SUPPLEMENTARY DATA 2: SEDIMENTOLOGY

METHODS

Outcrop at the Barrio de las Minas typically comprises short (< 3 m) sections exposed within man-made hillside caves (Supp. Data 2, Fig. 1). Where possible, the approximate abundance of shelly bioclastic elements, seeds, twigs and leaves was classified using the following scale: (1) very rare (occurring as isolated elements); (2) rare (majority of elements > 50 mm apart); (3) common (majority of elements 10-50 mm apart); and (4) abundant (majority of elements < 10 mm apart) (Supp. Data 2, Fig. 2A-C). For elongate plant fragments ('plant hash') the fragments' abundance was recorded as follows: (1) very rare (occurring as isolated elements); (2) rare (majority of elements >10 mm apart); (3) common (majority of elements 3-10 mm apart); (4) abundant (majority of elements < 3 mm apart) (Supp. Data 2, Fig. 2D-F). Their average length was recorded as follows: (1) very fine (< 1 mm in length); (2) fine (1-3 mm in length) (3) intermediate (3-5 mm in length); (4) coarse (> 5 mm in length) (Supp. Data 2, Fig. 2G-I).

RESULTS

The following facies were identified in the bituminous-calcareous subunit: (1) charophyte-rich packstones and wackestones; (2) charophyte-rich siltstones; (3) bioturbated mudstones; (4) weakly banded marls; and (5) laminated mudstones. Representative lithological sections are shown in Supp. Data 2, Fig. 3. Summaries of the lithologies of the non-laminated facies are presented in Supp. Data 2, Table 1. The sedimentology of the laminated mudstones, the fossil-bearing facies, is considered in detail below. Percentages quoted in the text for the abundance of microscopic components are based on 250-300 point counts.

Detailed description of lamina types

Micritic laminae (Supp. Data 2, Fig. 4A, D) comprise predominantly micrite (80-93%) and bioclasts (5-15%). Macroscopic bioclasts (*Planorbis* and bivalves) are usually absent, but occur rarely as dense concentrations with adjacent bioclasts in contact. *Ruppia* seeds and very fine to fine plant hash are rare or absent. Microscopic bioclasts are predominantly sponge spicules (83%); fragmented ostracods, gastropods and charophyte stems are rare. Microfloral remains comprise chrysophycean algae and cysts, and rare, highly fragmented frustules of the diatoms *C. placentula*, *N. radiosa* and *S. construens* (Supp. Data 2, Fig. 4D).

Organic-rich laminae (Supp. Data 2, Fig. 4B, E-G) comprise predominantly organic matter (72-83%) and micrite (8-20%). Macroscopic bioclasts (*Planorbis* and bivalves) are often absent but, where present, are common to abundant. *Ruppia* seeds are usually present and can occur in very dense concentrations, often with adjacent seeds in contact. Fine to intermediate plant hash (1-5 mm long) is common. Microscopic bioclasts are rare to absent (0-2%) and comprise predominantly sponge spicules (85-99%); ostracods (1-15%) also occur. Accumulations of well-preserved (i.e. exhibiting little fragmentation and/or dissolution) examples of the diatoms *C. meneghiniana, S. leptostauron* and *S. pinnata* occur as both near-monospecific, and mixed, assemblages within many of the dark gray laminae (Supp. Data 2, Fig. 4E-F) and are often associated with a structureless carbonaceous material. Fragments of *B. braunii* colonies are rare (Supp. Data 2, Fig. 4G).

Silty laminae (Supp. Data 2, Fig. 4C, H-I) comprise organic matter (48-69%), micrite (20-32%), detrital quartz (5-8%) and bioclasts (12-20%). Macroscopic bioclasts are predominantly concentrations of highly fragmented shelly material (each fragment < 2 mm); entire examples of *Planorbis* are common, and bivalves rare. *Ruppia* seeds are often present and, if so, usually common. Plant hash is almost always present, and is usually intermediate to coarse and common to abundant. Microscopic bioclasts are primarily sponge spicules; chrysophycean algae and fragmented frustules of the diatoms *S. construens* and *N. radiosa* are rare (Supp. Data 2, Fig. 4H). As in the dark gray laminae, these diatoms can be associated with structureless carbonaceous material (Supp. Data 2, Fig. 4I). Silt-sized monocrystalline and polycrystalline detrital quartz is typically concentrated in discrete horizons at, or close to, the base of these laminae.

