Geochemical characterization of source rocks, crude oils and gases of Northwest Cuba

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Abstract

In order to characterize the Cuban petroleum system, we analyzed source rocks as well as crude oils and gases from carbonate producing reservoirs from northwest onshore Cuban fields. The geochemical oil and gas characterizations were based on bulk analyses, chromatography (liquid and gas), mass spectrometry and mass spectrometry-coupled to stable carbon isotope ratio determination. Analyses of sampled source rocks intervals were performed with Rock Eval 6 and provided valuable data for the appraisal of the active petroleum system in Northwest Cuba. Our paper focused on the genetic semblance between carbonate reservoired crude oils and source rocks to assess the offshore extension of the onshore Cuban carbonate petroleum system. Crude oils-source rocks and crude oils-offshore DSDP source rocks were correlated to a common type of carbonate depositional environment. An understanding of the geochemical compositions of carbonate crude oils, grouped in families including heavy sulphur rich oils, light or mixed and biodegraded fluids helped to underline the different processes operating in the reservoirs today. The high degree of anoxicity during the organic matter deposition in the Upper Jurassic and Lower Cretaceous was confirmed. We observed low hydrocarbon cracking levels and non biodegraded fluids of Type II/IIS organic matter in Upper Jurassic and Lower Cretaceous reservoirs, while shallower Tertiary reservoirs, although more mature were altered by microbial biodegradation. There, reservoirs with temperatures below 80 °C provided a good habitat for aerobic/anaerobic bacteria and the possibility of hydrocarbon biodegradation.
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Keywords: Cuba; Organic geochemistry; Carbonate reservoired crude oils; Gases; Source rocks

1. Introduction

Renewed interest in the offshore exploration of north Cuba initiated collaborative work between CUPET, IFP, oil companies and geophysical service companies, to provide new insights on the petroleum system and help define prospective exploration targets. This involved interpreting new sedimentological depositional models, looking at new 2D seismic data and understanding the composition of fluids in the reservoirs which are dominated by carbonate lithologies. Gaumet and Letouzey (2002) have published the sedimentological and seismic aspects of the discussed Northern Cuban petroliferous zone. This paper addresses the petroleum potential of carbonate source rocks, and summarizes the composition of sampled reservoired crude oils and associated gases.

In Cuba, the geology is complex and involves oil traps in highly thrusted zones, likely explaining some confusing geochemical information on the existing composition of hydrocarbons that may be found. Part of the study has been focused on a study of outcrops and selected rock samples whose information served as input to geochemical modelling that simulate the generation and expulsion of the source rocks in 1D and 2D by Moretti et al. (2004). The rest of the geochemical characterization study concentrated on the origin and maturity levels of gases and oils, and their relationships to a Lower Cretaceous offshore DSDP well impregnated limestone sampled years ago (Herbin, Deroo, & Roucaché, 1984; Rüllkotter, Mukhopadhyay, Hartung, & Welte, 1984). The present newly collected source rocks, gases and oil samples were chosen because they belonged to producing intervals mostly in Upper Jurassic and Lower Cretaceous sedimentary sequences. The geochemistry of liquid and gas hydrocarbons is presented in terms of source

of the organic matter, thermal evolution, and degree of secondary alterations. Gas analyses, not reported so far in literature from Cuba, will be shown to provide valuable geochemical information for the understanding of the compositions and origin of C_1-C_4 hydrocarbons. Although most oils and associated gases were collected in U. Jurassic and L. Cretaceous intervals, a handful in shallower Tertiary clastics reservoirs were included to differentiate the depositional environments of the organic matter. Finally, this paper discusses the relatively low thermal maturities of hydrocarbons in carbonate reservoirs, the possible occurrence of biodegradation and hydrocarbon mixing.

2. Geological setting

As mentioned in papers by Denny, Austin, and Buffler (1994) and Sanchez and Tenreyro (1996), the island of Cuba is the result of the collision between Cretaceous Volcanic Arcs and the extensional continental margin of Northern America. The Gulf of Mexico region experienced rifting episodes of late Triassic and Middle Jurassic ages, represented by the half graben morphology in the North Cuban provinces. An important geological event is the deposition of a salt layer across the basin. This salt is thick and continuous in the northwest Gulf of Mexico but remains limited in the eastern part (sequence 'B'). The rifting led to the creation of a Late Jurassic oceanic crust during 10 My. Southward, the proto-Caribbean rifted more or less at the same time but structures in horsts and grabens are dominant. The early post rift sedimentation during the thermal subsidence consisted of a shallow marine platform developed along the northern proto-Caribbean margins whose build-up took place from the Oxfordian to the lower Tithonian. The end of the normal fault activity remains questionable and may not be synchronous all over the area but by the Berriasian, a flooding event sealed most of the tilted blocks (Sequence 'F'). Rapid thermal subsidence then led to the deposition of stacked shallow water platforms (Sequences 'G1, G2, G3 and H'). The total thickness of this second carbonate build-up phase may reach 1.5 km. The central basin deepened from the Aptian after a phase of relative starvation. The Middle Cretaceous Unconformity (MCU) started during the Cenomanian and continued up to the Maastrichian or later. The greater Antilles Arc collided with the Bahamas margin in the late Paleocene (Gordon, Mann, Caceres, & Flores, 1997) and since then clastic flysch deposited in the flexural basin (sequences 'J' and 'K') whereas platforms persisted in Yucatan and south Florida. The last thrusts are Eocene in age, with the filling of the basin continuing up to the Quaternary due to sediment influx resulting from mountain belt erosion. A new and updated stratigraphic column and cross section is provided for the northern Cuba sedimentary record (Fig. 1) that gives the correlated sedimentary units detailed by Gaumet and Letouzey (2002) with estimated depths for each units.

3. Geochemical background

Published studies on the geochemistry of source rocks and crude oils of Cuba remain scarce. To expand the database, new samples (outcrops, source rocks, oils and gases) were collected in the Northwest area within the context of the carbonate depositional models of the onshore and its extension to the offshore area (Gaumet and Letouzey, 2002; Moretti et al., 2004). New samples were obtained from the Jurassic section in Western Cuba and from the syn-rift stage, with the distinction of syn-rift (clastic deposits), post rift (regional carbonate platform) and the Cretaceous Bahamas channel deposits.

With respect to oil compositions, prior studies suggested that different petroleum provinces existed with complex migration routes in Cuba (Maksimov et al., 1986). Based on wide differences in fluid properties, the crude oils span from very light, mature low sulphur crudes, to high sulphur, NSO rich and biodegraded heavy oils, with sporadic occurrences of remigrated hydrocarbons, in particular in the center of Cuba. It appeared that more than one type of source rock could be invoked in the petroleum system of Cuba: A silico clastic with sometimes traces of terrigenous input in west and central Cuba and anoxic marine source rocks in the northwest carbonate sediments.

Because this study concentrated on the geochemistry of hydrocarbons in the northern carbonate reservoirs, particular attention was paid to the U. Jurassic and L. Cretaceous units, in order to detect and understand the heterogeneity in petroleum composition often encountered. For instance, within a few tens of kilometers along the northern coast, equivalent age-producing levels showed either good quality oils ($> 20°$ API), or highly viscous oils ($< 15°$ API) enriched in sulphur, metals and NSO compounds. We attempted to better understand these bulk properties by looking at the molecular information provided by biomarkers and carbon isotopes, in the aim of gaining more adequate thermal cracking evaluations, and molecular insights of depositional environments.

4. Database and methods

The database is composed of literature information, CUPET proprietary and new field acquisition of outcrops in western Cuba, with two wells studied in details. Oils and gases were collected in three areas, in the west of La Habana City at Martin Mesa (M. Mesa), then northwest in nearby fields at Via Blanca (V. Blanca), Boca Jaruco (B. Jaruco) and Yumuri. The last considered area is few tens of kilometers east, at Varadero, Marbella-Mar (M. Mar) and Cantel. The location of the new samples is given in Fig. 2 with a zoom of the northern coast line. Table 1 provides geological and general information on samples collected.
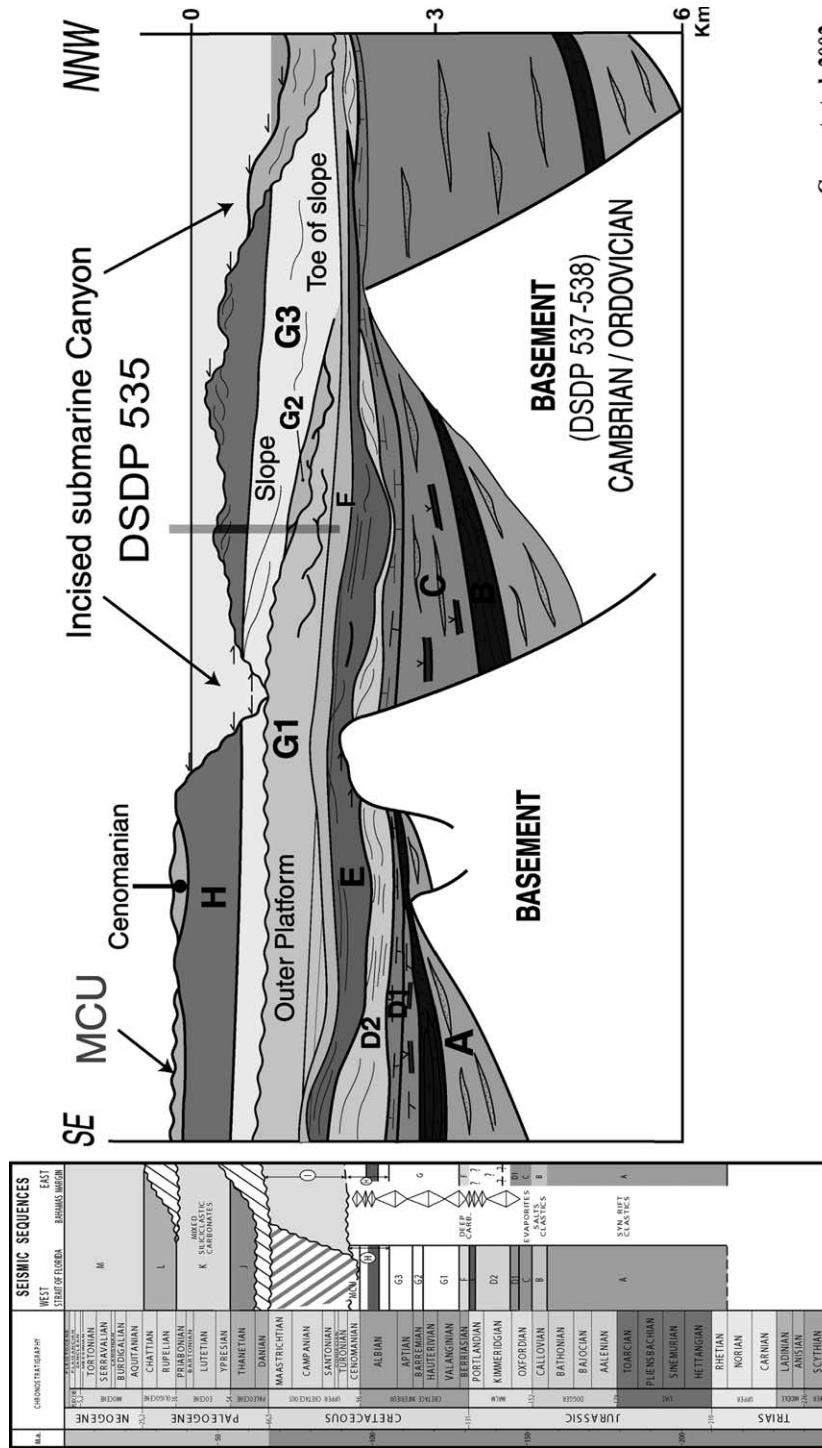
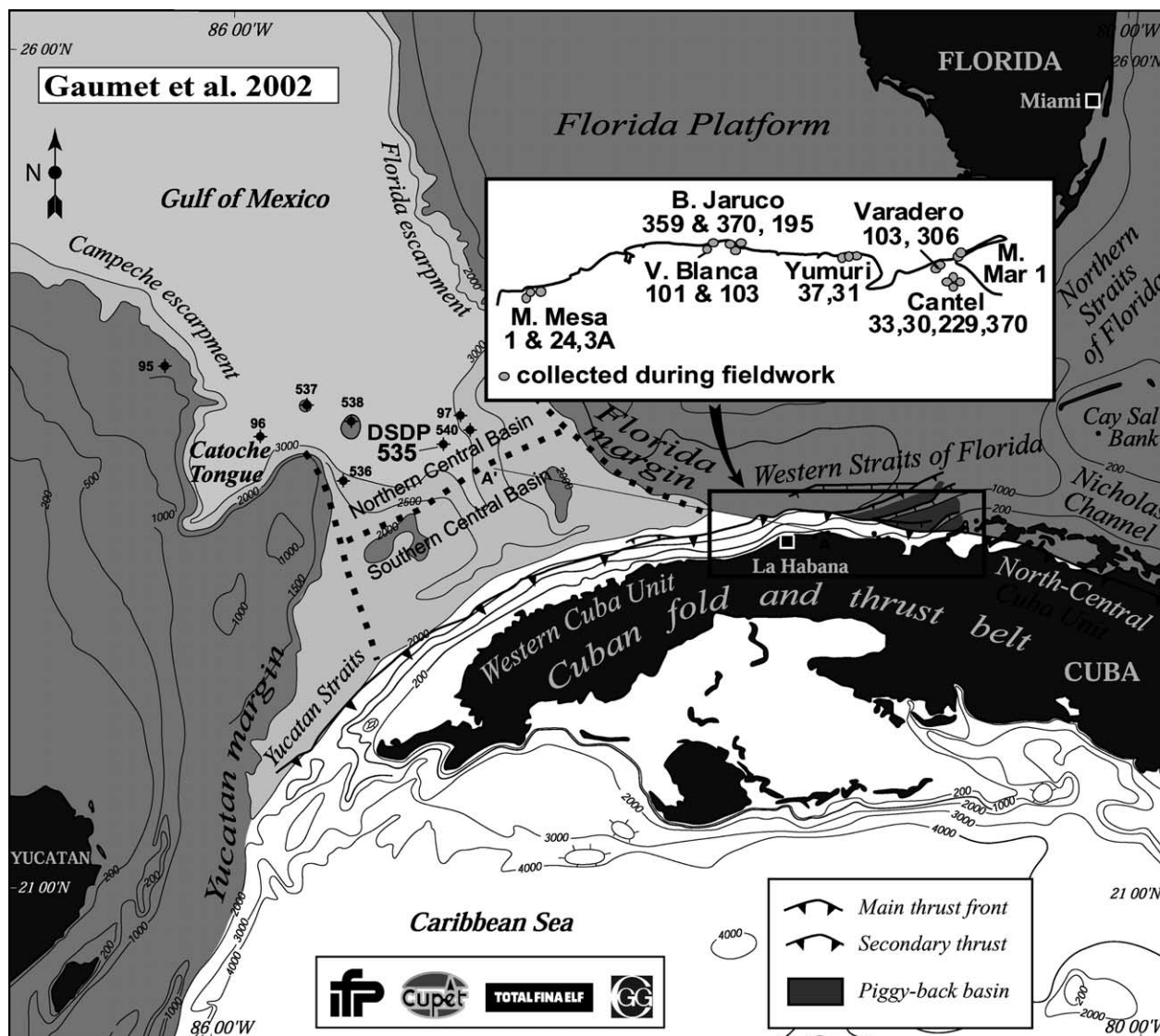Fig. 1. Schematic stratigraphic column of the northwest Cuban area.

