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Black pebbles of the Purbeckian (Swiss and French Jura): lithology, geochemistry and origin

By ANDRÉ STRASSER and ERIC DAVAUD[1])

ABSTRACT

Purbeckian black pebbles and associated features in the Swiss and French Jura Mountains are described and analyzed. They occur mostly in intertidal carbonate sediments, but also in subtidal and lacustrine sequences. Black pebbles consist of micritized subtidal, intertidal and supratidal carbonates. They are mostly reworked and often concentrated in storm deposits and beach conglomerates.

Their black colour is due to impregnation by organic matter (sometimes with the addition of pyrite) in a low Eh/high pH environment or microenvironment. Dissolved, colloidal or very finely particulate organic substances were fixed by adsorption on carbonate-crystal surfaces and by early vadose or phreatic freshwater cementation. The organic matter was derived from algae and higher terrestrial plants.

Due to their early cementation, blackened spots or levels were more resistant to erosion than their host sediment. Reworking by waves, tides and storms led to pebble formation.

Black pebbles can be useful indicators of terrestrial or associated environments.

ZUSAMMENFASSUNG

Aus dem Purbeck des schweizerischen und des französischen Juras werden schwarze Lithoklasten («black pebbles») beschrieben und analysiert. Sie treten vor allem in intertidalen, aber auch in sub- und supratidalen Karbonatsedimenten auf. Die geschwärzten Lithoklasten selbst bestehen aus mikritisierten subtidalen, intertidalen oder supratidalen Kalken. Meist sind sie aufgearbeitet und häufig in Sturmablagerungen und Strandkonglomeraten konzentriert.

Die schwarze Färbung entstand durch Imprägnierung mit organischem Material (manchmal zusätzlich noch mit Pyrit) in einem anoxischen und leicht alkalischen Milieu oder Mikromilieu. Gelöste, kolloidale oder sehr feinkörnige organische Substanzen wurden durch Adsorption an Karbonatkristallen und durch vadose oder phreatische Zementation gebunden. Das organische Material stammt von Algen und von höheren terrestrischen Pflanzen.

Durch ihre frühe Zementierung waren geschwärzte Zonen und Lagen erosionsresistenter als das umgebende Sediment. Aufarbeitung durch Wellen, Gezeiten und Stürme führte zur Bildung von Lithoklasten.

«Black pebbles» liefern nützliche Hinweise auf Landgebiete oder Landnähe.

RÉSUMÉ

Les «galets noirs» du Purbeckien du Jura suisse et français sont décrits et analysés. On les observe essentiellement dans des faciès carbonatés intertidaux mais également dans des séquences subtidales ou lacustres. Ils sont constitués de carbonates subtidaux à supratidaux micritisés. Ils sont la plupart du temps remaniés et souvent concentrés dans des dépôts de tempête ou des conglomérats de plage.

La couleur des galets noirs est due à une imprégnation de matière organique dans un milieu ou micromilieu caractérisé par un pH élevé et un Eh bas. La matière organique dérivait d'algues et de

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plantes terrestres supérieures. La fixation de substances organiques dissoutes, colloïdales ou particulaires s'est effectuée par adsorption à la surface des cristaux de carbonate et par cimentation précoce vadose ou phréatique.

La formation des galets noirs s'est opérée lors de l'érosion des côtes sous l'action des vagues, des marées et des tempêtes par remaniement des zones ou des niveaux noircis qui résistaient mieux à l'abrasion que les sédiments encaissants du fait d'une cimentation différentielle précoce.

Les galets noirs indiquent la proximité stratigraphique et/ou géographique de terres émergées.

1. Introduction

Stratigraphic and regional setting

Due to the scarcity of good paleontological markers, the biochronological position of the so-called "Purbeckian" in the Swiss and French Jura Mountains has not yet been established. Some authors place it in the uppermost Jurassic (e.g. JOUKOWSKY & FAVRE 1913, AUBERT 1943, PERSOZ & REMANE 1976), others in the lowermost Cretaceous (e.g. BENOÎT 1879, HÄFELI 1966, STEINHAUSER & CHAROLLAIS 1971). We therefore use "Purbeckian" strictly as a term describing a *facies association* and attribute it to the Jurassic-Cretaceous boundary (GIRARDOT 1885, DONZE 1958).

The Purbeckian sediments consist of *shallow marine, brackish and lacustrine* limestones. Frequent intercalations of marls lead to rather poor outcrops in mostly vegetated trough-shaped depressions.

A dolomite sequence often marks the passage from the Portlandian to the Purbeckian facies. Evaporite-bearing marls and algal mats indicate a supratidal environment while characeans are evidence of freshwater ponds. Sterile mudstones and the local abundance of monospecific faunas (miliolids, ostracods, gastropods) suggest restricted water circulation and salinities different from normal marine. Much of the Purbeckian, which usually displays fenestrae and desiccation polygons, was deposited in the *intertidal to supratidal* zones. Storm deposits and conglomerates are common. Oolites and grainstones of subtidal origin yield a normal shallow marine flora and fauna (dasycladaceans, echinoderms, large agglutinating foraminifera). The Purbeckian is capped by subtidal carbonates of Berriasian age.

The extent of Purbeckian outcrops in the Swiss and French Jura is indicated in Figure 1. Boreholes have proven that the facies continue under the Bresse and Molasse basins (DONZE 1958, FISCHER & LUTERBACHER 1963).

Three selected schematic sections (Fig. 2) demonstrate the oscillations from shallow subtidal to supratidal environments. The Mont-Salève section is dominated by limestone and contains several caliche horizons. The latter indicate subaerial exposure (STRASSER & DAVAUD 1982), probably on a barrier-island system separating the Purbeckian lagoonal environment to the WNW from the subalpine deepwater area to the ESE (DAVAUD et al. 1983). The section at Poizat shows a more internal facies development: dolomites are overlain by marls and often brecciated intertidal and supratidal sediments. Finally, at Cornaux, evaporite-bearing marls rich in plant material are dominant.

"Black pebbles" are angular to rounded, sand- to cobble-sized lithoclasts which are found in varying amounts in most of the studied sections (Fig. 1). Their most

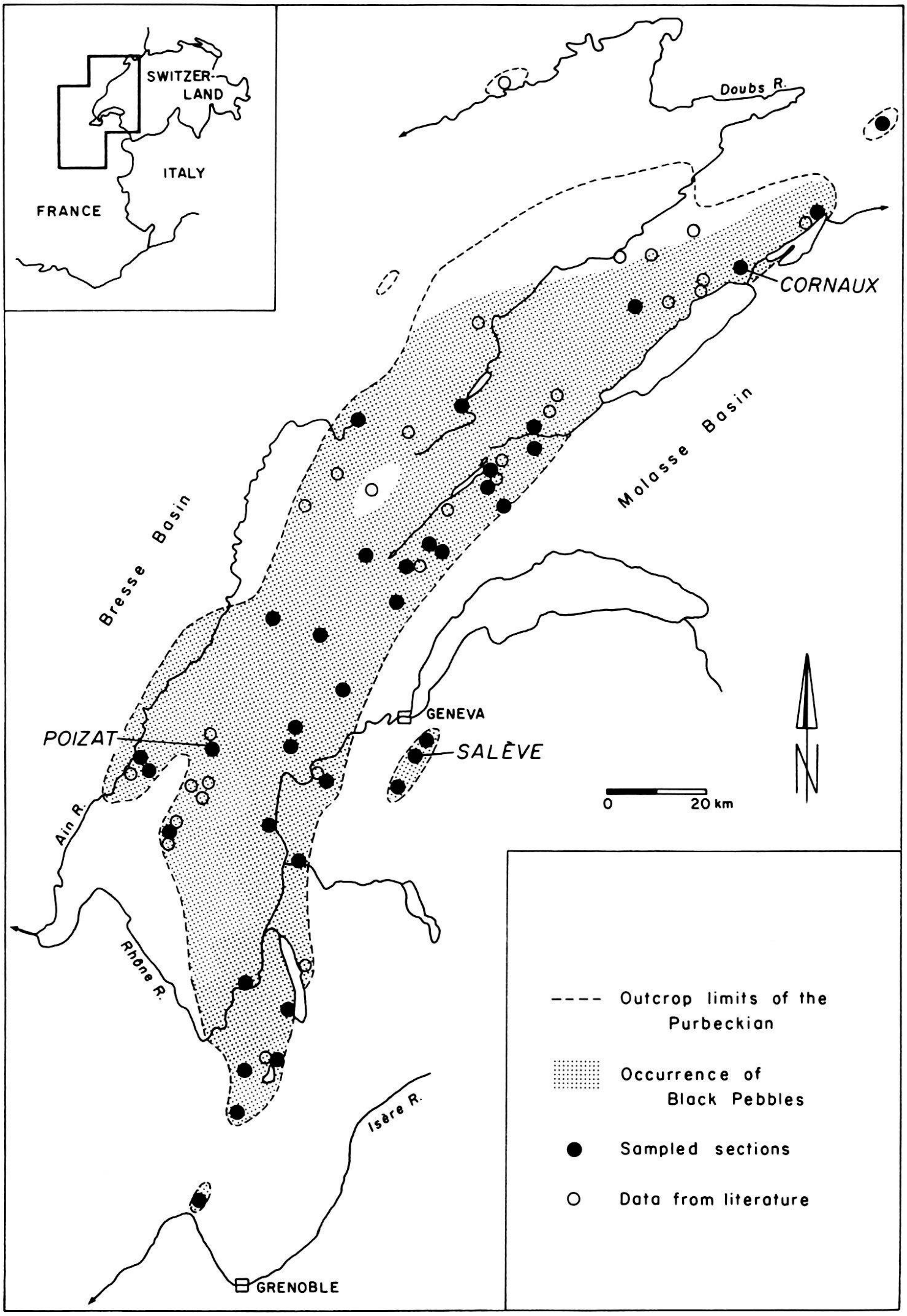

Fig. 1. Occurrence of black pebbles in the Purbeckian of the French and Swiss Jura. Literature data are from Ainardi (1973), Carozzi (1948), Donze (1958), Girardot (1885), Loriol (1954), Loriol & Jaccard (1865) and Maillard (1884).