Alternations of these lamina types define the following facies. Facies A comprises intervals (typically 0.6-1.5 m thick) of white to light gray laminated mudstones (Fig. 3A, main manuscript). Micritic laminae predominate and average 2.4 mm thick; silty and organic-rich laminae are rare and, when present, very thin (≤ 0.3 mm thick). Facies B comprises intervals (typically 0.4-1 m thick) of organic-rich laminated mudstones (oil shales) that are characterized by the high abundance of organic-rich laminae (Fig. 3B, main manuscript). Subdivision of this facies is based on the relative abundance of micritic and silty laminae. Facies B_1 comprises alternations of micritic (average 0.83 mm thick) and organic-rich laminae (average 0.65 mm thick) (Fig. 3C, main manuscript); silty laminae are rare. Facies B_2 comprises alternations of silty (average 0.96 mm thick) and organic-rich (average 0.75 mm thick) laminae (Fig. 3D, main manuscript); micritic laminae are rare and, if present, less than 1.2 mm thick.

Interpretation of Depositional Environment

Seasonal origin of lamination: Laminated mudstones are characteristic of deep-water lacustrine sedimentation and routinely form the basis of high-resolution (on annual to sub-annual scales) studies of climatic, ecological and environmental variability (e.g. Burke et al., 2002; Chang et al., 2003; Pietras et al., 2003; Ojala et al., 2005; Stebich et al., 2005). Not all laminated sediments, however, archive seasonal deposits: certain pulses of open-water sedimentation, e.g. storm deposits, can be independent of seasonal cycles (Dean et al., 1999). Before laminated lacustrine sediments may be considered to archive seasonal events, the depositional environment must exhibit the following characteristics (Cohen, 2003): (1) a stratified water column; (2) minimal bioturbation; (3) minimal bottom currents; (4) minimal disturbance of lamination by microbially-induced gas formation; (5) a seasonally variable flux of epilimnetic material; and (6) rates of particle settling sufficiently high to permit transmission of this seasonal signal to the lake floor.

Several factors indicate that the Libros lake was strongly and permanently stratified during deposition of the laminated mudstones. Delicate millimetric- and sub-millimetric lamination is preserved, indicative of environmental anoxia (preventing infaunal colonization) and weak to absent bottom currents. The carbonates associated with the oil shales exhibit depressed $\delta^{13}C$ values (Anadón et al., 1992), characteristic of extensive sulfate reduction. The oil shale hopanoid distribution comprises predominantly odd alkylthiophenes, including gammacerane and trisnorhopane, biomarkers characteristic of a stratified water column, and a sulfidic monimolimnion, respectively (Sinninghe Damsté et al., 1995; de las Heras et al., 2003). These factors, combined with faunal and microfloral data, indicate that the Libros laminated mudstones represent profundal sediments deposited under dysoxic to anoxic conditions during periods of stable lake stratification in an oligohaline, oligotrophic lake phase.

Alternation of laminae of varying composition (rhythmites) is characteristic of hemipelagic deposition in which the flux of minerals and organic matter to the lake floor is seasonally variable (Chang et al., 1998; Dean et al., 1999; Chang et al., 2003; Cohen, 2003). Carbonate / organic couplets similar to Facies B_1, and clastic/organic examples similar to Facies B_2, have been recorded in numerous other modern, and fossil, non-glacial lakes and considered to be seasonal in origin (e.g. Donovan, 1980; Anadón et al., 1989; Lamb et al., 1995; Utrilla et al., 1998). These factors, combined with the presence of well-preserved monospecific diatom assemblages (indicative of high rates of particle settling), confirm that the Libros laminated mudstones can be considered to archive seasonal events.