Fig. 2. Physiographic features of northwestern Cuba. Zoom of the area where oil and gas samples were collected.

4.1. Source rocks

Over a hundred rocks were selected in the northern zone from the syn-rift, post-rift sequences, and the Upper Cretaceous sequences (Moretti et al., 2004). The rock samples were crushed and pyrolised by the Rock Eval 6 apparatus and found adequate for carbonate samples (Lafargue et al., 1998). The advantage lies in the quantification of both mineral (MINC) and organic carbon (TOC) which can be done simultaneously by the pyroliser. Two U. Jurassic source rocks were used to attempt oil-source rock correlation, one from the well Yumuri-35 (Cifuentes Fm. at an interval between 2118–2121 m), and Varadero-23 (Cifuentes Fm. 1478–1480 m). For estimating the continuity of the source rocks offshore, DSDP wells and subsequent data from literature were used, in particular

the DSDP 535 well, drilled during Leg 77 at a water depth of 3455 m in the Straits of Florida.

4.2. Crude oils, gases

Fifteen crude oils and thirteen gases were collected at wellheads in producing fields (Table 1). The gas samples are solution gases from the producing oil column. Four oils and gases were collected in Tertiary silico-clastic reservoirs in M. Mesa and Cantel fields. The main sampling for crude oils and associated gases was made in limestone reservoirs at V. Blanca, B. Jaruco, Yumuri, M. Mar and Varadero, Cantel (Fig. 2). Gas chromatography was used to quantify the C_1–C_4 content in molar percent, and the specific compound isotopic carbon ratios ($^{13}C/^{12}C$) were evaluated in triplicates with the Micromass

Table 1
Crude oils and gases sampled in Northwest Cuba

Crude oils well	Depth top	Depth bottom	Geological formation	AGE	°API	%S	Ni	V	%ASPH	%RES	%ARO	%SAT
MARTIN MESA 1	824	807	Manacas	Lower Eocene	23.2	1.38	24	12	6	30.6	25.3	34.1
MARTIN MESA 24	733	773	Manacas	Lower Eocene	18.6	0.69	13	5	1.4	21.5	31.9	40.8
CANTEL 30	477	683	Serpentinite	Paleocene	12.0	1.17	na	na	3.7	38.9	27.3	27.4
CANTEL 229	424	488	Serpentinite	Paleocene	14.1	na	49	41	3.6	38.8	28.1	27.0
CANTEL 33	1140	1172	Carmita	U. Cretaceous	na	na	na	na	5	30.1	27.1	34.2
VIA BLANCA 101	1380	1410	Carmita	L. Cretaceous	33.7	2.9	9	14	2.3	28.1	23.7	38.9
BOCA JARUCO 370	1306	1356	Carmita	L. Cretaceous	na	3.53	23	23	1.2	31.8	30.1	31.2
MARBELLA MAR 1	2550	2565	Paraiso	L. Cretaceous	11.0	5.69	45	76	19	38.6	20.6	15.6
YUMURI 31	1487	1467	Ronda	L. Cretaceous	10.4	4.6	31	58	24.8	35.9	21.4	10.1
YUMURI 37	1280	1350	Cifuentes	U. Jurassic	6.3	5.96	70	111	10.9	43.4	25.0	13.3
YUMURI X	2321	3600	Cifuentes	U. Jurassic	na	7.64	54	102	16	45.0	20.2	13.2
VIA BLANCA 103	1989	2054	Cifuentes	U. Jurassic	21.1	6.2	30	50	11.3	34.3	22.1	26.4
BOCA JARUCO 359	1826	1857	Cifuentes	U. Jurassic	na	6.76	28	47	9	42.7	23.4	19.3
VARADERO 103	1690	1720	Cifuentes	U. Jurassic	9.6	8.9	68	109	19.6	46.8	18.0	11.0
VARADERO 306	1613	1645	Cifuentes	U. Jurassic	10.4	8.9	66	105	20.2	46.4	17.7	10.7
MARTIN MESA 24	733	773	Manacas	Lower Eocene								
MARTIN MESA 3A	1340	1350		Lower Eocene								
CANTEL 229	424	488	Serpentinite	Paleocene								
CANTEL 370	882	1062	Serpentinite	Paleocene								
BOCA JARUCO 370	1306	1356	Carmita	L. Cretaceous								
VIA BLANCA 101	1380	1410	Carmita	L. Cretaceous								
MARBELLA MAR-1	2550	2565	Paraiso	L. Cretaceous								
YUMURI X	2321	3600	Cifuentes	U. Jurassic								
VIA BLANCA 103	1989	2054	Cifuentes	U. Jurassic								
BOCA JARUCO 195	1500	1549	Cifuentes	U. Jurassic								
BOCA JARUCO 359	1826	1857	Cifuentes	U. Jurassic								
VARADERO 103	1690	1720	Cifuentes	U. Jurassic								
VARADERO 306	1613	1645	Cifuentes	U. Jurassic								

% ASPH: asphaltenes; % RES: resins; % ARO: aromatics; % SAT: saturates.

VG-Isochrom Optima apparatus. The units for carbon isotopic ratios are per mil (°/°°) and given in comparison to the PDB international standard. The analytical tools for the characterization of crude oils included ICP measurements for metal content, calorimetry for sulphur, HPLC/MPLC for hydrocarbon separations of chemical families (SARA), and High Resolution Gas Chromatography (GC) for oil fingerprints (C_5-C_{35}). Mass Spectrometry in line with a Gas Chromatograph (GC-MS, GCMS-MRM) was used for molecular assessment (biomarker analysis).

5. Source rock evaluation

Rock Eval 6 was used to characterize the source rocks and outcrops from field work. More details on the samples are largely documented and published by Moretti et al. (2004). The plot of Rock Eval S2 as a function of Total Organic Carbon (%) resulting from a large data base of rock samples is illustrated on Fig. 3.

The Syn-rift clastic deposits (GROUP 'A/B/C') are largely represented by the San Cayetano Fm. and Francisco Fm. The net thicknesses for each units are provided but one should keep in mind that the depths vary across the studied area. The facies occur in small basins and half grabens as found offshore. Thin intercalations of shale and sandstone are found within a main silico-clastic sequence. The average values of the Total Organic Content (TOC) averages 1%

(Max. 3.2%), and the rocks are overmature with Tmax averaging 495 °C (Max. 610).

Post-rift group ('D to G') found in most basinal facies in onshore Cuba show numerous good source rock levels from the U. Jurassic to the L. Cretaceous, characterized as marine source rocks associated with carbonated facies. The source rock levels are often overmature. Best potentials have been found in the Cifuentes Fm. (Tithonian, GROUP 'E') where the values are rather homogeneously distributed. Tmax ranges between 410–420 °C with average TOC at 1.0% and HI values above 400 mg HC/g rock, with very rich levels having up to 600 mg HC/g rock.

GROUP 'F' which has been extensively studied as an organic rich section includes the Neocomian Trumba-dero, Sumidero and Ronda Formations (Berriasian to Valanginian). The Neocomian section in western Cuba is rich as in central Cuba: the best S2 values are around 26 mg HC/g rock, HI around 600 and TOC sometimes >6% for immature samples. Averages values are however not always representative of the initial potential of the formations.

