

AN UNUSUAL OCCURRENCE OF AMBER IN LAMINATED LIMESTONES: THE CRATO FORMATION LAGERSTÄTTE (EARLY CRETACEOUS) OF BRAZIL

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Abstract: Sub-ellipsoidal to irregular clasts of amber occur within millimetrically laminated limestones of the Nova Olinda Member, Crato Formation (Early Cretaceous, ?Aptian) of the Araripe Basin in Ceará, north-east Brazil. The amber is associated with resin-filled cones, foliage and palynomorphs attributed to the Araucariaceae and may be referred

to *Brachyphyllum* sp., cf. *Wollemlia* sp. and cf. *Agathis* sp. Irregular, septate tubular structures may represent micro-inclusions and are considered to be fungal hyphae.

Key words: Amber, Araucariaceae, *Agathis*, *Brachyphyllum*, *Wollemlia*, Cretaceous, Crato Formation, Brazil.

THE Crato Formation Konservat Lagerstätte of the Araripe Basin in north-east Brazil represents one of the most diverse fossil assemblages for the Early Cretaceous. Only the lowest part of the formation, the Nova Olinda Member, yields the famous well-preserved fauna and flora, although the entire formation is fossil-bearing (Martill and Wilby 1993). The assemblage of the Nova Olinda Member is largely allochthonous or parautochthonous, but transport appears to have been a result of floating in surface waters, and consequently it is extremely well preserved.

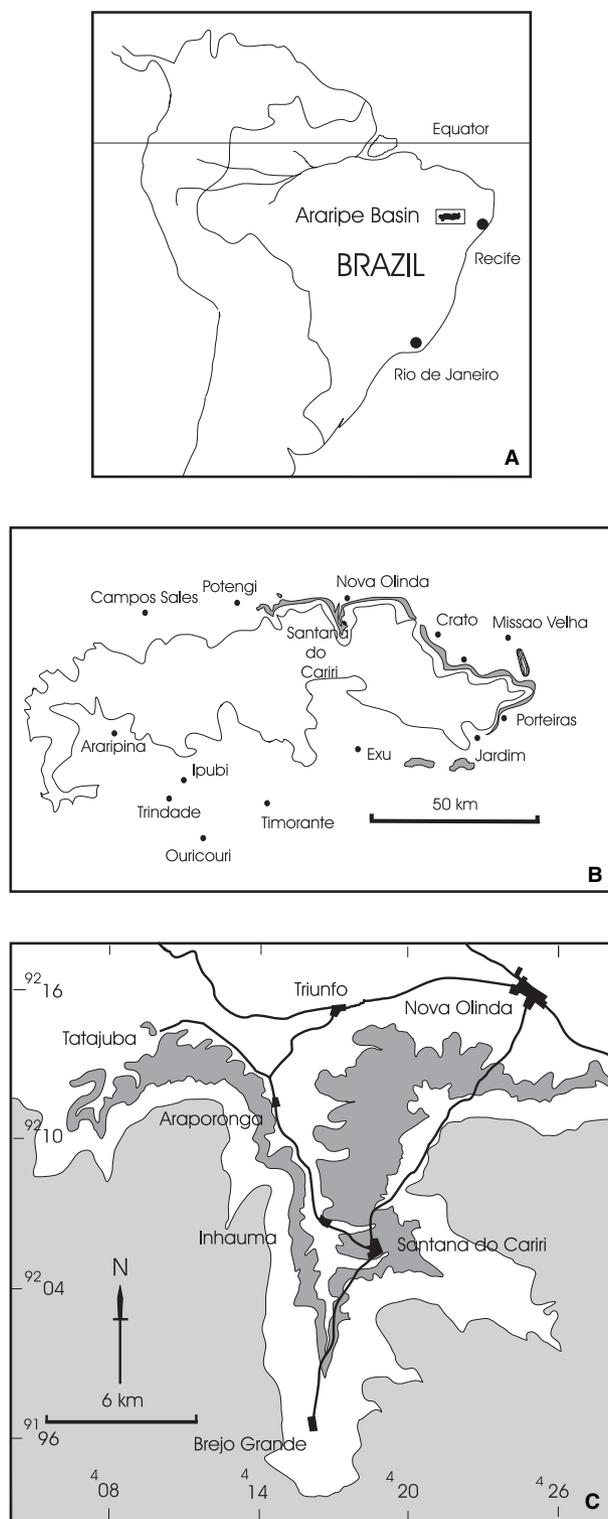
The assemblage includes vertebrates (Martill 1993), imago and larval insects (Martins-Neto 1992) and a variety of terrestrial arthropods including centipedes, spiders, scorpions and rarer aquatic crustaceans. Molluscs are absent from the Nova Olinda Member, but do occur elsewhere within the Crato Formation. By far the most abundant component of the macrofossil assemblage are remains of the small goniorhynchiform fish *Dastilbe crandalli* (Davis and Martill 1999). Other vertebrates are much rarer, but bizarrely, pterosaurs are the second most abundant.