Origins of the different lamina types: Micritic laminae represent deposition during low rates of organic matter accumulation and/or production. High rates of carbonate production are typical of early spring in subtropical lakes: photosynthetic abstraction of CO_2 by phytoplankton results in increases in pH and calcium carbonate saturation, and induces carbonate precipitation (Talbot and Allen, 1996). The paucity of diatoms and the presence of chrysophycean cysts indicate deposition during winter and/or early spring (Chang et al., 2003; Pla and Catalan, 2005). The most plausible origin for these laminae is therefore deposition of epilimnetic carbonate precipitates during early spring.

Organic-rich laminae represent deposition during high rates of organic matter production and/or accumulation. Near-monospecific accumulations of well-preserved *C. meninghiana* and *F. pinnata* represent rapidly sedimented late summer / autumn blooms (Gil, 1986; Leland et al., 2001; Margalef, 1947); mixed diatom assemblages are also characteristic of late summer/autumn deposition (Dean et al., 1999; Chang et al., 2003). Seed production in *Ruppia* occurs primarily between May and August (Malea et al., 2004) (although small quantities of seeds can be present on plants throughout the year (Cho and Poirrier, 2005)). Hence the organic-rich laminae are best interpreted as deposition during mid-summer to autumn. The oil shale kerogen is predominantly of algal origin, with inputs from higher plants and bacteria (de las Heras et al., 2003); the organic matter in these laminae (and the carbonaceous amorphous material often associated with the diatoms) most likely represents an organic "sludge" (Trewin and Davidson, 1996, p. 238) derived from decay of autotrophic algae.

Silty laminae reflect increases in precipitation and/or runoff into the lake during autumn/winter flood and/or storm events, as evidenced by their high terrigenous component and the paucity of diatoms and micrite.

Facies interpretations: Facies A is almost completely devoid of organic matter, and organic-rich laminae are rare; deposition occurred during periods of low primary productivity, and/or low rates of accumulation of organic matter. The latter could result from high oxygen levels, resulting in elevated oxygenation of organic matter, and/or from increased grazing by epilimnetic zooplankton. Neither scenario is plausible, however: low oxygen levels during deposition of Facies A are implied by the preservation of fine lamination in the sediments, and there is no evidence (such as a high abundance of faecal pellets) for increased grazing by zooplankton. Lower primary productivity during the deposition of this Facies is therefore the most likely. *B. braunii* is the primary contributor to sedimentary organic matter in many Phanerozoic lacustrine deposits (Derenne et al., 1988, 1997; Grice et al., 1998; Sinninghe Damsté et al., 1993). It is widespread in modern, oligotrophic to eutrophic, temperate and tropical (and, to a lesser extent, arctic) habitats (Wolf and Cox, 1981). The high abundance of *B. braunii*-derived amino acids in the Libros kerogen (del Río et al., 2004), and the presence of *B. braunii* colonies in the organic-rich laminae, suggests that this alga may have contributed significantly towards the formation of the Libros kerogen. Despite its broad temperature (20-27ºC) and salinity (0-8.8%) optima (Qin, 2005), growth of *B. braunii* is depressed at pH >8-8.5 (Belcher, 1968; Sánchez et al., 2005). These conditions could have been achieved during deposition of Facies A by evaporitive concentration of lake waters during relatively warm, arid, climatic periods. Carbonate precipitation would have been enhanced during deposition of this Facies: high temperatures and solute concentrations (especially carbonate) each reduce the solubility of calcium carbonate. As a euryhaline and eurythermal species, *Ruppia* would have been little affected by fluctuations in temperature and/or salinity. The paucity of *Ruppia* seeds in Facies A likely reflects restricted littoral habitats, i.e. low lake levels.

