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The massive sulphide event in the Iberian Pyrite Belt: confirmatory evidence from the Sotiel-Coronada Mine

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Abstract – Well-preserved miospore and organic-walled microphytoplankton assemblages have been recovered from the black shaly series hosting the massive sulphide deposits of Sotiel-Coronada Mine (Iberian Pyrite Belt). The productive samples yielded miospore assemblages representing the uppermost Famennian *Retispora lepidophyta–Verrucosisporites nitidus* (LN) miospore Biozone of Western Europe. This palynological evidence has important implications for the local geology, constraining the commencement of the volcanic activity and corroborating the previously-defined local tectonic style. Moreover, at regional scale, the new data permit correlation of the black shaly series (which hosts the mineralization in all the sulphide deposits hitherto dated in the region), reinforcing the hypothesis of an anoxic event occurring in the Iberian Pyrite Belt in the latest Devonian times.

Keywords: Devonian, Iberian Pyrite Belt, Sotiel-Coronada Mine, Spain, palynology, black shales.

1. Introduction

The Sotiel-Coronada Mine occurs within the Iberian Pyrite Belt, which is located in the southwestern corner of the Variscan Iberian Massif. This basin is defined by a belt 200 km long and 40 km wide comprising Middle Devonian–lower Pennsylvanian sedimentary, volcanic and volcaniclastic rocks which, following the Variscan main structures, extends from south of Lisbon in Portugal to Seville in Spain (Fig. 1). The Iberian Pyrite Belt is considered one of the major massive sulphide provinces in the world, including more than 80 known deposits, whose estimated original reserves exceed 1700 Mt. It contains giant and supergiant deposits as well known as Riotinto, Tharsis, Aznalcóllar, Sotiel-Coronada, Aljustrel and Neves-Corvo. The massive sulphide deposits of the Iberian Pyrite Belt are of Iberian type (Sáez *et al*. 1999), and most of them are commonly included within, or closely associated with, black shale levels. Both black shales and sulphide sedimentation were strongly controlled by the palaeogeography and palaeoenvironment conditions of the basin (Moreno, Sierra & Sáez, 1996).

The initial effects of the Variscan orogeny in the Iberian Pyrite Belt occurred throughout the Late Devonian–Early Carboniferous. At this time the basin was affected by an extensional tectonic regimen and an unusual high geothermal gradient, possibly related to the activity of a mantle plume developed under SW Iberia (Simancas *et al*. 2003). This situation resulted in intense bimodal magmatic activity, the compartmentalization of the former basin, and the subsequent development of sub-basins with different subsidence and fill rates and disparate environmental conditions (Moreno, Sierra & Sáez, 1996). Some of these sub-basins reached the optimum conditions for the sulphides to be deposited and preserved (Almódovar & Sáez, 2004). Considering the close relationship between black shales and massive sulphide deposits, the study of these rocks is essential for understanding the complex palaeogeography of the basin, the space–time distribution of the massive sulphide deposits, and the environmental conditions prevailing at the onset of sulphide sedimentation. Previous data obtained in different parts of the basin reveal that the black shales correlate laterally, comprising an anoxic sequence that hosts the mineralization (Moreno *et al*. 2003). The palynomorph assemblages recovered from three deposits included in this sequence (Aznalcóllar (Pereira *et al*. 1996); Neves-Corvo (Oliveira *et al*. 1997); and Tharsis (González *et al*. 2002)) provide a similar late Famennian age that supports both the coeval nature of the mineralization suggested by Relvas *et al*. (2001) and also the hypothesis of a latest Devonian geological event in the Iberian Pyrite Belt.

The present paper, devoted to the palynostratigraphy of the black shales associated with the Sotiel-Coronada Mine deposits, provides new data reinforcing this hypothesis, and emphasizes the role of the palynostratigraphy in the understanding of the palaeogeographical evolution of the Iberian Pyrite Belt basin.

2. Geological setting

The Devonian–Carboniferous record of the Iberian Pyrite Belt has traditionally been divided into three lithostratigraphical units (Schermerhörn, 1971). From footwall to hanging wall these are: the Phyllite–Quartzite Group, the Volcano-Sedimentary Complex, and the Culm Group.