The palaeoflora is diverse, comprising abundant macrophyte material as well as spores and pollen grains

(de Lima 1979). The flora has not been studied in detail, but it includes complete plants (roots, stems, leaves and fruiting bodies) of a variety of pteridophytes, gymnosperms, cycads, gnetaleans and angiosperms (Crane and Maisey 1991). Isolated fragments and worn pebbles of wood also occur, but large tree trunks are not reported, although they are known from other formations within the Araripe Basin (Martill 1993). We describe here the first substantiated occurrence of fossil resins, herein called amber, associated with araucarian macrophyte remains in the Nova Olinda Member.

LOCALITY

The amber specimens described here were obtained from quarry workers in the vicinity of Nova Olinda, Ceará, north-east Brazil (Text-fig. 1). Nova Olinda is one of several centres for stone production using laminated limestones that are quarried along an escarpment at the edge of a dissected plateau beneath the Chapada do Araripe (Martill 1990). Currently all of the working quarries are in the lowest limestone (Nova Olinda Member), but it is



TEXT-FIG. 1. A, map of northern South America showing location of Araripe Basin in north-east Brazil. B, outcrop of the Crato Formation around the Chapada do Araripe. C, map showing the outcrop of the Crato Formation in the vicinity of Santana do Cariri, Nova Olinda and Tatajuba in southern Ceará, from previously unpublished data.

not possible to give a precise location for the source of the specimens. Aspects of the matrix colour and texture can be somewhat informative. Dark grey to bluish grey rock is currently being excavated in quarries owned by Sr Anton Philippe near Tatajuba (GPS reading 07 07' 35"S, 039 49' 37"W), but it is also occasionally excavated in quarries in the valley of the Riacho Jacu between Nova Olinda and Santana do Cariri. Elsewhere in the region quarries are in weathered, buff-coloured rock. We were told that some of the specimens came from the Tatajuba quarries.

STRATIGRAPHY

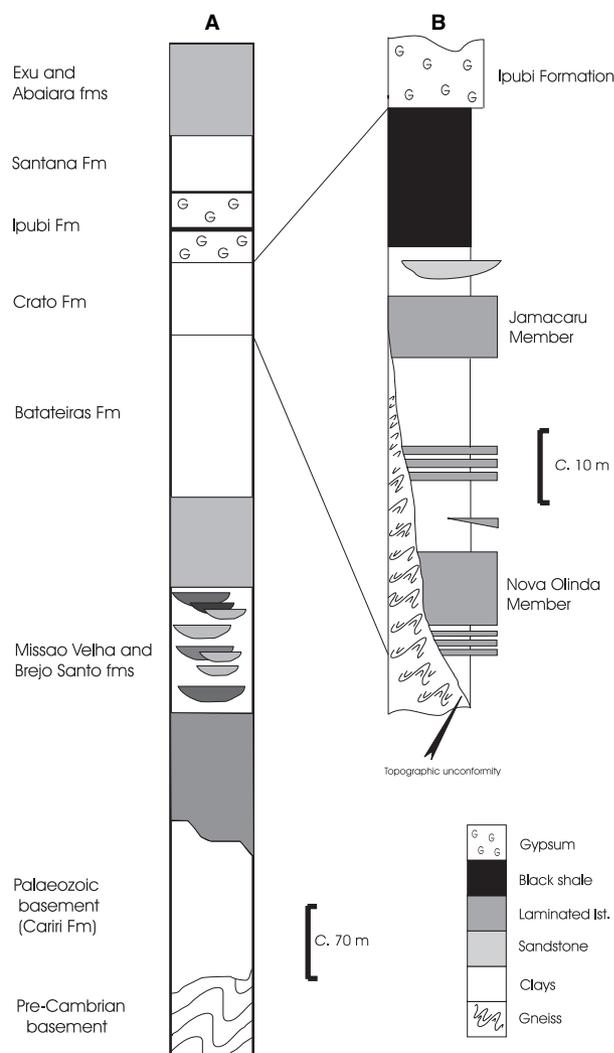
The Nova Olinda Member (*sensu* Martill and Wilby 1993) is the basal member of a series of interbedded limestones, clays, silts and fine sandstones (Text-fig. 2). Together they comprise the Crato Formation *sensu* Beurlen (1963) and were dated as Aptian by Pons *et al.* (1990). The Nova Olinda Member has a near-continuous outcrop on the northern and eastern flanks of the Chapada do Araripe, but its presence in the south and west of the Araripe Basin remains to be established (lithologically similar limestones are present in the south at Mamelocou, Pernambuco, but a direct correlation remains to be confirmed).