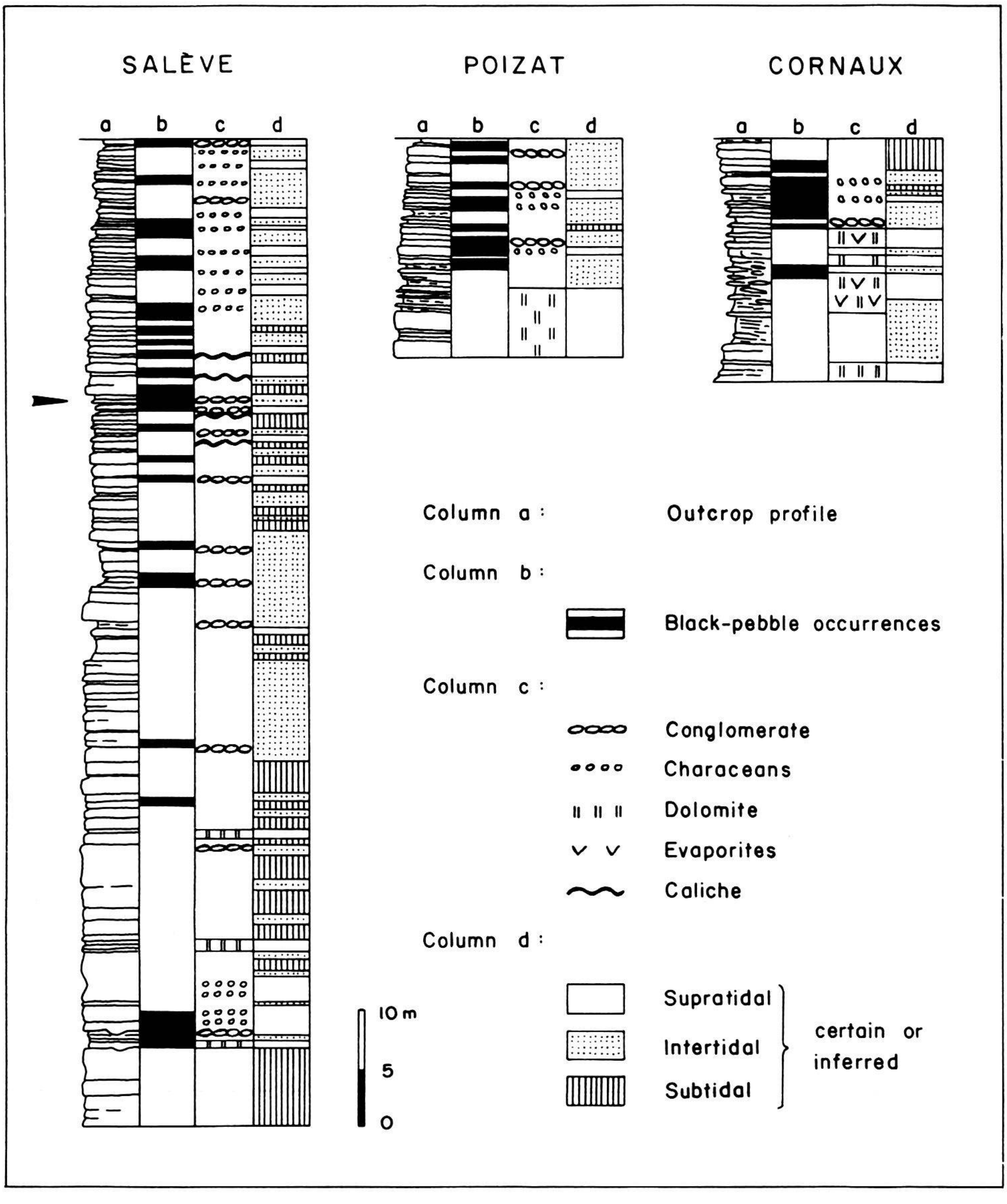

Fig. 2. Schematic sections showing vertical facies development (for location see Fig. 1). Arrow marks position of Figure 3.

striking appearance is in graded multicoloured breccias where they contrast with white, light brown and yellowish clasts. Black pebbles occur in subtidal, intertidal and supratidal, marine, brackish, freshwater and evaporitic sediments (Fig. 2). Almost every type of Purbeckian facies occurs as black pebbles. The black pebbles may or may not be of the same lithology as the host sediment.

The purpose of this paper is to analyze the composition, facies relationships, genesis and environmental significance of the black-pebble deposits.

History of research

Ever since the Purbeckian has been studied, its conspicuous black pebbles[2]) have intrigued geologists. There had always been a search for a "black-pebble source rock" but it could never really be found. The scientists tried to explain the preservation of the black pebbles and the "disappearance" of the source rock by differential oxidation, or by total erosion of the source material.

L. VON BUCH (1803) was the first to describe black pebbles. He found them in the Neuchâtel Jura and concluded that they had to be allochthonous. LORY (1857) supposed that the pebbles had derived from black Alpine limestones. As there was no evidence for extensive emersion of the Alpine area at the time of Purbeckian sedimentation, GILLIÉRON (1873) proposed temporary exposure and erosion of the Tithonian of the Fribourg Alps, which lie much closer to the Purbeckian outcrops. MAILLARD (1884) rejected this theory because it was proven that the Tithonian didn't emerge at that time. He suggested a northern origin: fluvial transport of Lower and Middle Jurassic clasts from the Vosges.

CHOFFAT (1877) was the first to see the origin of the black pebbles in the Purbeckian itself. He found black lacustrine limestones as well as freshwater fossils in the black pebbles. He also noted black rock fragments in Kimmeridgian, Portlandian and Berriasian formations.

According to GIRARDOT (1885), the black pebbles formed syngenetically by "agglomeration" in the mud, or they derived from the underlying beds. In 1913, JOUKOWSKY & FAVRE described the decomposition and erosion of oolite beds and the formation of a multicoloured breccia containing black oolite fragments. The black mudstone pebbles in the same breccia would have derived from nearby mudstone beds. The dark colour was explained by finely dispersed pyrite, and a layer of cement around the pebbles would have prevented their oxidation. The two authors also postulated that, because of complete oxidation, the source rocks of the black pebbles became indistinguishable from other rocks. FAVRE & RICHARD (1927) linked the presence of black pebbles to vertical oscillations of the seafloor, sometimes leading to emersion. For them, too, the black pebbles were reworked from underlying sediments and protected from oxidation by a layer of cement.

A detailed study of the Purbeckian black pebbles was undertaken by CAROZZI in 1948. He concluded that they had not been reworked from underlying beds, but from contemporaneous layers of freshwater sapropelite. He postulated also black oolites, now bleached by oxidation, as source rock. He attributed the black colour of the pebbles to a certain content of organic matter, which was protected from oxidation by the cement enclosing the pebbles. The multicoloured breccias were interpreted as deposits of subaquatic slides and "turbidity currents" (CAROZZI 1951).

DONZE (1958) distinguished between in situ black spots with diffuse boundaries, and exogenic clasts of fresh- or brackish-water sediments. Hydrocarbons derived

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[2]) The commonly used French terms for "black pebbles" are "galets noirs" and "cailloux noirs".

from decomposed organic matter gave the pebbles their dark colour. He also connected the occurrence of black pebbles with emersion.

COTILLON (1960) thought that pebbles lying in a locally anoxic marine environment were pigmented by colloidal humic substances derived from terrestrial vegetation.

Extensive research on black pebbles was carried out by HÄFELI (1966). He listed the presence of black pebbles from varying environments (shallow marine to fresh water) in Upper Oxfordian, Kimmeridgian, Portlandian, Purbeckian, Berriasian and Tertiary strata. He could prove that the black colour is due to small amounts of organic matter. HÄFELI believed the source sediment to be a lime-gyttja which was reworked by waves, tides or currents, thus destroying the primary evidence.

BARTHEL (1974) compared Purbeckian black pebbles with sub-Recent occurrences on the Florida Keys. He noted their association with caliche crusts and mentioned the possibility of forest fires as blackening agent.

The most recent studies dealing with black pebbles of the Jura Mountains have been made by BLÄSI (1980). He interpreted the black-pebble horizons as storm deposits in a supratidal or intertidal environment. To explain the black coloration, he discussed subaerial weathering on one side, infiltration of organic matter and iron compounds in a reducing environment on the other.

2. Lithologic description

In this chapter, blackened fossils, black peloids, ooids and oncolites, black caliche crusts, black spots and blackened limestone beds, as well as the black pebbles sensu stricto (i.e. lithoclasts), are described. Data from all the Purbeckian samples studied are compiled in Table 1.

Reliable identifications of subtidal, intertidal and supratidal environments are often not possible, especially when criteria such as specific cements, fenestral structures or typical faunal associations are missing. In these cases, the environment is speculatively inferred from the stratigraphic context.

Black fossils

Blackened characeans, bivalve and ostracod shells are common (Fig. 4a, 4b). Black foraminifera (Fig. 4e) and serpulid tubes are also found. The fossils show all shades from grey to black, and the coloration often outlines the shell structure (Fig. 4b)[3]. In some gastropods, micritic chamber fillings are blackened, whereas the

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[3] This penetrative coloration has to be distinguished from the superficial blackening by blue-green algae and bacteria, as often observed in Recent environments.

Table 1: *Purbeckian black pebbles and associated features: data from all studied samples. Results from type sections in Figure 2 are shown separately. Represented are number and frequency of samples containing listed components or features (one sample may contain several features). Subtidal, intertidal and supratidal environments are certain or inferred. Supratidal environment also includes lacustrine facies. As the sampling hasn't been systematic, the table indicates only general trends. For interpretation see chapter 5.*

	SALÈVE			POIZAT			CORNAUX			ALL SAMPLES		
		%	%		%	%		%	%		%	%
Total samples	195	100		20	100		32	100		608	100	
Isolated black fossils	11	6		3	15		8	25		71	12	
Characeans	2			1			2			25		
Ostracods	0			1			4			29		
Lamellibranchs	1			2			7			49		
Forams	6			0			2			10		
Others	3			0			1			8		
Black peloids	5	3		5	25		8	25		49	8	
Black ooids	2	1		0	0		5	16		28	5	
Black oncolites	9	5		0	0		3	9		14	2	
Black lithoclasts total	59	30	100	12	60	100	9	28	100	210	35	100
in supratidal environment	8		14	2		17	2		22	50		24
clasts with supratidal facies	0			2			2			30		
intertidal	4			0			0			10		
subtidal	1			0			0			1		
not definable	3			0			0			13		
in intertidal environment	42		71	9		75	5		56	129		61
clasts with supratidal facies	1			1			2			15		
intertidal	32			7			4			105		
subtidal	12			0			0			12		
not definable	7			1			0			14		
in subtidal environment	9		15	1		8	2		22	31		15
clasts with supratidal facies	0			0			0			3		
intertidal	1			0			1			8		
subtidal	8			1			1			20		
not definable	1			0			0			3		
frequency: less than 1 %	17		29	3		25	4		44	55		26
1 - 5 %	29		49	8		67	4		44	110		52
6 - 10 %	8		14	1		8	1		11	32		15
11 - 20 %	5		8	0		0	0		0	10		5
over 20 %	0		0	0		0	0		0	3		1
mean size: less than 1 mm	32		54	3		25	4		44	93		44
1 - 5 mm	20		34	6		50	5		56	90		43
6 - 10 mm	3		5	2		17	0		0	17		8
11 - 20 mm	2		3	1		8	0		0	7		3
over 20 mm	2		3	0		0	0		0	3		1
shape: angular	2		3	0		0	0		0	5		2
angular to rounded	8		14	4		33	1		11	60		29
rounded	49		83	8		67	8		89	145		69
clasts with pyrite	9		15	1		8	2		22	24		11
Multicoloured breccias	5	3		3	15		0	0		31	5	
Conglomerates total	13			3			1			36		
with black pebbles	5	3		2	10		0	0		21	3	
Black caliche	1	1		0	0		0	0		5	1	
Black spots total	5	3		4	20		1	3		19	3	
in supratidal environment	1			1			1			6		
intertidal	4			3			0			13		
subtidal	0			0			0			0		
with pyrite	5			2			0			14		
Black limestone beds total	0	0		1	5		16	50		54	9	
supratidal facies	0			0			5			16		
intertidal facies	0			1			6			20		
subtidal facies	0			0			5			18		
with pyrite	0			0			2			7		
matrix lighter than particles	0			1			13			35		
matrix darker than particles	0			0			3			19		

shell has been replaced by clear sparry calcite (solution cavity fill, common for molluscan shells, never displays blackening).