The Lower Cretaceous sections represented by the Pons and Carmita Fm. (Aptian/Albian) for GROUP 'G' were not very well studied previously. The new results show the existence onshore Cuba of a Lower Cretaceous additional source rock level which is synchronous with the source rocks of the DSDP wells. In the outcrops, best values for S2 are 9 mg HC/g, Hydrogen Index (HI) is 280 and more;
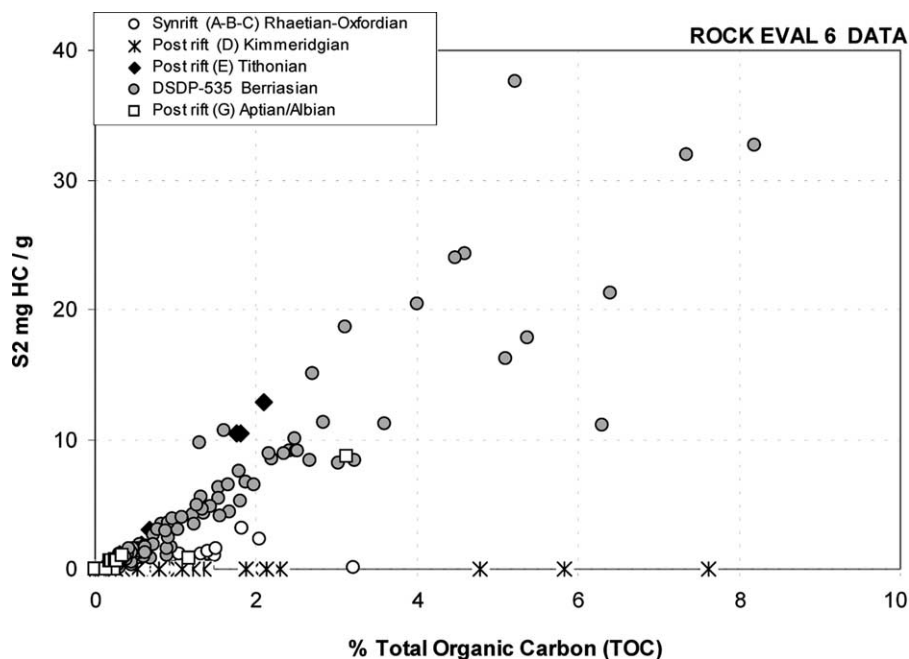
Fig. 3. Rock Eval 6 data of onshore outcrops and source rocks, with offshore DSDP-535.

mature sample with high TOC (3.1%) whose production index is 0.8, could have had an initial S2 over 30 mg HC/g.

Finally, the Upper Cretaceous section represented in GROUP 'H' for Cenomanian/Turonian sediments, also have good organic richness levels. That sequence is however partially eroded or missing due to the lack of sedimentation after the MCU. The source rocks are characterized as carbonate, affiliated with marls and mudstones deposited in restricted lagoonal environments. Total Organic Content values reach 3% and HI surpasses 610 for immature samples, and Tmax average 426 °C with a maximal at 500.

5.1. Literature information of the offshore DSDP-535 sample

Five Lower Cretaceous formations were described in the literature as levels of distinct and variable organic rich sediments (Herbin et al., 1984; Rüllkotter et al., 1984). The published data mentions bioturbated organic matter in limestones and marl layers with unusual asphalt filled fractures and adjacent oil stained levels, where the host rocks contain small levels of unstained organic carbon with very different genetic and thermal signatures. Herbin et al. (1984) gave evidences that the Lower Cretaceous sequence in the Neocomian from the offshore DSDP-535 well contained hydrocarbon rich levels with good source rock potential and with average TOC superior to 2.3%, and S2 values between 4 and 20 mg HC/g rock. The Aptian/Albian sequence had 1.3% TOC and S2 values above 7 mg/g. The offshore DSDP-535 rock samples were also interpreted as source rocks with marine Type II organic matter. The samples were characterized as immature.

5.2. Summary of the petroleum systems and exploration implications

The conclusions from Moretti et al. (2004) suggest that two and perhaps three petroleum systems are found in northwest Cuba. The first petroleum system is syn-rift and linked to a deep half graben with Jurassic silicoclastic source rocks with overmature source rocks (San Cayetano Fm.). A second petroleum system is related to post-rift deep water carbonate deposits and the resulting migrated hydrocarbons are currently found in the thrusted belts of the Northern Cuban island. It is divided into two facies, the basinal starved carbonate sediments and the shallow water carbonate platforms and ramps. The deep water facies are rich in organic matter whereas the shallow carbonates are poorer source rocks. Another petroleum system may be considered in the Lower Cretaceous section deposited during the Valanginian to Albian.

6. Geochemical characterization of gases and crude oils

Data for the hydrocarbon gases are given according to the reservoir age, i.e. Tertiary, Cretaceous and Jurassic (Table 2).

6.1. Gases

The data gives normalized $C_1–C_4$ hydrocarbon content and the respective $\delta^{13}C$ isotopic ratio for natural gases and acid gases. Stable carbon isotope values are reported for gases according to reservoir ages and summarized in Fig. 4. First of all, the molar compositions of the gases reveal that they are wet gases, an information that is considered to reflect low thermal

Table 2
Molar and carbon isotopic data for gases

Field	Depth top	Depth bottom	Geological formation	Age	Molar composition (normalized)					Carbon isotopic values					Others		
					%n-C1H4	%n-C2H6	%n-C3H8	%n-iC4H10	%n-nC4H10	dC13-C1	dC13-C2	dC13-C3	dC13-iC4	dC13-nC4	%-CO2	dC13-CO2	%-H2S
MARTIN MESA 24	733	773	Manacas	Lower Eocene	99.00	0.75	0.09	0.12	0.04	−45.62	−24.39	−12.41	−20.57	−17.36	10.29	20.39	
MARTIN MESA 3A	1340	1350	Manacas	Lower Eocene	99.12	0.78	0.08	0.02	0.00	−45.27	−26.21	−23.58	−26.29	−26.75	0.34	-12.21	
CANTEL 229	424	488	Serpentinite	Paleocene	99.29	0.35	0.04	0.01	0.31	−45.53	−26.21	−24.76	−28.62	−26.65	1.22	6.12	
CANTEL 370	882	1062	Serpentinite	Paleocene	97.45	2.15	0.20	0.14	0.05	−44.49	−26.32	−18.98	−25.80	−24.08	1.42	1.83	
BOCA JARUCO 370	1306	1356	Carmita	L. Cretaceous	92.37	4.01	2.26	0.45	0.91	−40.43	−30.08	−27.57	−26.20	−25.83	1.75	14.00	
VIA BLANCA 101	1380	1410	Carmita	L. Cretaceous	92.32	4.10	2.26	0.49	0.83	−40.12	−28.72	−25.54	−25.32	−24.31	0.15	−15.00	
MARBELLA MAR-1	2550	2565	Paraiso	L. Cretaceous	92.65	3.64	2.41	0.38	0.92	−43.65	−32.82	−29.33	−28.50	−27.26	13.60	−6.12	
YUMURI X	2321	3600	Cifuentes	U. Jurassic	88.07	5.94	3.65	0.84	1.50	−46.78	−32.20	−28.38	−26.92	−26.92	13.17	−1.33	1.82
VIA BLANCA 103	1989	2054	Cifuentes	U. Jurassic	91.59	5.06	2.40	0.36	0.60	−45.61	−33.29	−28.91	−28.55	−27.59	6.52	−7.55	
BOCA JARUCO 195	1500	1549	Cifuentes	U. Jurassic	94.24	3.22	1.58	0.29	0.67	−43.32	−30.08	−26.43	−27.00	−25.86	2.71	−6.67	
BOCA JARUCO 359	1826	1857	Cifuentes	U. Jurassic	93.53	3.90	1.72	0.29	0.56	−43.08	−29.48	−26.22	−25.94	−24.73	2.72	−1.06	
VARADERO 103	1690	1720	Cifuentes	U. Jurassic	91.57	4.96	2.48	0.34	0.63	−43.81	−28.85	−26.88	−27.76	−26.48	3.46	0.19	
VARADERO 306	1613	1645	Cifuentes	U. Jurassic	91.55	4.74	2.55	0.42	0.74	−49.06	−31.58	−27.10	−27.54	−25.55	10.83	−7.71	

maturity hydrocarbons. Most gases sampled in Jurassic and Cretaceous reservoirs have isotopically speaking thermogenic signatures with carbon isotopic values for methane between -40 to $-47°/°°$. No biogenic methane is evidenced in those reservoirs. The U. Jurassic Varadero-306 sample is perhaps the only gas with a methane carbon isotope value at -49°/°° and very high CO_2 content (10%) which could be inferred to be linked to early biogenic methane production. Evidence of gas biodegradation is more easily seen by isotope ratios when heavier isotopic values of n-C_3 and nC_4 isomers are found compared to the iC_4 species (Prinzhofer and Huc, 1995). For example, Tertiary reservoired gases (M.Mesa-24, Cantel 229 and Cantel-370 except M. Mesa-3A) may be interpreted as biodegraded gas, although it does not appear that biogenic methane has been produced. Biodegradation using higher molecular weight homologs is illustrated on the graph plotting the molar C_2/iC_4 ratio (x-axis) versus the molar C_2/C_3 ratio (y-axis) as described by Lorant et al. (1998). M. Mesa-24 and Cantel samples clearly plot in the biodegraded zone (Fig. 5a). Fig. 5b is a projection of the data for the assessment of the gas maturation level. This was made, without the biodegraded gas from Cantel and M. Mesa-24. Only the unbiodegraded Tertiary and M. Mesa-3A appears as a dry and mature gas (no C_{3+} specie) with a heavy $^{13}δ$ isotope signature. Most Jurassic and Cretaceous reservoired gas samples plot in the primary cracking zone. The small spread of the maturity values between Jurassic and Cretaceous samples however makes it difficult to assess a difference among those gases.

A relative assessment of the distances of migration from source rock to reservoirs from the hydrocarbon gas data by statistical analysis and principal components was made as described in Prinzhofer et al. (2000). Results show that low migration distances from source rocks to reservoirs are occurring.

The data set also had samples with substantial acid gases with percent composition ranging from 1 to 14% of CO_2, and one sample with more than 1% H_2S (Yumuri). The high positive $δ^{13}C$ values ($>10°/°°$) in the CO_2 isotopic signatures ($+4$ to $-5°/°°$) are not typical of carbonate decomposition. On the other hand, values for carbon isotopic ratios between values of $-15°/°°$ and $-12°/°°$ for the CO_2 reflect the thermal degradation of organic matter. One should note that biodegraded gases sampled in Tertiary reservoirs (except for the mature M. Mesa 3A) have also unusually high positive carbon isotopic ratios for CO_2, and an overall higher CO_2 content, likely resulting from the biodegradation process.