The Nova Olinda Member limestone comprises between as little as 2 m and as much as 13 m vertical thickness of fine millimetrically laminated limestones of alternating organic-rich and organic-poor lithologies. The sequence is uninterrupted except for occasional slumped horizons and irregular metre-scale, diagenetically altered zones that may represent subaqueous sulphur-rich springs.

The palaeoecology has been examined by a number of workers, but no consensus on the nature of the water body has emerged. Martill (1993) argued for a stratified water body with hypersaline bottom waters, while Maisey (1990), considered the limonitic style of preservation to be of primary origin, and consequently argued for a freshwater body to explain the presence of fossils preserved in hydrated iron oxides.

DESCRIPTION

Three pieces of amber were found during our field expedition of 2004 and four pieces were found by one of us (JAFGdeA) in 2003. The specimens described here are accessioned in the collection of the School of Earth and Environmental Sciences, University of Portsmouth, numbers prefixed UOP BR. The amber occurs as subspherical to angular clasts or as irregular masses between 7.5 and



TEXT-FIG. 2. A, schematic stratigraphic log for the Araripe Basin, Ceará, Brazil, based mainly on Martill and Wilby (1993). B, simplified stratigraphic log for the Crato Formation in the Nova Olinda/Santana do Cariri area. This sequence is highly variable within the Araripe Basin.

15 mm in diameter (Pl. 1, figs 1–6). They have been flattened slightly due to compaction. The colour varies from amber to dark brown, ruby-port coloured, and some appear black. Most pieces are crazed and have brittle fractures, some of which are conchoidal. In some examples the amber has become milky at the margins. Small fragments of black carbonaceous plant material occur in the surfaces of some pieces, while debris a few tens of micrometres in size can be seen in some of the clearer examples. Some fractures appear to be filled with white crystalline calcite. All of the pieces have been flattened somewhat in the plane of the bedding. Although no macroscopic inclusions have been located in the few pieces that can be examined internally, micro-inclusions of possible fungal hyphae occur in the

form of irregular, elongate, anastomosing septate tubes (Pl. 1, fig. 6).

ANALYSIS

The amber was examined under a stereomicroscope and using a petrological microscope on a ground section. The ground section was made by embedding a piece of amber within its matrix in a synthetic resin on one side and grinding down from the other side until the resin clast appeared as a ‘window’ in the piece of limestone. The grinding was repeated until the amber clast became transparent. One piece of amber was analysed by Fourier Transform Infra Red Spectroscopy (FTIR) in an attempt to characterise the fossil resin as outlined below.

Method. The amber was prepared by scraping pieces from a sample using a dental tool. The entire sample (185 mg) was then mixed with KBr and ground to a powder for pressing into a pellet, which was placed in a spectrophotometer and analysed. The pellet was then halved and reground with more KBr and reanalysed. This was repeated for a third time.

Results. An FTIR spectrograph was produced for sample UOP BR03cp/2004 and the results for wave numbers from 500 to 4000 are presented in Text-figure 3. Significant absorption occurred at wave numbers (cm^{-1}) 3412, 2925, 2860, 1710, 1626, 1450, 1245, 1190, 1025, 965, 860, 810 and 720. As in the Cretaceous ambers from Alava, northern Spain (Alonso *et al.* 2000) and the Isle of Wight, southern England (Nicholas *et al.* 1993), the Nova Olinda amber spectrum is dominated by C-H stretching bands near 2950 cm^{-1} and C-H bending between 1470 and 1380 cm^{-1} . Furthermore, prominent absorption due to carbonyl bands occurs near 1700 cm^{-1} and hydroxyl bands at 3500 cm^{-1} . Thus the FTIR spectrum of the Nova Olinda amber compares well with other Cretaceous ambers considered to be derived from representatives of the Araucariaceae. Significant differences in absorption properties between spectra from extant araucariacean resins and fossil ambers are attributed to diagenetic maturation (Alonso *et al.* 2000). We also draw comparison with published spectra from *Agathis* and *Pinus* species (Nicholas *et al.* 1993) and note that the Nova Olinda amber shows absorption at 3500 (Text-fig. 3A), which also occurs in *Agathis* (Text-fig. 3B), but is represented by a shoulder in *Pinus* (Text-fig. 3C).

