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Organic geochemistry, maturity, palynofacies and palaeoenvironment of Upper Kimmeridgian and Lower Tertiary organic-rich samples in the southern Jura (Ain, France) and subalpine massifs (Haute-Savoie, France)

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Key-words: Palynofacies, Palaeoenvironment, Thermal maturity, Biomarkers, Source rocks, Petroleum, Kimmeridgian, Early Tertiary, Jura, Prealps, France.

ABSTRACT

Investigations on palynofacies and geochemical fossils (biomarkers) carried out on three organic-rich rock units in the Geneva area indicate:

– Kimmeridgian platy limestones in the southern Jura were formed mainly under anoxic conditions developing in backreef lagoonal "cuvettes". They are thermally immature. Locally they form a rich, type II, oil-prone source rock (e.g. at Orbagnoux).

– Coaly beds of the Upper Eocene Diablerets Layers in the Bornes Massif were deposited in narrow grabens where mildly anoxic marine to brackish conditions prevailed. They may form a mixed type II/III source rock.

– Lower-Middle Oligocene Meletta Shales, restricted to the Bornes Massif, are a mixed type II/III source rock formed under dysaerobic conditions. They are so far the only described occurrence of mild anoxia in Oligocene flysch-like sediments of the Delphino-Helvetic realm in France and Switzerland. Their eastwards increase in thermal maturity, leading locally to expulsion of oil, is probably due to the overburden of the prealpine nappes. In the west, they are immature, but they may bear the geochemical signature of short-lived thermal anomalies along the Vuache wrench fault.

RÉSUMÉ

L'étude des palynofaciès et fossiles géochimiques (biomarqueurs) effectuée sur trois niveaux riches en matière organique dans les environs de Genève a confirmé que:

– Les calcaires en plaquettes du Kimméridgien du Jura méridional se sont formés dans des conditions généralement anoxiques qui se développaient dans des dépressions localisées d'un lagon d'arrière-récif. Ils sont thermiquement immatures. Ils forment localement une excellente roche-mère à huile de type II (par exemple à Orbagnoux).

– Les charbons éocènes des Couches des Diablerets dans le Massif des Bornes se sont déposés en milieu marin à saumâtre dans d'étroits grabens favorisant le développement de conditions dysaérobiques. Ils forment probablement une roche-mère de type mixte II/III.

– Le faciès des Schistes à Meletta d'âge Oligocène inférieur à moyen semble limité au Massif des Bornes. Ces schistes forment une roche-mère de type mixte II/III déposée dans des conditions dysaérobiques. Ils représentent la seule indication d'anoxie modérée décrite à ce jour dans les dépôts oligocènes de type flysch du domaine delphino-helvétique franco-suisse. Leur maturité thermique augmente vers l'est où ils ont produit probablement de l'huile. Ce gradient géothermique est probablement dû à la surcharge des nappes préalpines. A l'ouest du Massif, les schistes sont immatures, mais l'analyse des biomarqueurs suggère qu'ils pourraient avoir été exposés à de brèves anomalies thermiques liées à la faille décrochante du Vuache.

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ZUSAMMENFASSUNG

Die Untersuchung der Palynofazies und der geochemischen Fossilien (Biomarkers) dreier, an organischer Substanz reicher Gesteinseinheiten aus der Umgebung von Genf brachte folgende Ergebnisse:

– Die Kimmeridge-Plattenkalte des südlichen Jura wurden hauptsächlich unter anoxischen Bedingungen in flachen Becken einer Hinterriff-Lagune gebildet. Sie haben eine geringe thermische Reife. Lokal bilden sie ein reiches Erdölmuttergestein des Typs II (z.B. bei Orbagnoux).

– Die kohligen Lagen der obereozänen Diablerets-Schichten im Bornes Massiv wurden in engen Gräben unter dysaeroben, marinen bis brackischen Bedingungen abgelagert. Sie können einem gemischten Typ II/III-Muttergestein zugerechnet werden.

– Die unter- bis mitteloligozänen Meletta-Schiefer beschränken sich auf das Bornes Massiv und bilden ein gemischtes Typ II/III-Muttergestein. Sie wurden unter dysaeroben Bedingungen abgelagert. Dies ist das einzige bis jetzt beschriebene Vorkommen von semi-anoxischen Schichten in oligozänen, Flyschähnlichen Sedimenten des delphino-helvetischen Raums in Frankreich und der Schweiz. Ihre nach Osten zunehmende thermische Reife ist wahrscheinlich auf die Auflast, der sie überlagernden, präalpinen Decken zurückzuführen. Ihre thermische Reife im Westen ist gering; hingegen können sie geochemische Merkmale kurzfristiger, thermischer Anomalien entlang der Vuache-Transformstörung aufweisen.

Introduction

A preliminary survey of some potentially organic-rich rocks was undertaken in areas surrounding Geneva, i.e. in the southern Jura, the Mt. Salève and the Bornes and Aravis subalpine massifs (Fig. 1). Some 40 samples (Table 1) were collected in terrains spanning the Jurassic, Cretaceous and Early Tertiary (Fig. 2).

Sampling was not exhaustive and not aimed at establishing a continuous geochemical log of the area from the Jurassic to the Tertiary. It was designed to locate some organic-rich horizons on which combined geological and geochemical methods of investigation could be applied in order to better understand their environment of deposition and their thermal history.

All collected samples were screened using Rock-Eval pyrolysis (Espitalie et al. 1985–1986), which determines, among other things, their content in total organic carbon (TOC). Only three rock units showed a significant TOC content (Table 1):

– the Kimmeridgian platy limestones in the southern Jura,
– the Upper Eocene Diablerets Layers in the Bornes Massif,
– the Lower-Middle Oligocene Meletta Shales in the Bornes Massif.

These samples were consequently analysed by the geological and geochemical methods described below.

Methods of investigation

Geological microscopic studies and Rock-Eval pyrolysis

Samples 1011 (Kimmeridgian platy limestones), 6691 (Diablerets Layers) and 6686, 6687 and 6695 (Meletta Shales) were prepared palynologically, i.e. all mineral constituents were destroyed by hydrofluoric acid and the remaining organic matter (OM) was observed under the microscope in transmitted light to determine the *palynofacies* of the samples (Combaz 1980). Observation of the various organic constituents (amorphous or figured) and determination of their relative abundance are useful com-

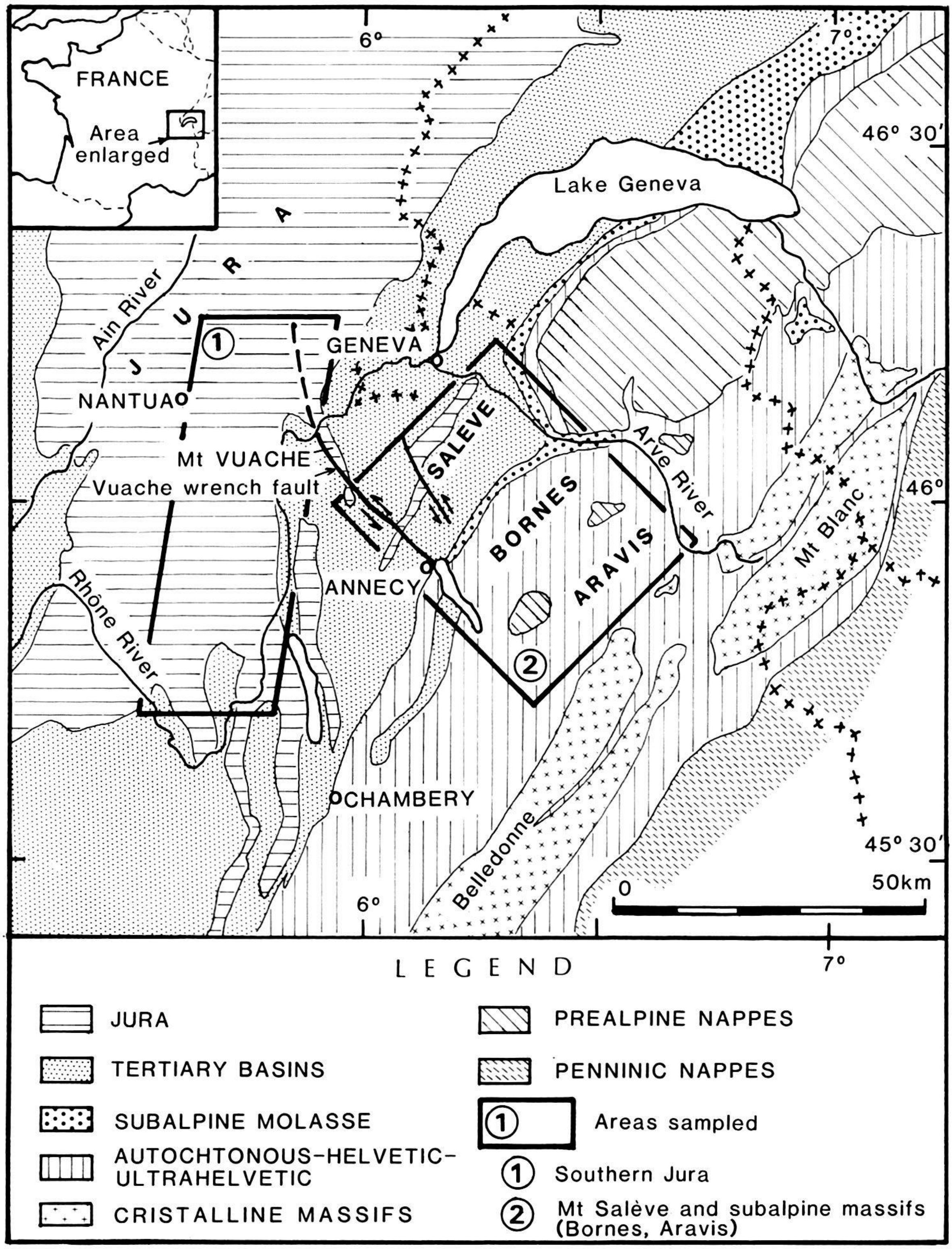


Fig. 1. Location map and tectonic framework.