6.2. Crude oils

Crude oils originate mainly from limestone reservoirs, although a handful were from Tertiary silico-clastic intervals (Table 1). The latter were compared with other samples to
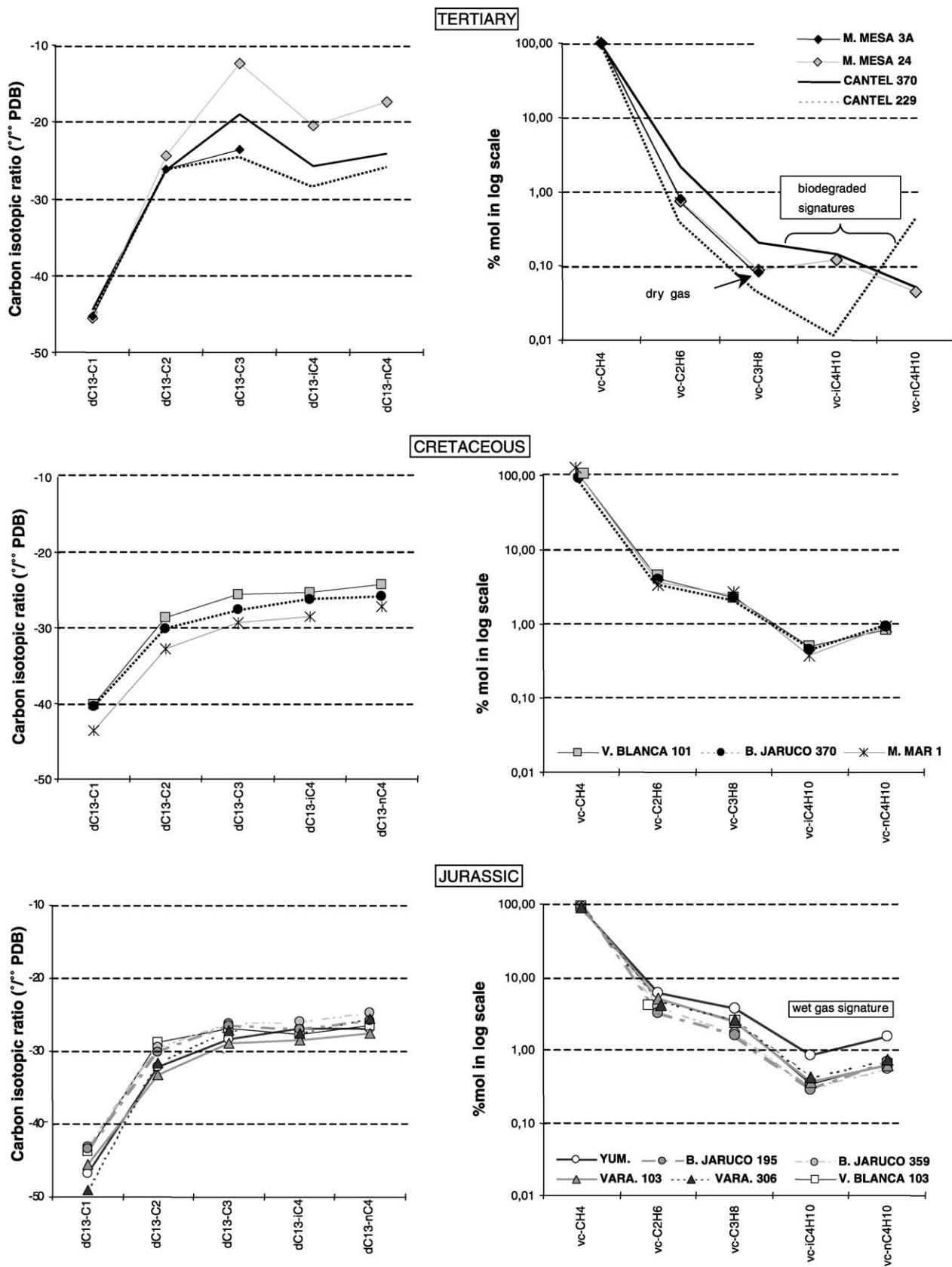
Fig. 4. Carbon isotope ratios (left) and molar compositions (right) of gases in Tertiary, Lower Cretaceous and Upper Jurassic reservoirs.
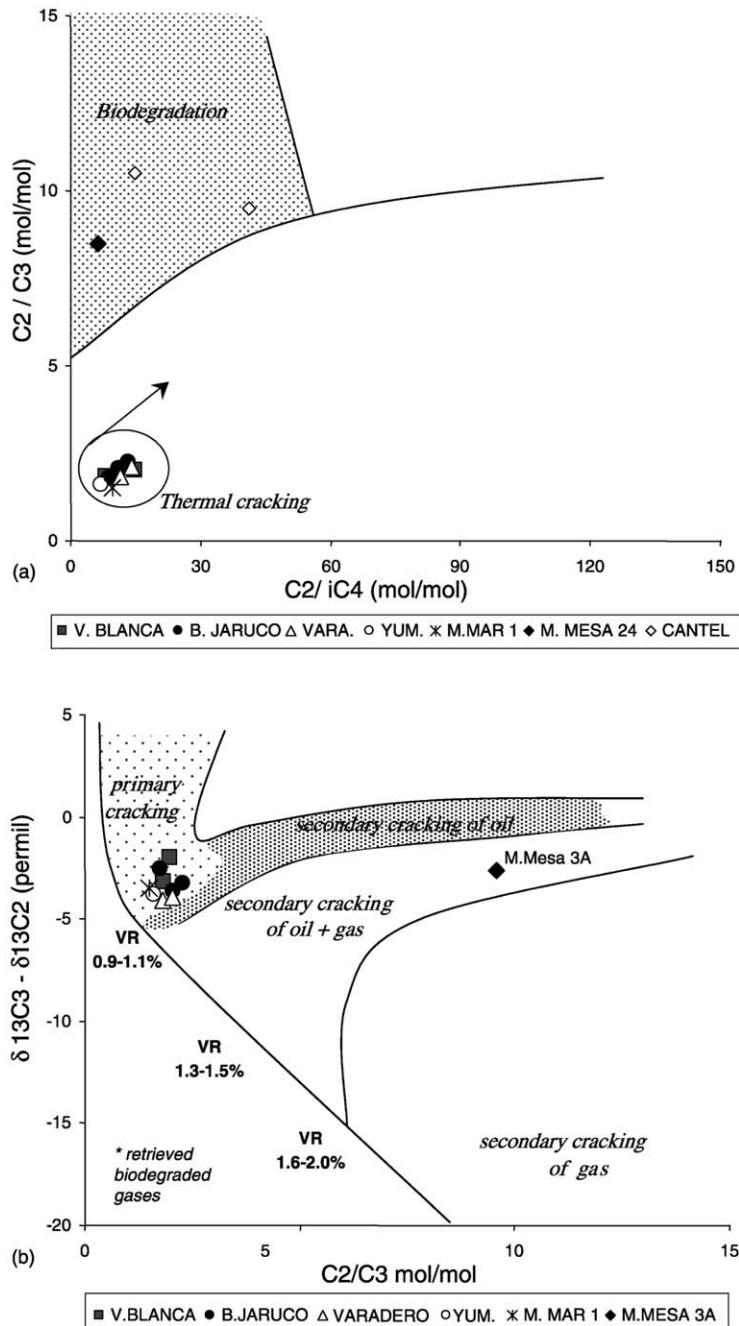
Fig. 5. Biodegradation (top) and maturity (bottom) plots from gas isotopic and molar analysis.

check expected geochemical compositional differences. Bulk oil properties represented by the API gravity and SARA chemical proportions described a bimodal distribution. Some crude oils showed consistent low API gravities concomitant with high sulphur ($>1\%$) and high Nickel and Vanadium content in the Jurassic and Cretaceous intervals (Fig. 6), with the Tertiary oils plotting on a different slope. A direct relationship was established between NSO content (resins and asphaltenes) and the percent sulphur as both

increase together. The origin and cause for low API gravities (below $15°$ API), high sulphur and high metal content hydrocarbons may be due to the alteration of fluids by biodegradation or in situ products of early cracking of the source rocks, or a mixture of both processes. One of the aims of the study was to decipher which process was dominant in the present northern Cuban oils. Consequently, we attempted to classify the different compositions with regards to their belonging to normal oils, and oils with traces of secondary
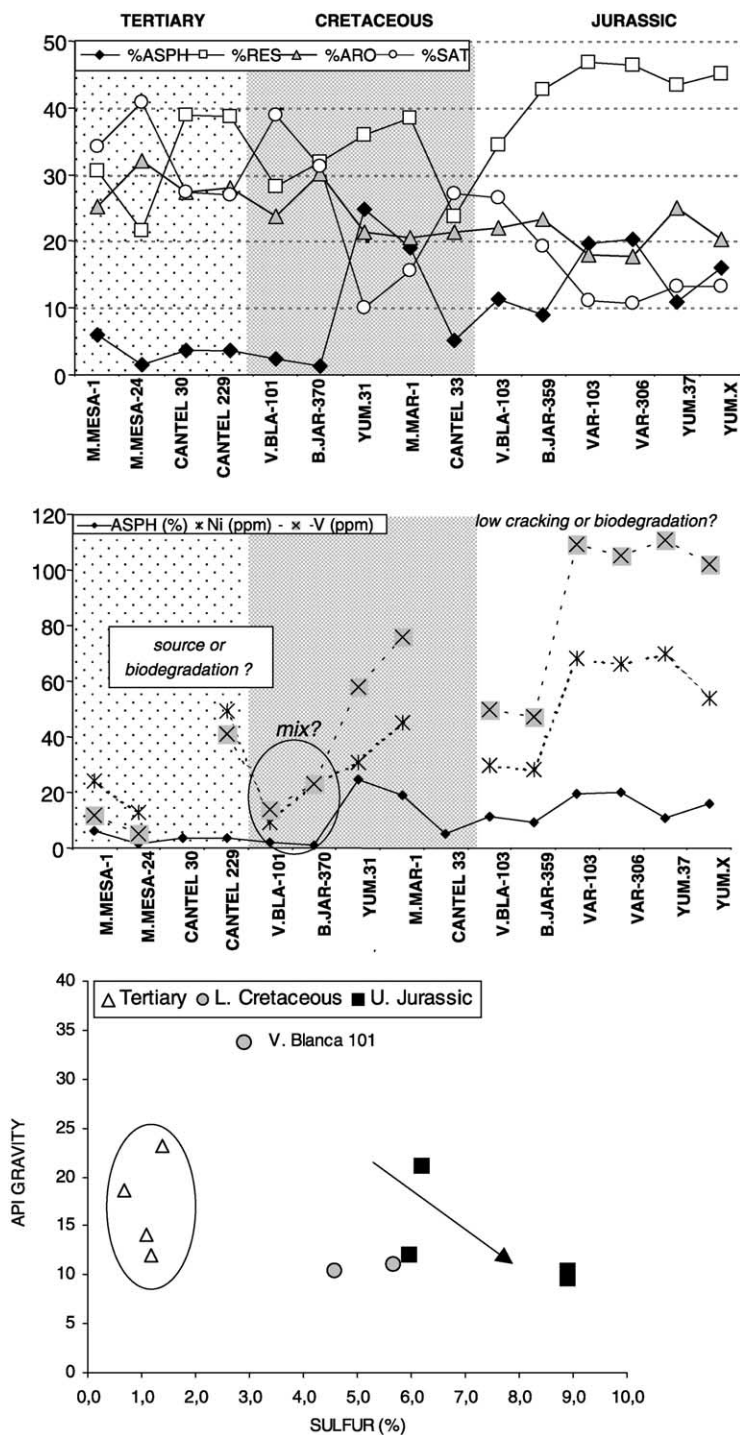
Fig. 6. The top graph shows the distribution of chemical families in oils (asphaltenes, resins, aromatics and saturates). The middle graph shows the proportion of metals (nickel and vanadium) and percent of asphaltenes. The bottom graph is a plot of the bulk API gravity of the oils as a function of the percent sulphur.

affects such as biodegradation and co-mingling of crude oils within a same reservoir unit.

6.3. High resolution gas chromatography of whole oils

High resolution gas chromatography analyses (GC) of whole oils give the distribution of hydrocarbons from $C_5–C_{35}$ range, and we classified the oils as follows (Fig. 7):

GROUP 1 contains not degraded oils with relatively 'normal' GC traces: It includes only one of the Tertiary reservoir (M. Mesa-1), two Cretaceous reservoired fluids (V. Blanca-101 and M. Mar-1), and three U. Jurassic producing oils (V. Blanca-103, Varadero-103, B. Jaruco-359).