TAPHONOMY

The presence of allochthonous amber clasts in a mid-basin laminated limestone is problematic. While it is

possible that resin-producing plants may have overhung the margins of the Nova Olinda Lagoon, currently all stone quarries are some distance from the palaeoshoreline (c. 1 km minimum for quarries near High Tatajuba; >2 km for quarries at Nova Olinda), and therefore an autochthonous/para-autochthonous origin can be ruled out. Fossil resins (amber) most commonly occur either as allochthonous primary deposits, having washed in from their immediate source area, or as secondary allochthonous deposits, having been reworked from forest soils (Rasnitsyn and Quicke 2002). The occurrence of allochthonous clasts of amber within a laminated limestone is, therefore, unusual and has not been reported previously.

While the Crato Formation comprises a heterolithic sequence representing lacustrine, fluvial and terrestrial environments, a feature of amber-bearing sequences (Grimaldi *et al.* 2000; Plint and Wadworth 2003), the Nova Olinda Member is uniform and represents a single sedimentary setting with little or no lateral lithological variation, at least in the Nova Olinda region. Most analyses conclude that the Nova Olinda Member was deposited in a lacustrine or lagoonal setting and represents an extensive body of water (Maisey 1990; Martill 1993). The Crato lagoon/lake was at least 75 × 50 km in the eastern Araripe Basin, and may have been considerably larger in extent. Controversy concerns mainly aspects of salinity, oxygen levels and water depth (e.g. contrast Martill and Wilby 1993 with Maisey 1990). Of concern here is how clasts of resin came to be deposited within a non-clastic sediment that was laid down some distance from a shoreline, and was not under the influence of current activity.

The Nova Olinda Member contains little or no clay material, and coarser clasts are absent, except at a few horizons close to the unconformity with the Proterozoic basement (Martill and Wilby 1993). There is no clastic component associated with the amber specimens described here other than an abundance of fossil material derived from the land (wingless terrestrial arthropods and plants). It therefore can be assumed that the amber clasts

must have either floated into the basin or drifted in with associated floating plant material. This latter mode of incorporation into the rock record has been documented for a number of primary amber occurrences where fossil resins occur in fluvial sediments (Nicholas *et al.* 1993). None of the pieces of resin found in the Nova Olinda Member is in intimate association with macro-plant remains, although some macrophyte material contains amber within its internal structure (Pls 2–3). Some of this material is extremely well preserved, and at least one cone contains amber within its resin canals (Pl. 2, figs 1–2). Large pieces of woody material are, however, rare. We suggest that although the resin described here could have been derived from pieces of wood that had undergone significant decay, with the amber dropping readily from the rotten plant tissue, flotation is also a possible mechanism.