Sample no.	Geographical location	Age	Rock Unit	Lithology	Rock Eval TOC (% weight)
SOUTHERN JURA MOUNTAINS					
6698	Mt. Vuache	Mid. Berrias.	Vions Fm.	gy-brn sandy lst	< 0.25
6696	Chésery	Earl. Berr.	Purbeck. facies	gy lst	ca. 0
6697	"	" "	" "	blk pebble cgl	"
6699	Mt. Vuache	" "	" "	" " "	"
1007	**Lac Armailles**	**Late Kimmer.**	**"Platy limest."**	**gy-brn lamin. lst**	**< 0.25**
1008	**Forens**	**" "**	**" "**	**" " " "**	**"**
1009	**Saint Champ**	**" "**	**" "**	**light brn lamin. lst**	**"**
1010	**Cerin**	**" "**	**" "**	**gy-brn " "**	**"**
1011	**Orbagnoux**	**" "**	**" "**	**" " " "**	**2.09**
1006	Montanges	Late Bathon.		brn bioclastic lst	ca. 0
1005	Forens	" "		gy fetid lst	ca. 0
1004	Monnetier	Late Bajoc.		gy bioclastic lst	< 0.25
1003	Ruty	Aalenian	"Cancelloph. mrl"	dk brn argill. lst	< 0.25
1002	Champfromier	Pliensbachian		dk gy mrl	0.28
1001	"	Hettangian		dk gy lst	< 0.25
MT. SALEVE					
6680-81	La Croisette Rd	Earl. Hauter.	Hauterive Marls	gy-blue sandy marl	< 0.25
S 812	Etiollets	Earl. Berrias.	Purbeck. facies	gn marly lst	ca. 0
S 875	"	" "	" "	blk pebble in bc	"
SUBALPINE MASSIFS					
BORNES					
6688	Dessy	Mid. Oligoc.	Subalp. flysch	fine sst	ca. 0
6700	**Pré-Vernay**	**E.-M. Oligoc.**	**Meletta Shales**	**platy gy-blue mrl**	**0.83**
6695	**Nanoir**	**" "**	**" "**	**" " " "**	**1.32**
6686	**Dessy**	**" "**	**" "**	**" " " "**	**1.27**
6687	**"**	**" "**	**" "**	**" " " "**	**1.18**
6693-94	Grand Bornand	Late Eocene	"sm. Nummul. lst"	gy lst	< 0.25
6683-84	Delaire	" "	Diablerets Layers	coaly argill. lst	< 0.25
6691	**Cenise**	**" "**	**" "**	**blk coaly lst**	**7.42**
6682, 6690	Delaire	Barremian	Urgonian facies	brn lst	ca. 0
6692, 6706	Petit Bornand	Hauterivian	"nod. sandy lst"	light gy lst	ca. 0
6689	Andey Plateau	Late Valang.	"altern. mrl-lst"	blue-gy argil. lst	< 0.25
6707	Entremont	Late Berrias.	"shaly marls"	dark brn mrl	< 0.25
6704-05	Petit Bornand	" "	" "	gy lst	< 0.25
ARAVIS					
6708-09	Aravis Pass	Late Jurass.	"Tithon. escarpm."	dark gy lst	ca. 0
6711	Pont de Manant	Call.-Oxford.	"shales"	blk slaty sh	< 0.25
6710	La Giettaz	Bajoc.-Bath.	altern. mrl-lst	gy argil. lst	< 0.25

Colours: brn=brown, blk=black, gn=green, gy=grey
Lithologies: bc=breccia, cgl=conglomerate, lst=limestone, mrl=marl, sh=shale, sst=sandstone

Table 1. List of samples investigated by Rock-Eval pyrolysis.

plements to standard sedimentology (Batten 1982, Habib 1983, Hart 1986 and Bustin 1988). They allow to identify the source of OM and to characterise its palaeoenvironment. These observations also help evaluating the degree and nature of diagenetic influences the OM has been exposed to (e.g. by determining sporomorph colour and preservation, Gutjahr 1966, Combaz 1980, Hart 1986). Qualitative observation of the fluorescence of the same palynological constituents in UV light gives another indication to the thermal maturity level of the sediments (Robert 1985, Hart 1986). Moreover, in immature to early mature OM, UV fluorescence allows a better definition of the nature of amorphous organic matter (AOM). In our microscopic observations

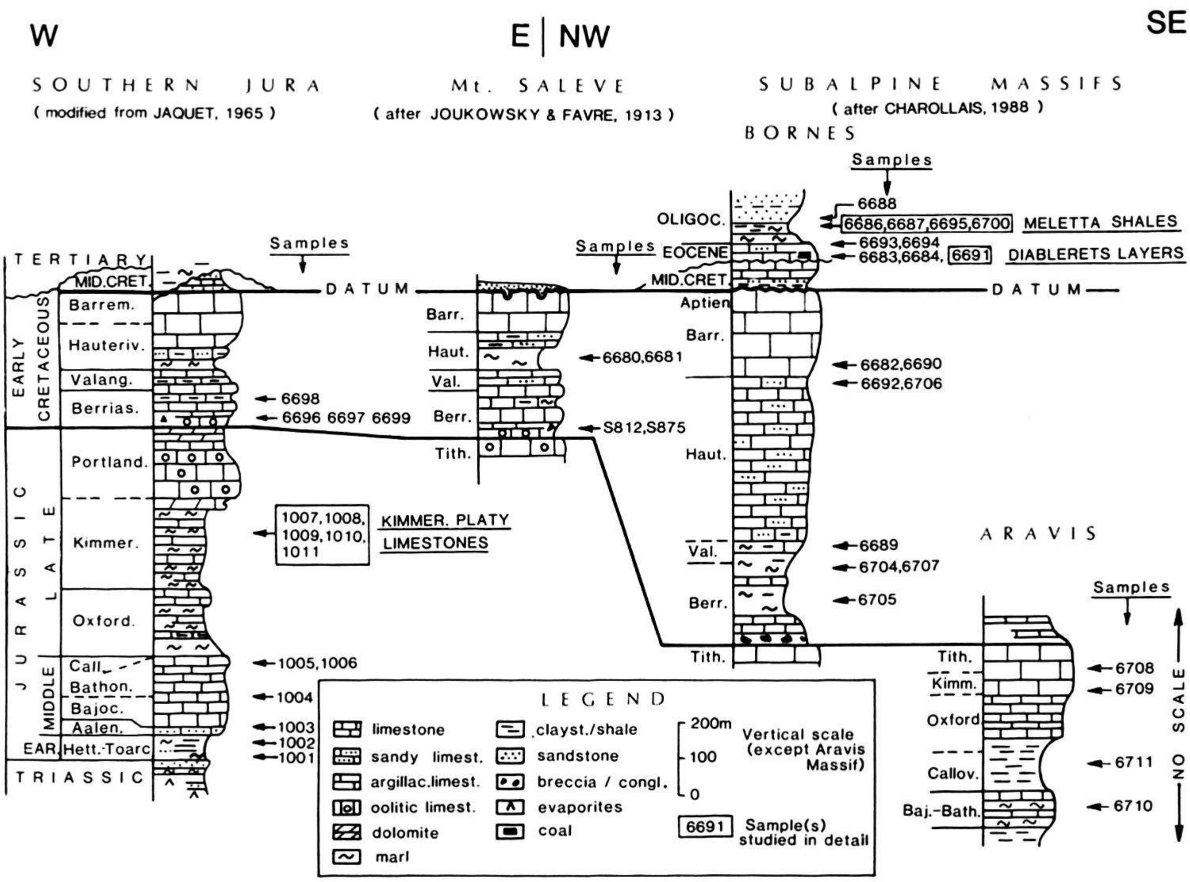


Fig. 2. Stratigraphical distribution of investigated samples.

we have used the thermal alteration index (TAI) of Staplin (1969). Its correlation with the vitrinite reflectance (VR) scale is given on Fig. 3 (after Hood et al. 1975).

The concept of the "oil window" is well summarized in Thomas et al. (1985): the onset of OM transformation to oil (the upper limit of the oil window) is thought to lie between VR = 0.5% and 0.7%, depending on the type of OM. The oil floor, representing the lower (deepest) limit of thermal conditions suitable for the preservation of liquid hydrocarbons, is reported by various authors to lie between VR = 1.2% and 1.35%. The peak of oil generation varies depending on type of OM: it lies at around VR 0.8% for a type II OM, and around VR 0.9% for a type III OM (Tissot & Welte 1984). This broadly-defined oil window (VR = 0.5% to 1.35%), representing a generalised observation from many source rocks and in numerous basins, is correlated with the TAI on Fig. 3. In this paper, we shall use the following threefold subdivision when referring to potential hydrocarbon generation (Fig. 3):

- immature VR < 0.5%
- oil zone VR = 0.5% to 1.35%, or mature
- gas zone VR > 1.35%, or overmature

Rock-Eval pyrolysis gives rapid information (Fig. 4) on the depositional environment and maturation of samples with a significant (i.e. greater than 0.25%) TOC content (Espitalie 1986, Espitalie et al. 1985–1986).

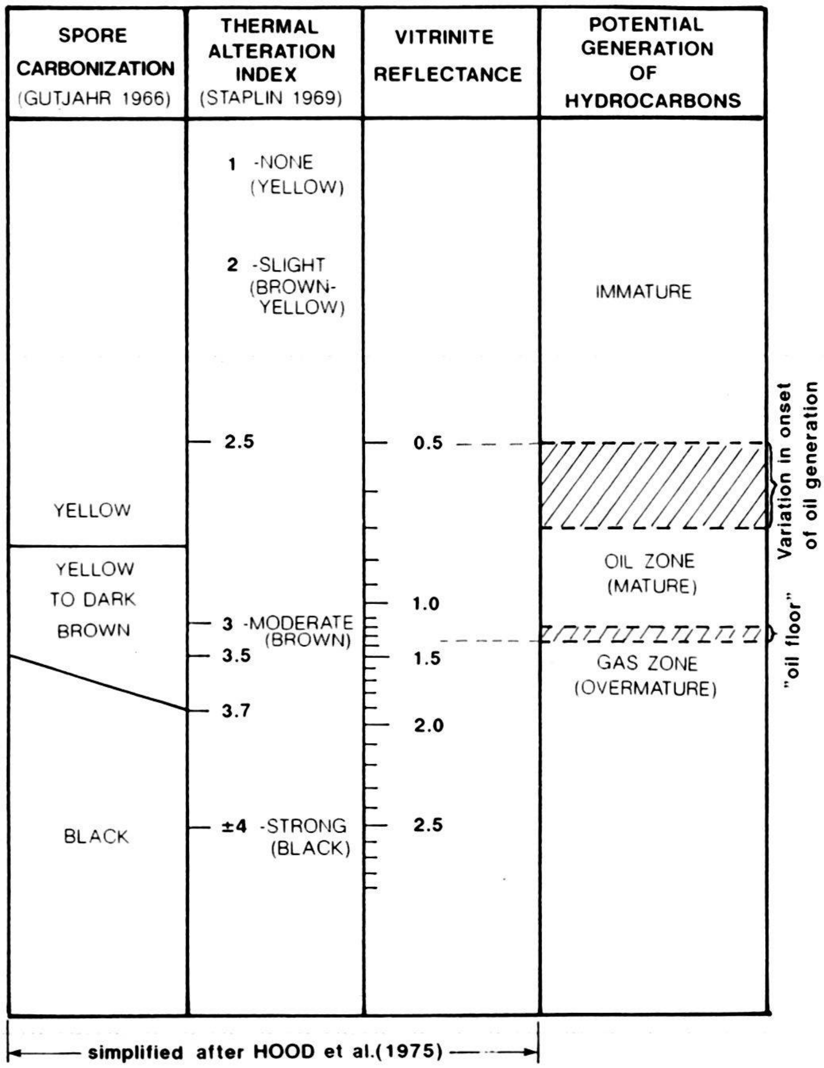


Fig. 3. Scales of organic metamorphism (simplified after Hood et al. 1975) and potential hydrocarbon generation.