GROUP 2 is composed of crude oils with biodegraded or odd hydrocarbon profiles. All the GC traces have a predominant 'unresolved hump' in the $C_{26}–C_{35}$ range
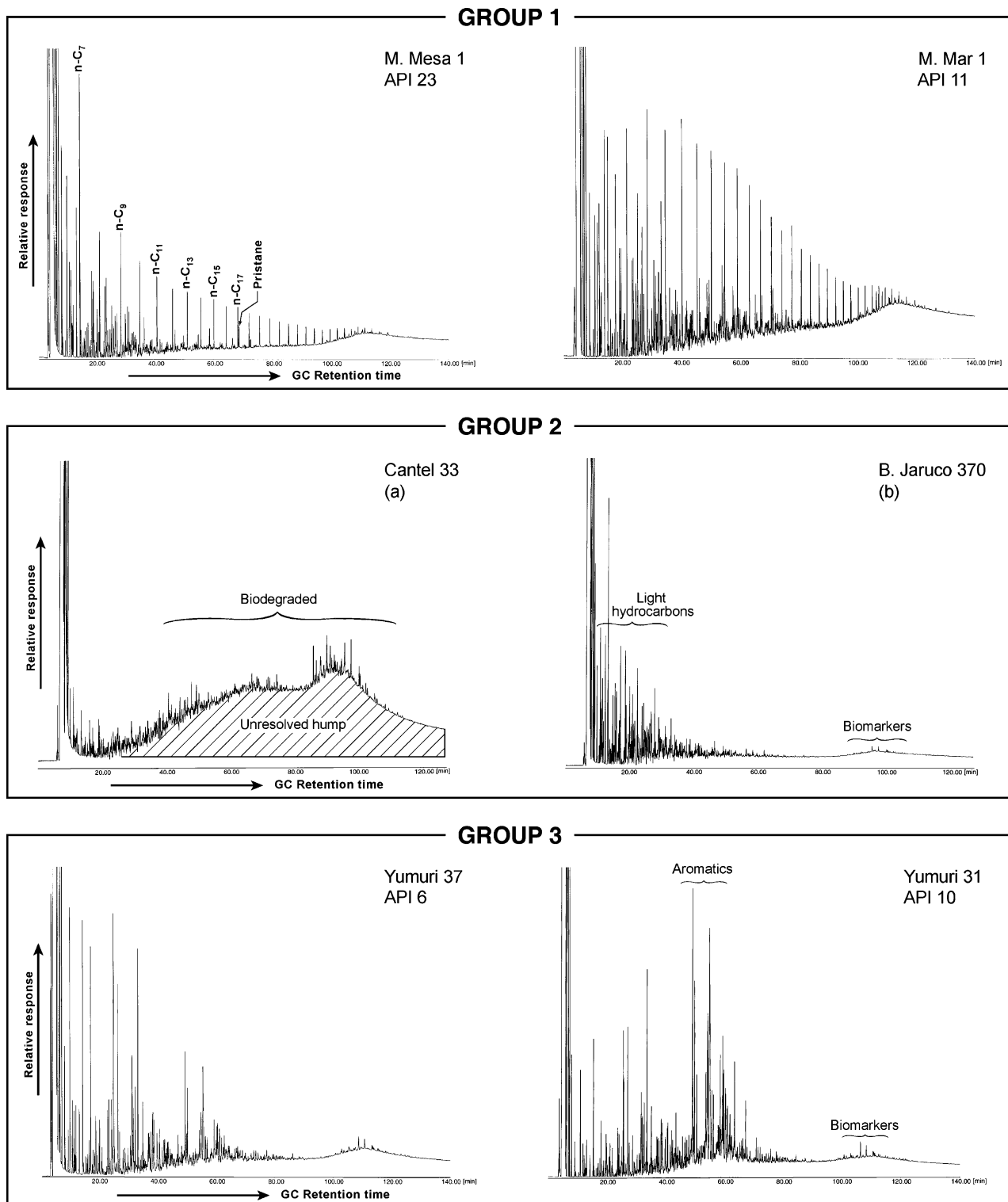
Fig. 7. High resolution gas chromatography hydrocarbon profiles of representative whole oils (GROUP 1 = normal; 2 = biodegraded profiles; 3 = semi normal).

with visible biomarkers, and a total absence of C_{15+} n-alkanes as in Cantel-30, Cantel-229, M. Mesa-24 oils (Tertiary reservoirs) and Cantel-33 oil (U. Cretaceous reservoir GROUP 2a oils left, Fig. 7). Included in

the group are three 'odd' samples, B. Jaruco-370 (L. Cretaceous) and two U. Jurassic Varadero-306 and Yumuri-X oils (GROUP 2b oils right, Fig. 7). These crude oils have been included in GROUP 2 because of their C_{12+} n-alkanes

depletion, although the chromatograms show the presence of some light hydrocarbons. Light ends in crude oils are generally easily degraded from bacteriae, making GROUP 2b atypical for biodegraded crude oils.

GROUP 3 contains two samples, Yumuri-37 oil (U. Jurassic reservoir) and Yumuri-31 oil (L. Cretaceous reservoir) distinguished by their GC trace. In this restricted group, the hydrocarbons have ample C_5–C_{20} compounds, including minor low molecular weight straight chain n-alkanes, and 1other peaks with strong FID signal response, that elute at different retention times than regular n-alkanes. Based on full scan mass spectrometry, these compounds were identified as aromatic compounds: benzene, toluene, ethylbenzene, xylenes, napthtalenes, alkyl benzenes, alkylated thiophenes and in minor amount dibenzothiophenes, phenanthrenes. These GC profiles are comparable to published data from early generated hydrocarbons issued from artificial maturation experiments by closed system pyrolysis reported by Sinninghe Damsté, Eglinton, De Leeuw, and Schenk (1989) and Tomic et al. (1995) of type IIS kerogen. It is unclear, however, if in the two Yumuri oils of GROUP 3, low thermal cracking is the only process that could provide such profiles, but we can mark out biodegradation as no significant traces of it may be seen in the n-alkane distribution.

The information obtained by gas chromatography thus confirmed that crude oils in Cretaceous and Jurassic reservoirs have semi-normal patterns of hydrocarbon distribution, that are associated to marine anoxic depositional environments of the organic matter. The source affiliation is made from the GC profiles of oils with visible n-alkanes and where ratios of pristane/phytane are ≤ 1, and sometimes minor even to odd carbon preference. Finally, oil fingerprints show the possibility of mixed biodegraded reservoired fluids that contain light ends. That point will be discussed however in more details later.

6.4. Gas chromatography-mass spectrometry

The C_{15+} saturated hydrocarbon fractions were analyzed by full scan GCMS and parent mode MRM gas chromatography-mass spectrometry (GCMSMS). Typical MRM transitions for steranes and hopanes are reported for tricyclic, tetracyclic and pentacyclic terpanes (m/z191 → parent ion) and for steranes (m/217 → parent ion). For information, the MRM transitions for those oils shown on Fig. 7 are reported in Fig. 8.

6.5. Secondary alterations (biodegradation)

Prior to the source and maturity assessment of C15 + hydrocarbons, the data was screened using full scan GCMS for oil quality control and molecular C15 + hydrocarbon biodegradation. GROUP 2 oils seem to suggest the occurrence of biodegradation because of the hydrocarbon profiles. That was investigated by GCMS

which may ascertain the degree of biodegradation on a molecular level and on a relative scale as discussed by Peters and Moldowan (1993). After extensive biodegradation, biomarkers generally change in spatial conformation (stereochemistry) and proportions. A well recognized biodegradation related biomarker, 25-norhopane is often taken as evidence of extensive bacterial activity. 25-norhopane without the full homolog serie, was however only found in low amount in an non biodegraded M. Mesa-1 sample (GROUP 1 oil) limiting the idea of very extensive biodegradation in north Cuban reservoired fluids. As a matter of fact, we believe that in this particular case, its presence in this seemingly normal oil sample is unusual, and would require obtaining some sampling of the source rocks which we do not have available at this time. We cannot however rule out the possibility of a left over biodegraded fluid mixed with a more recent pulse, as no other geochemical evidence was found for that scenario. The M. Mesa-24 sample from the same field (Tertiary reservoir), although it features a depleted n-alkanes GC trace and is seemingly more biodegraded, contains non altered C15 + biomarkers and no 25-norhopanes.

We can thus assume that biodegradation, as shown from the GCMS data available, has not very much affected the C15 + saturated fractions from producing Jurassic and Cretaceous intervals belonging to GROUP 1 oils (V. Blanca-101 and 103, B. Jaruco-359, M. Mar-1, Varadero-103), GROUP 2b (B. Jaruco-370, Yumuri-X and Varadero-306), nor any of GROUP 3 oils (Yumuri-31 and 37).

In GROUP 2a however, Tertiary oils have lost most of their n-alkanes and the molecular GCMS data shows that some hydrocarbons contain regular patterned pentacyclic hopanes, and somewhat regular steranes with substantial rearranged steranes (diasteranes). The only sample that is genuinely biodegraded in terms of biomarkers is from the U. Cretaceous reservoir, Cantel-33 oil (1140–1172 m) depleted in saturated C_{5+} hydrocarbons (no n-alkanes and a significant unresolved C_{28}–C_{35} hump), showing a normal trend of pentacyclic hopanes but quite an altered and non recognizable sterane distribution together with a strong presence of diasteranes (m/z 217 trace). Although the contribution of diasteranes can be sometimes linked to the source or elevated maturity, it is very likely to be due here to biodegradation. Therefore, ratios for the maturity evaluation based on the steranes in Cantel-33 were excluded for maturity evaluation, and so were the maturity ratios measured in Cantel-30, Cantel-229 oils (biodegraded hydrocarbons).

GCMS results from those oils belonging to GROUP 2b bring evidences that the unusual hydrocarbon distributions provided by high resolution whole oil gas chromatography are not signs of fluids affected by microbial action, but on the contrary underline the action of another secondary process other than biodegradation (the presence of
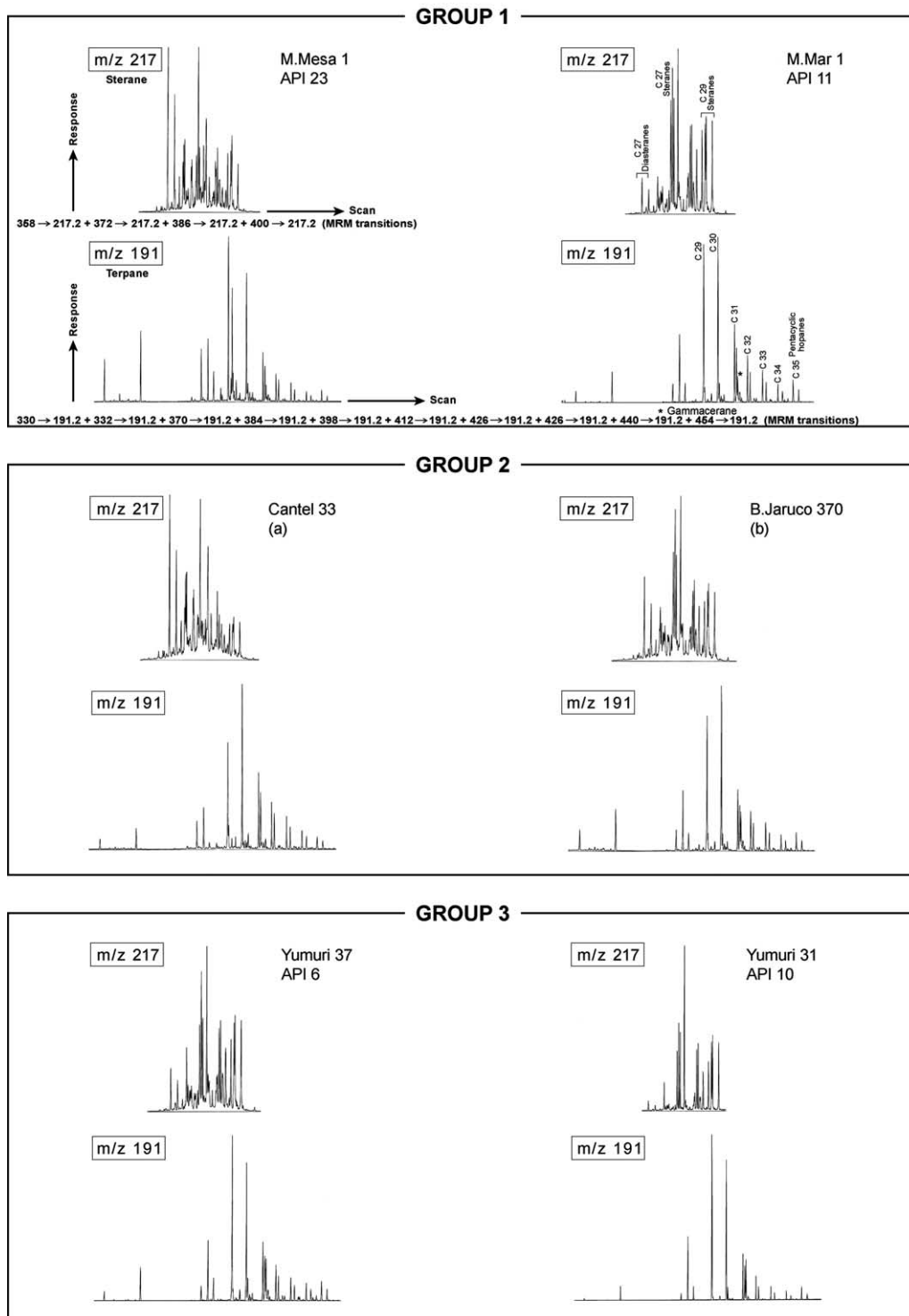
Fig. 8. Gas chromatography mass spectrometry data (MRM transitions) of selected samples.