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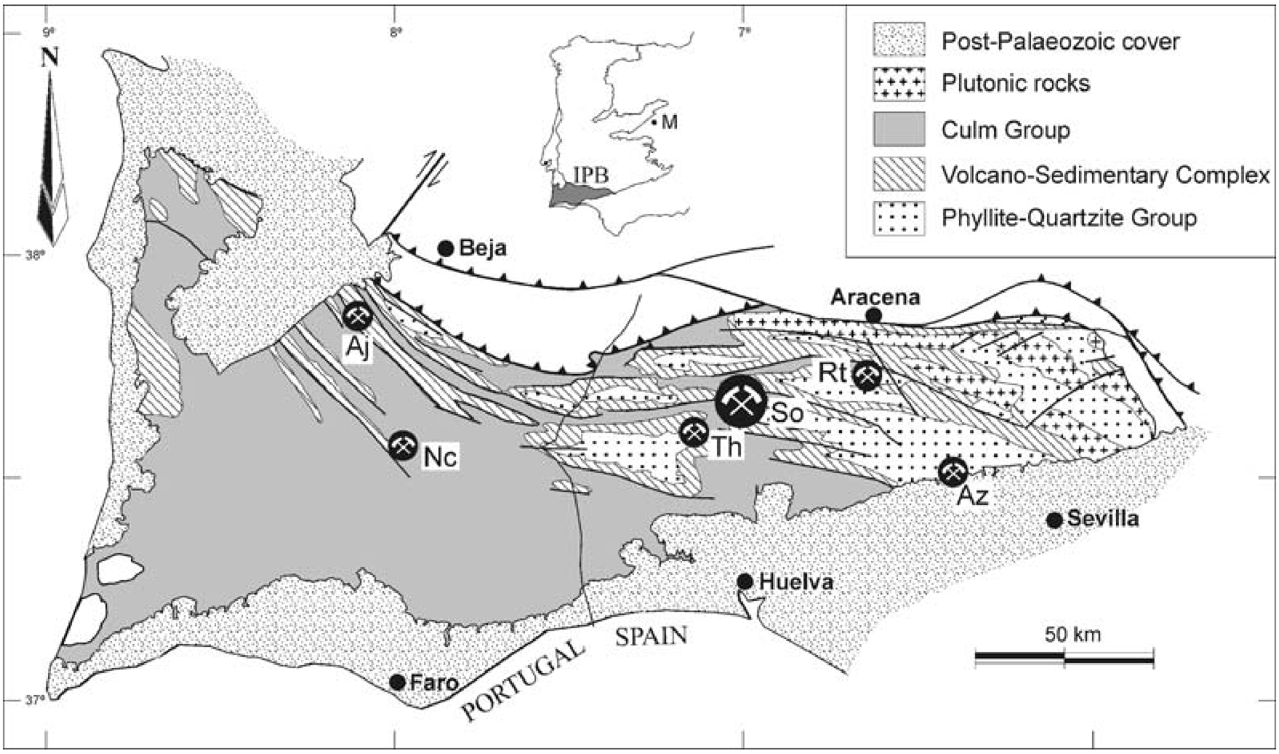

Figure 1. Geological map of the Iberian Pyrite Belt showing the location of the Sotiel-Coronada Mine and the main mining districts. Abbreviations: IPB – Iberian Pyrite Belt; M – Madrid; Aj – Aljustrel; Nc – Neves-Corvo; Th – Tharsis; So – Sotiel-Coronada; Rt – Riotinto; Az – Aznalcóllar.