The combined informations of Rock-Eval analyses and microscopic studies lead to the definition of organic facies and environments as defined in the Jurassic sediments of the North Sea by Thomas et al. (1985). The three rock units we studied were all deposited under marine conditions. Their OM consists essentially of type II and type III as defined by Tissot & Welte (1984). Type II OM (sapropelic) is an autochtonous OM derived mainly from phytoplankton and microorganisms (bacteria) and deposited in a reducing environment, defined as "anoxic aquatic" by Demaison & Moore (1980). Under aerobic conditions, OM, particularly that of marine origin, is quickly oxidised and destroyed, leaving only inert remnants (inertinite) and more-resistant material of largely terrestrial origin (vitrinite), which form type III OM. A mixed type II/III will occur under mildly anaerobic (i.e. dysaerobic) conditions. A dysaerobic environment reflects the condition where the oxic/anoxic interface lies close to the sedimentary surface, providing a variable and imperfect condition for organic preservation (Thomas et al. 1985).

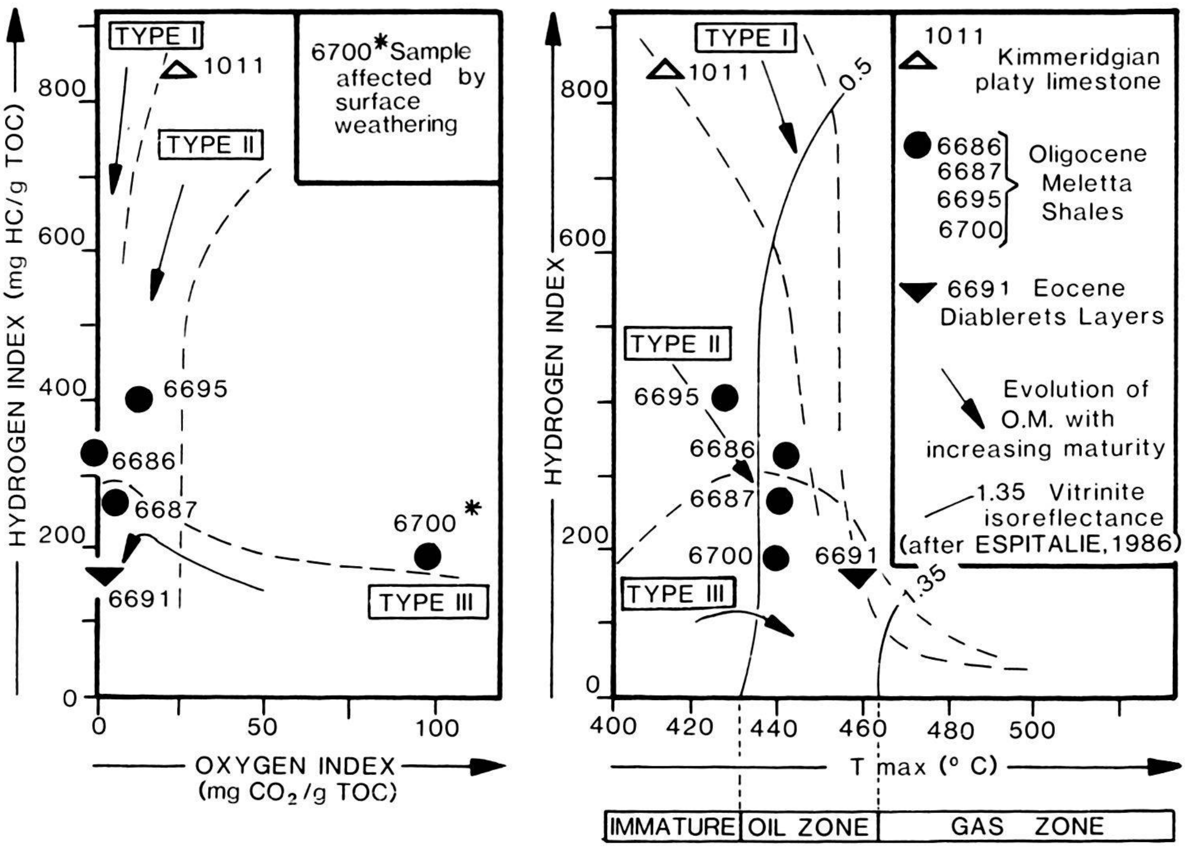


Fig. 4. Rock-Eval pyrolysis of selected samples: classification of organic matter in HI-OI and HI-T_{max} diagrams.

Geochemical studies

Geochemical investigations were performed on biological markers (organic compounds with the same skeleton as their natural precursors) using gas chromatography (GC) and mass spectrometry (MS) techniques. Biological markers such as isoprenoid, triterpenoid and steroid hydrocarbons show different distributions in the sediments according to the depositional environment and the physico-chemical reactions during and after burial. GC-MS techniques need only very small amounts of OM and can be readily applied to samples with less than 0,25% TOC. Consequently, in order to get a better picture of the Kimmeridgian platy limestones, the five samples 1007 to 1011 were analysed, although only sample 1011 showed a significant content in OM (Table 1).

The study of sterane distribution is one of the best methods for measuring thermal stress (Seifert 1980, McKenzie et al. 1980, 1982). Isomerisation of the biologically-derived 20R-5α(H),14α(H),17α(H)-steranes at C-20 and transformation of the (20R+20S)-5α(H),14α(H),17α(H)-steranes to the more stable (20R+20S)-5α(H),14β(H),17β(H)-steranes isomers are two reactions depending on maturity. Moreover, the isomerisation of the 17α(H),21β(H)-hopanes at C-22 in the range C_{31} to C_{35}, for which equilibrium is reached more rapidly and is completed before oil generation (22S/22R > 1) (MacKenzie et al. 1982), is also a maturity parameter (Seifert &

MOLDOVAN 1980), as well as the "Carbon Preference Index" (CPI) (TISSOT & WELTE 1984).

Aromatisation of ring-C monoaromatic steroid hydrocarbons (MA) to triaromatic steroid hydrocarbons is more temperature-dependent than time-dependent as is the isomerisation of steranes (MACKENZIE & MACKENZIE 1983); thus, a comparison between aromatisation and isomerisation gives some information about the temperature reached during burial.

Parameters such as pristane/phytane ratio (POWELL & MCKIRDY 1973), measuring the oxicity of depositional environment, relative quantities of C_{27}-, C_{28}- and C_{29}-steranes (Fig. 5) showing the type of organic input (MACKENZIE et al. 1983) and diasteranes/steranes ratio which is determined by the acidity of the depositional environment (SEIFERT & MOLDOVAN 1978), are used as ecological parameters.

Experimental procedure for GC-MS analyses

After crushing, rock samples were hydrolysed at reflux in HCl/MeOH 6N. Residues were filtered and ultrasonically extracted with methanol and twice with methylene chloride. The liquid phase was extracted twice with methylene chloride. After solvent evaporation, the organic extracts were separated by column chromatography (Silica gel 60, 70–230 mesh, Merck). The first fraction (hydrocarbons), eluted with hexane, was analyzed by GC and GC-MS.

The GC analyses were performed on a Carlo Erba Fractovap 4160 instrument equipped with a FID detector and a 25 m × 0.3 cm i.d. OV-1 WCOT Pyrex capillary column made in our laboratory. Samples were injected in splitless mode in hexane. A typical oven temperature program was: 60 °C for 1 min, from 60 to 150 °C at 15 °C/min, from 150 to 280 °C at 3 °C/min and finally held at 280 °C until the elution was completed. The GC-MS analyses were performed on a VG 7070E mass-spectrometer coupled with a Dani 3800 GC and a VG 11/250 data system. The GC conditions for GC-MS analysis (on the same column) were: 60 °C for 1 min, from 60 to 150 °C at 30 °C/min, from 150 to 280 °C at 2.5 °C/min and finally held at 280 °C until the elution was completed. The fragmentograms were obtained by electron impact at 70 eV with the Selected Ion Recording (SIR) mode at a resolution of approximately 3500.