C_5-C_{20} hydrocarbons together with a important GC hump). The presence of light hydrocarbons (carbon range between C_5-C_{14}) will be discussed later, and could be linked to the stage of the hydrocarbons thermal maturity.

The levels of biodegradation given from the relative scale of alteration described by Peters and Moldowan (1993) are provided in Table 3 for a summary. As a reminder, pentacyclic hopanes, tricyclics and tetracyclic terpanes, and steranes are estimated to begin to be altered at level 5 (out of a maximum of 10). At level 5, the oils have lost n-alkanes, and are depleted in isoprenoids (phytane and pristane for example).

Table 3
Gas chromatography-mass spectrometry data for biomarker analysis

Crude oils well	Depth top	Depth bottom	Age	Maturity ratios							
				C27 steranes (S/R + S)	C27 steranes αββ (/ααα + αββ)	C27 diasterane index	C27 steranes (Ts/Ts + Tm)	C29 steranes (S/R + S)	C29 streanes (αββ(/ααα + αββ)	C29 diasterane index	C29 steranes (S/R)
MARTIN MESA 1	824	807	L. Eocene	0.444	0.498	1.247	0.46	0.552	0.563	0.806	1.234
MARTIN MESA 24	733	773	L. Eocene	0.462	0.500	0.965		0.532	0.537	0.625	1.137
CANTEL 30	477	683	Paleocene	na	na	na	na	na	na	na	na
CANTEL 229	424	488	Paleocene	na	na	na	na	na	na	na	na
CANTEL 33	1140	1172	U. Cretaceous	0.474	0.439	2.411	0.41	0.488	0.499	1.740	0.954
VIA BLANCA 101	1380	1410	L. Cretaceous	0.318	0.355	0.413	0.30	0.318	0.400	0.301	0.466
BOCA JARUCO 370	1306	1356	L. Cretaceous	0.356	0.408	0.321	0.25	0.401	0.486	0.232	0.669
MARBELLA MAR 1	2550	2565	L. Cretaceous	0.409	0.467	0.117	0.20	0.466	0.514	0.070	0.873
YUMURI 31	1487	1467	L. Cretaceous	0.293	0.403	0.053	0.15	0.436	0.550	0.045	0.774
YUMURI 37	1280	1350	U. Jurassic	0.354	0.444	0.161	0.16	0.444	0.521	0.151	0.798
YUMURI X	2321	3600	U. Jurassic	0.391	0.435	0.174	0.21	0.431	0.510	0.117	0.759
VIA BLANCA 103	1989	2054	U. Jurassic	0.420	0.470	0.234	0.31	0.466	0.534	0.144	0.874
BOCA JARUCO 359	1826	1857	U. Jurassic	0.386	0.449	0.188	0.25	0.457	0.521	0.120	0.841
VARADERO 103	1690	1720	U. Jurassic	0.378	0.466	0.116	0.17	0.473	0.547	0.088	0.899
VARADERO 306	1613	1645	U. Jurassic	0.388	0.472	0.139	0.19	0.450	0.559	0.077	0.817

| Crude oils well	Depth Top	Oil group	Source ratios								Biodegradation level
			Gam/H30	H35/H34	H29/H30	Tet24/H30	Tet24/Tri26	Tri C23/H30	Gam/Tri23	30nor/Gam	1-normal 10-biodeg.
MARTIN MESA 1	824	1	0.120	1.000	0.883	0.221	0.889	0.379	0.473	1.538	1–2
MARTIN MESA 24	733	2a	0.100	0.800	0.688	0.188	1.500	0.325	0.308	0.000	4–5
CANTEL 30	477	2a		na	na	na	na	na	na	na	7–8
CANTEL 229	424	2a		na	na	na	na	na	na	na	7–8
CANTEL 33	1140	2a	0.090	0.800	0.062	0.074	1.500	0.136	0.273	1.000	7
VIA BLANCA 101	1380	1	0.240	0.985	0.813	0.073	1.467	0.107	2.250	0.472	1–2
BOCA JARUCO 370	1306	2b	0.190	1.050	0.939	0.085	1.750	0.146	1.167	0.643	1–2
MARBELLA MAR 1	2550	1	0.196	1.241	0.970	0.083	1.474	0.107	1.833	0.758	1–2
YUMURI 31	1487	3	0.310	1.229	1.030	0.104	2.833	0.116	4.316	0.402	1–2
YUMURI 37	1280	3	0.329	1.273	1.186	0.100	1.750	0.129	2.556	0.435	1–2
YUMURI X	2321	2b	0.313	1.280	0.753	0.048	1.000	0.096	3.250	0.308	1–2
VIA BLANCA 103	1989	1	0.380	1.069	0.675	0.066	1.158	0.114	3.316	0.317	1–2
BOCA JARUCO 359	1826	1	0.259	1.063	0.835	0.071	1.333	0.118	2.200	0.477	2–3
VARADERO 103	1690	1	0.291	1.261	0.848	0.061	1.667	0.073	4.000	0.458	1–2
VARADERO 306	1613	2b	0.291	1.269	0.771	0.065	1.375	0.106	2.750	0.384	2–3
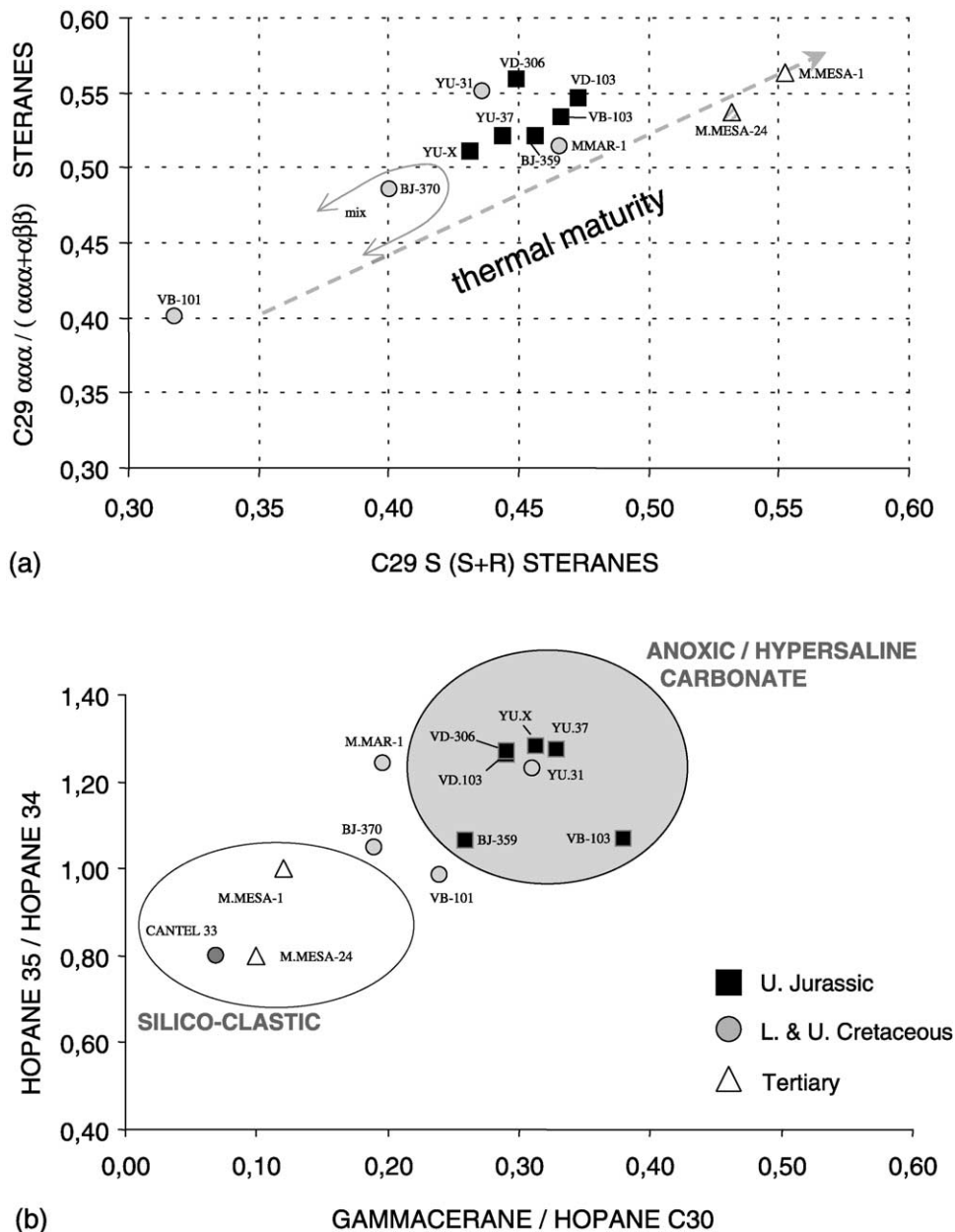
Fig. 9. (a) Thermal maturity ratios from biomarker analyses. (b) Plot of source indicators showing the hypersaline depositional environments of the crude oils.

6.6. Thermal maturity of crude oils

The thermal assessment of the hydrocarbons was calculated and discussed for the gaseous fractions. The average Cretaceous and Jurassic reservoired gases were found to have moderately low thermal maturities. Tertiary gases were biodegraded so that the C_1–C_4 maturity parameters were not computable, except for one producing interval (M. Mesa-3A) with very dry and mature gas (no associated oil). Using Full Scan MS and MRM transitions to exclude possible mass interferences, the thermal maturity of C_{15+} hydrocarbons of the crude oils was then assessed. The most mature hydrocarbons are once more encountered in

the Tertiary reservoirs in M. Mesa-1 and M. Mesa-24 (biomarkers not biodegraded) with ratios of the C_{29} (S/S + R) steranes between 0.54 and 0.56, signifying that the C_{29} sterane isomers reached equilibrium (Fig. 9a). However, the maturity ratios of C_{29} (S/S + R) steranes for most U. Jurassic and L. Cretaceous reservoired hydrocarbons range between 0.43 and 0.50, apart from two samples V-Blanca-101 and B. Jaruco-370 with ratios at 0.32 and 0.40, respectively. This signifies that most oils are found in the early hydrocarbon generation window (steranes are still dominated by the 20R stereochemistry, % vitrinite reflectance equivalence of 0.6–0.8). Note also that Yumuri oils systematically appear in the lower end of the maturity

trend as compared to most samples. This confirms other geochemical observations made so far about type IIS organic matter hydrocarbons, if we consider that Yumuri oils are more type IIS than strictly type II marine organic matter sourced. Considering regionally the Jurassic and Cretaceous reservoired oils, a minor trend in maturity is observed from East to West (V. Blanca > B. Jaruco > Yumuri, and in the eastern part, M. Mar > Varadero).