The Phyllite–Quartzite Group consists of a thick and homogeneous detrital sequence of shales and quartz-arenites of Middle to Late Devonian (late Givetian–late Famennian) age deposited on a shallow marine platform, eventually affected by storm activity. This pre-orogenic sedimentation turns rapidly at the top into an intricate sequence of shales, sandstones, and conglomerates, including mega debris-flow and fan-delta deposits, interpreted as the earliest effects of the Variscan orogeny in the region (Moreno, Sierra & Sáez, 1996). Sparse carbonate lenses yielding conodonts of late Famennian age also appear at the top of this unit (Boogaard, 1967). The base of the succeeding Volcano-Sedimentary Complex is constituted by the anoxic sequence of organic-rich black shales hosting the sulphide mineralization. This sequence, of latest Devonian age, represents the conformable boundary between the Phyllite–Quartzite Group and Volcano-Sedimentary Complex; it may also include volcanic rocks, and commonly displays facies and thicknesses that are highly variable laterally and vertically. Above the anoxic sequence the Volcano-Sedimentary Complex displays a diversity of Lower Carboniferous volcanic, sedimentary, and volcaniclastic rocks, with frequent lateral and vertical facies changes and thickness variations, representing early synorogenic sedimentation in a heterogeneous and compartmentalized basin. The contact between the Volcano-Sedimentary Complex and the overlying Culm Group is represented by another boundary sequence, formally defined as the Basal Shaly Formation (Moreno & Sequeiros, 1989). It consists of an upper Viséan volcanodetrital and shaly sequence composed of the reworked material of the upper Volcano-Sedimentary Complex levels, and the first manifestations of the post-volcanic turbiditic activity in the region. Above the Basal Shaly Formation, the Culm Group, of late Viséan to late Bashkirian age (Oliveira, 1990), comprises a succession of shales, litharenites and rare conglomerates, displaying turbiditic facies, interpreted as the post-volcanic, late synorogenic infill of the Iberian Pyrite Belt basin (Moreno, 1993). The entire succession was folded and thrusted during the Asturian phase of the Variscan orogeny, giving rise to a thin-skinned tectonic style (J. B. Silva, unpub. Ph.D. thesis, Univ. Lisbon, 1983).

In the Sotiel-Coronada area all three lithostratigraphical units of the Iberian Pyrite Belt are well represented (Fig. 2). The Phyllite–Quartzite Group sequence, frequently intersected by mafic sills and veins of the feeding stockwork mineralization, displays at its top various conglomerate levels, representing the best exposures of gravity-flow deposits in the region (Moreno, Sierra & Sáez, 1995), and scattered, small-sized carbonate lenses. The overlying Volcano-Sedimentary Complex contains at its base a thick level of felsic volcanic and coarse- to fine-grained volcaniclastic rocks (Va_1), whose thickness decreases laterally. Above these volcanic rocks, the black shales of the anoxic sequence host the mineralization (Fig. 2). The anoxic sequence in Sotiel-Coronada Mine is

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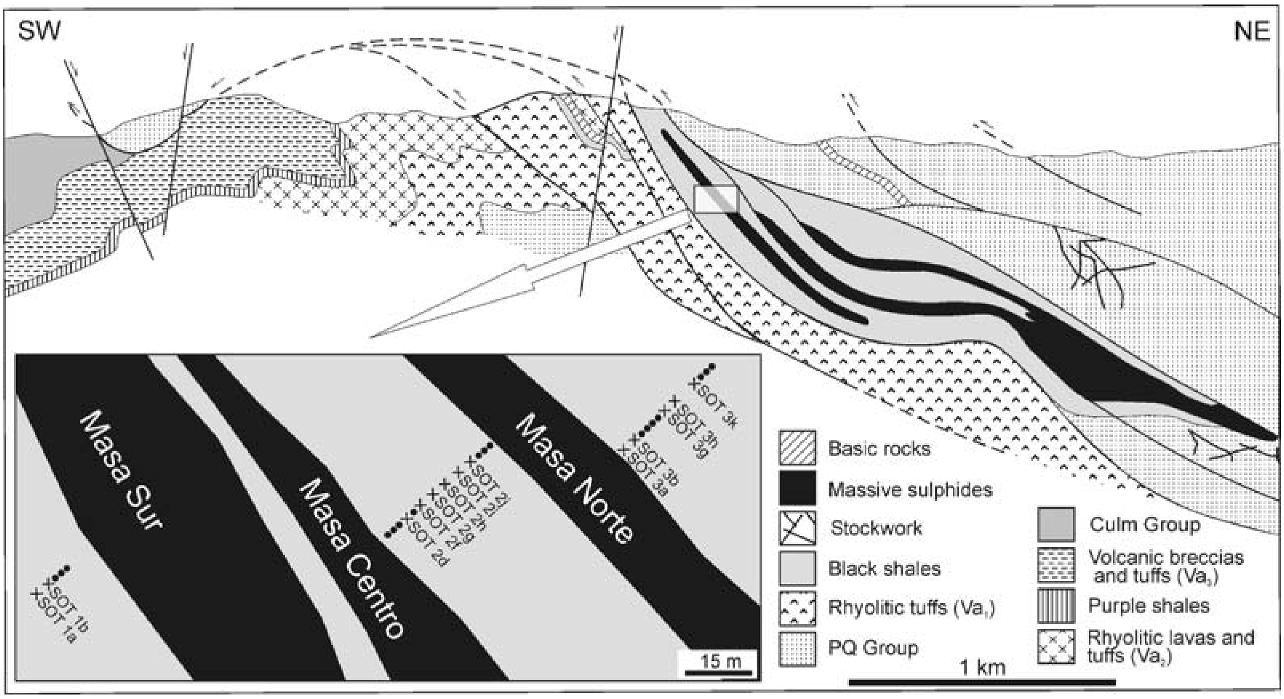