Black fossils occur in supratidal, intertidal and subtidal environments and are often associated with black pebbles. They occur as black dots in light or grey sediments, or as black fragments inside a black pebble. Generally they make up less than 1% of the sediment, but concentrations of up to 20% exist.

Very few black fossils show signs of transport. They coexist with uncoloured fossil fragments.

Black peloids, ooids and oncolites

In sediments of subtidal or intertidal origin, peloids, ooids and oncolites are sometimes blackened. The ooids show very distinct differential colouring (Fig. 4d):

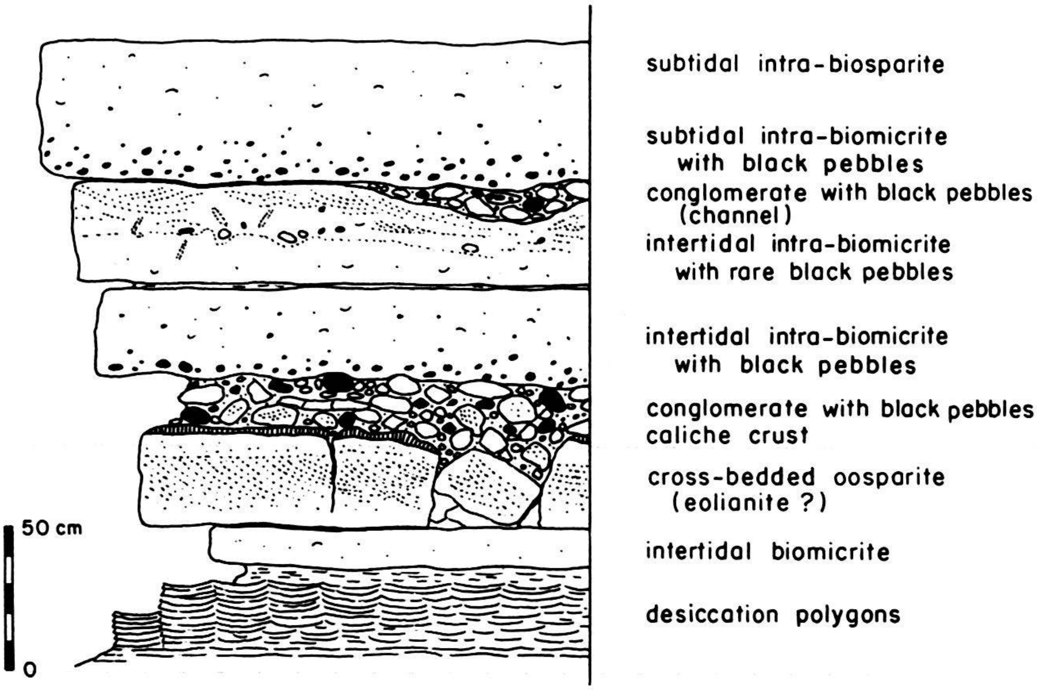


Fig. 3. Detail of Salève section (Fig. 2). For discussion refer to chapters 2 and 5.

Fig. 4. (Purbeckian; polished sections.)

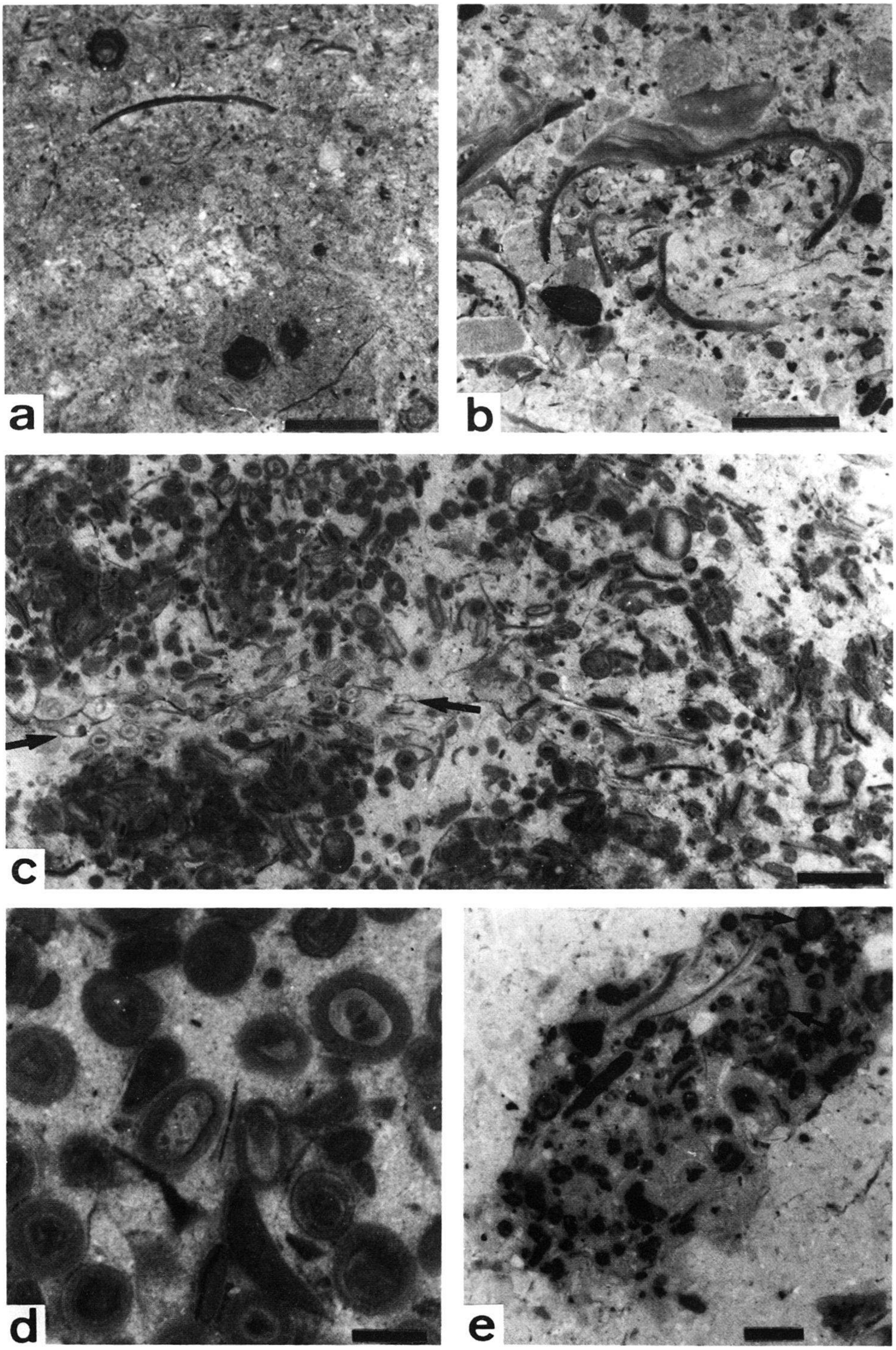
a: Black characeans and ostracods in a brown fresh- to brackish-water limestone (scale 2 mm).

b: Differential blackening outlining the shell structures of bivalves. Multicoloured breccia, interpreted as storm deposit (scale 5 mm).

c: Grey packstone with black ooids and bioclasts. Oxidation along fractures (arrows) (scale 2 mm).

d: Detail of Figure 4c: differential blackening of ooids (scale 0.5 mm).

e: Burrow filled with black lithoclasts and black agglutinated foraminifera. Note marginal blackening of some lithoclasts (arrows) (scale 2 mm).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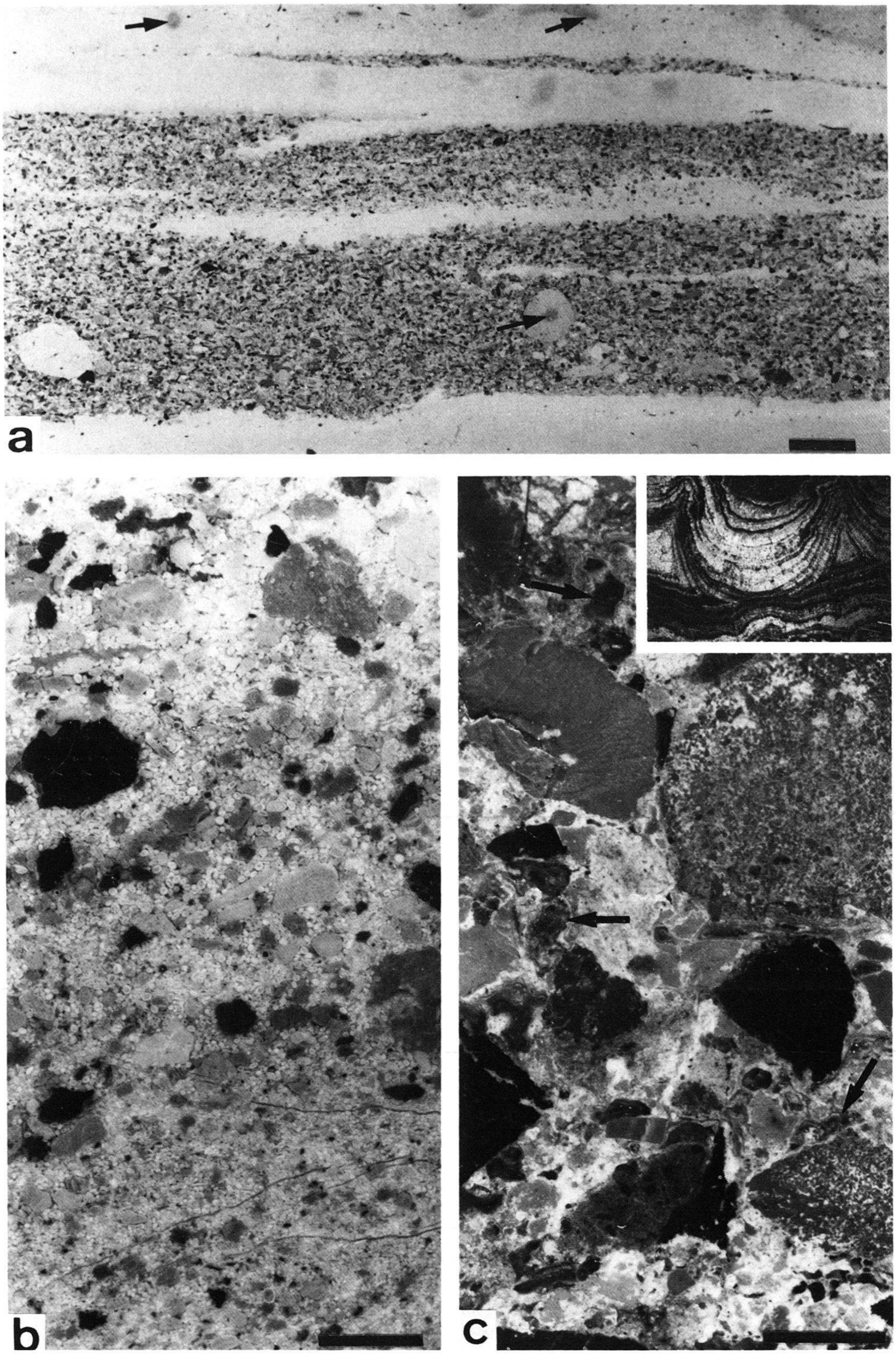
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black core - light cortex, black core - light rim - black cortex, light core - black cortex, black core - black cortex. The particles are embedded in a lighter-coloured sediment, or just the opposite, the matrix is darker than the particles (Fig. 8a). Black fossils serving as ooid cores are usually much darker than other cores.