Results

Kimmeridgian platy limestones in the southern Jura

The Kimmeridgian carbonate platform in the southern Jura has been well studied: in the established model (BERNIER & ENAY 1972, BERNIER & COURTINAT 1979, BERNIER 1984), the Kimmeridgian platy limestones were deposited in a lagoonal environment protected to the east and south by a reefal barrier (interpreted from the patch reefs found in massive limestones, Fig. 6). The organic microplankton of these finely laminated, locally bituminous limestones has been originally described by DEFLANDRE (1941). More recently BERNIER & Courtinat (1979) have described the organic facies of these sediments in the light of the palaeogeographical model mentioned above. Their study points to the development of shallow, confined "cuvettes", where stratification of

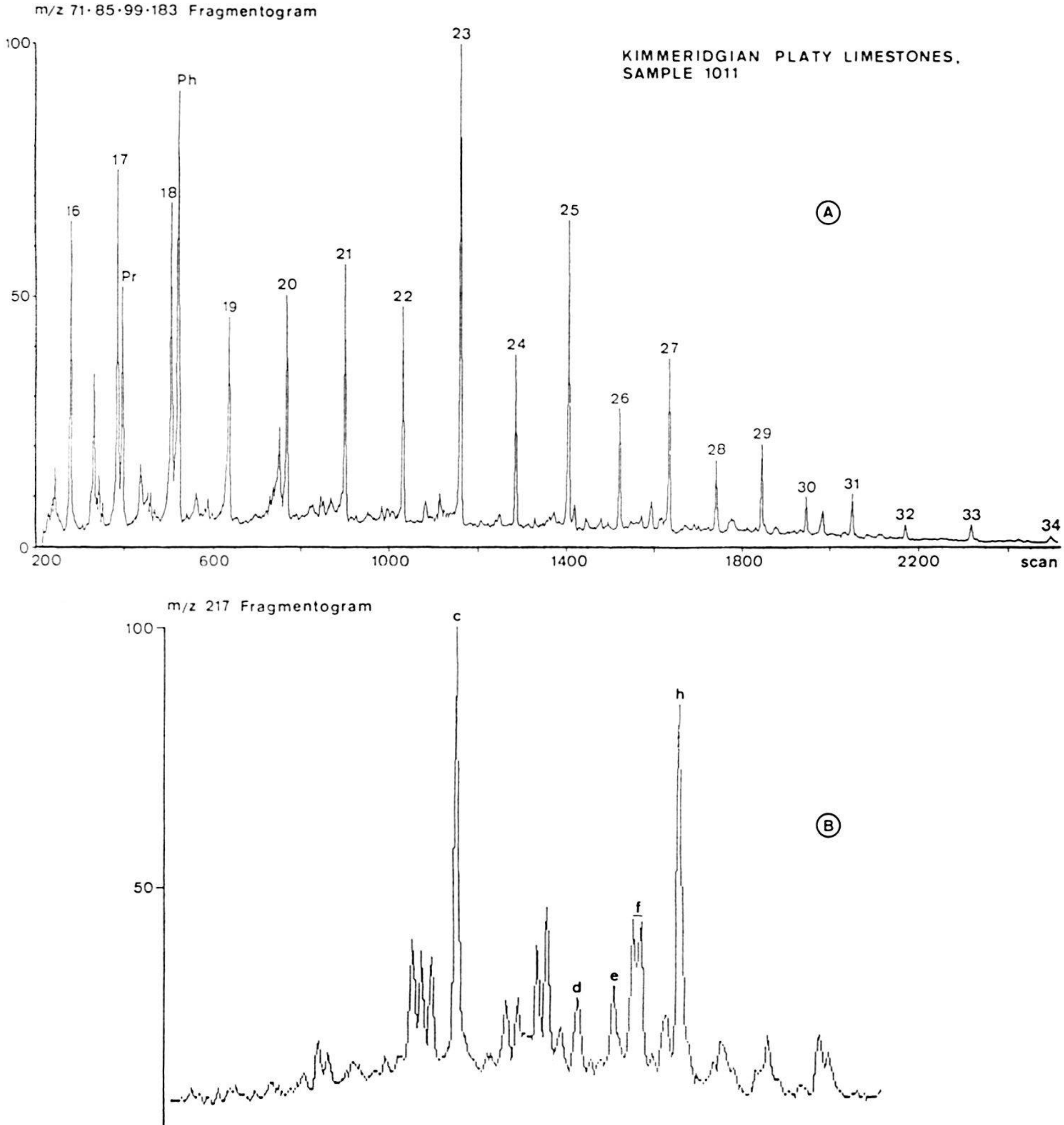


Fig. 5. Sample 1011, Kimmeridgian platy limestones (southern Jura):
(A) Mass fragmentogram of m/z 57+71+85+183 for n-alcanes and isoprenoids distribution (normal scan mode). Pr=pristane, Ph=phytane.
(B) Partial mass fragmentogram of m/z 217 for sterane distribution (SIR mode).

List of compounds:
a 20S-13β(H),17α(H)-diacholestane (C$_{27}$)
b 20R-13β(H),17α(H)-diacholestane ($_{27}$)
c 20R-5α(H),14α(H),17α(H)-cholestane (C$_{27}$)
d 20R-5α(H),14α(H),17α(H)-methylcholestane (C$_{28}$)
e 20S-5α(H),14α(H),17α(H)-ethylcholestane (C$_{29}$)
f 20R-5β(H),14α(H),17α(H)-ethylcholestane (C$_{29}$)
g 20S+20R-5α(H),14β(H),17β(H)-ethylcholestane (C$_{29}$)
h 20R-5α(H),14α(H),17α(H)-ethylcholestane (C$_{29}$).

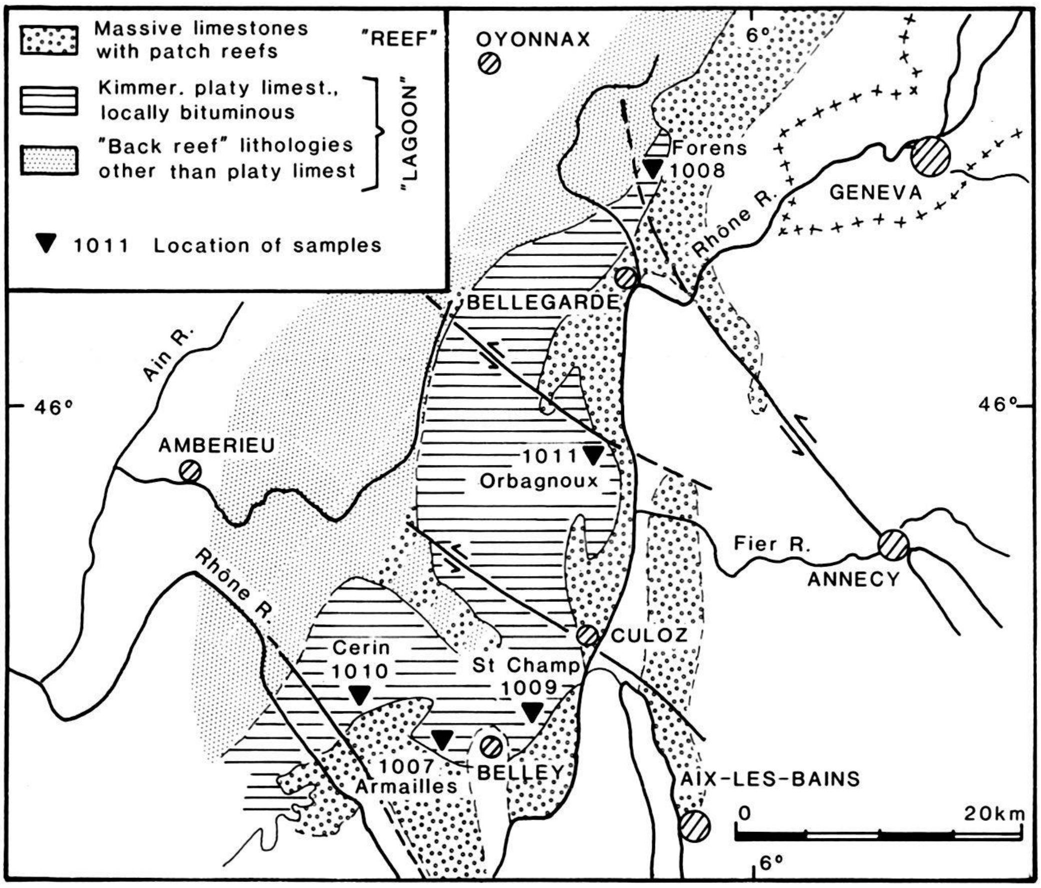


Fig. 6. Late Kimmeridgian palaeogeographic framework of southern Jura (modified from Bernier & Courtinat 1979) and location of selected "Kimmeridgian platy limestones" samples (see Table 2 for coordinates).

the marine waters permitted the development of anoxic conditions. The alternation of lighter and darker laminae in the limestones indicates that this anoxia was not permanent but rather episodic (Bernier 1984). Amount and nature of OM may vary considerably from one "cuvette" to the other (Bernier & Courtinat 1979) and a detailed study of each outcrop would be necessary to precisely describe the organic facies of the platy limestones. Therefore, the five samples examined in our study can in no way be representative of the whole rock unit. Each sample provides some extra data that should be looked at in the light of the existing environmental model.

The five samples were collected in different "cuvettes" in the back of the reefal barrier at Forens, Orbagnoux, St. Champ, Lac d'Armailles and Cerin (Fig. 6, Tables 1 and 2). Although the five of them were lithologically very similar (grey-brown, finely laminated limestones), only that collected at Orbagnoux (sample 1011) has a significant TOC and can be considered as an excellent oil source rock (Table 2). Out of the five, it is the only one that can be qualified as "bituminous". Its high hydrogen index indicates a type II (to I) OM (Fig. 4), i.e. a lipid-rich kerogen made of algal-planktonic OM deposited in an anaerobic environment where bacterial reworking is important. This is confirmed by the palynofacies study (Plate 1, Figs. 1–3): some 70% of the OM is amorphous (OM "grumeleuse" of Bernier & Courtinat 1979), the rest being mainly or-

Sample no.	Location	Coordinates (Lambert proj., zone 2)	TOC (% weight)	ROCK-EVAL RESULTS Tmax (°C)	Hydrogen I. (mg HC/g TOC)	Oxygen I. (mg CO_2/g TOC)
SOUTHERN JURA						
KIMMERIDGIAN PLATY LIMESTONES						
1007	Armailles	856.93/2090.62	<0.25			
1008	Forens	871.40/2139.90	<0.25			
1009	St. Champ	865.05/2093.67	<0.25			
1010	Cerin	850.29/2091.68	<0.25			
1011	Orbagnoux	866.91/2118.55	2.09	415	853	29
BORNES MASSIF						
EOCENE DIABLERETS LAYERS						
6691	Cenise	916.20/2119.40	7.42	460	169	0
OLIGOCENE MELETTA SHALES						
6686	Dessy	912.50/2124.84	1.27	442	323	0
6687	"	" "	1.18	440	254	6
6695	Nanoir	899.91/2109.60	1.32	428	400	15
6700	Pré-Vernay	897.50/2108.15	0.83	440	191	97

Table 2. Rock-Eval pyrolysis results of selected samples.

ganic microplankton of algal origin (leiospheres and dinoflagellates). Debris of continental origin (e.g. vitrinite) are rare. In UV light the AOM appears brightly fluorescent and made of fragments that probably result from bacterial reworking of microplankton (leiospheres, dinoflagellates). This facies is interpreted as an indication of possible "algal blooms" forming in the low energy facies of the Kimmeridgian lagoon, similar to those observed nowadays on carbonate platforms in the Bahamas and the Persian Gulf. This sample is thermally immature, as shown by Rock-Eval pyrolysis (Fig. 4) and by palynomorph coloration and fluorescence (Table 3).