Facies B_1 represents seasonal fluctuations in environmental conditions between periods of low organic input (micritic laminae: early spring deposition) and periods of high organic input (organic-rich laminae: late summer-autumn deposition). This could originate via seasonal variation in runoff, and thus precipitation. This facies represents deposition during relatively humid, seasonal periods: high precipitation levels result in a high lake level, stabilising stratification and positioning the anoxic-oxic boundary within the water column; these features facilitate accumulation of organic matter on the anoxic lake floor (Anadón et al., 1991).

During deposition of Facies B_2, organic matter flux to the lake floor was consistently high (relative to Facies A and B_1) throughout the year. The high abundance of speckled gray laminae indicates flooding and / or storm events were common during both spring and autumn/winter. Littoral zones were extensive, as evidenced by the high abundance of *Ruppia* seeds. Hence this Facies represents deposition during more humid conditions than Facies A, in a relatively deeper, more dilute lake. The absence of carbonate laminae from the annual dark gray-speckled laminae couplets could reflect lower rates of carbonate production and/or accumulation of carbonate on the lake floor. Lower temperatures and dilution of the lake waters would have increased the solubility of calcium carbonate, mitigating against its precipitation; further, the presence of a deeper (relative to Facies A and B_1), acidic, monimolimnion would have enhanced dissolution of carbonate precipitates as they sank from the epilimnion. The similar composition and abundance of bioclasts in each facies reflects the euryhaline nature of the former (Berezina, 2003).

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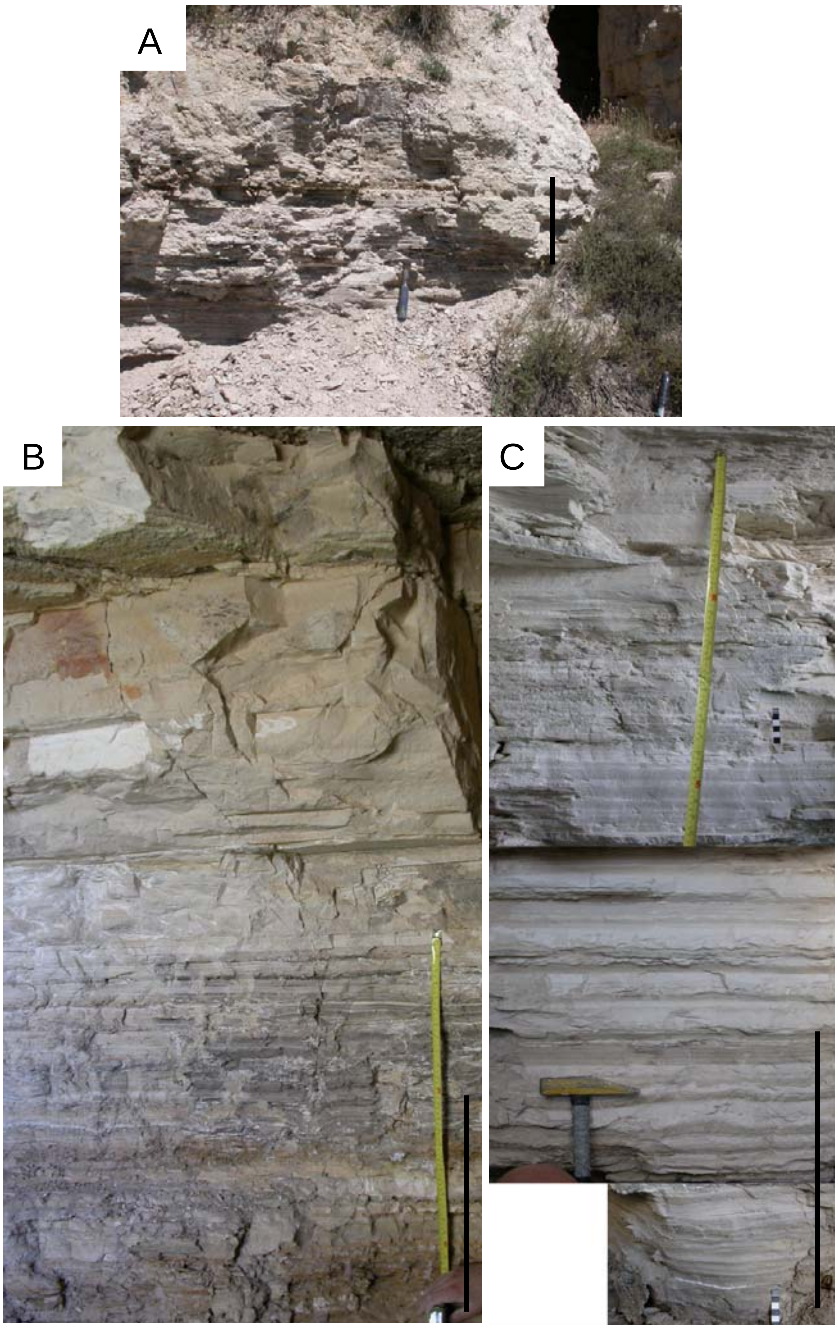