Like ooids, oncolites also show very clear differential coloration.

Black lithoclasts

The term "black pebble" usually refers to dark lithoclasts. They contrast with the generally light-coloured Purbeckian sediments. The sizes of black lithoclasts range from 0.1 mm up to 35 cm (Häfeli 1966), the average being around a few millimetres. Most black lithoclasts are rounded or subrounded, sometimes with lobate contours (Fig. 6b), but sharply angular fragments also occur. They are found either as isolated pebbles or may account for up to 50% of the sediment (in the multicoloured breccias). Graded bedding is frequent (Fig. 3).

Most black lithoclasts are strongly micritized[4]), so that in many cases their original facies cannot be determined. Pure micrites and pelmicrites, sometimes containing ostracods and characeans, are common. They are derived from an intertidal to supratidal, brackish to lacustrine environment. They may contain birdseye structures or pedogenic features such as fractures (Fig. 6b) and root casts (Fig. 6c). Black oomicrites and pelmicrites bearing marine fossils represent a subtidal facies.

Lithoclast shades range from grey to black and zoning is often displayed (Fig. 6d). Particles can be lighter or darker than the matrix. Some lithoclasts show perforations with oxidation rims (Fig. 6d). In some black pebbles, small pyrite grains are concentrated at the centres (Fig. 5a). In dolomitic sediments, black pebbles may contain dolomite crystals.

Composite black lithoclasts also exist (Fig. 6b). Small black pebbles float in the dark matrix of a larger lithoclast. Accordingly, any clast of dark limestone is defined as black pebble.

Multicoloured breccias ("brèches multicolores")

These breccia layers, which range in thickness from a few centimetres to tens of centimetres, contain not only black pebbles, but also yellowish, brownish and other

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[4]) "Micritization" in this paper means: obliteration of pre-existing structures (probably by differential dissolution of carbonate crystals in acid meteoric waters, Purser 1980), and filling of intergranular and intragranular voids by microcrystalline cements.

Fig. 5. (Purbeckian; polished sections.)

a: Fine-grained multicoloured breccia, interpreted as distal tempestite. Arrows point to black spots with finely disseminated pyrite (scale 5 mm).

b: Inversely graded multicoloured breccia. The matrix is oolitic grainstone (scale 5 mm).

c: Coarse multicoloured breccia with black pebbles of various lithologies and degrees of rounding. Cavities (arrows) are filled with iron-rich, often stalactitic, calcite cement (inset) (scale 5 mm, thin section inset 3.5 mm wide).

light-coloured lithoclasts. Rounded and angular clasts exist next to each other. Some clasts are fractured. Grain sizes range from a few millimetres to tens of centimetres, the average being about 1 cm. Most of the breccia horizons show normal or (rarely) inverted graded bedding (Fig. 5a, 5b, 5c, 6a).

The matrix of the breccia is mostly fine-grained, but in one case pores are filled with vadose cement (Fig. 5c).

Many multicoloured breccias show *storm-deposit* features (BLÄSI 1980, AIGNER 1982). Others are probably collapse breccias (see chapter 5).

Conglomerates

Conglomerates (2–80 cm thick beds) are very typical for the Purbeckian sequence. Black pebbles, slightly flattened but well rounded, occur in some of them. The sizes range from a few millimetres to 20 or 30 cm (average a few centimetres). The conglomerates usually overlie a depositional unconformity, or in some cases a caliche crust (Fig. 3). The pebble components have been reworked from underlying sediments. The matrix generally consists of greenish marls.

Black caliche

Some black pebbles consist of, or contain, black laminated calcrete (Fig. 7a). In one case (Grande Varappe near Salève section) black caliche encrusts a channel bottom.

Black spots

Some Purbeckian limestones show black spots or patches of various sizes and shapes which usually display diffuse boundaries. They occur mostly in mudstones and wackestones, but sometimes also in packstones and grainstones. Concentration of small pyrite crystals commonly exists in black-spot areas.

In several cases, black spots seem to have originated from bioturbation (Fig. 7d). There is an evident relationship also between black spots and root casts: immediate surroundings of rootlets are usually the darkest (Fig. 7b, 7c). Blackening may also be more pronounced along fractures.

Black limestone beds

Besides pebbles and spots, relatively large areas in bedded sediments may show various degrees of blackening. They are generally made of packstones and wacke-

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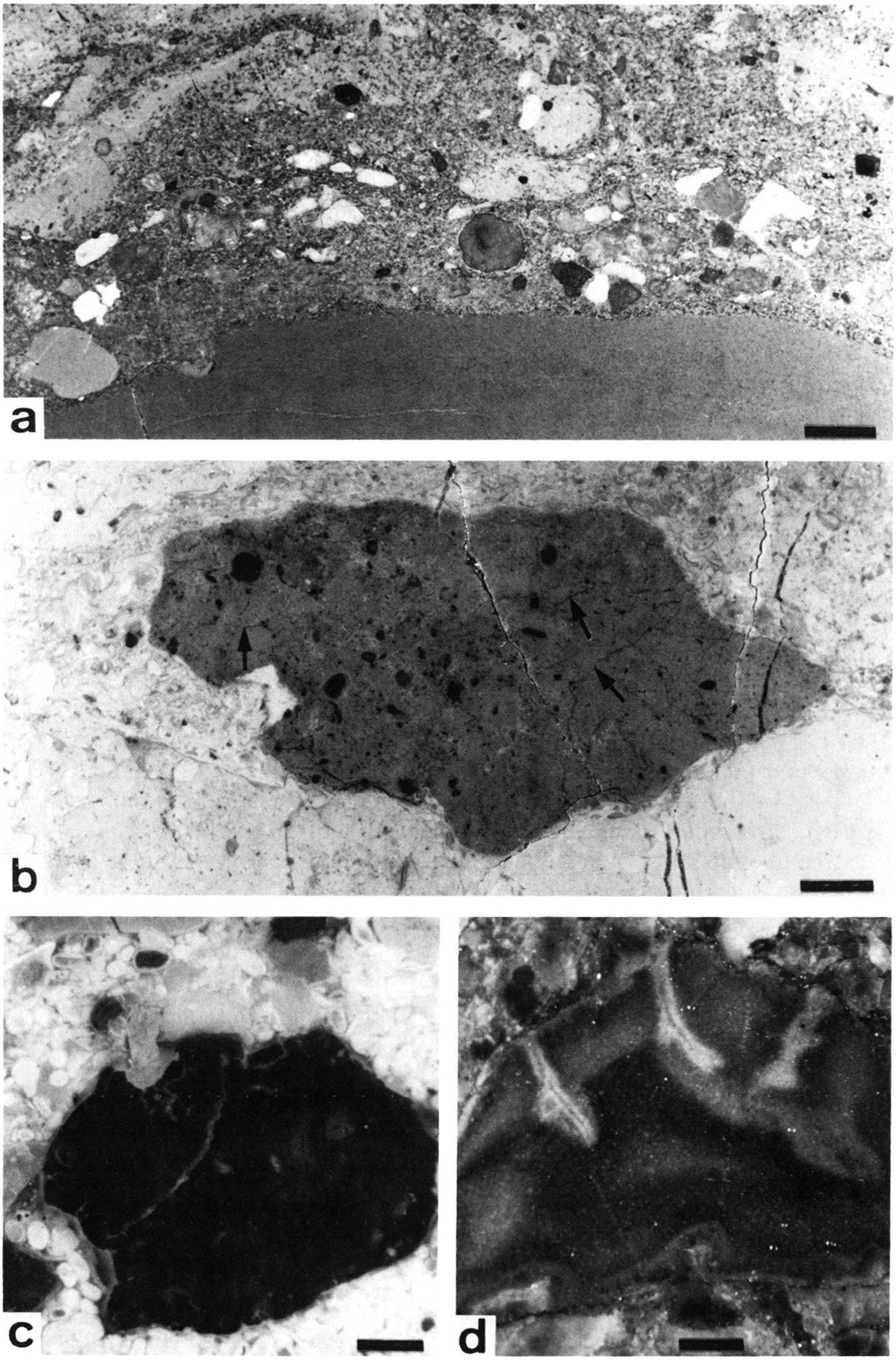
Fig. 6. (Purbeckian; polished sections.)

a: Graded multicoloured breccia with a matrix of grey pellet-packstone. Erosional contact to fine-grained laminated organic-rich wackestone (scale 5 mm).

b: Composite black pebble: dark grey matrix with black lithoclasts, characeans and ostracods. Fractures preceding transport are indicated by arrows (scale 2 mm).

c: Detail of Figure 5b: micritized and blackened oolite. Fractures are due to penetrating rootlets (structure is visible in large fracture) (scale 1 mm).