Geochemical GC-MS analyses show that the five samples have a high predominance of phytane over pristane (Fig. 5 and Table 4) that reflects a strongly anoxic depositional environment (Didyk et al. 1978) typical of carbonate rocks. Moreover, absence or very low concentration of diasteranes (Table 4, % Dia) tend to confirm the purely carbonate (non acidic) character of these samples. On a triangular diagram (Fig. 7), Kimmeridgian platy limestones fall in an area which implies rather near-continental conditions than open marine ones. The steranes distribution shows the predominance of the biologically-derived 20R-α(H),14α(H),17α(H)-steranes, which is typical of immature sediments. This observation is confirmed by the maturity parameters given in Table 3. Sample 1009 is slightly more mature than the others and has a CPI lower than 1 (predominance of nC_{22}). If the original OM in sample 1009 had the same composition as that in the other samples, these parameters may reflect, together with a low pristane to phytane ratio, a hypersaline environment (Jiamo et al. 1986, Tissot & Welte 1984). This fits well within the model of confined, relatively shallow and warm water masses where salinities are likely to be above average (Bernier & Courtinat 1979, Bernier 1984).

Therefore, although their content in TOC varies greatly, the five samples give indications of anoxic conditions developing in a purely carbonate, near-shore marine envi-

Samples No.	CPI[1]	Pr/Ph[2]	% 20S[3]	% $\beta\beta$[4]	% Tri[5]	% Dia[6]	% 22S[7]
SOUTHERN JURA							
KIMMERIDGIAN, PLATY LIMESTONES							
1007	1.09	0.35	34.20	50.28	0.00	0.00	56.70
1008	1.19	0.48	14.33	43.07	0.72	3.52	39.16
1009	0.77	0.25	43.26	51.06	11.94	0.00	57.31
1010	1.34	0.62	32.99	54.86	2.63	4.52	50.86
1011	1.26	0.76	20.80	53.06	3.42	0.00	27.43
BORNES MASSIF							
EOCENE, DIABLERET LAYERS							
6691	1.05	1.52	51.64	66.41	100.00	46.57	64.16
OLIGOCENE, *MELETTA* SHALES							
6686	1.05	2.07	41.75	68.51	88.27	53.47	65.30
6687	1.08	2.14	59.74	74.55	76.03	68.76	61.09
6695	1.25	1.67	8.22	nd[8]	67.55	12.75	42.80
6700	1.22	0.49	8.28	26.77	84.41	19.47	48.45

[1] Carbon Preference Index measured as :
$[2\cdot(nC_{23}+nC_{25}+nC_{27}+nC_{29})+nC_{21}+nC_{31}]/[2\cdot(nC_{22}+nC_{24}+nC_{26}+nC_{28}+nC_{30})]$

[2] Pristane/Phytane

[3] % (20S)-/(20S+20R)-5α(H),14α(H),17α(H)-24-ethylcholestane

[4] %(20S+20R)-5α(H),14β(H),17β(H)- /(20S+20R)-5α(H),14β(H),17β(H)- and (20S +20R)-5α(H),14α(H),17α(H)-24-ethylcholestane

[5] % C_{28}-triaromatics / (C_{28}-triaromatics + C_{29}-5α-monoaromatics ring-C)

[6] % 20R-13β(H),17α(H)-diacholestanes / 20R-13β(H),17α(H)-diacholestane and 20R-5α(H),14α(H),17α(H)-cholestane

[7] % (22S)- / (22S+22R)-17α(H),21β(H)-bishomohopane

[8] Not determined because of coelution with other products

Table 3. Summary of maturity indicators of selected samples.

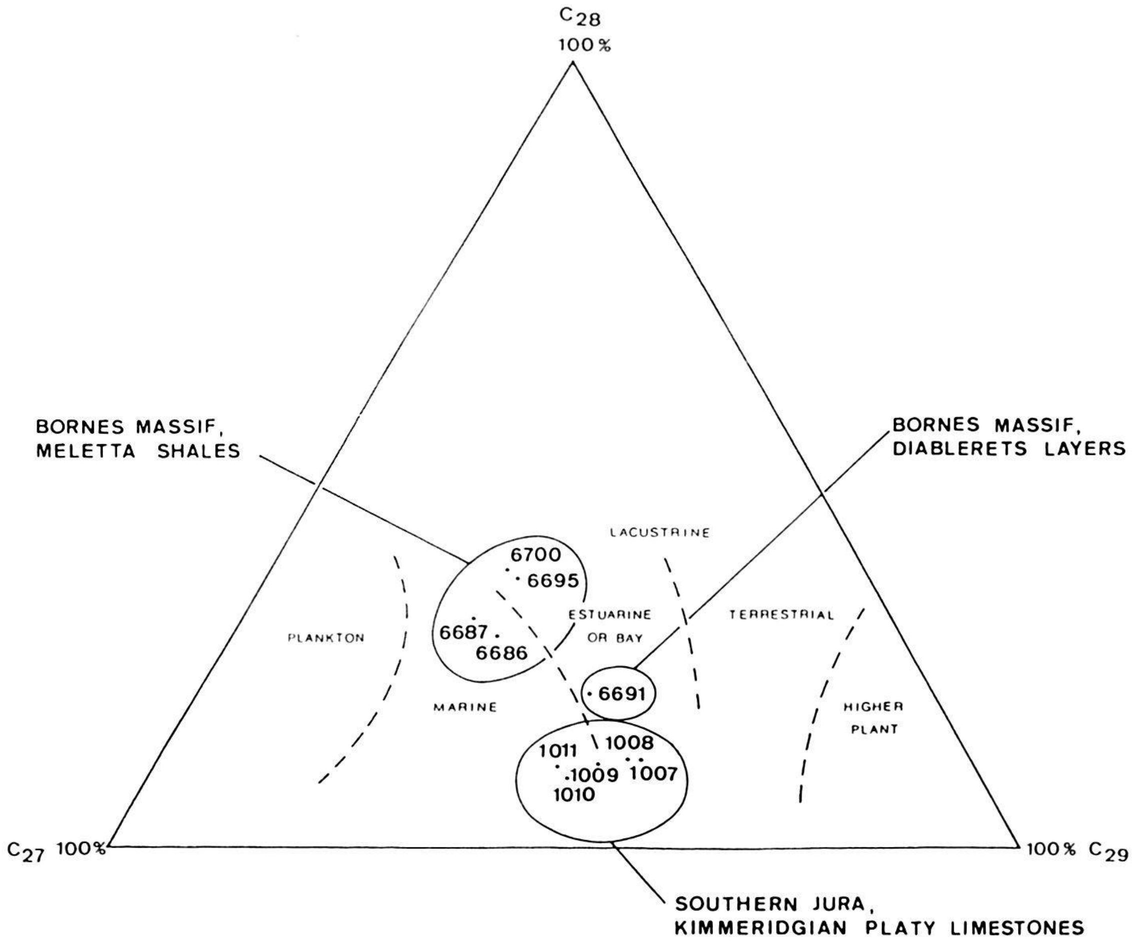


Fig. 7. Triangular diagram of relative abundances of 20R-5α(H), 14α(H), 17α(H)-steranes (C_{27}–C_{29}) in Kimmeridgian and Lower Tertiary samples.

ronment. All geochemical parameters show the platy limestones to be immature (Table 3). These results are in good agreement with the palynofacies of sample 1011 at Orbagnoux. All observations concur with the palaeoenvironmental model summarised on Fig. 6.

Bornes subalpine Massif

Rock-Eval pyrolysis analyses performed on Cretaceous and Lower Tertiary rocks of this Massif (Gorin 1988) have revealed a significant amount of OM in two Tertiary rock units:

- the Upper Eocene Diablerets Layers,
- the Lower-Middle Oligocene Meletta Shales.

Detailed stratigraphic and lithological information on these rock units can be found in Charollais (1963), Charollais et al. (1979) and Charollais et al. (1988).

Upper Eocene Diablerets Layers

This unit unconformably overlies Middle Cretaceous sediments (Fig. 2). It is not continuous in the subalpine massifs, but is confined to narrow grabens that were progressively invaded by the Late Eocene sea. It is up to 50 metres thick and consists of sandy to argillaceous, locally coaly, limestones containing fossils of oysters, polyps, calcareous algae, bryozoans and numerous foraminifers. The environment of deposition is interpreted as confined marine to brackish.

Centimetre-thick coaly layers extend over a few hundred metres to kilometres, some of them having been exploited in the 18th century. Sample 6691 comes from such a coaly layer in the Cenise syncline (Fig. 8 and Table 2). Measurements of vitrinite reflectance carried out on this layer have shown R_0 values of 0.8 to 0.85% (Kübler et al. 1979).

The high T_{max} obtained by Rock-Eval pyrolysis indicates a level of maturity in the upper end of the oil zone (Fig. 4). Because the hydrogen index decreases with increasing maturity, this prevents typing of the OM in a Rock-Eval diagram (Fig. 4). Microscopic observations of palynological slides indicate a thermal alteration index (TAI) of 3–3.5. A large part of the organic residue consists of thermally more resistant humic material, but ghosts of originally hydrogen-rich components can be observed: dinoflagellates (Plate 1, Figs. 4 and 5), spores, bisaccate pollens, cutinite debris and amorphous organic matter (AOM), which display dark brown to black colours. The proportion of AOM is relatively low, but may have been originally higher, because AOM becomes rapidly thermally degraded to very fine particles that are lost while filtering the palynological residue. The palynofacies of sample 6691 indicates the original kerogen was probably of mixed type II/III and deposited in a dysaerobic marine environment with strong continental influences.

On a triangular diagram (Fig. 7) sample 6691 falls in an area which implies near-continental marine conditions. The pristane to phytane ratio (increasing with maturity, Fig. 9) represents an environment with reducing conditions (Didyk et al. 1978). Geochemical parameters show this sample to be very mature, i.e. the maximum of oil generation seems to be passed since all maturity parameters have reached their maximum value (Table 4).

Therefore, these observations indicate that the coal beds in the Diablerets Layers were deposited in marine waters close to vegetated land. Environmental parameters point to reducing dysaerobic conditions. This fits well with the model of narrow grabens where confined marine to brackish conditions prevailed. Maturity reached by the coal is higher than that measured by Kübler et al. 1979 (Table 3). A possible explanation for this high level of thermal maturity is given below, when discussing the Meletta Shales. The coaly layers may be of average source rock quality.