Figure 2. Cross-section showing the lithostratigraphical and structural interpretation of the Sotiel-Coronada area (modified from Santos, Prada & Rosales (1993)), and the positions of the samples studied (crosses represent productive samples and dots represent barren samples). Abbreviations: Va_1 – first felsic volcanic episode; Va_2 – second felsic volcanic episode; Va_3 – third felsic volcanic episode.

laterally correlatable with the shaly sequences described in other mining areas of the Iberian Pyrite Belt as, for example, in the Tharsis mining district, where the volcanic activity is only represented by a thin horizon of fine-grained volcaniclastic felsic rocks, included within the black shales (González *et al*. 2002). Succeeding the anoxic sequence, the local series at Sotiel-Coronada is followed by basic rocks and tuffs, felsic volcanic and volcaniclastic rocks and tuffs (Va_2), purple shales, and a succession of volcanic breccias and reworked tuffs (Va_3). At the top, the shales and greywackes of the Culm Group complete the stratigraphic sequence of Sotiel-Coronada.

As throughout the Iberian Pyrite Belt, all the Sotiel-Coronada rocks were intensely deformed by the Variscan orogeny, resulting in an imbricate thrust system (Fig. 2) that overthrusts the Volcano-Sedimentary Complex onto itself, and the Phyllite–Quartzite Group onto the black shales including the massive sulphides (Santos, Caballero & Prada, 1996).

The mineralization in the Sotiel-Coronada Mine comprises several lensoidal to irregular-shaped ore bodies tectonically stacked into the black shales of the anoxic sequence. Its estimated resources exceed 130 Mt of massive sulphides and an uncertain amount of stockwork-type mineralization. Average ore grades are approximately 0.7 % Cu, 1.24 % Pb and 2.76 % Zn (Santos, Prada & Rosales, 1993). The sulphide deposits have been grouped into three mining areas: Sotiel, Sotiel-Este and Migollas. The present study is focused on the biostratigraphical analysis of the black shales hosting the mineralization in the Sotiel area, where the ore bodies Masa Sur, Masa Centro and Masa Norte have been exploited by means of underground mining operations (Fig. 2).

3. Biostratigraphy

All the samples analysed in the present study were obtained from the mining galleries of Sotiel-Coronada Mine. Thirty samples were collected from the black shales hosting the Masa Norte, Masa Centro and Masa Sur ore bodies (Fig. 2), of which thirteen proved to contain moderately-well-preserved palynomorphs. Two of the productive samples were collected from the shales at the base of the Masa Sur ore body (SOT1), six more productive samples were from the shales delimiting Masa Centro and Masa Norte (SOT2), and the last five samples containing palynomorphs were collected from the shales overlying Masa Norte (SOT3). Consequently, the palynostratigraphical analysis described here comprises the black shales located immediately below and above the mineralization. Standard techniques for extraction and concentration of organic-walled microfossils were used, employing hydrochloric and hydrofluoric acids to remove the inorganic content of the sediments and Fuming Schulze Solution to oxidize the organic debris. This oxidation procedure was critical, as the time required to clarify the palynomorphs was high and varied considerably