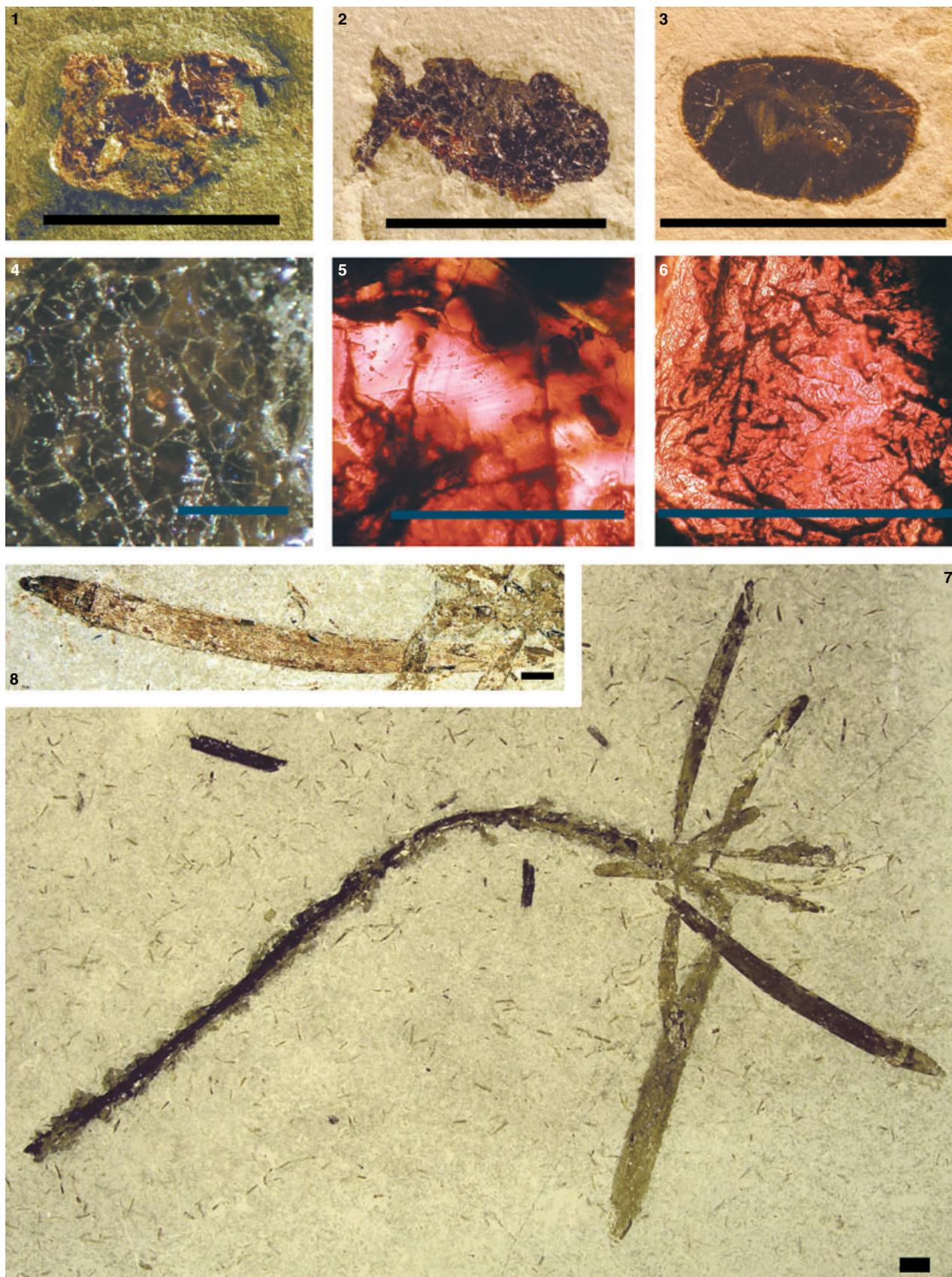
Floating resin may have drifted into the lagoon in a wind-generated current or under a weak fluvial influence. Furthermore, the amber probably floated in the surface waters, there being little evidence for benthic currents (contra Maisey 2000). This latter conclusion has implications regarding the salinity of the Nova Olinda Member (NOM) lagoon. Most resins sink in fresh water, but amber is known to float in sea-water, aided by its low-density composition and the presence of trapped gas bubbles (Rasnitsyn and Quicke 2002). In an experiment, Recent leguminous resin sank rapidly in hypersaline water. This might suggest that it is only amber that floats in sea-water, whereas prefossil resins sink. There is evidence to support a water body of elevated salinity for the NOM lagoon, including the absence of freshwater taxa such as unionid bivalves (these occur in the Crato Formation above the NOM), viviparid and other pulmonate gastropods, cypridid ostracods (which are also abundant both above and below the NOM), and conchostracans (abundant above and below the NOM); the super-abundance of the gonorhynchiform fish *Dastilbe* (Recent gonorhynchiforms are tolerant of high salinities) and the occurrence of rare fish genera that are elsewhere known from marine strata, including the ichthyodectid

EXPLANATION OF PLATE 1

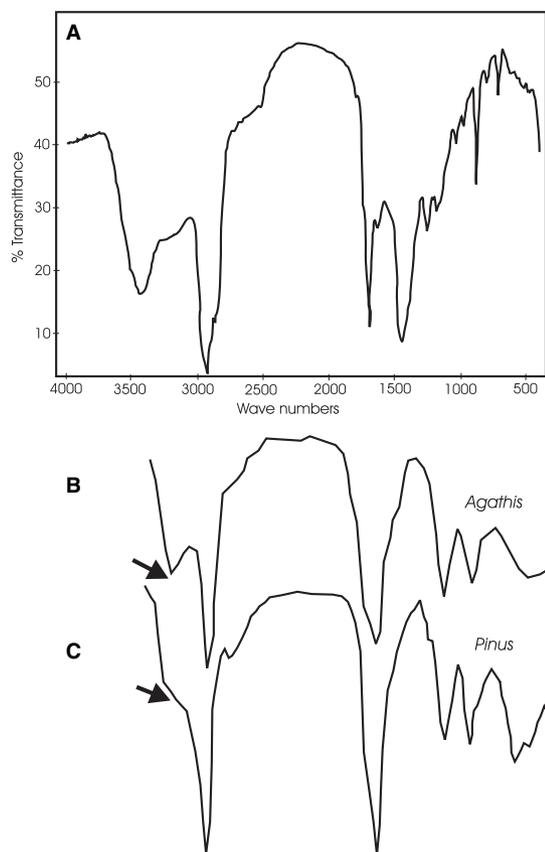
Figs 1–6. Amber clasts in laminated limestone of the Nova Olinda Member, Crato Formation (Early Cretaceous, Aptian), of southern Ceará, north-east Brazil. 1, UOP BR03cp/2004; 2, UOP BR02/2004; 3, UOP BR01/2004; 4, close-up of UOP BR02/2004 showing crazed surface; 5–6, UOP BR02cp/ts/2004, thin section seen in plane polarised light. In 6 a series of irregular tubular structures with cell-like septae may represent microinclusions of fungal hyphae. Scale bars represent 10 mm in 1–3, 1 mm in 4–6.