FIGURE 1–Photographs of selected sections of the bituminous-calcareous subunit of the Libros Gypsum Unit, Barrio de las Minas. Scale bars, 500 mm.

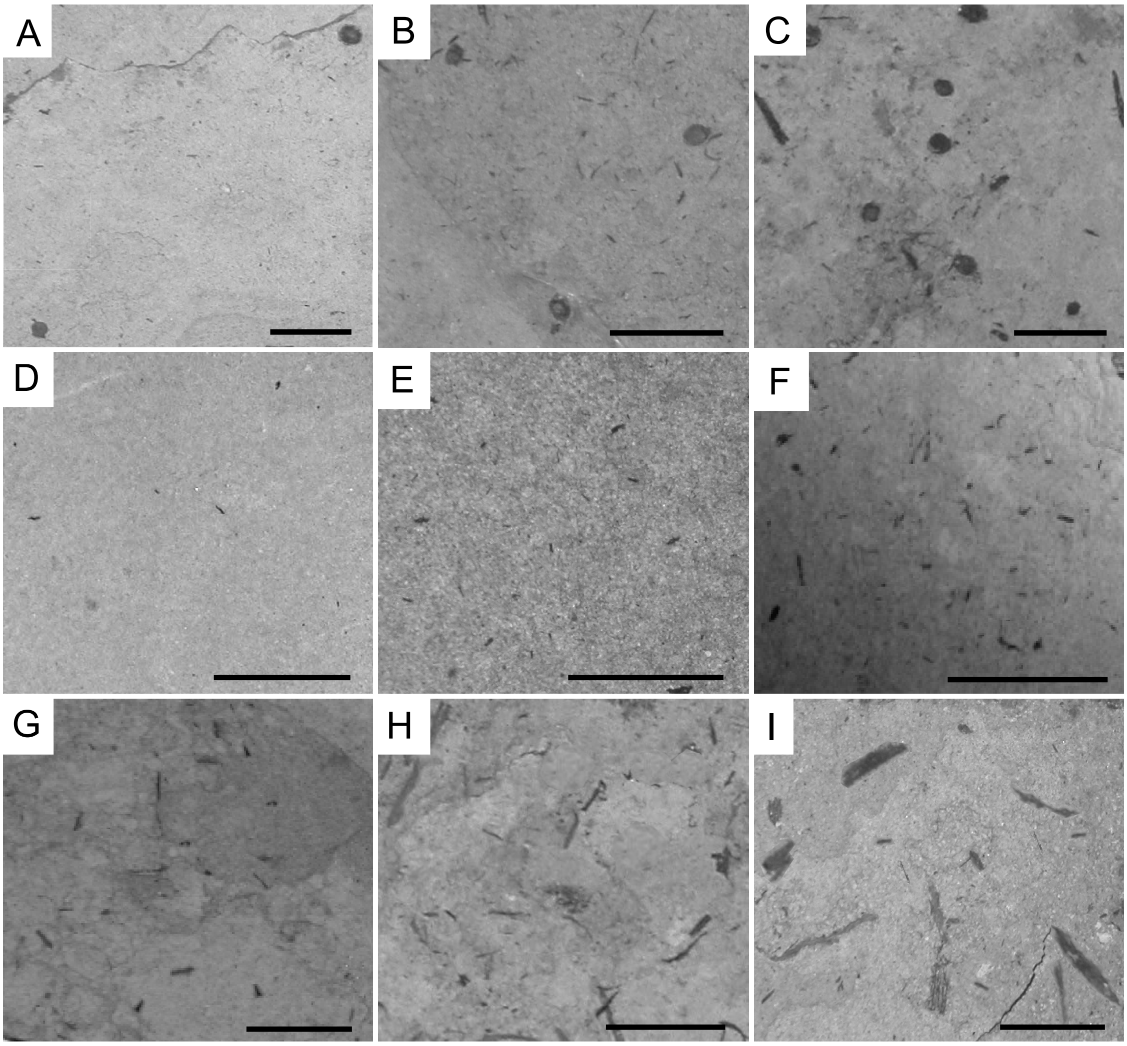

FIGURE 2—Classification scheme for the abundance and, for plant hash, fragment size, of macroscopic bioclastic and vegetal material present in the calcareous-bituminous sub-unit. A-C: Abundance of a discrete category of bioclast or plant material (here *Ruppia* seeds); rare (A); common (B); abundant (C). D-F: Abundance of plant hash; fragments in each of D-F are very fine in size and are rare (D), common (E) and abundant (F) respectively. G-I: Hash fragment size; fragments in each of G-I are common and are fine (G), intermediate (H) and coarse (I) in size. Scale bars, 10 mm.