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MIOSPORE SPECIES / SAMPLES	SOT1a	SOT1b	SOT2d	SOT2f	SOT2g	SOT2h	SOT2i	SOT2j	SOT3a	SOT3b	SOT3g	SOT3h	SOT3k
Auroraspora macra Sullivan	•	•	•	•	•	•	•	•	•	•		•	•
Cordylosporites cf. *C. marciae* Playford & Satterthwait		•	•				•	•			•		
Cristatisporites triangulatus (Allen) McGregor & Camfield				•	•		•						
Densosporites cf. *D. spitsbergensis* Playford	•			•			•			•			•
Diducites versabilis (Kedo) Van Veen			•										
Emphanisporites annulatus McGregor												•	
Emphanisporites hibernicus Clayton, Higgs & Keegan			•										
Emphanisporites rotatus McGregor emend. McGregor			•	•			•	•		•	•	•	
Endosporites tuberosus González, Playford & Moreno			•			•	•		•				
Epigruspora regularis González, Playford & Moreno			•			•	•		•				
Geminospora spongiata Higgs, Clayton & Keegan			•	•	•	•	•		•	•	•		
Grandispora cornuta Higgs	•		•			•	•	•					
Grandispora echinata Hacquebard emend. Utting		•	•		•	•	•						
Indotriradites diversispinosus González, Playford & Moreno			•									•	
Indotriradites explanatus (Luber) Playford			•		•	•					•	•	
Knoxisporites cf. *K. literatus* (Waltz) Playford					•								
Plicatispora scolecophora (Neves & Ioannides) Higgs *et al.*			•										
Punctatisporites planus Hacquebard			•				•		•				
Pustulatisporites dolbii Higgs, Clayton & Keegan							•			•		•	
Punctatisporites solidus Hacquebard		•											
Retispora lepidophyta (Kedo) Playford		•	•	•	•	•	•	•	•				
Retusotriletes incohatus Sullivan	•	•	•	•	•	•	•	•	•	•	•	•	•
Retusotriletes crassus Clayton, Johnston, Sevastopulo & Smith	•												
Retusotriletes rotundus (Streel) Streel	•		•				•			•		•	
Rugospora flexuosa (Jushko) Streel		•	•	•	•	•							
Vallatisporites vallatus Hacquebard	•	•	•	•		•	•	•	•	•			
Vallatisporites verrucosus Hacquebard	•					•	•	•	•				
Verrucosisporites cf. *V. scurrus* (Naumova) McGregor & Camfield			•										
Verrucosisporites nitidus Playford			•	•			•						
MICROPHYTOPLANKTON SPECIES													
Leiosphaeridia spp.						•		•					
Tasmanites spp.			•	•	•	•	•	•			•	•	•
Cymatiosphaera multisepta Deunff							•						
Cymatiosphaera tenuimembrana González, Moreno & Playford				•			•		•				
Dictyotidium araiomegaronium Hashemi & Playford			•										
Dictyotidium sp. A							•						
Maranhites brasiliensis Brito			•	•		•	•	•	•				
Maranhites gallicus Taugourdeau-Lantz			•		•								
Maranhites mosesii (Sommer) Brito			•		•	•			•		•		
Maranhites multioculus González, Moreno & Playford			•		•	•	•						
Maranhites perplexus Wicander & Playford						•	•						
Chomotriletes vedugensis Naumova			•	•	•			•					•
Dupliciradiatum crassum González, Moreno & Playford			•	•	•		•			•			
Dupliciradiatum tenuis González, Moreno & Playford			•										
Gorgonisphaeridium ohioense (Winslow) Wicander			•		•		•						
Gorgonisphaeridium plerispinosum Wicander				•		•	•						
Gorgonisphaeridium sp. A			•										
Micrhystridium stellatum Deflandre		•					•						
Umbellasphaeridium deflandrei (Moreau-Benoit) Jardiné et al.			•	•									
Veryhachium trispinosum (Eisenack) Stockmans & Willière					•	•	•						
Veryhachium valiente Cramer							•						
Winwaloeusia repagulata González, Moreno & Playford			•		•		•						