Lower-Middle Oligocene Meletta Shales

Meletta Shales form the base of flysch-like sedimentation in the Bornes Massif to which they seem to be restricted. Their maximum thickness is 30 metres. They differ from the underlying Lower Oligocene "Foraminifers Marls" by their richness in fish scales and other fish debris, by the disappearance of benthic and large planktonic foraminifers and by the predominance of small planktonic foraminifers. This is interpreted as an evolution of the basin towards confinement (Charollais et al. 1979).

Four samples were analysed, two in the eastern part of the Massif near Bonneville and the other two in the west, near Annecy (Fig. 8). Rock-Eval pyrolysis (Table 2)

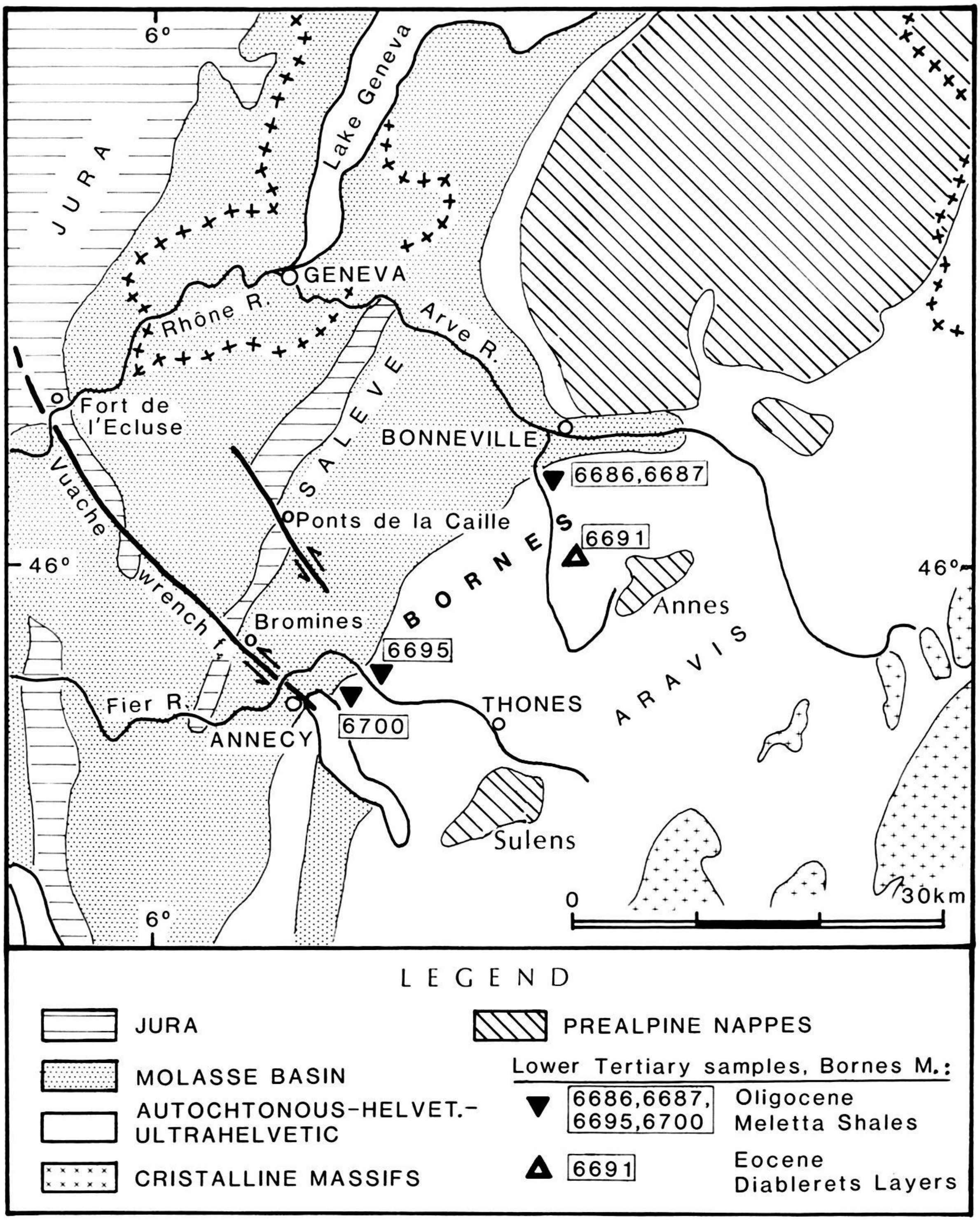


Fig. 8. Location of selected Lower Tertiary samples in the Bornes Massif (see Table 2 for coordinates). The higher marurity (see Table 4) of samples 6686, 6687 and 6691 to the east may reflect the overburden of the now-eroded prealpine nappes. Immature samples 6695 and 6700 to the west are located in the vicinity of a regional wrench fault zone, the reactivation of which may have resulted in short episodes of high heat flow affecting some geochemical parameters of the samples.

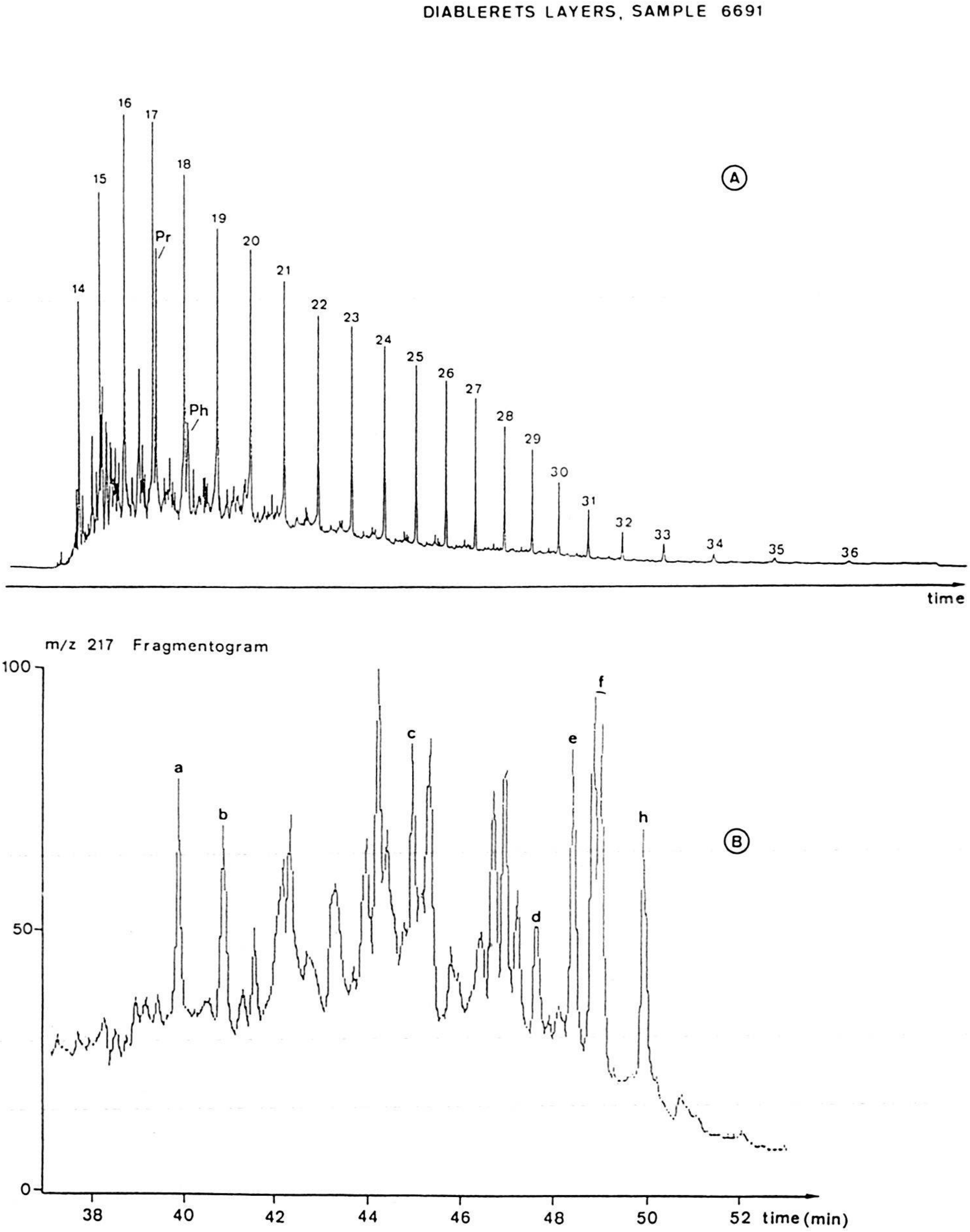


Fig. 9. Sample 6691, Eocene Diablerets Layers (Bornes Massif):
(A) Gas chromatogram of hydrocarbon fraction, Pr = pristane, Ph = phytane.
(B) Partial mass fragmentogram of m/z 217 for steranes distribution (SIR mode). Compounds *a* to *h*, see caption Fig. 5.

Sample no.	Rock-Eval Tmax (ESPITALIE 1986)	Thermal alteration index (STAPLIN 1969)	Exinite Fluorescence (ROBERT 1985)	CPI (TISSOT & WELTE 1984)	Sterane distribution (SEIFERT 1980)	Vitrinite reflectance (% R) (KÜBLER et al. 1979)
SOUTHERN JURA						
KIMMERIDGIAN PLATY LIMESTONES						
1007	n/a	n/a	n/a	immature	immature	n/a
1008	"	"	"	"	"	"
1010	"	"	"	"	"	"
1009	"	"	"	immature-mature	"	"
1011	immature	2 (immature)	bright yellow (immature)	immature	"	"
BORNES MASSIF						
EOCENE DIABLERETS LAYERS						
6691	oil zone	3-3.5 (oil-gas zone)	no Fluor (oil-gas zone)	oil zone	oil zone	0.8-0.85
OLIGOCENE MELETTA SHALES						
6686	oil zone	2.5-3 (oil zone)	yellow-orange (oil zone)	oil zone	oil zone	ca. 0.6
6687	" "	2.5-3 (oil zone)	" "	" "	" "	" "
6695	immature	2 (immature)	bright yellow (immature)	immature	immature	n/a
6700	(oil zone)*	n/a	n/a	"	"	"