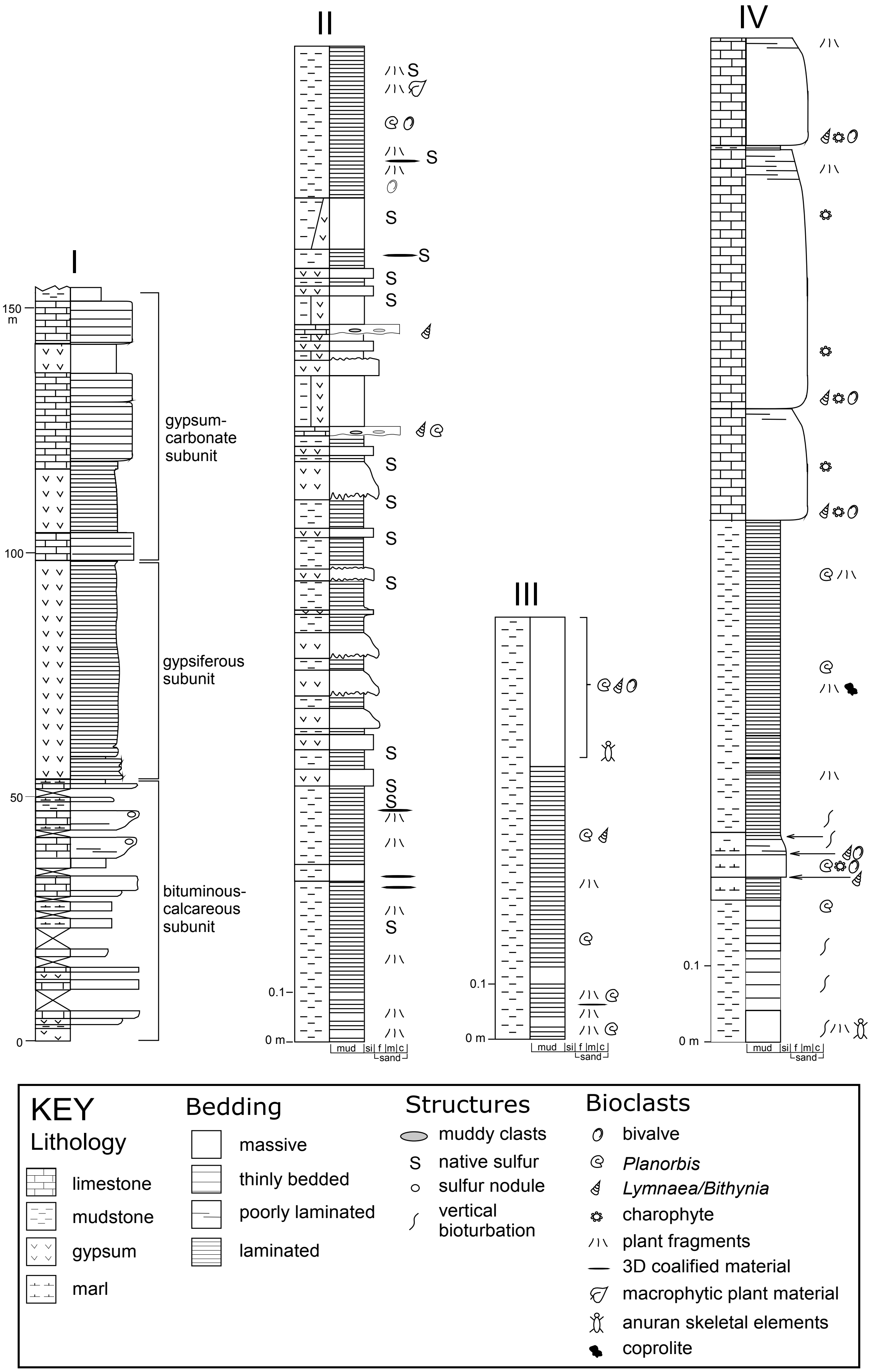

FIGURE 3-Representative lithological sections of the bituminous-calcareous subunit. Section I is modified from Fig. 4 in Ortí et al. (2003). Sections II, III and IV are shown in Supplementary Data 2 Fig. 1A, 1B and 1C, respectively.