Fig. 7. UOP BR11p/2004, *Agathis*-like foliage on branch from the laminated limestone of the Nova Olinda Member. Scale bar represents 10 mm.

Fig. 8. UOP BR11cp/2004, counterpart of the darkest leaf seen in 7, to show the parallel veining. Scale bar represents 10 mm.



MARTILL *et al.*, 'Agathis', amber clasts



TEXT-FIG. 3. Infra red spectra for fossil amber and Recent *Agathis* and *Pinus* species for comparison. A, IR spectrum from fossil amber from the Nova Olinda Member of the Crato Formation (Aptian, Early Cretaceous), Araripe Basin, north-east Brazil. Analysis performed on specimen UOP BR03p/2004. B, IR spectrum for *Agathis australis* (Auracariaceae). C, IR spectrum for *Pinus echinata* (Pinaceae). IR spectra in B and C modified from Nicholas *et al.* (1993).

Cladocylus, also support an elevated salinity for the water body. While not conclusive, the restricted aquatic assemblage suggests a water body of unusual chemistry. Thus it is possible that the amber in the Nova Olinda Member represents an allochthonous secondary deposit that was reworked in a fluvial setting and introduced into the NOM lagoon at a delta. Being a very light material, the

amber would have drifted as part of the bed-load by gentle saltation. On entering the lagoon it may have floated in the hypersaline water body. The sediment load however, would have flocculated in the saline water and fallen out in the distal parts of a delta (a silty mud rich in plant debris occurs underneath a thin development of the NOM at Cascata, near Crato). The amber would have continued to float until it became thoroughly water-logged. Alternatively the resin pieces may have drifted down gentle slopes towards basin lows by the effects of gravity.

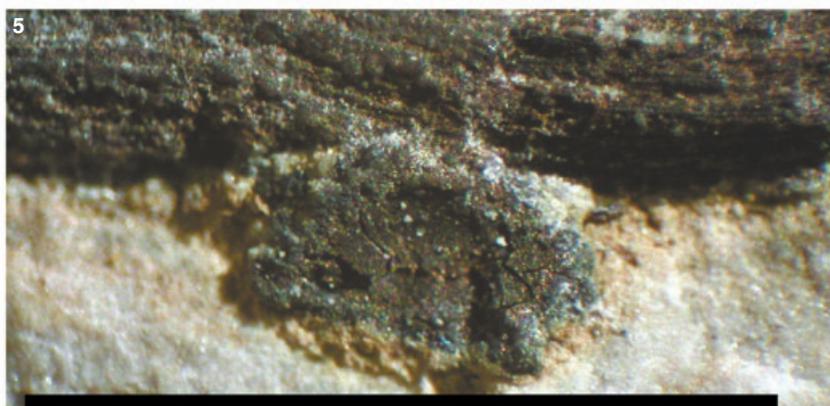
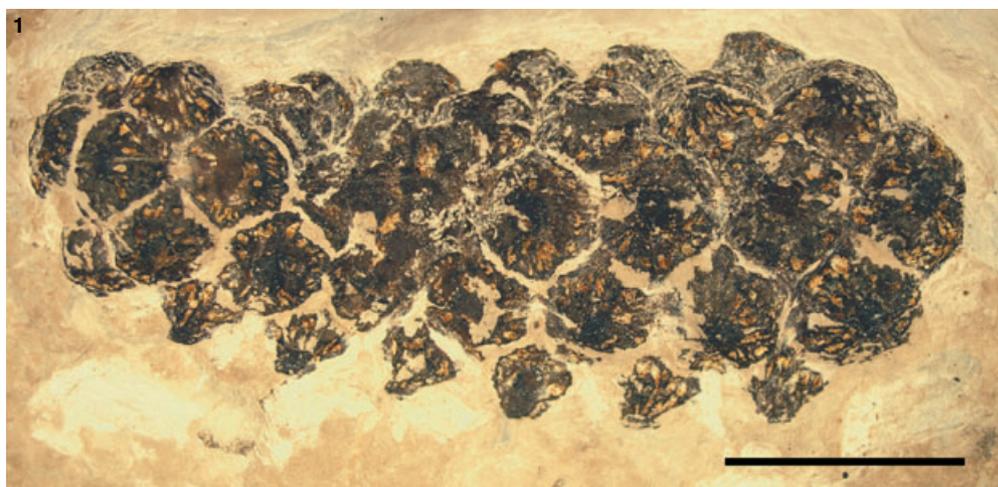
ASSOCIATED MACROPHYTE REMAINS