Two samples both from L. Cretaceous reservoirs however were noted to have unusually low thermal maturities: V. Blanca-101 and B. Jaruco-370 (two wells that are geographically close). Moreover, that information is contradictory to the preliminary information gathered from the light and bulk fluid composition that would indicate that the oils are light and mature.

6.7. Source of hydrocarbons

GC-MSMS is a powerful tool to ascertain the different depositional environments of the source rocks that generate hydrocarbons, and allows us to compute isomerization stage of specific compounds that have been correlated to vitrinite reflectance evolution and consequently may be directly linked to the thermal maturation of hydrocarbons. As long as biodegradation has not altered the composition of the $C_{26}–C_{35}$ fractions (the latter are more resistant) this information is taken to be accurate. Source data for the samples analyzed is summarized in Table 3. Two main families exist in the data set, one that includes Tertiary oils with a silico-clastic source (M. Mesa and Cantel wells), and another which includes all the oils from U. Jurassic, L. Cretaceous (B. Jaruco, V. Blanca, Yumuri, M. Mar, Varadero). The source rock for the latter are characterized as marine carbonates with organic matter deposited in hypersaline/anoxic cycles. The marine influence on the composition of the crude oils from carbonate producing intervals is confirmed by the presence of specific biological tracers. Pentacyclic steranes C_{30}, abundant tricyclics terpanes, C_{24} tetracyclics have been related to such environments by Moldowan (1985). The higher proportion of C_{27} relative to C_{29} steranes (Huang and Meinschein, 1979), gammacerane (Ten Haven, De Leeuw, & Schenck, 1985), C_{30} norhopanes (Peters & Moldowan, 1993), and
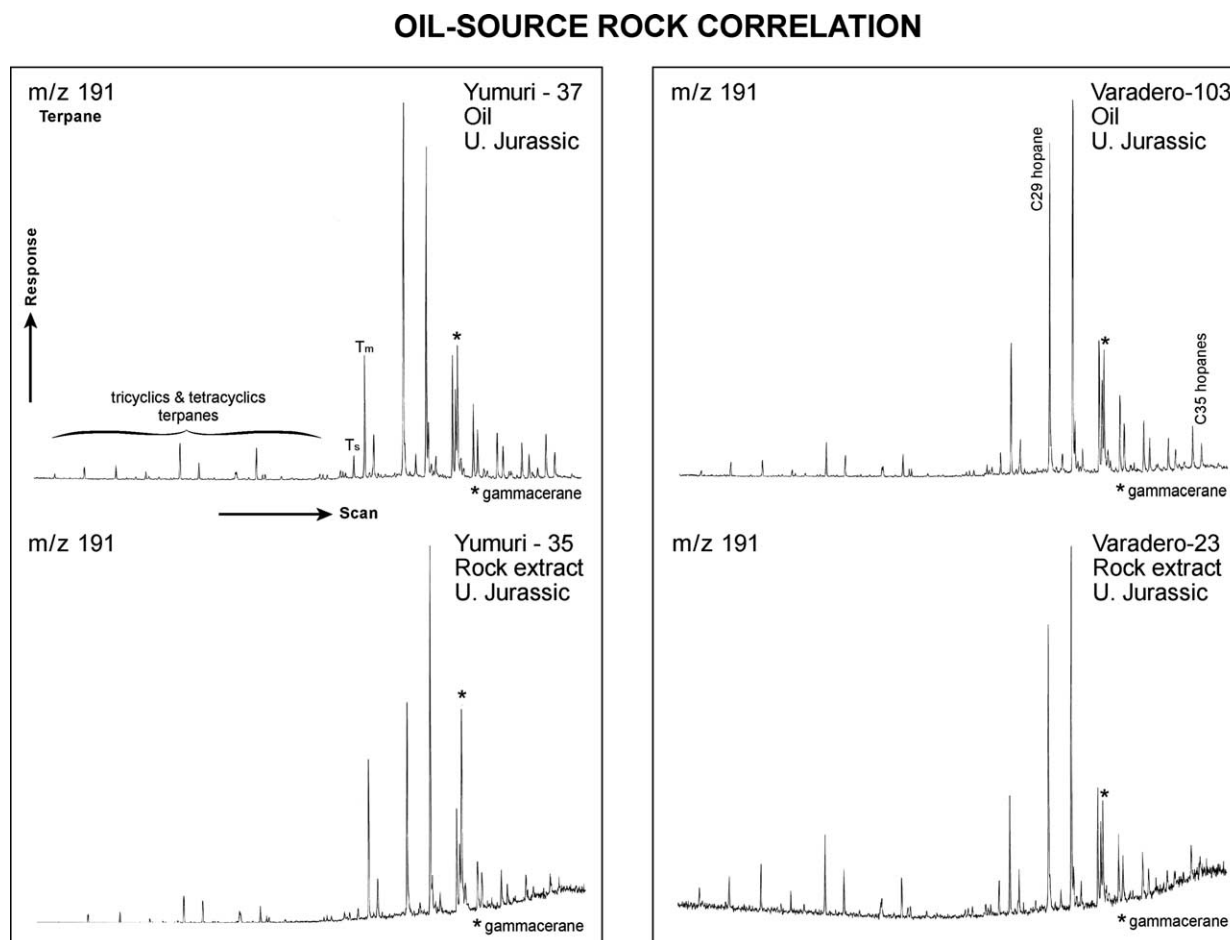
OIL-SOURCE ROCK CORRELATION

Fig. 10. Correlation of rock extracts and crude oils from the Upper Jurassic sediments.

28–30 bisnorhopane in moderate concentration (Mello, 1988) are also biological markers from marine deposits. A high ratio of C_{35}/C_{34} pentacyclic hopanes (>1) often infers a carbonate hypersaline/anoxic depositional environment. When plotting two 'carbonate' indicators for this data set, the gammacerane/hopane C_{30} ratio vs. the hopane C_{35}/hopane C_{34} ratio, two oil families may be observable in northern Cuban hydrocarbons (Fig. 9b). On the one hand, the silico-clastic oil family including M.Mesa-1 (lower content of gammacerane and a C_{35}/C_{34} pentacyclic hopane ratio <1), the partially biodegraded M. Mesa-24 (heavy oil, depleted GC trace but normal biomarkers) and the Cretaceous Cantel-33 sample (the pentacyclic hopanes are not degraded), and on the other, the grouping of the other L. Cretaceous and U. Jurassic reservoired oils. A trend shows that U. Jurassic hydrocarbons (V-Blanca-103, B-Jaruco-359 and Varadero) are somewhat more hypersaline than L. Cretaceous oils (i.e. B. Jaruco-370 and V. Blanca-101, M.Mar-1). The distinction is however not so clear with L. Cretaceous Yumuri-31 oil which is very similar to the other two available U. Jurassic reservoired Yumuri oils.

6.8. Oil-source rock correlation

A good agreement is seen between source rocks and crude oils with the *m/z* 191 trace, as shown with the Yumuri-35 source rocks (2118–2121 m, U. Jurassic) and compared to the producing Yumuri-37 oil (1280–1350 m, U. Jurassic). A positive correlation was also established between the U. Jurassic Varadero-23 rock extract (1478–1480 m) and the U. Jurassic Varadero-103 crude oil. The correlation between U. Jurassic source rocks and crude oils is illustrated on Fig. 10.

The DSDP-535 L. Cretaceous sample described by Palacas et al. (1984) was tentatively correlated to Cuban northern oils and to Sunniland oils from offshore Florida. Some of the difficulties arose when comparing published GCMS fingerprints from twenty years ago and the present high resolution gas chromatography analyses. However, our results show that the DSDP-535 impregnated L. Cretaceous limestone sample may be better affiliated, from its distribution and proportion of C15 + hydrocarbons, to the Varadero and M. Mar-1 crude oils producing from U. Jurassic reservoirs. The marine carbonate origin of the hydrocarbons was confirmed with the ubiquitous presence of gammacerane and high C35-hopane signal. It is interesting to note that the correlation was in fact better with the Cuban onshore carbonate reservoired oils than with a Sunniland oil (offshore Florida, Palacas et al., 1984), as the latter did not show the presence of 28,30 bisnorhopane, nor a similar distribution of tricyclics (Fig. 11). The good agreement between recent rock extracts, published DSDP rock extracts and the new northern Cuban U. Jurassic oils do indeed suggest that a common type of depositional
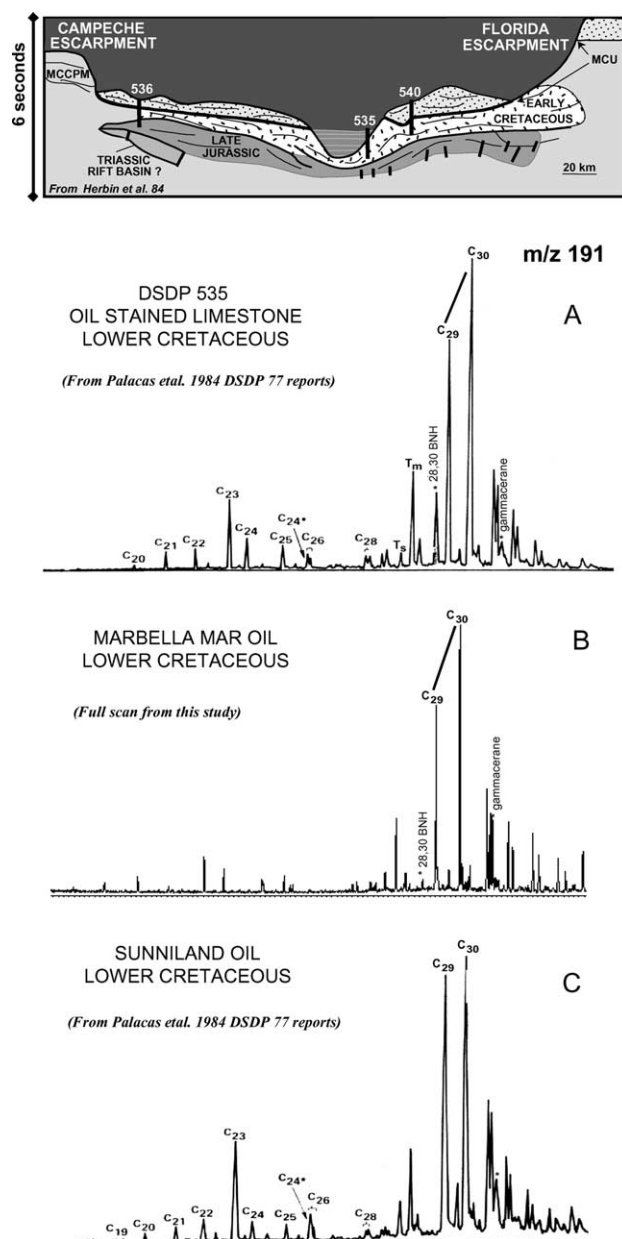

Fig. 11. Comparison of Lower Cretaceous biomarker distribution (*m/z* 191) from the offshore DSDP-535 core, the Cuba M. Mar-1 and offshore Sunniland oils.

environment and a common petroleum system for carbonate derived oils exist in the southern part of the Gulf of Mexico.