Figure 3. Compositions of the palynomorph assemblages obtained from the Sotiel-Coronada Mine.

between samples. The nature and amount of organic debris destroyed during oxidation also varied and was undeterminable for each sample. Therefore, quantitative analysis of the assemblages recovered was not possible.

Most of the assemblages yield terrestrial and marine palynomorphs (miospores and organic-walled microphytoplankton). Only the worst preserved assemblage (SOT1a) is represented exclusively by terrestrial components. The state of preservation was generally deficient, and depended on the location of the samples. Those samples of groups SOT1 and SOT3, located respectively below Masa Sur and above Masa Norte, were the worst preserved, and yielded the lowest diverse and productive assemblages. By contrast, the samples situated between Masa Centro and Masa Norte (SOT2) were better preserved and yielded more diverse and productive assemblages. The dependence of these parameters of diversity and productivity on the preservational state of the assemblages is not exclusive to the study area, as it is confirmed in the whole Iberian Pyrite Belt. The complete composition of each assemblage is indicated in Figure 3.

Characteristic palynological assemblages of Sotiel-Coronada are dominated by the miospores *Auroraspora macra, Cordylosporites* cf. *marciae, Densosporites* cf. *spitsbergensis, Emphanisporites rotatus, Geminospora spongiata, Grandispora cornuta, Grandispora echinata, Indotriradites explanatus, Retispora lepidophyta, Retusotriletes incohatus, Retusotriletes rotundus, Rugospora flexuosa, Vallatisporites pusillites, Vallatisporites verrucosus*, and *Verrucosisporites nitidus.* The additional organic-walled microphytoplankton suite is represented mainly by acritarchs and prasinophyte phycomata of the genera *Gorgonisphaeridium, Veryhachium, Tasmanites* and *Maranhites.* A selection of the most representative taxa of both terrestrial and marine palynomorphs is illustrated in Figure 4.

The systematic descriptions of the complete miospore assemblage, including the newly instituted taxa, *Endosporites tuberosus*, *Epigruspora regularis*