* = sample affected by surface weathering
n/a = data not available

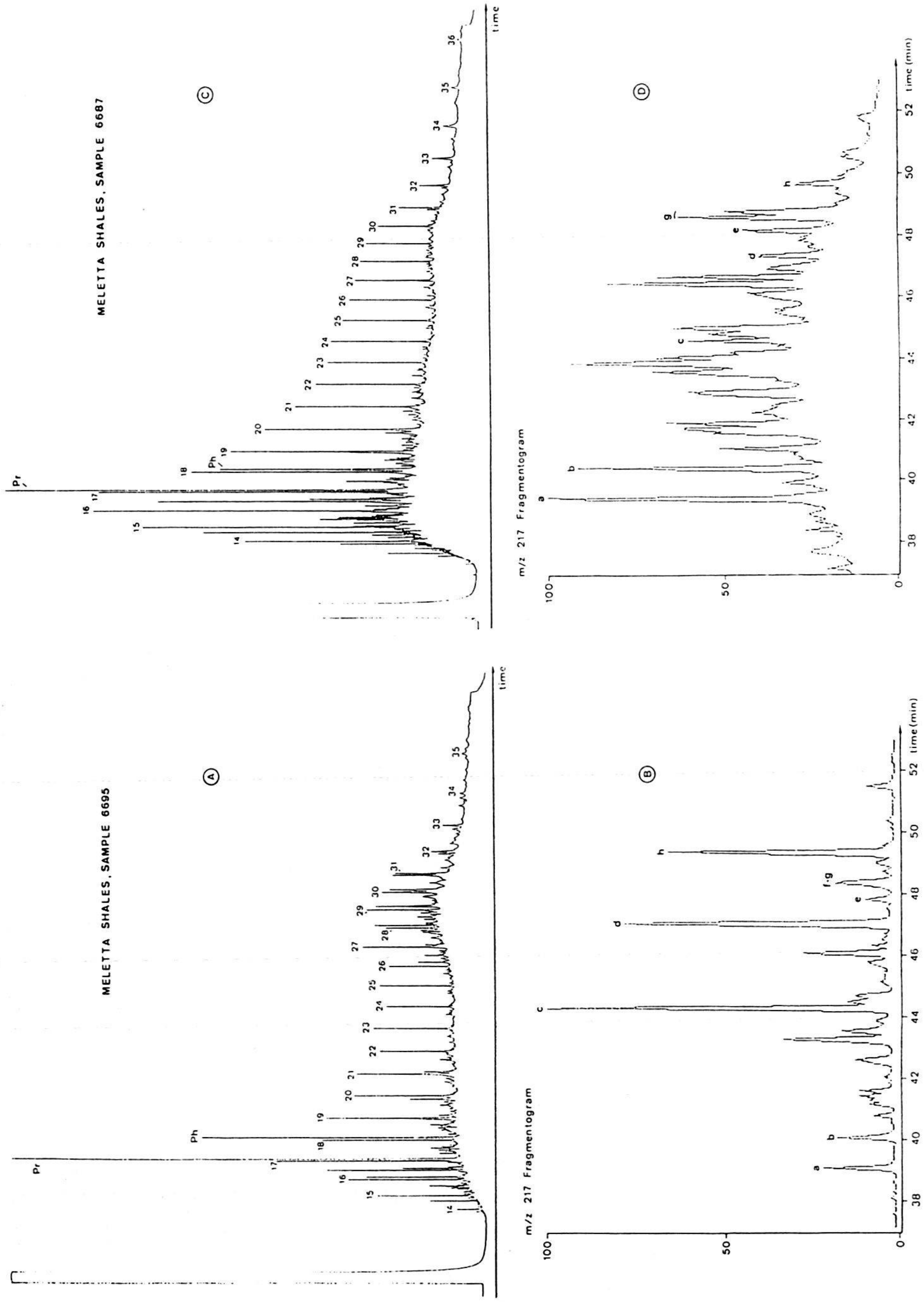
Table 4. Geochemical parameters of selected samples.

shows a TOC content above 1% (except sample 6700) and the OM to be of mixed type II/III (Fig. 4). The high oxygen index of sample 6700 is interpreted as en effect of surface weathering (Espitalie et al. 1985–1986) and Rock-Eval parameters (hydrogen index and T_{max}) are not reliable.

Palynofacies (Plate 1, Figs. 6 to 9) confirm the mixed type of the OM: it is mainly amorphous (50–90% AOM), together with varying amounts of palynomorphs (dinoflagellates, pollens and spores described in Charollais et al. 1979) and marine algae like *Pterospermella* sp. This marine plankton seems more abundant in the east (20–30%) than in the west. Vitrinite and inertinite fragments do not exceed 10%. Under UV light, some 20% of the AOM fragments fluoresce, indicating hydrogen-rich OM. The largest part of the AOM consists of cellulosic-lignitic terrestrial material degraded by bacteria and fungi (Hart 1986). Sporomorph colour, preservation and fluorescence show that sample 6695 (Plate 1, Figs. 6 and 7) is thermally immature, but that samples 6686 and 6687 at Dessy are in the oil-generating zone (Plate 1, Figs. 8 and 9).

In GC-MS analyses, samples 6695 and 6700 show a n-alcanes distribution (CPI = 1.25 and 1.22) typical of non-mature sediments while the two others, 6686 and 6687 (Fig. 10) have reached maturity (CPI≈1) and present a relative increase of the n-alcanes concentration. Pristane to phytane ratios show some differences; sample 6700 was deposited in a totally anoxic environment, while the value for sample 6695 represents anoxic to oxic fluctuations; for the two mature samples, higher values of this ratio is due rather to maturation than to depositional environment.

The sterane triangular diagram (Fig. 7) suggests that samples 6686 and 6687 at Dessy are more marine than the other two. This is in agreement with the increase in


MELETTA SHALES, SAMPLE 6695
MELETTA SHALES, SAMPLE 6687
m/z 217 Fragmentogram
m/z 217 Fragmentogram
time
time (min)

marine plankton from west to east (see above). The sterane distribution confirms that samples 6686 and 6687 have reached values which permit oil generation. It is interesting to note that the aromatisation is about the same for these four samples; this may be interpreted as an indication that the maximum temperature reached by the two immature samples in the west may have been considerably greater than that reached by the two mature ones in the east, but only during a very short time.

Therefore, palynofacies and organic geochemistry demonstrate that Meletta Shales were deposited in a mainly dysaerobic marine environment in agreement with the confined nature of the basin. It is suggested that more open marine conditions may have existed in the eastern part of the basin. Meletta Shales are of average source rock quality and have reached maturity in the eastern part of the Bornes Massif, where they probably sourced the oil impregnating sandstones of the subalpine flysch in the Dessy area.

Environment and thermal maturity of the Meletta Shales

Charollais et al. 1979 point out that it is not yet possible to conceive a precise palaeogeographical model of the Helvetic domain in the subalpine massifs during the Early-Middle Oligocene. Nevertheless field observations, micropalaeontology, organic facies and geochemistry confirmed the limited and confined nature of the Meletta Shales. Tectonic studies (Bergerat 1985, Pfiffner 1986) suggest that extensional faulting affected the alpine foreland at that time. Meletta Shales were deposited in the elongated basin fringing the alpine front (Fig. 11 and Caron et al. 1989). To our knowledge they represent the only occurrence of mildly anoxic conditions described so far in Oligocene flysch-like sediments of the Delphino-Helvetic realm in France and Switzerland. Transition from the underlying "Foraminifers Marls" seems gradational. The reason why this part of the basin evolved towards confinement remains an open question: progressive tectonically or eustatically induced isolation of a relatively shallow mass of water, local deepening through extensional faulting (e.g. as in a half-graben), etc.?

The higher thermal maturity of samples in the eastern part of the Massif (Fig. 8, samples 6686, 6687 and coal sample 6691 of the Diablerets Layers) is in agreement with the regional geothermal gradient observed by other authors in the Bornes-Aravis area (Martini & Vuagnat 1965, Kübler et al. 1974, Sawatzki 1975, Deconinck & Charollais 1986). This eastward increase in maturity may reflect the overburden of the prealpine nappes that once covered the whole area (as evidenced by the Annes and Sulens klippes, Fig. 8).

Fig. 10. Oligocene Meletta Shales (Bornes Massif):
(A) Sample 6695: Gas chromatogram of hydrocarbon fraction.
(B) Sample 6695: Partial mass fragmentogram of m/z 217 for steranes distribution (SIR mode).
(C) Sample 6687: Gas chromatogram of hydrocarbons fraction.
(D) Sample 6687: Partial mass fragmentogram of m/z 217 for sterane distribution (SIR mode).
In (A) and (C): Pr = pristane, P = phytane.
In (B) and (D): Compounds *a* to *h*, see caption Fig. 5.

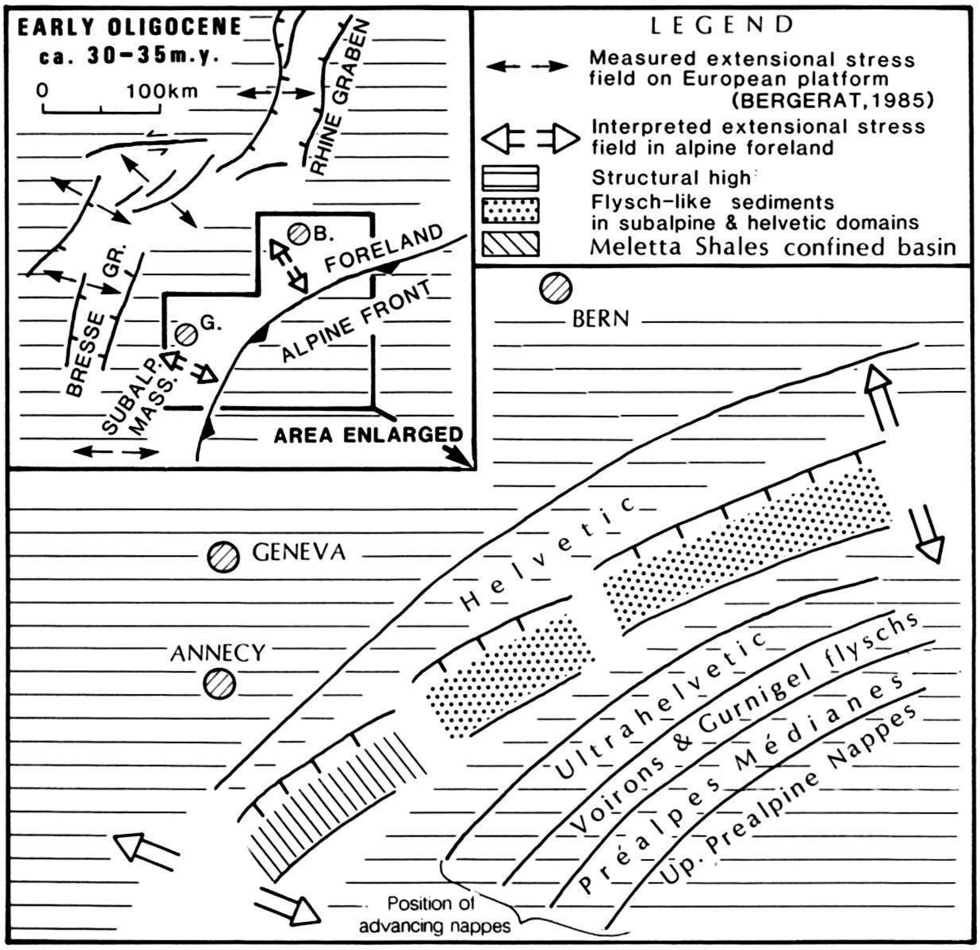


Fig. 11. Palaeogeographical sketch at Early Oligocene times showing the possible structural setting of the confined Meletta Shales sedimentation (modified from CARON et al. 1989).