6.9. Is hydrocarbon mixing occurring in cretaceous reservoirs in north Cuba?

The contradictory bulk compositions with respect to the C15 + thermal maturity estimations in the samples V. Blanca-101 and 103 were then questioned (Fig. 12). V. Blanca-101 oil has a 34 API gravity in a reservoir located at a depth of 1380-1410 m (L. Cretaceous), while

GC WHOLE OIL DATA
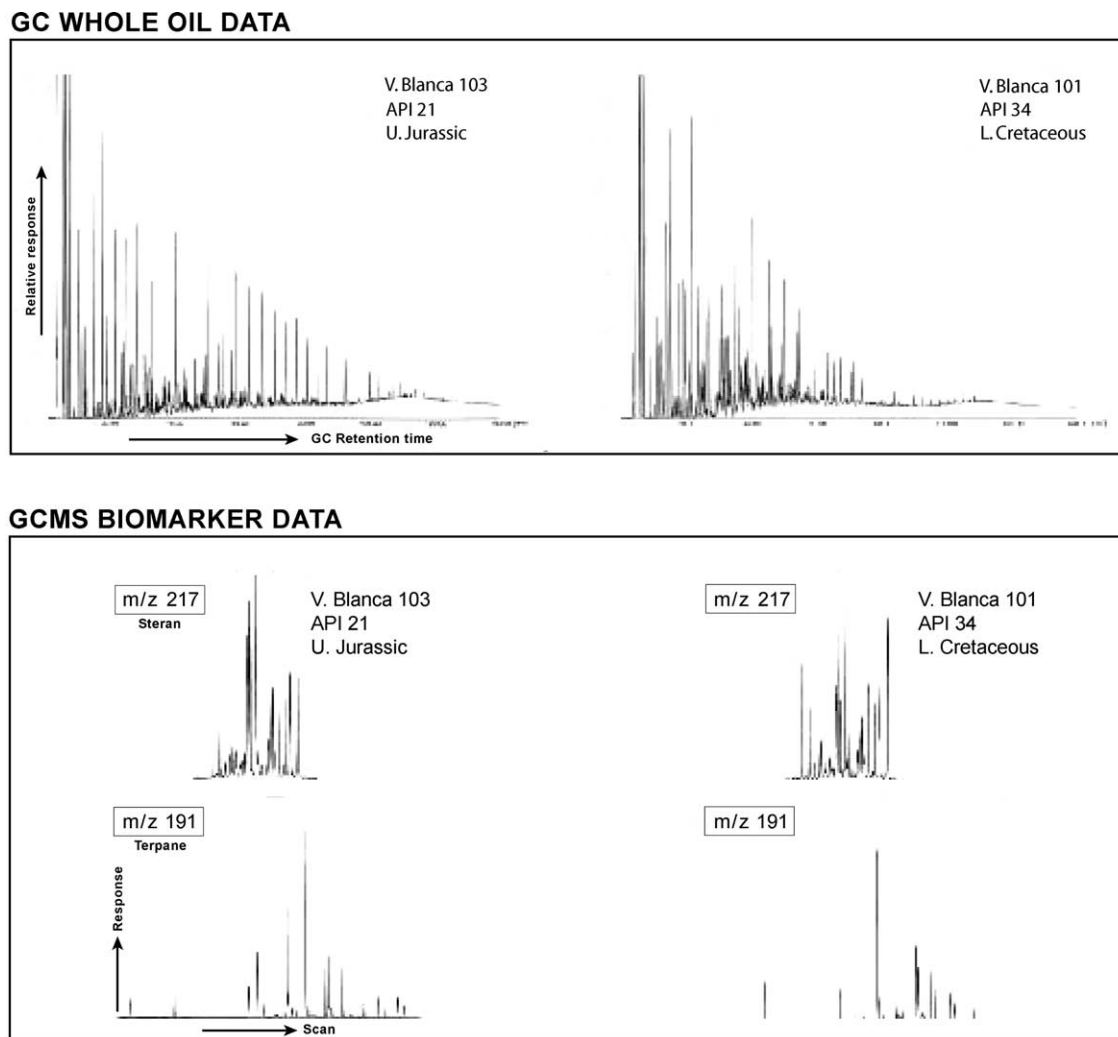

GCMS BIOMARKER DATA

Fig. 12. Secondary effects in crude oils: Example of hydrocarbon mixing in the Via Blanca field shown by whole oil gas chromatography (top) and biomarker data (bottom).

V. Blanca-103 has a 21 API gravity at a 1989–2054 m producing interval (U. Jurassic). As the deeper V. Blanca-103 reservoir hydrocarbons have higher % sulphur, high metal content and substantial content of NSO compounds (includes asphaltenes and resins), and considering that both samples have no traces of biodegradation from the gas and oil characterization (GROUP 1), the authors assumed that these properties were due to low thermal maturities of the V. Blanca-103 hydrocarbons. However, contradicting information was provided by the maturity data in the different hydrocarbon fractions, e.g. API gravities and gas data (C_1–C_4 specie) on the one hand and anomalous C_{15+} maturity data on the other. In other words, V. Blanca-101 seemed more mature than V. Blanca-103 from the API gravity and gas data, but V. Blanca-101 had less mature biomarkers than V. Blanca-103 (C_{29} (S/S + R) sterane ratio equal to 0.32 and 0.47, respectively). The authors accredit this oddity to a probable dilution effect occurring in the shallower

V.Blanca-101 Cretaceous reservoir caused by a mixing effect from more mature and lighter hydrocarbons (higher API gravity) that have not completely washed away the preexisting viscous and immature $C15+$, non biodegraded hydrocarbons. The immature biomarker $C15+$ hydrocarbons in the sample V.Blanca-101, are thus not representative of the present oil in place which are low viscous, light fluids. As for the V.Blanca-103 hydrocarbons, producing in Jurassic reservoirs, the level of maturity and the distribution of the hydrocarbons are in fact very much comparable to the other Jurassic oils from this study, and should be considered as normal hydrocarbons of relatively low thermal maturities. This scenario of multi pulse and mixing could also be occurring in the L. Cretaceous reservoir B. Jaruco-370, because of the low measured thermal level of biomarkers, the abundance of light ends and gases in the fluids, all elements which are in disagreement with a strict (1) biodegradation effect and (2) low maturation effect. Recharging of the accumulation

could consequently explain the bulk properties with low proportion of polar components (i.e. dilution of NSO compounds, sulphur and metal).

7. Discussion

The extensive study of source rocks showed that numerous good source rocks levels were present in the U. Jurassic and L. Cretaceous sediments. The selected source rocks were characterized as having marine carbonate facies inherently rich in sulphur and with high Total Organic Carbon content and low Tmax. Two samples from source rock extracts were genetically linked to oils sampled from carbonate reservoirs in upper Jurassic producing intervals, and to an offshore DSDP lower Cretaceous impregnated limestone rock. The positive molecular correlation between various onshore reservoired oils and an offshore DSDP core from L. Cretaceous sediments suggest a genetic tie in the organic matter found in both. Low API gravity fluids containing high sulphur, high metal content, and high NSO compounds compose the majority of the U. Jurassic and Cretaceous reservoired fluids, easily differentiable from Tertiary samples. The Jurassic and Cretaceous reservoired fluids are predominantly from marine hypersaline depositional environments, with minor degrees of anoxicity during the lower Cretaceous (lower anoxic period). Most of the U. Jurassic and L. Cretaceous carbonate reservoirs contain thermogenic gases of low thermal evolution. The distances of gas migration from source to accumulation are estimated as short and suggest a source rock deposition very close to the actual reservoirs. GROUP I oil samples producing in Jurassic and Cretaceous reservoirs present no clear evidences of bacterial biodegradation in gases and none in crude oil fractions, and consequently could be considered representative of fluids originating from only one early mature hydrocarbon pulse (M. Mar-1, V. Blanca-103, Varadero-103, B. Jaruco-359). Moderate alteration due to biodegradation are denoted in Tertiary silico-clastic reservoirs (M. Mesa-24, Cantel-30, Cantel-229) and one U. Cretaceous

producing interval (Cantel-33). Although, the tertiary reservoirs seem to have accumulations of the most mature fluids and gases, due to their shallow depths and likely fractured setting, they also contain the most extensively biodegraded hydrocarbons. GROUP 2b hydrocarbons contain small concentrations of normal n-alkanes and no C15 + hydrocarbon biodegradation, a composition of oils that might result from low thermal maturities of hydrocarbons derived from type IIS organic matter. The two Yumuri oil samples classified in GROUP 3 may also be included as hydrocarbons of marine depositional environments rich in sulfur and very immature. The contradictory geochemical maturity information found in a seemingly normal high API gravity crude oil producing in a L. Cretaceous reservoir, V.Blanca-101 (GROUP 1) is interpreted as resulting from an event of recharging of the reservoir with lighter and mature fluids. The same kind of multi filling scenario could provide an answer for the geochemical composition of B. Jaruco-370 (GROUP 2b) in view of the bulk, gas and liquid information obtained.

8. Conclusion

The geochemical study of outcrops and source rocks on the one hand, and crude oils and associated gases from producing wells on the other allowed us to furnish an updated database for the interpretation of the petroleum potential of northwest and offshore Cuba. One of the main result of this geochemical work was to bring growing support to an extension of the petroleum system from the Cuban folds and thrust belts located onshore to the offshore area crossing through the unexplored foreland basin to the platform area. This may involve a deep source rock system from post-rift deepwater carbonates of U. Jurassic and L. Cretaceous age, as schematically proposed on Fig. 13 (Gaumet & Letouzey, 2002). The interpretation is in agreement with the properties of the Jurassic and L. Cretaceous reservoir fluids. Furthermore, the oil and gas characterization confirms that northwest Jurassic and Cretaceous carbonate reservoirs have oils with no
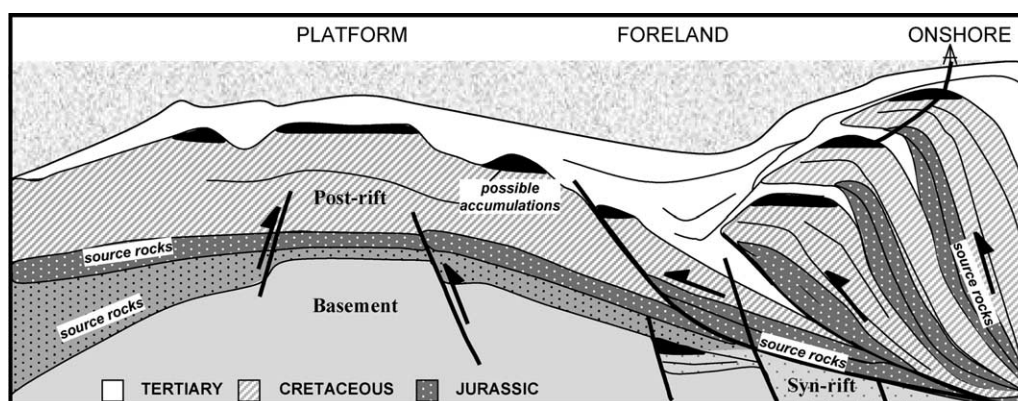

Fig. 13. Schematic illustration of the possible hydrocarbon migration pathways and petroleum systems in Northwest and offshore Cuba (adapted from Gaumet & Letouzey, 2002).

significant traces of biodegradation and that the hydro-
carbons have in average low to early thermal maturation
levels.

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