d: Irregularly blackened lithoclast in dark packstone showing oxidation rims around perforations (root tubes?) (scale 0.5 mm).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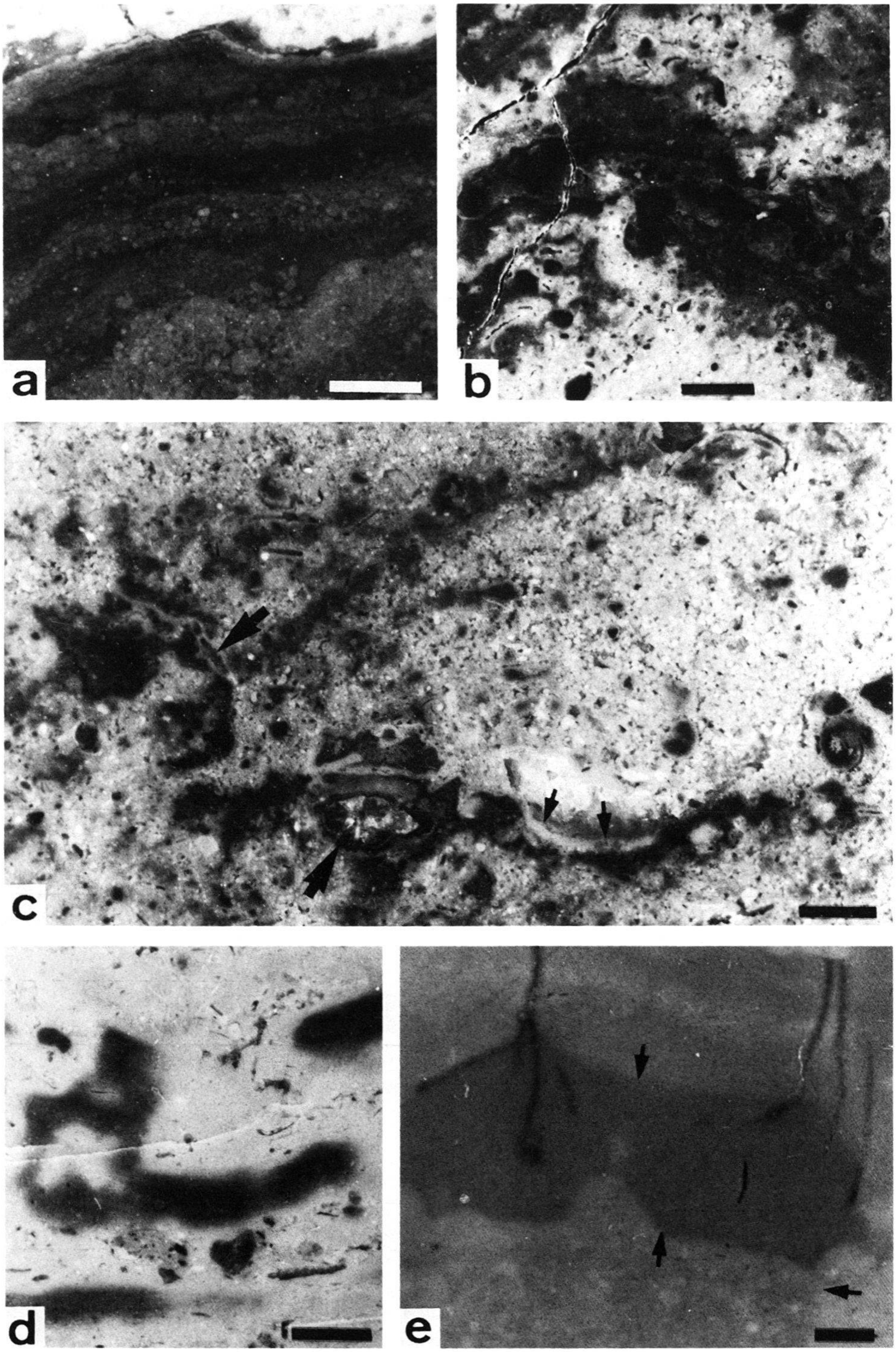
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stones with black particles (fossils, peloids, ooids, oncolites, lithoclasts) in a grey matrix, and penetrate into fractures or burrows of the underlying sediment (Fig. 4e). Bioturbation leads to differential coloration (Fig. 8b). Mudstones, in which the blackening may accentuate laminations, have sometimes been disrupted to form grey or black flat pebbles.

Some of these limestones have a dark brown colour. The particles are often lighter than the matrix (Fig. 8a, 8b).

Black limestones have sometimes been oxidized to a yellow-brown colour at the surface of the outcrop and along fractures (Fig. 4c, 7e).

Stratigraphic position of the black pebbles

Although isolated black pebbles occur in any stratigraphic position in the Purbeckian, they usually concentrate in certain facies and are linked to certain sedimentological features (Fig. 2).

- Most black pebbles are found in intertidal to supratidal, brackish to freshwater facies.
- High concentrations of large black pebbles occur in multicoloured breccias and in conglomerates.
- High concentrations of small and medium-sized black pebbles occur at the base of beds overlying a conglomerate or diastem.

3. Other occurrences

Black pebbles, which are so characteristic of the Purbeckian in the French and Swiss Jura, are also found in other areas and in other geological epochs.

Europe

Black pebbles and multicoloured breccias are reported from the Purbeckian in the southeast of France (GIGNOUX & MORET 1937, DONZE 1958, COTILLON 1960, 1974), and in Estremadura, Portugal (REY 1972).

In the French and Swiss Jura, black pebbles occur in the Upper Oxfordian, Kimmeridgian and Portlandian strata and in the Purbeckian-Berriasian transition

Fig. 7. (Purbeckian; polished sections.)

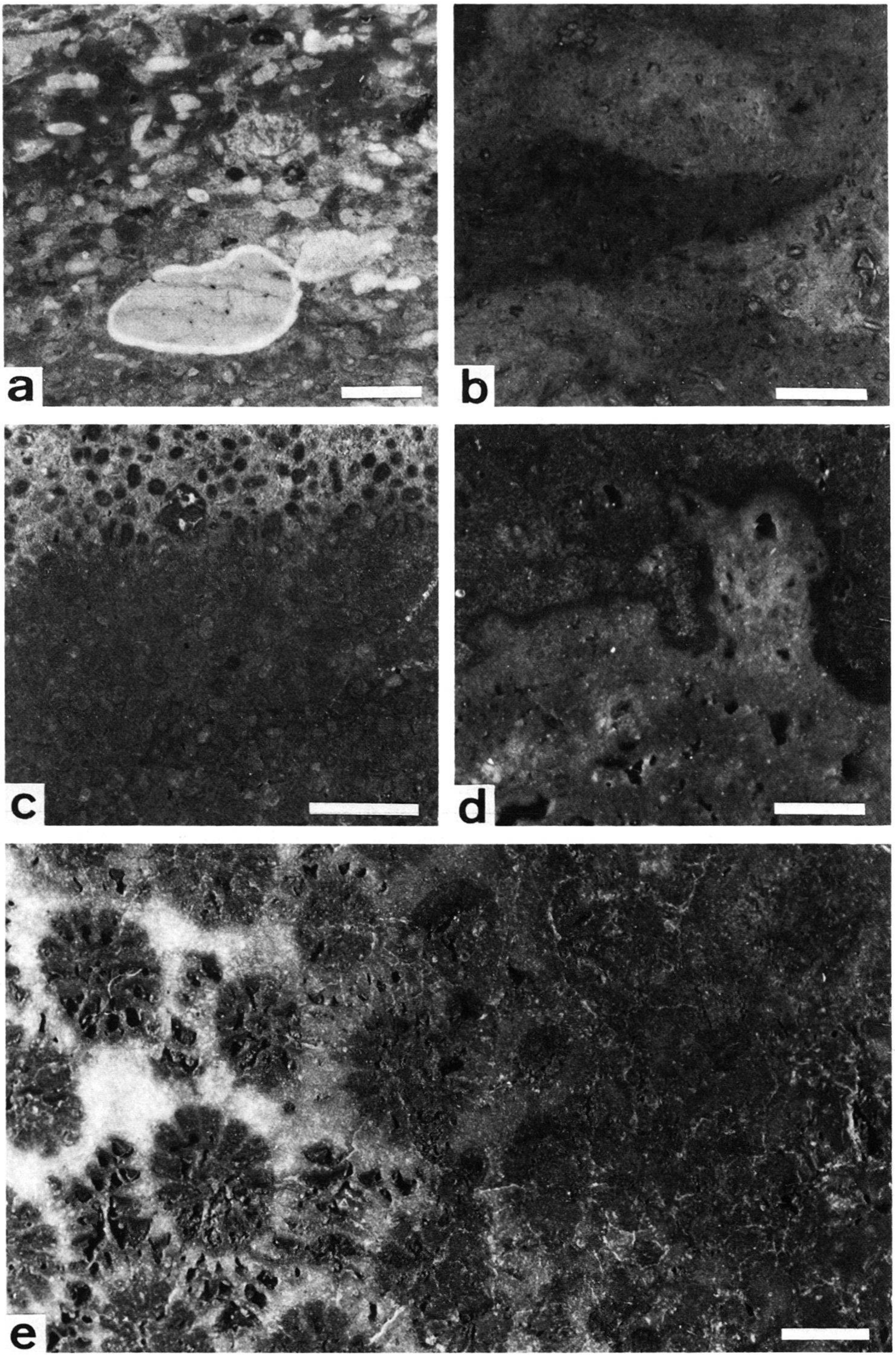
a: Black caliche clast. Trapped particles are lighter than laminated matrix. Photo is upside-down, showing overturned clast in upright position (scale 2 mm).

b: Concentration of blackening around root casts. Locally occurring fine-grained pyrite does not show in this photograph (scale 2 mm).

c: Black spots, partly pyritic, were probably arranged along decayed rootlets. Some root traces are preserved (large arrows). Small arrows point to geopetal filling of a void, possibly also a root cast (scale 2 mm).

d: Black spots with finely disseminated pyrite, interpreted as bioturbation (scale 2 mm).

e: Dark grey wackestone showing pyrite impregnation along fissures, which were probably initiated by rootlets. Several fronts of oxidation are marked by arrows (scale 2 mm).


a
b
c
d
e

beds (several authors; compilation in Häfeli 1966 and Enay 1980). These black pebbles are found in subtidal, intertidal and supratidal-lacustrine paleoenvironments.

Tertiary black pebbles in the Jura Mountains are of lacustrine origin (Häfeli 1966). A borehole near Basle (Brianza et al. 1983) furnished small grey lithoclasts with black spots, black peloids and black ostracods from a Sannoisian (Lower Oligocene) freshwater limestone. Also of Sannoisian age are dark lacustrine limestones with black gastropod shells at Le Puy, southeastern France. Black and grey pebbles occur in Oligocene freshwater limestones (Calcaire de Beauce) in central France (Freytet 1982). Montenat (1981) found black caliche pebbles cemented into a Pleistocene caliche crust in the southeast of Spain.

Enay (1980) cites multicoloured breccias of Upper Oxfordian age in the Aquitaine (France) and in the central and eastern Pyrenees. Seyfried (1980) describes breccias with black pebbles from the Jurassic of the Betic Cordilleras (southeastern Spain). There they are associated with green marls and constitute part of a cyclothem similar to the Lofer cyclothems in the Triassic of Austria (Fischer 1964).

Black pebbles also form part of the Bleiberg cyclothems in the Carnian Wetterstein Formation of Austria (Eppensteiner 1965, Bechstädt 1975, 1979). An idealized section includes, from bottom to top: subtidal deposits - intertidal deposits - an erosion surface - green marls with black breccia components - a breccia with black pebbles - supratidal deposits - intertidal deposits - subtidal deposits. The black pebbles often have lobate shapes and some contain birdseye structures. Piller (1976) describes black breccia components in the neighbourhood of algal stromatolites and green marls from the Upper Triassic Dachstein Limestone of Austria.

Black pebbles and breccias occur in the Upper Cretaceous of southern Yugoslavia (Jelaska et al. 1983).

Florida and Bahamas

Black lithoclasts and black fossils in the Florida Keys have been cemented into Holocene caliche crusts which cap Pleistocene reef limestones (dating: Robbin & Stipp 1979). The clasts are angular to rounded, their sizes range from a few millimetres to several centimetres. Recrystallized subtidal grainstones and coral debris are common (Fig. 8e). Some pebbles show borings.