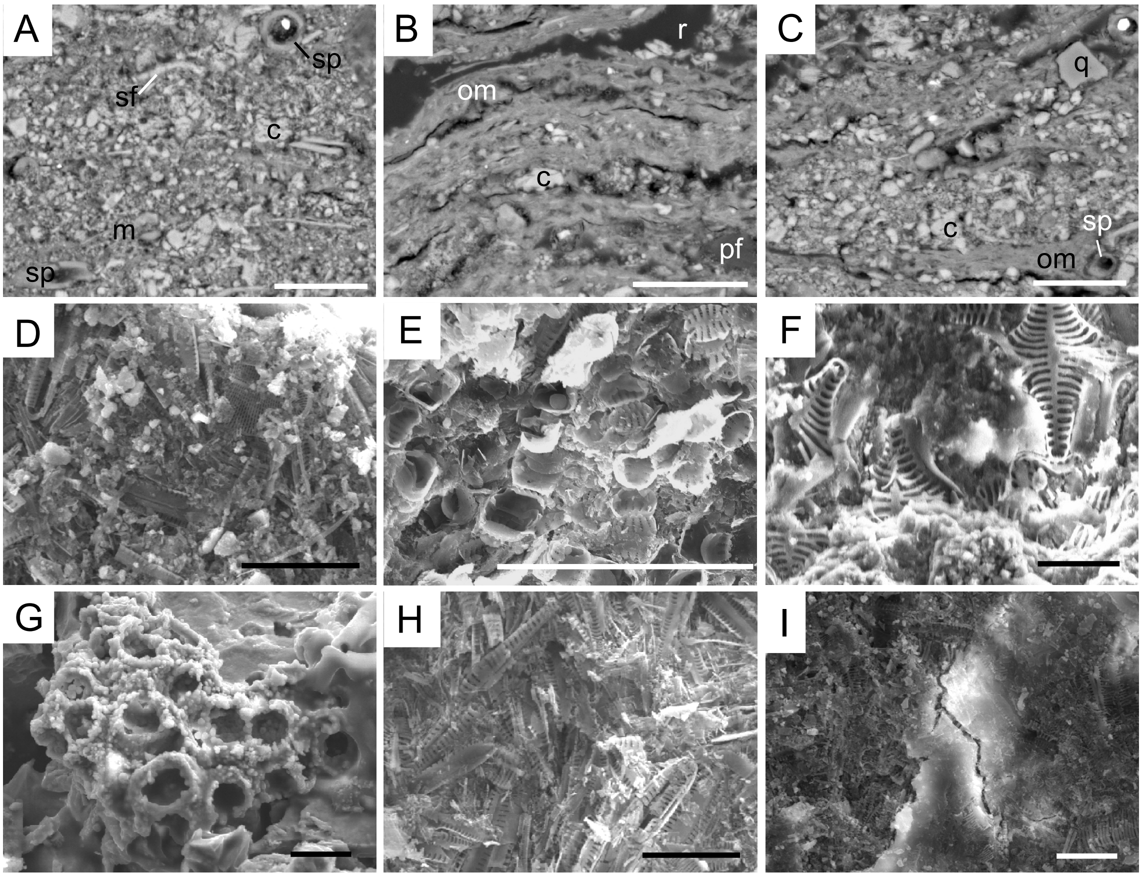

FIGURE 4. Lamina types (A-C) and microfloral remains (D-I) of the laminated mudstones. A-C: Backscattered electron micrographs of micritic (A), organic-rich (B) and silty (C) laminae. c, carbonate grain; m, micrite; om, organic matter; pf, carbonaceous plant fragment; q, quartz grain; sf, shell fragment; sp, sponge spicule; r, resin. D-I: Scanning electron micrographs of microfloral remains. D: Fragmented frustules of the diatoms *Cocconeis placentula* and *Navicula radiosa* from a micritic lamina. E, F: Well-preserved, monospecific assemblages of *Staurosira construens* (E) and *Staurosirella leptostauron* (F) from an organic-rich lamina. G: Fragment of a *Botryococcus braunii* colony from an organic-rich lamina. H, I: Fragmented frustules of *N. radiosa* from silty laminae; associated amorphous material charging in (I) is carbonaceous in composition. Scale bars, 20 µm.

Facies	*Colour*	*Bedding*	*Primary components*	*Other features*	*Interpretation*
Bioclastic packstones and wackestones	Cream	Thin to thick Fine upwards Erosively based	Micrite (12% - 85%) Bioclasts (12-40%; charophyte stems >> gastropods)	Porous (3-22% porosity) with vugs up to 8 mm diameter Muddy intraclasts up to 12 mm diameter Rare sulphur nodules	Turbiditic underflow deposits derived from a more proximal charophyte bench during floods / storms