Larger plant remains can be abundant in the Nova Olinda Member, though few have been described in detail. The flora includes a diversity of gnetaleans, gymnosperms and angiosperms (see Mohr and Friis 2000; Mohr and Rydin 2002; Mohr and Eklund 2003; Rydin *et al.* 2003). Although most plant remains are oxidised, orange-brown and rather powdery, black carbonised remains occur occasionally in the unweathered rock. Although plant fragments are usually encountered, more complete remains occur frequently, including stems with foliage and fruiting bodies. Entire plants are also found, and rarely a plant may occur in which soil is still attached to the root ball (Martill, pers obs.). Although the macrofloral assemblage is diverse, few species have been described. By far the most abundant and readily identifiable remains are portions of branches of the araucarian *Brachyphyllum* (Duarte 1985, 1989). These occur as broken tips of branches, usually bearing several smaller branches clad in tightly pressed, 'fleshy' leaves (Pl. 3, figs 1–4). Despite the abundance of this taxon, we have only discovered one piece of *Brachyphyllum* bearing resin (Pl. 3, figs 3–4). We have been able to recognise several types of cone, including both male and female forms (Pl. 3, figs 1–4), which could be attributed to the Araucariaceae, although not to any particular genus with certainty. Foliage comprising narrowly lanceolate, parallel-veined leaves spirally attached to the distal portions of bare stems resembles the foliage of the extant araucariacean *Agathis australis* (Kauri Pine) (Pl. 1,

EXPLANATION OF PLATE 2

Figs 1–2. UOP BR09/2004, male cone of *Agathis*-like araucariacean gymnosperm with amber in resin canals. 1, complete cone preserved probably prior to release of pollen. 2, detail of two cone scales showing yellowish amber within the resin canals. Scale bars represent 10 and 1 mm respectively.

Figs 3–5. UOP BR10/2004, female cone of possible araucariacean gymnosperm. 3, stem with terminal cone and two parallel-veined leaves. 4, detail of cone. 5, detail of part of stem with possible exudation of resin. Scale bars represent 10 mm.



MARTILL *et al.*, coniferalean cones

figs 7–8). The stem of this specimen (Pl. 1, fig. 7) contains small quantities of resinous material. The foliage also resembles that of *Podocarpus* and the ginkgoalean *Zamia*, but the preservation of resin canals implies affinities with the Araucariaceae, and more specifically *Agathis* sp. We have been unable to find cones and leaves associated. On one staminate cone, resin is preserved as yellowish to black amber in resin canals within the cone scales (Pl. 2, figs 1–2).

A single female cone with stem and small parallel veined leaves (Pl. 2, fig. 3) morphologically resembles the recently discovered extant Australian araucarian *Wollemia* (Chambers *et al.* 1998). There are very small quantities of resin in both the cone scales and the stems and leaves. A patch of organic material adjacent to the main stem (Pl. 2, fig. 5) appears slightly resinous, and may have been an exudate.

Besides macrophyte material, de Lima (1979) recorded three species of pollen attributed to the Araucariaceae; *Araucariacites australis* Cookson, *A. limbatus* (Balme) Habib and *A. guianensis* van der Hammen and Burger, but it is unclear exactly in which horizon these pollen occur.

DISCUSSION

Amber has been reported previously from the Araripe Basin. Castro *et al.* (1970) noted it from an unspecified locality and horizon near Porteirias at the eastern end of the Chapada do Araripe, while Cardoso *et al.* (1999) described amber which they claimed came from the Crato Formation of Santana do Cariri. No figures or reliable evidence has been supplied to support either of these claims.

The botanical source of the Nova Olinda Member amber is most likely to be the Araucariaceae. Typically, Cretaceous ambers derive from araucarian trees such as *Agathis* sp. (Langenheim 1995), although Cretaceous occurrences of amber derived from dicotyledonous angiosperms have been reported (Grimaldi *et al.* 2000; Langenheim 2003). The common occurrence of araucariacean branches and fruiting bodies in the same strata as the isolated amber, and the occurrence of resin within a cone, are good evidence for an association

and, therefore, an araucarian origin for the amber. Although it remains to be demonstrated which cones match the foliage specimens, it is likely that the elongate leaves resembling *Agathis* sp. belong with the highly resinous cone in Plate 2, figures 1–2 on the basis of their morphology, size and the presence of resin in the leaves.

Descriptions of the occurrence of Early Cretaceous ambers are few, though there are many accounts of fossil inclusions within them (e.g. Azar *et al.* 1998; Poinar and Milki 2001). The most notable occurrences are those of the Lebanon and Jordan (Cano *et al.* 1993; Azar *et al.* 1998), and the Nograro Formation, Alava, Basque Country (Alonso *et al.* 2000). Fossil-bearing ambers also occur in the Early Cretaceous (Barremian) Wessex Formation of the Isle of Wight, England (Nicholas *et al.* 1993; Selden 2001).