Fig. 8. (Polished sections.)

a: Multicoloured breccia with predominantly white lithoclasts in dark matrix (Purbeckian; scale 2 mm).
b: Bioturbated dark brown characean limestone (Purbeckian; scale 2 mm).
c: Differential colouring of ooids along front of blackening (Bimini, Bahamas; scale 2 mm).
d: Detail of contact between black pebble above and caliche crust below, showing front of blackening (Crawl Key, Florida; scale 0.5 mm).
e: Coral fragment displaying preferential blackening of porous and permeable chambers (Crawl Key, Florida; scale 2 mm).

On Bimini (Bahamas) black pebbles are embedded in caliche crust which is now situated in the intertidal zone. They accumulated in bowl-shaped depressions. Some of the in situ caliche has also been blackened. Loose, well-rounded black pebbles are found in the intertidal zone on the western and northern sandy shores. Average pebble size is a few centimetres (the largest one found measures 14 cm in diameter). They consist of micritized black oolitic grainstones (Fig. 8c) derived from the Pleistocene and Holocene carbonate rocks forming the Bimini Islands (dating: GIFFORD 1973). Compared to the porosity of the unaltered limestones, the porosity of the black pebbles is much lower. Here, as in the Purbeckian, black and unblackened lithoclasts of the same facies and size range are found next to each other.

On the wide carbonate tidal flats along the western coast of Andros Island, black pebbles are found in the intertidal and supratidal zones. They are rounded, micritized packstones of 1 to 3 cm diameter. The black coloration often is patchy. Black-impregnated bivalve shells are also common. Loose and porous pebbles in a layer of black mud on the bottom of a channel show black spots at their centres.

BEACH & GINSBURG (1980) described black lithoclasts on Pliocene-Pleistocene subaerial exposure surfaces underlying the Great Bahama Bank.

Tunisia

Black pebbles occur in the intertidal and supratidal zones on the islands of Kerkenna, situated east of Sfax on the shallow carbonate platform of the Pelagian Sea. They are found as moderately rounded clasts a few centimetres in diameter which lie on the beach or are cemented into a pisolitic limestone crust. They are blackened and micritized fragments reworked from the Tyrrhenian (uppermost Pleistocene) biogenic, quartz-rich limestones. The blackening often is patchy. Black pebbles coexist with light-coloured pebbles of the same facies and size range.

In the area of Zarzis (southern Tunisia), sub-Recent black pebbles occur in the intertidal and supratidal zones. They are quartz-rich limestones showing patchy blackening. Beachrock underlying rotting seagrass *(Posidonia)* displays incipient blackening.

Black foraminifera, bryozoans and shell debris occur in several dark, organic-rich layers in the modern intertidal and subtidal muddy sediments. In off-shore drillholes, black fossils (foraminifera, molluscs, bryozoans) are found almost continuously from the Recent down to the Upper Pleistocene (courtesy L. Blanc-Vernet).

Other areas

Blackened Pleistocene eolianite and black caliche pebbles have been reported near hypersaline pools and lakes on Isla Mujeres, Yucatán (WARD et al. 1970).

WILSON (1967) found black lithoclasts related to subaerial exposure in the Pennsylvanian limestones of southern New Mexico.

Shells and carbonate particles are blackened in Holocene shallow-water sediments of the southwestern Persian Gulf (KENDALL & SKIPWITH 1969). Black-stained foraminiferal tests are found in the southern part of the Great Barrier Reef, Australia (MAIKLEM 1967).

4. Mineralogical and geochemical analysis

Methods

The mineral composition of selected samples was determined by X-ray powder diffractometry. Both bulk samples and insoluble residue (decarbonated with 10% hydrochloric acid) have been analyzed.

The composition and state of the organic matter in black pebbles and some associated sediments was evaluated using two methods of pyrolysis and subsequent analysis of the volatilized compounds. For differential thermoanalysis (DTA), samples were decarbonated with 10% formic acid. The residue was heated from 25 to 1000 °C in air, whereby organic and other compounds were oxidized. The amount of H_2O, CO_2 and SO_2 released was measured by mass-spectrometry as a function of temperature. For the present study, CO_2 curves only were used, and this qualitatively, since it is their shape which best characterizes the organic material. Whole-rock analysis followed the standard Rock-Eval pyrolysis method developed by ESPITALIÉ et al. (1977).

Results

The mineral composition of black pebbles implies that, in most cases, it is *organic matter* which causes the blackening. Pyrite may add to the colouring, but in most black pebbles it is not present. The same holds for clay minerals. Two samples have been analyzed by microprobe: iron and manganese are present, but only locally and in traces, so that a significant contribution to the colouring can be excluded. (WARD et al. 1970 found that iron and manganese are less abundant in black limestones than in brown ones.) In both samples phosphorus is absent.

Organic matter as blackening agent has already been proposed by CAROZZI (1948). HÄFELI (1966) undertook detailed chemical analyses of the organic content of Purbeckian black pebbles and concluded that, according to the H/C ratio and to sedimentological criteria, the organic matter must have been derived from plants.

In order to determine the state and origin of the organic matter, selected samples of black pebbles, black caliche, black limestones and black marls were analyzed (Table 2). Two samples from the Bahamas have been included for comparison. Table 2 demonstrates that relatively small amounts of organic carbon (less than 0.1% of the total rock) may already cause blackening. Light-coloured rocks contain only traces of organic matter, or none at all.

The Rock-Eval analysis shows that the H/C and O/C ratios of all samples (Fig. 9) lie in the field of vitrinites (VAN KREVELEN 1961), indicating that the organic matter was derived from land plants. The extremely low H/C ratios point furthermore to alteration of the organic material by oxidation and transport "as inertinite-type particles and/or soil humic acids" (TISSOT & WELTE 1978, p. 145–146).

In Figure 10, the CO_2 curves obtained from the differential thermoanalysis are compared. Purbeckian samples P1, P2, P3 and P4, as well as sub-Recent sample B1 from the channel floor on Andros, show peaks at low temperatures, indicating young, little-altered kerogene, probably derived from algae (G. Kahr, pers. comm.). With aging of the kerogene, the peaks shift towards higher temperatures.

Table 2: *Description of samples analyzed for organic matter.*

Purbeckian		% $CaCO_3$	% org. C*
P1	Black-spotted limestone (Fig. 7b): dark grey pelmicrite with desiccation fissures. Irregular black spots, often around root casts, together with pyrite.	94.3	0.09
P2	Black spots (bioturbation?) with disseminated pyrite in micrite (Fig. 7d).	92.3	0.10
P3	Laminated dark grey micrite with finely disseminated pyrite underlying a grey multicoloured breccia (Fig. 6a).	91.3	0.16
P4	Black marls with grey limestone nodules and pyrite.	83.7	0.31
P5	Black pebble in conglomerate: black calcrete with abundant root casts filled with white spar.	98.0	0.19
P6	Black pebble in conglomerate: dark grey oomicrite with desiccation features and stringers of coaly substance.	94.5	0.12
P7	Black pebble in conglomerate: dark grey pelmicrite with black lithoclasts and peloids; root traces.	95.7	0.13
P8	Dark brown heterogeneous marly limestone with large limestone pebbles and pieces of charred wood.	69.8	2.63
P9	Black pebble in conglomerate: dark grey bioturbated pelmicrite with layer of black calcrete.	98.5	0.08
P10	Black laminated calcrete: dark grey and black laminae.	96.2	0.16
Bahamas			
B1	Porous pebble with a blackened and partly pyritic centre. From layer of soft black mud at bottom of channel on Andros Island.	88.0	0.50
B2	Black laminated calcrete, now in the upper intertidal zone at the northern shore of North Bimini Island.	97.3	0.10

* Values from Rock-Eval analysis.

In the other analyzed samples, the peak of young kerogene is only hinted at (P7, P8, P9) or lost altogether. The CO_2 curves each show one pronounced peak around 500 °C and steep right flanks, indicating *coalification* of the organic matter (compare with curve of commercial charcoal). Blackened calcretes from the Purbeckian (P10) and from the Bahamas (B2) show a shift of the right flank towards higher temperatures (i.e. the organic matter is in a more stable, graphitic state). Coalification due to weak Alpine metamorphism can thus be excluded, since Purbeckian and sub-Recent samples show similar curves.

Organic material derived from higher terrestrial plants tends to coalify, whereas algal matter leads rather to kerogene formation (G. Kahr, pers. comm.). In fact, most samples containing root casts (P5, P7) or caliche (P9, P10, B2) suggest coalification (the organic matter contained in brown caliche crusts apparently has not been coalified). The resemblance between the CO_2 curves of black pebble P5 and charcoal is striking. Forest fires probably furnished burnt organic matter for the coloration of black pebbles (HARRIS 1958 found charcoal produced by forest fires in the Mesozoic).

Purbeckian black pebbles and calcretes therefore seem to be blackened mostly by material derived from *higher plants* (which may have been burnt), whereas black spots and blackened limestones owe their coloration rather to *algal matter and pyrite.* Mixing from both sources is common, as in samples P7, P8 and P9.

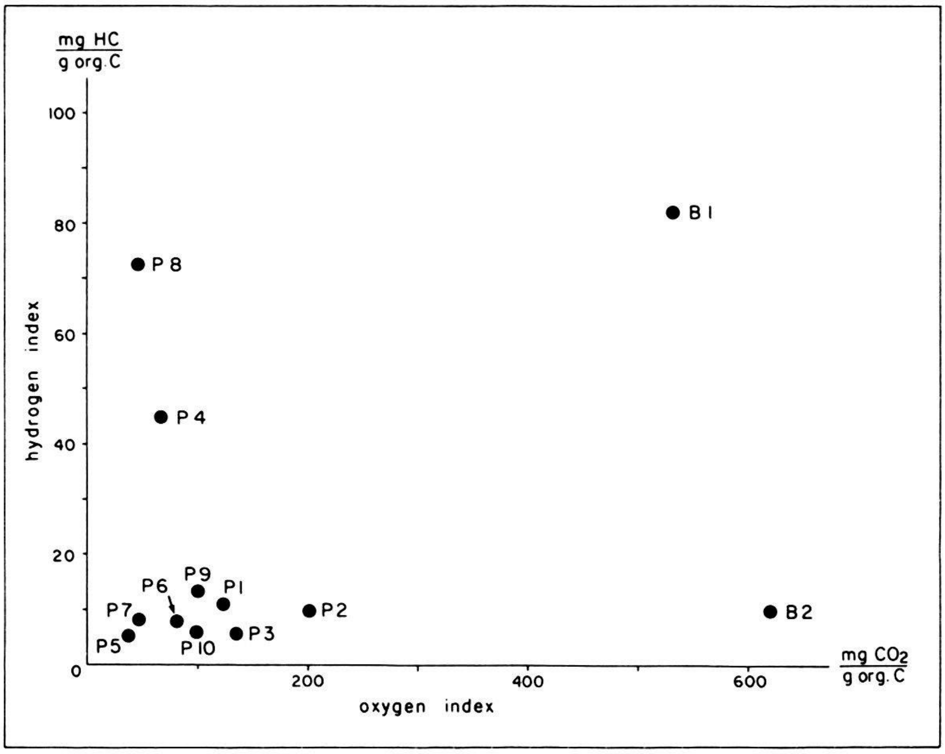


Fig. 9. Results of Rock-Eval analysis, diagram after TISSOT & WELTE (1978). The low hydrogen values indicate relationship to vitrinites. Note high oxygen ratio of young Bahamian samples.