Bioclastic siltstones	Light green to reddish brown	Thin Fine upwards Erosively based	Silt-sized carbonate (82-91%) Bioclasts (8-15%; charophytes > plant fragments) Silt to sand-sized quartz (0-2%) Finely disseminated haematite (0%-6%)	Very dense accumulations of fragmented charophyte stems (up to 15 mm length) and overlapping shell fragments at bases of beds	Terrigenous input during flood / storm events
Bioturbated mudstones	Light green-brown to cream	Intervals 0.3 - 0.7 m thick	Micrite (62%-96%) Bioclasts (0%-6%) Finely disseminated haematite (16-30%) Silt-sized quartz (0%-2%)	Abundant vertical burrows (0.5-2.2 mm diameter) overprint a homogenised texture Common horizontal burrows (0.5-2 mm diameter) Rare isolated anuran skeletal elements	Background deposition during relatively arid periods when low lake levels precluded stable lake stratification and promoted oxygenation of the lake floor; some terrigenous input
Banded marls	Cream	Thin to medium	Micrite (90%-94%) Bioclasts (2%-5%)	Banding (10-15 mm thick bands) reflects variations in bioclastic content, amount of calcite cement and porosity	Background deposition during a low lake level phase without terrigenous input

TABLE 1—Non-laminated facies in the bituminous-calcareous subunit.