on charts for decades after 1848, indicates the minimum extent of the original contiguous marshland in the recent past. According to U.S. Corps of Engineers tidal gage records (Mobile District chart data), sea level rise even at Biloxi, a location not affected by the intensive delta-flank subsidence experienced in the Hancock area, amounted to at least 18 cm since 1895. Subsidence and concomitant marsh erosion is expected to accelerate in the future, due to man-influenced global climatic factors.

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