A simple test had been carried out by WARD et al. (1970) on sub-Recent black pebbles of Yucatán, Mexico. Powdered black limestone would not react to bleaching unless the carbonate was dissolved. This is true also for the Purbeckian and Bahamian black pebbles and limestones, except for the black marls (P4). According to SUESS (1970, 1973) and CARTER (1978), organic matter is *adsorbed* as molecular layers on the surface of micritic carbonate crystals. This explains the observed resistance to bleaching as long as the carbonate is not dissolved. On the other hand, clay minerals fix the organic matter in their lattice spaces (FU JIAMO 1980), which must have been the case in the marls of sample P4. Black pebbles are also relatively resistant to natural oxidation, even if subaerially exposed for long periods of time.

Under the scanning electron microscope, even at a magnification of 200,000, the organic matter on and between the carbonate crystals is not visible. But after treatment with oxygen plasma, which removed the organic material completely, crystals in blackened areas show corroded and pitted surfaces, whereas crystals in unblackened areas apparently stay unaffected (Fig. 11).

Adsorption of organic matter on carbonate-crystal surfaces also explains why blackening occurs preferentially in fine-grained carbonates (ch. 2): the overall larger crystal surface can adsorb more organic matter. How much influence the presence of organic matter has on recrystallization and micritization of its host material is uncertain. However, Bahamian black pebbles consist entirely of low Mg-calcite, whereas their host rock (of same facies and age) contains aragonite and high Mg-calcite. Inversion of aragonite to calcite, along with a reduction of grain size, favours

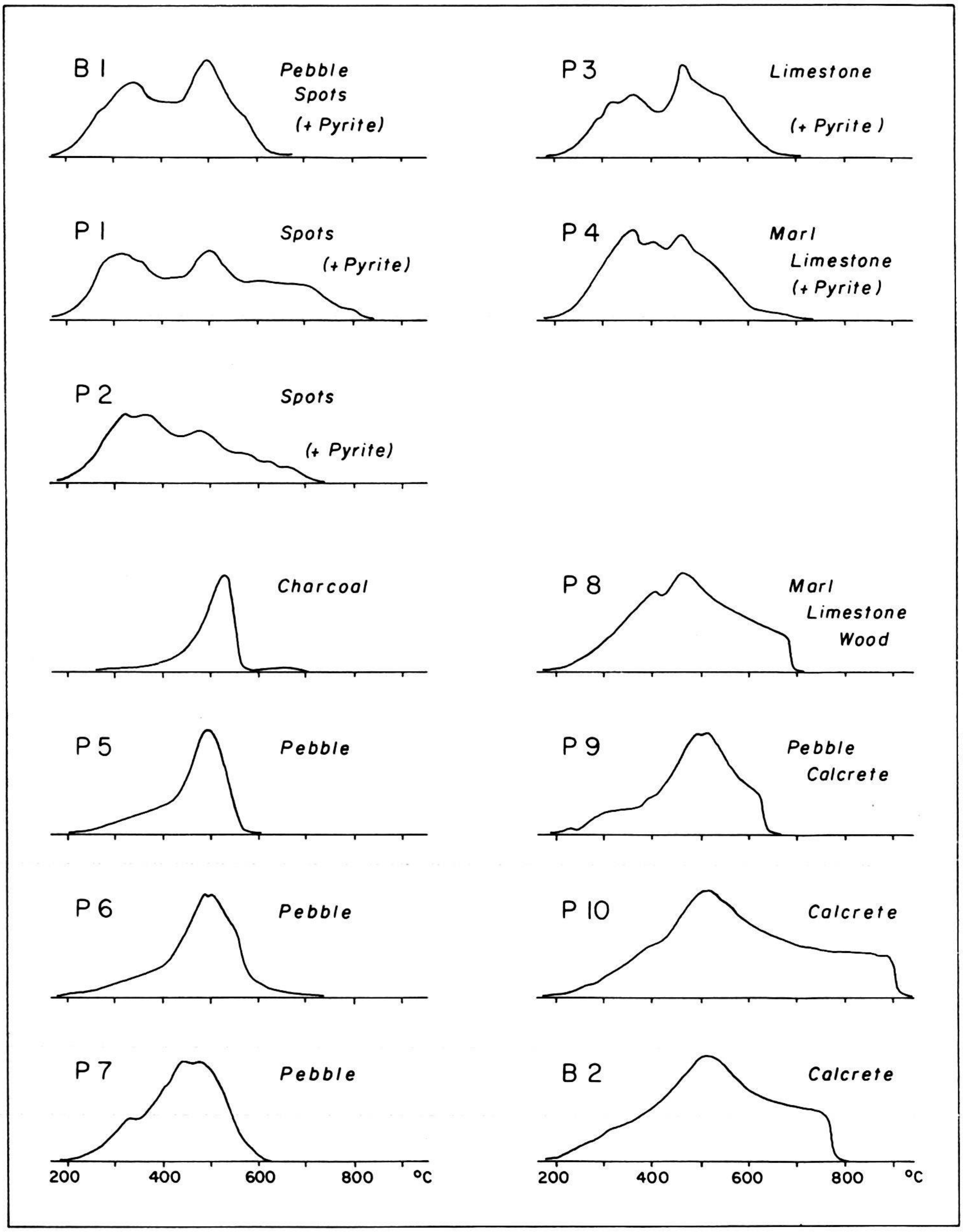


Fig. 10. Comparison of the shapes of CO_2 curves (obtained from DTA analysis), characterizing the organic matter. For explanation see chapter 4.

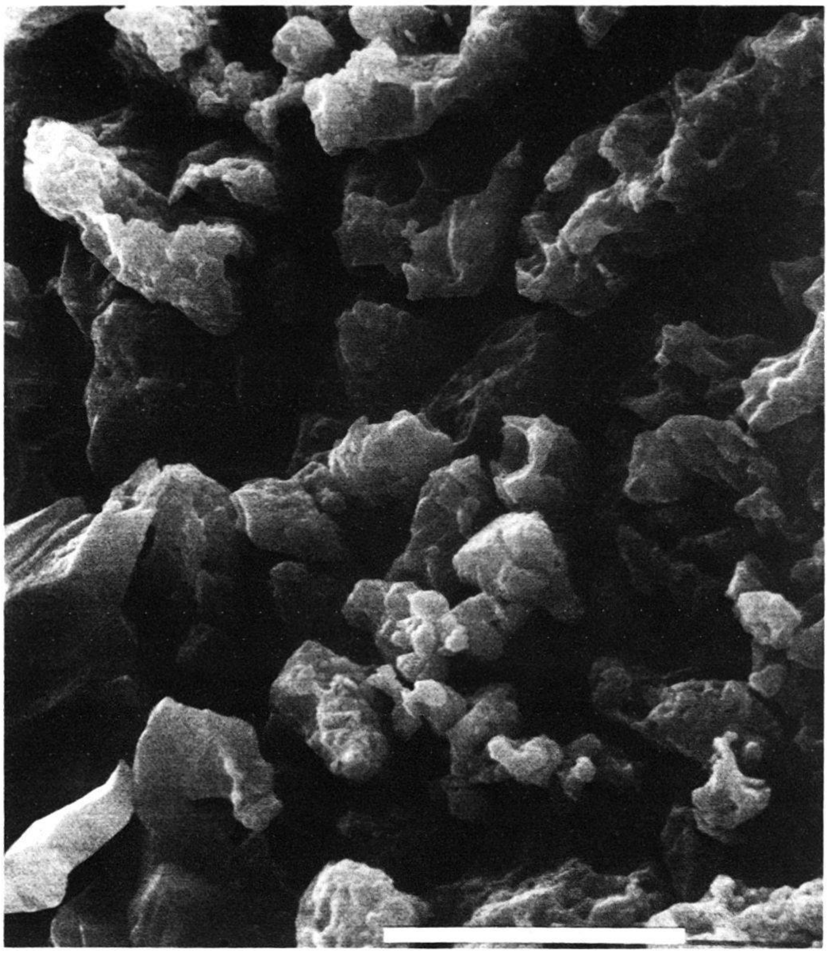


Fig. 11. SEM photograph of a Purbeckian black-pebble surface, etched with diluted HCl and treated with cold oxygen plasma to destroy the organic matter. The picture shows the contact of dark black area (upper right) to lighter area (lower left). Pitted crystal surfaces at upper right result from removal of organic matter (scale 5 µm).

blackening. Due to the larger crystal size, fillings of fractures and solution cavities are never blackened.

Blackening also appears to be a function of porosity and permeability: around root casts (Fig. 7b) and along fissures blackening is often stronger. Differential blackening is shown very clearly in a coral fragment of the Florida Keys (Fig. 8e) as well as in oolitic limestones (Fig. 8c). Later on, oxidation may proceed along the same paths (Fig. 4c, 6d).

In several cases, an advancing front of blackening is suggested (Fig. 8c, 8d). In the same way, oxidation may later bleach the black limestones (Fig. 7e). In order to penetrate fissures and pores, the organic matter has to be in a mobile (i.e. dissolved, colloidal or extremely finely particulate) state. Except for rare calcified algal filaments in sub-Recent black pebbles, no plant fibres or particles have been observed in the blackened areas.

5. Interpretation

Geochemistry

The degradation of dead plant material is carried out mainly by fungi and bacteria. Even after transport and sedimentation of decayed organic matter, the bacterial attack continues under aerobic or anaerobic conditions and leads to particulate, colloidal or dissolved organic compounds (MOORE 1969). Decay of terrestrial plants will thus eventually produce humic substances (fulvic and humic acids; STEVENSON & BUTLER 1969) as well as pigments, waxes, fatty acids, etc. Organic substances such as amino acids (derived from proteins) are found in skeletal carbonates of worms, corals, foraminifera, molluscs and algae, as well as in carbonate mud and oolites (MITTERER 1972).

SUESS (1970) showed experimentally that dissolved organic compounds in seawater (pH = 8.3) interact with carbonate-crystal surfaces to form organo-carbonate complexes. He used stearic acid, which reacted preferentially with calcite and formed monomolecular layers on the crystal surfaces. The amount of adsorbed organic carbon increased with decreasing grain size of the calcite. Later he confirmed his findings in a study of Recent sediments (SUESS 1973). CARTER (1978) demonstrated that, under slightly alkaline conditions, calcite preferentially adsorbs organic matter rich in aspartic acid.

Organic acids are not only adsorbed on carbonate-crystal surfaces, but also have a certain influence on *neomorphism*. According to JACKSON & BISCHOFF (1971), alkaline and neutral amino acids accelerate the conversion of aragonite to calcite, whereas acidic amino acids inhibit this reaction. Organic coatings may also hinder crystal growth during diagenesis (CHAVE & SUESS 1970), which could explain the predominance of microcrystalline cements in the black pebbles.