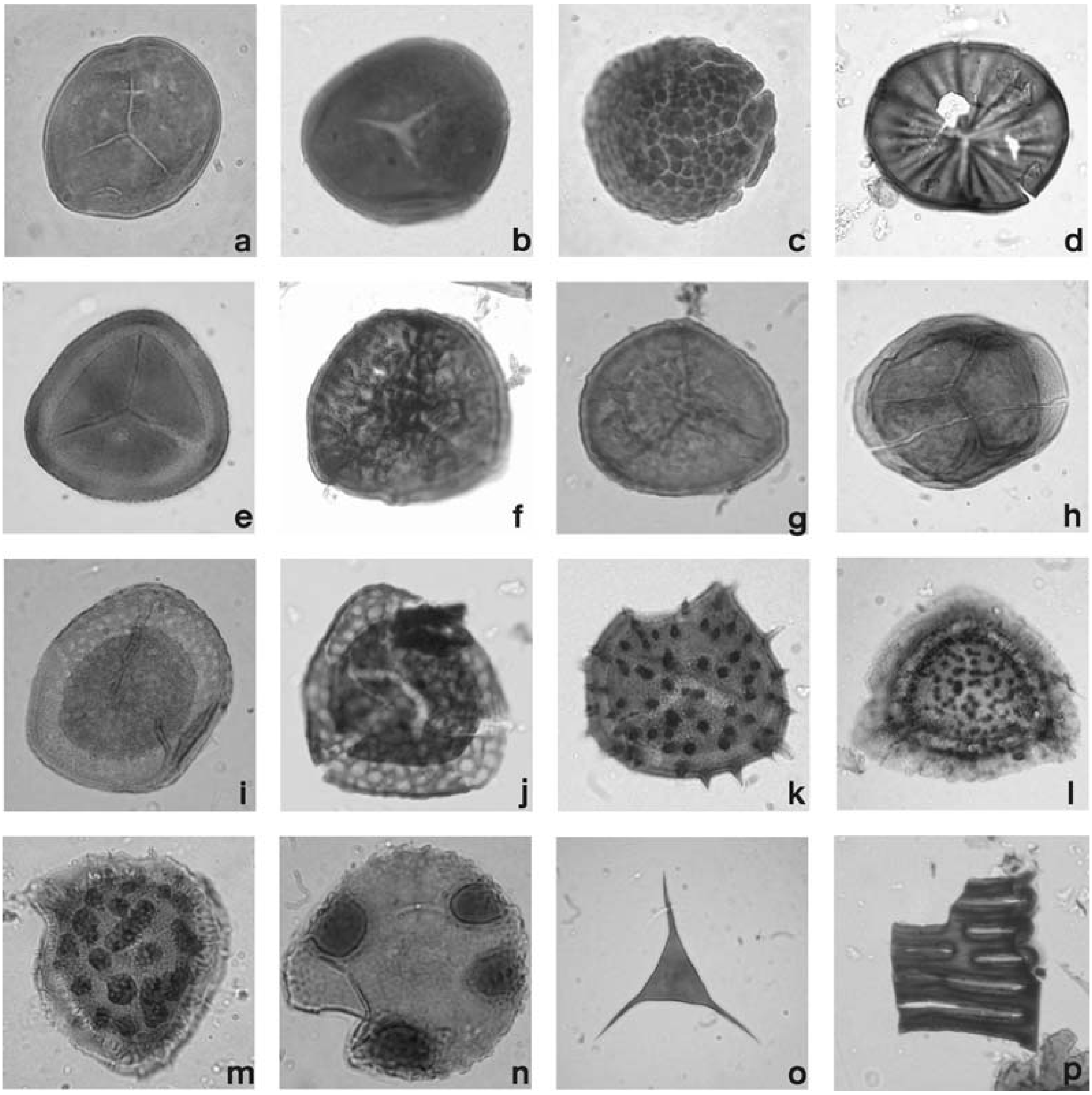

Figure 4. Selected palynomorphs from the black shales of the Sotiel-Coronada Mine. Magnification × 700. (a) *Retusotriletes incohatus* Sullivan (K218B-0860620). (b) *Retusotriletes incohatus* Sullivan (K222B-0953490). (c) *Verrucosisporites nitidus* Playford (K214D-0990270). (d) *Emphanisporites rotatus* McGregor emend. McGregor (K45C-0822456). (e) *Geminospora spongiata* Higgs, Clayton & Keegan (K215D-0960510). (f) *Rugospora flexuosa* (Jushko) Streel (K217D-0960620). (g) *Rugospora flexuosa* (Jushko) Streel (K215D-0980500). (h) *Auroraspora macra* Sullivan (K215D0960595). (i) *Retispora lepidophyta* (Kedo) Playford (K215D-0940520). (j) *Retispora lepidophyta* (Kedo) Playford (K45B-0850535). (k) *Grandispora cornuta* Higgs (K45E-0970490). (l) *Vallatisporites vallatus* Hacquebard (K45B-0990655). (m) *Vallatisporites verrucosus* Hacquebard (K218C-0910540). (n) *Maranhites mosesii* (Sommer) Brito (K215D-0910400). (o) *Veryhachium trispinosum* (Eisenack) Stockmans & Willière (K45B-0990360). (p) Structured phytoclast (K45B-1005494).

and *Indotriradites diversispinosus*, can be followed in González, Playford & Moreno (2005), whereas the systematic description of the organic-walled microphytoplankton, including the new species *Cymatiosphaera tenuimembrana*, *Maranhites multioculus*, *Dupliciradiatum crassum*, *D. tennuis*, and *Winwaloeusia repalgulata*, is found in González, Moreno & Playford (2005).