Samples 6695 and 6700 in the west (Fig. 8) are immature, but geochemical evidence suggests they may have been exposed, for a very short time, to a much higher temperature than samples in the east. Both immature samples are located on the trend of a major sinistral wrench fault zone affecting the Mt. Salève and the Mt. Vuache (Fig. 8). Recent seismicity associated with these faults is well documented (AMATO 1983, CHAROLLAIS et al. 1983 and AMATO et al. 1987). Sudden increase in heat flow associated with sulfurous vapors was reported along the Vuache wrench fault at Fort de l'Ecluse (Fig. 8) during an 1840 earthquake (PERREY 1845). Sulfurous warm springs are known today near Bromines and Ponts de la Caille (Fig. 8). These observations support the possibility of having sudden short-lived increases of local heat flow associated with movements of deep-seated basement faults in this area. Such thermal events may affect surrounding sediments and could be an explanation of the geochemical signature observed on samples 6695 and 6700. Nevertheless, further detailed studies would be needed to confirm such an hypothesis.

Conclusions

Palynofacies, Rock-Eval pyrolysis and GC-MS geochemical studies of three organic-rich intervals in the southern Jura and the Bornes Massif lead to the following conclusions:

1. Kimmeridgian platy limestones, Southern Jura

a. They were formed in an anoxic, carbonate, restricted marine environment. Their content in total organic carbon (TOC) is highly variable depending on the location. In the "bituminous" limestones at Orbagnoux, the organic matter (OM) is of type II (to I) and is essentially amorphous (AOM). This AOM results from bacterial reworking of marine phytoplankton (leiospheres and dinoflagellates).
b. All observations fit with the existing depositional model of a partly confined lagoon fringed to the east and south by a succession of patch reefs (Bernier 1984).
c. Kimmeridgian platy limestones are locally (e.g. at Orbagnoux) an excellent immature oil-prone source rock.

2. Coaly beds in Upper Eocene Diablerets Layers, Bornes Massif

a. They were formed in mildly anoxic restricted marine to brackish conditions. Prior to thermal degradation the OM was probably of mixed type II/III with a strong continental influence.
b. Field observations (Charollais et al. 1979 and 1988) showed that they were deposited in narrow grabens, where dysaerobic conditions could ideally develop, the vegetated areas bordering the grabens providing abundant terrestrial OM.
c. Their high level of thermal maturity (TAI = 3–3.5) is interpreted as the effect of overburden associated with the now-eroded prealpine nappes. These coaly beds may be of average source rock quality.

3. Lower-Middle Oligocene Meletta Shales, Bornes Massif

a. They were formed in dysaerobic confined marine conditions. Their OM is of mixed type II/III with a large proportion of terrestrial material reworked by bacteria and fungi (AOM). Sterane distribution and richness in phytoplankton suggest conditions may have been more open marine to the east.
b. They are limited to the Bornes Massif (Charollais et al. 1979) and represent so far the only known occurrence of mild anoxia in Lower-Middle Oligocene flysch-like sediments deposited in the Delphino-Helvetic realm in France and Switzerland.
c. They represent a source rock of average quality, immature in the western part of the Massif, but mature in the east, where they probably sourced the oil locally impregnating flysch sandstones (grès du Val-d'Illiez). The higher maturity is explained (as for the Diablerets Layers) by the overburden of the now-eroded prealpine nappes. In the west, short-lived increases in heat flow associated with the activity of the Vuache wrench fault have left a geochemical signature in these sediments.

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Plate 1

Figs. 1, 2, 3. Palynofacies of Kimmeridgian platy limestones at Orbagnoux (southern Jura), samples 1011, slides 1011a (Fig. 1) and 1011b (Figs. 2 and 3). This sample is thermaly immature (TAI = 2).

a = *Leiosphaeridia* spp. (marine microplankton of probable algal origin)
b = cf. *?Pterocystidiopsis angulosa* DEFLANDRE (marine microplankton of probable algal origin)
c = proximate dinoflagellate cyst
d = vitrinite
e = amorphous organic matter (AOM). This AOM has a brigth fluorescence in UV light (i.e. it is hydrogen-rich) and consists mainly of phytoplankton debris reworked by bacteria.

Figs. 4, 5. Palynofacies of coals in Upper Eocene Diablererts Layers at Cenise (Bornes Massif), sample 6691, slide 6691a. The thermal maturity level is in the upper end of the oil zone (TAI = 3–3.5).

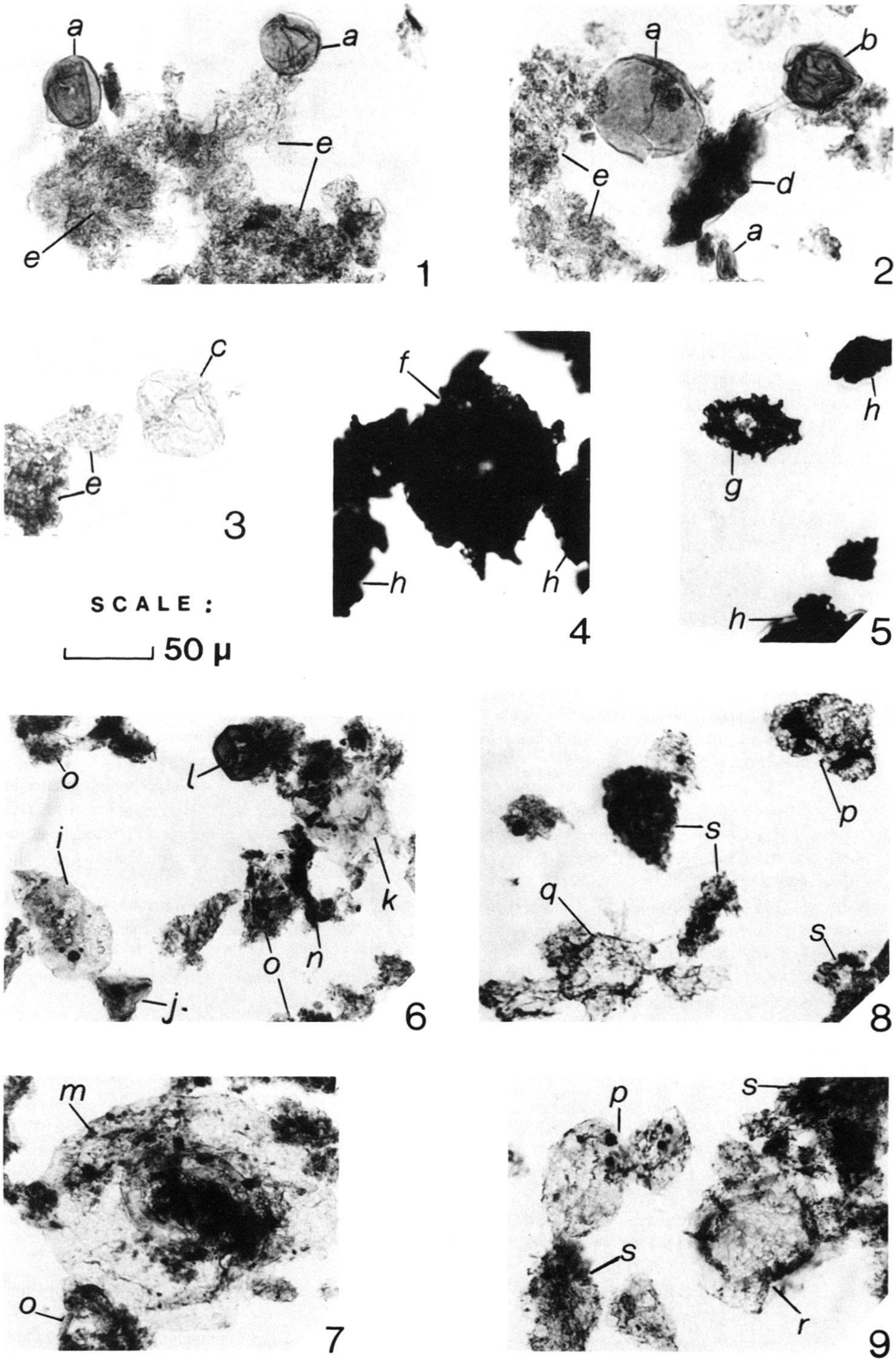
f = thermally altered, peridinoid dinoflagellate cyst, cf. *Deflandrea* sp.
g = thermally alterated dinoflagellate cyst
h = blackened organic matter, mainly vitrinite and inertinite.

Figs. 6, 7. Palynofacies of Lower-Middle Oligocene Meletta Shales at Nanoir (Bornes Massif), sample 6695, slide 6695a. This sample is thermally immature (TAI = 2–2.5).

i = gymnosperm pollen
j = spore
k = peridinoid dinoflagellate cyst, cf. *Deflandrea spinulosa* ALBERTI
l = fungal spore
m = *Pterospermella* sp., a marine alga
n = vitrinite
o = amorphous organic matter (AOM). In UV light, this AOM shows only a few fluorescent hydrogen-rich fragments and a high proportion of non fluorescent residual primary fragments of terrestrial origin.

Figs. 8, 9. Palynofacies of Lower-Middle Oligocene Meletta Shales at Dessy (Bornes Massif), sample 6687, slide 6687b. The thermal maturity level is in the oil zone (TAI = 2.5–3).

p = darkened, thermally altered gymnosperm pollens
q = darkened, thermally altered chorate dinoflagellate cyst
r = darkened, thermally altered alga (*Pterospermella* sp.)
s = amorphous organic matter (AOM), same characteristics as AOM on fig. 6 and 7.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