A detailed biochemical analysis of the organic matter in the Purbeckian black pebbles lies outside the scope of this paper. We may assume, however, that organic compounds, derived from algae and higher terrestrial plants, and transported by water, infiltrated the porous limestone and were adsorbed on the crystal surfaces. At the same time they promoted recrystallization, aragonite/calcite inversion or replacement, and microcrystalline *cementation* which led to reduced porosity (as observed in sub-Recent black pebbles, ch. 3) and thus to an additional fixation of the organic matter. This sealing process further prevented the access of oxidizing solutions.

The coexistence of calcium carbonate and organic matter requires *anoxic and slightly alkaline* conditions: Eh below zero, pH greater than 7.8 (KRUMBEIN & GARRELS 1952). For pyrite formation, Eh has to be less than −200 mV and sulfate reducing bacteria have to be active. In the above-mentioned stability fields, salinity has no influence (KRUMBEIN & GARRELS 1952).

To what extent the above cited published data may be applied to the problem of Purbeckian black pebbles is uncertain. The relationship between adsorption of organic substances and neomorphism especially is not clear. According to CARTER (1978), quartz has only a slightly smaller adsorption capacity than calcite, but black pebbles occur (to our knowledge) only in carbonate environments. Early cementation and neomorphism of carbonates might therefore be an important factor.

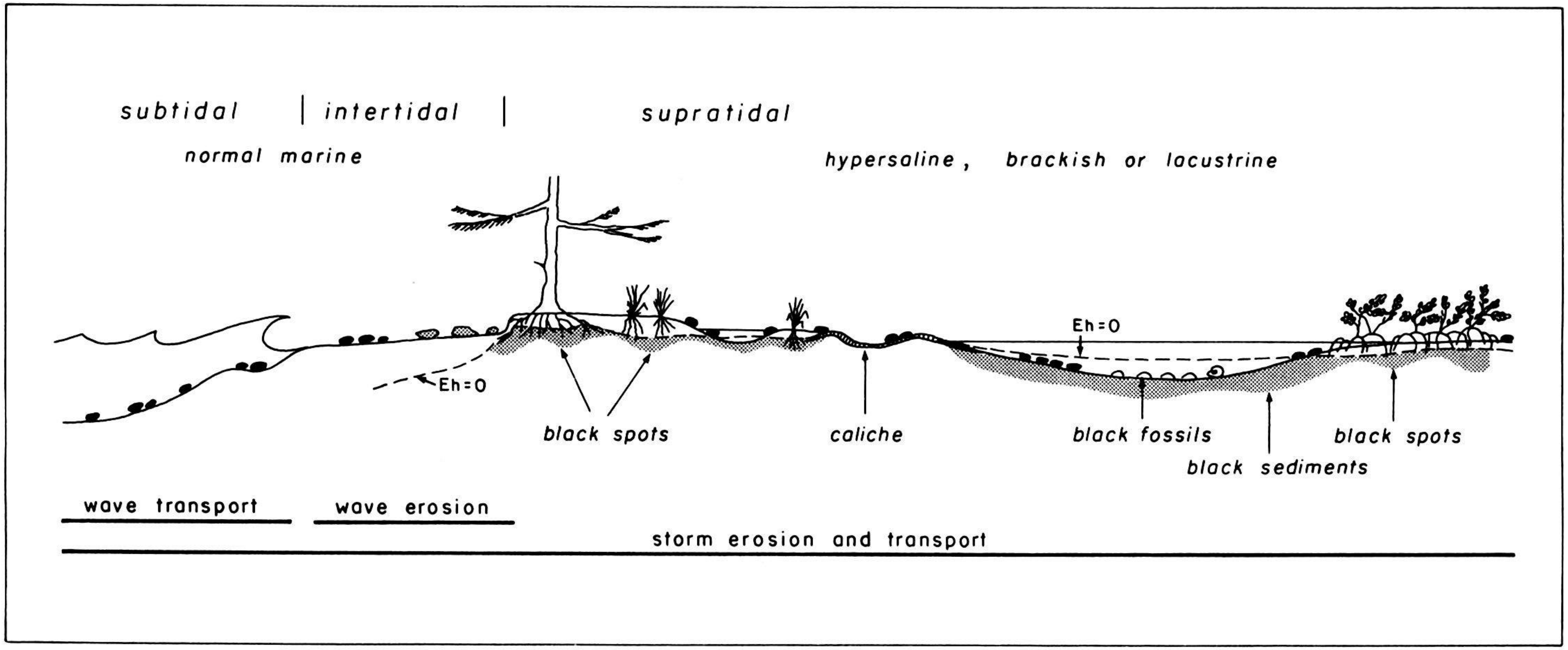


Fig. 12. Sketch of various possible environments which favour black pebble formation and deposition. For explanation see chapter 5.

Facies and sedimentology

Black pebbles are sometimes associated with caliche, or contain root casts. Black spots form around root molds, and pedogenic features are frequent (ch. 2 and 3). In this clearly *terrestrial* environment, a supply of decaying (and maybe burnt) plant material and favourable pH/Eh ranges for the blackening of sediments can be expected (Fig. 12). Microorganisms around decaying roots create anoxic conditions and cause patchy blackening. Reduced water circulation in freshwater or brackish pools favours blackening of bioclasts, limestone particles and submerged caliche crust. Reducing conditions and high pH are also found in hypersaline ponds (WARD et al. 1970). Dropping of ground water level can lead to desiccation and probably to partial cementation of these blackened sediments.

In the *vadose zone* (beach levees, coastal dunes), the percolation of water charged with organic substances is facilitated. Vadose and freshwater phreatic conditions also favour diagenesis leading to stable carbonates, and through early cementation to an additional, diagenetic fixation of organic matter.

Variations in microenvironment, in supply of organic material, in primary texture and mineralogy, and in porosity and permeability lead to locally limited blackening (ch. 2). Differential blackening of carbonate particles may be due to the same factors.

These preferentially blackened and cemented zones are easily reworked by *coastal erosion,* whereas the surrounding less consolidated sediment may be removed. Black pebbles thus become *relic* sediment components. Most black pebbles are rounded (Table 1), indicating transport. Polyphase reworking is probably common, since the cemented black limestones are harder than the unblackened host sediment. Blackening of pre-existing lithoclasts is also possible if the conditions are suitable. Because of chemical adsorption, diagenetic fixation and maturation (coalification) the black organic matter is relatively resistant to oxidation and decoloration, unless oxidizing waters penetrate through fissures or perforations.

In the case of the Purbeckian, the final deposition of black pebbles took place in the shallow subtidal, intertidal and supratidal zones (Fig. 2). Together with unblackened lithoclasts, large black pebbles are found in locally channelized conglomerates, which are thought to have formed at or near a beach in a high energy zone. Quite often they overlie a caliche crust (Fig. 3). Storm events deposited the multicoloured breccias, which often show graded bedding and an erosive contact with the substrate. Low energy sediments overlying coarse storm or spring tide deposits often include a basal component of small reworked black pebbles (Fig. 3). Angular breccias, also containing black lithoclasts, could have formed through collapse caused by dissolution of underlying evaporites.

The sequence comprising intertidal or supratidal sediments - caliche crust - conglomerate with black pebbles and green marls, appears not only in the Purbeckian, but also in the Triassic of Austria and the Jurassic of Spain (ch. 3).

The genesis and significance of the green marls is not yet understood. In the Purbeckian, they consist essentially of calcite and illite, with additions of mixed-layer minerals. Green marls also occur independently from the conglomerates, but in most cases they include black pebbles. They always seem to mark a transgressive event. Black-pebble conglomerates and green marls have regional significance, especially in the upper part of the Purbeckian, where they can be correlated over tens of kilometres.

Paleogeography

Broadly speaking, the Purbeckian sediments were deposited on a very shallow, partly and ephemerally emergent platform, protected from the open sea (to the ESE) by barrier islands (Davaud et al. 1983). Caliche crusts formed on these islands. Conifer wood fragments indicate forestation. On the internal platform, small changes in topography (through sediment accumulation and transport) or in sea level led to fluctuations from normal marine to brackish to freshwater conditions (Fig. 2). Intertidal flats must have been very wide. The platform may also have been dotted with ephemeral, muddy, vegetated islands with freshwater or hypersaline ponds (as today in Florida Bay). In some areas, sabkhas developed (Cornaux, Fig. 2). All these facies zones were exposed to storms and spring tides, and mixing of sediment of various facies must have taken place. These settings were favourable for black-pebble formation.

On the Florida Keys, on the Bahamas and in Tunisia, sub-Recent black pebbles have been observed in similar environmental settings (ch. 3).

At the Jurassic-Cretaceous boundary, the Swiss and French Jura was situated at a paleolatitude of about 38 degrees north (Barron et al. 1981). The climate must have been warm, with periods of rain, since it supported vegetation and the formation of evaporites. Sub-Recent black pebbles and black limestone particles are found in humid tropical (Yucatán, Ward et al. 1970) as well as in arid climates (Persian Gulf, Kendall & Skipwith 1969), where they are always associated with carbonate sediments.

6. Summary and conclusions

In this paper we have attempted to contribute to the understanding of black-pebble genesis, facies and sedimentology. We believe the following points to be of importance:

1. Reworked black pebbles and blackened limestone particles are found in shallow subtidal to supratidal facies, but occur mainly in the intertidal zone. They accumulate in storm deposits, beach conglomerates and tidal channels, or are admixed to lower-energy sediments. Black pebbles consist of subtidal, intertidal and supratidal sediments. Facies vary from normal-marine to brackish to freshwater limestones.
2. Impregnation of pre-existing sediments by *organic matter* in a reducing, slightly alkaline environment or microenvironment is the cause of the blackening. Pyrite may be present. We propose that, in the vadose or freshwater phreatic zone, dissolved, colloidal or very finely particulate organic substances favour aragonite to calcite inversion or replacement, and that they are adsorbed on carbonate-crystal surfaces. The *interaction of adsorption and early cementation* produces a microcrystalline matrix which fixes the organic matter. This process, as well as the mature state of the organic matter, makes the black coloration relatively resistant to oxidation.
3. Preferential early cementation of the blackened zones leads to a higher *preserva-*

tion potential than that of the host sediment. This allows for reworking and pebble formation by coastal erosion.

4. Black pebbles occur exclusively in *carbonate environments,* which is probably due to the above proposed fixation process.
5. The organic matter derived mainly from *algae and higher terrestrial plants,* which may also have been burnt by forest fires. This and the facies relationships (point 1) are evidence for the terrestrial influence on black-pebble formation.

As some authors have already pointed out (COTILLON 1960, WARD et al. 1970, BARTHEL 1974, FLÜGEL 1978), we believe that black pebbles are valuable keys to the *interpretation of ancient carbonate environments.* Through their size and conspicuous colour they are easy to recognize. They always indicate the presence of vegetated terrestrial areas. Even if found in subtidal facies with no indication of subaerial exposure, a nearby island or coastline can be postulated. Black pebbles might be also the only relics of eroded intertidal and supratidal sediments.

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