Of note is the occurrence of this amber in the Southern Hemisphere. Grimaldi *et al.* (2002) pointed out that most major occurrences of Cretaceous amber are known from the Northern Hemisphere (Burma, Lebanon, New Jersey, Spain, western Canada, Siberia). Although the Lebanese ambers are of Gondwanan origin, the amber locality described here, although not representing a major occurrence, is a rare example of Cretaceous amber in Gondwana.

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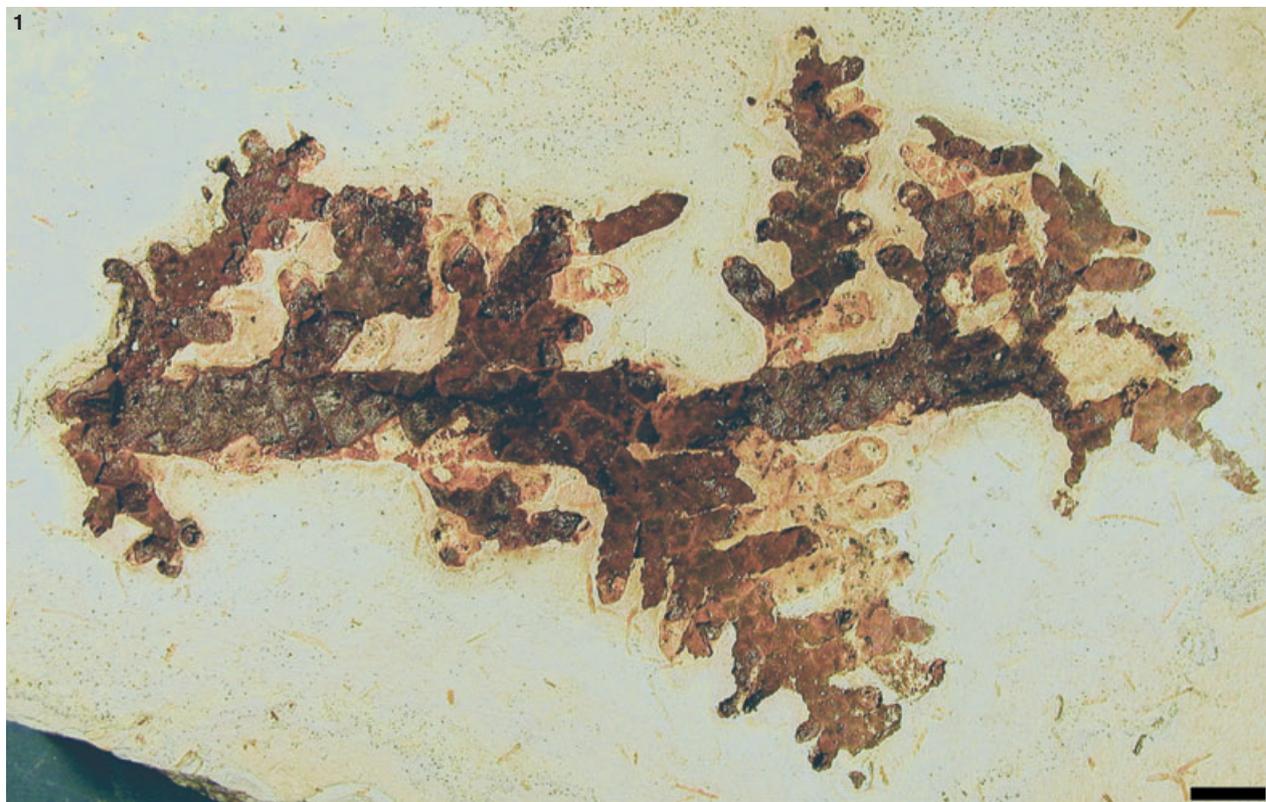
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EXPLANATION OF PLATE 3

Figs 1–2. *Brachyphyllum* sp. Nova Olinda Member, Crato Formation, Ceará, Brazil, UOP BR07/2004. Specimen preserved in highly oxidised state with no organic matter remaining. 1, terminal fragment of branch with numerous sub-branches. 2, detail of leaves. Scale bars represent 10 and 5 mm respectively.

Figs 3–4. *Brachyphyllum* sp. Nova Olinda Member, Crato Formation, Ceará, Brazil, termination of sub-branch preserved in unoxidised state with carbonaceous material and amber beneath leaves. 2, detail of area where several leaves have been removed to reveal amber. Scale bars represent 5 mm.



MARTILL *et al.*, *Brachyphyllum*

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