The joint occurrence of *R. lepidophyta* and *V. nitidus* has been used in Western Europe to define the *Retispora lepidophyta–Verrucosisporites nitidus* (LN) Biozone (Streel *et al.* 1987). In addition, most of the assemblages recovered include species such *D.* cf. *spitsbergensis* and *V. verrucosus*, which are typically recorded for the first time at the base of this Biozone (Higgs, Clayton & Keegan, 1988). Hence, these miospore assemblages are confidently assigned to the LN Biozone. The base of the LN Biozone is located within the upper Famennian, in Fa2d in terms of the Belgian succession (see Maziane, Higgs & Streel (1999), fig. 2), whereas the base of the succeeding *Vallatisporites verrucosus–Retusotriletes incohatus* (VI) Biozone approximately coincides with the Devonian–Carboniferous boundary (Higgs & Streel, 1984; Higgs, Clayton & Keegan, 1988; Streel *et al*. 1987).

In Western Europe, the LN–VI boundary, i.e. the Devonian–Carboniferous boundary, is marked by the extinction of a biostratigraphically important group of miospores including *Retispora lepidophyta* and *Rugospora flexuosa* (Higgs, Clayton & Keegan, 1988). Some of the assemblages recovered in Sotiel-Coronada (SOT1a, SOT3b, g, h, k) do not contains these two taxa, but the disparate stratigraphical position of such samples prevents any further biostratigraphical consideration regarding the location of the Devonian–Carboniferous boundary in the study area. Thereby the absence of the miospores previously cited is related to the deficient preservational state of the assemblages in question, which are the worst preserved of the samples analysed.

The shaly series hosting the massive sulphides in Sotiel-Coronada can be defined as latest Devonian (late Famennian) in age, although further analysis of the highest interval of the sequence is clearly necessary in order to locate the Devonian–Carboniferous boundary.

From a palaeogeographical standpoint, the most salient feature is the co-occurrence of miospores and organic-walled microphytoplankton in the majority of the assemblages analysed, evidence for a marine setting for the deposition of the shales. Moreover the predominance of the terrestrial input suggests a high continental influence on the depositional environment.

4. Conclusions

The palynostratigraphical analysis carried out in the Sotiel-Coronada Mine provides the following conclusions:

(1) The black shale sequence hosting the massive sulphide deposits in the Sotiel-Coronada Mine can be assigned to a late Famennian (latest Devonian) age on the basis of its palynological content.

(2) The volcanic activity in the Sotiel-Coronada area, represented by the felsic volcanic and coarse- to fine-grained volcaniclastic rocks at the base of the anoxic sequence, is presumably contemporaneous with the massive sulphide sedimentation, but more precisely, the commencement of the volcanic activity in this area can be considered to be no earlier than this time.

(3) The identical age data deduced for the whole shaly sequence hosting the mineralization, confirm the multiple repetition of the ore bodies, reinforcing therefore the imbricate thrust system deduced by Santos, Caballero & Prada (1996) for the Sotiel-Coronada area. Similar tectonic style has been defined and corroborated palynologically in other mining districts of the Iberian Pyrite Belt such Neves-Corvo and Aznalcóllar.

(4) The marine component of the palynological assemblages recovered in Sotiel-Coronada Mine, represented by acritarchs and prasinophyte phycomata, indicates a marine environment for the deposit of the black shales hosting the mineralization, but the recovered miospores, terrestrial in origin, suggest a high continental influence in the depositional environment, presumably in the vicinity of the shoreline.

(5) The late Famennian age recorded here fully coincides with the ages obtained in the sulphide deposits so far in the region, i.e. Aznalcóllar, Neves-Corvo and Tharsis. The coeval nature of the massive sulphide sedimentation can be related with the effects of an anoxic crisis occurring in latest Devonian time in the Iberian Pyrite Belt.

(6) The co-occurrence of the anoxic crisis, the rupture and compartmentalization of the basin, and the commencement of the volcanic activity, strongly support the hypothesis advanced by González *et al*. (2002) that the genesis of the massive sulphide deposits in the Iberian Pyrite Belt was intimately associated with a geological event occurring close to the Devonian–Carboniferous boundary.

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