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Large gypsum nodules in the Paleogene and Neogene evaporites of Spain: distribution and palaeogeographic significance

Federico ORTÍ, Laura ROSELL, Elisabet PLAYÀ and Javier GARCÍA-VEIGAS

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Gypsiferous units occur along the margins of some Paleogene and Neogene basins in Spain. These units accumulated in shallow saline lakes of low ionic concentration. Other gypsiferous facies constitute outer rings of the thick, highly-saline evaporite formations that occupy the central parts of the basins. In some of these gypsum units and rings, large nodules (from 0.5 m to several metres across) of secondary gypsum that originated as replacive or displacive nodular anhydrite are present. Although these occurrences usually show a stratiform arrangement, vertical geometries are observed locally, suggesting ascending circulation of anhydritizing flows. The common characteristics of the large gypsum nodules in the various occurrences indicate that the precursor anhydrite formed in burial conditions from shallow to moderate depths. A few occurrences of large, secondary gypsum nodules in the gypsum units are linked to deep faults or diapiric structures.

Federico Ortí, Laura Rosell and Elisabet Playà, Departament de Geoquímica, Petrologia i Prospecció Geològica, Universitat de Barcelona, Carrer de Martí i Franquès, 08028 Barcelona, Spain, e-mails: f.orti@ub.edu, lrosell@ub.edu, eplaya@ub.edu; Javier García-Veigas, Serveis Científicotècnics, Universitat de Barcelona, Carrer de Lluís Solé i Sabaris 1-3, 08028 Barcelona, Spain, e-mail: garcia_veigas@ub.edu *(received: December 29, 2009; accepted: August 25, 2010).*

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INTRODUCTION

Sabkha and deep burial settings are the most common sites where diagenetic anhydrite forms. In a sabkha setting, displacive facies (isolated nodules, bedded nodules, enterolithic levels) of early diagenetic or primary anhydrite are generated (Shearman, 1966; Hardie, 1967). These anhydrite facies are commonly found at the top of shoaling cycles representing the evolution from subaqueous depositional conditions at the base (carbonates, lutites) to exposure conditions at the top where interstitially-grown gypsum/anhydrite develops (sabkha cycles). In a deep burial setting, gypsum transforms totally to anhydrite with increasing temperature and lithostatic pressure (Murray, 1964). Although this mineral transformation usually preserves the depositional gypsum facies, a significant textural change is involved in other cases, resulting in replacive anhydrite with a nodular-mosaic or "chicken-wire" fabric (Warren, 2006). In the two settings, however, the size of the individual anhydrite nodules is relatively small, rarely reaching some tens of centimetres across. Moreover, bedding is preserved or little disturbed, although minor deformation is caused by the displacive sabkha nodules.

These two settings of anhydrite growth have been recognized in the assemblage of both marine and non-marine evaporite units of Mesozoic and Paleogene and Neogene age in the sedimentary basins of Spain (Ortí, 1992, 1997; Playà *et al.*, 2000). The present paper is concerned with a different setting of anhydrite growth occurring in the same geological domain. In exposure, the facies characterizing this setting is made up of large (>0.5 m up to several metres across) nodules of secondary gypsum (Fig. 1A). These nodules occur in the non-marine evaporite units that accumulated preferentially along the basin margins. Very often the anhydrite precursors of these nodules have been interpreted as sabkha products in the literature.

This paper gives an overview of these unusual gypsum nodules in some Iberian Paleogene and Neogene basins as well as a new interpretation of their genesis (Fig. 2). However,

Fig. 1. Large gypsum nodules in the proximity of the village of Fuentes de Jiloca (Yesera del Pilar, Calatayud Basin)

A – general view, scientist is Dr. Alicja Kasprzyk (November, 1989); **B** – detail of alabastrine core, individual porphyroblastic crystals, and porphyroblastic coating in a large nodule of secondary gypsum; hammer for scale

the occurrences present in the Ebro Basin will be dealt with elsewhere (Ortí *et al*., in prep.). Given their purity in calcium sulphate, all these facies have in the past been exploited economically for the production of both plaster of Paris and high-quality alabaster sculptures, in some cases, since Roman times.

STRATIGRAPHIC AND SEDIMENTOLOGIC SETTING

Non-marine evaporitic sedimentation was recorded in several Iberian basins during the Paleogene and Neogene. In some of these basins, a number of small saline lakes developed with their mother waters characterized by a low ionic concentration and a Ca-sulphate composition (Ortí *et al*., 1989*a*, *b*). The isotopic compositions of the gypsum units that derived from these lakes indicate that the sulphate in the mother waters mainly resulted from chemical recycling of the marine Mesozoic evaporites (Triassic, lower Liassic and Upper Cretaceous) present in the Alpine chains bounding the basins (Utrilla *et al*., 1991, 1992).

In the Iberian Paleogene and Neogene basins, gypsiferous facies also occur as outer rings of the thick, non-marine evaporite formations that occupy the basinal depocenters. These central formations are characterized by complex mineral parageneses derived from highly-saline mother brines (Na-chlorides, Na-sulphates; Ortí, 1989*b*).

Large gypsum nodules (between 0.5 m and several metres across) are found in some of the gypsum units and gypsiferous outer rings. They replace or displace the host lithologies, either evaporitic (gypsum/anhydrite) or non-evaporitic (lutites, carbonates). Petrographic evidence in these nodules such as the presence of diagnostic microscopic textures of secondary gypsum and abundant anhydrite relics indicate that these are secondary gypsum features that originated as anhydrite. Final exhumation of the gypsum units during the Pliocene–Pleistocene resulted in the anhydrite-to-secondary gypsum transformation of the large nodules.

The most common texture of secondary gypsum in these large nodules is alabastrine, but both porphyroblastic crystals and porphyroblastic coatings are often developed in the external boundaries (Fig. 1B). Some large nodules are also made up of megacrystals (“megacrystalline secondary gypsum”; Ortí, 1977).

OCCURRENCES

CALATAYUD BASIN (MIOCENE UNITS)

The intermontane Calatayud Basin is located in the central part of the Iberian Range (Figs. 2 and 3A). It has a graben structure elongated in a NW to SE direction, and is flanked by areas of high relief mainly composed of slates, quartzites and carbonate rocks of Paleozoic age. The basin, which formed during Miocene extensional tectonics, was filled with a thick (up to 1500 m), non-marine Neogene succession (Anadón and Alcalá, 2004). Three major lithostratigraphic units have been distinguished in this succession (Sanz-Rubio *et al*., 1997): the Miocene Lower Unit, which is detrital in the marginal zones and evaporitic in the centre; the Miocene Intermediate Unit, which is gypsiferous and marly-calcareous; and the Miocene Upper Unit, which is calcareous to siliciclastic in composition.

Two evaporite units have been distinguished in this Neogene succession (Ortí and Rosell, 2000): a lower evaporite unit that is composed of Na-sulphate (glauberite and thenardite) beds in the centre and gypsum beds at the margins. This unit corresponds to the evaporitic facies that occupy the

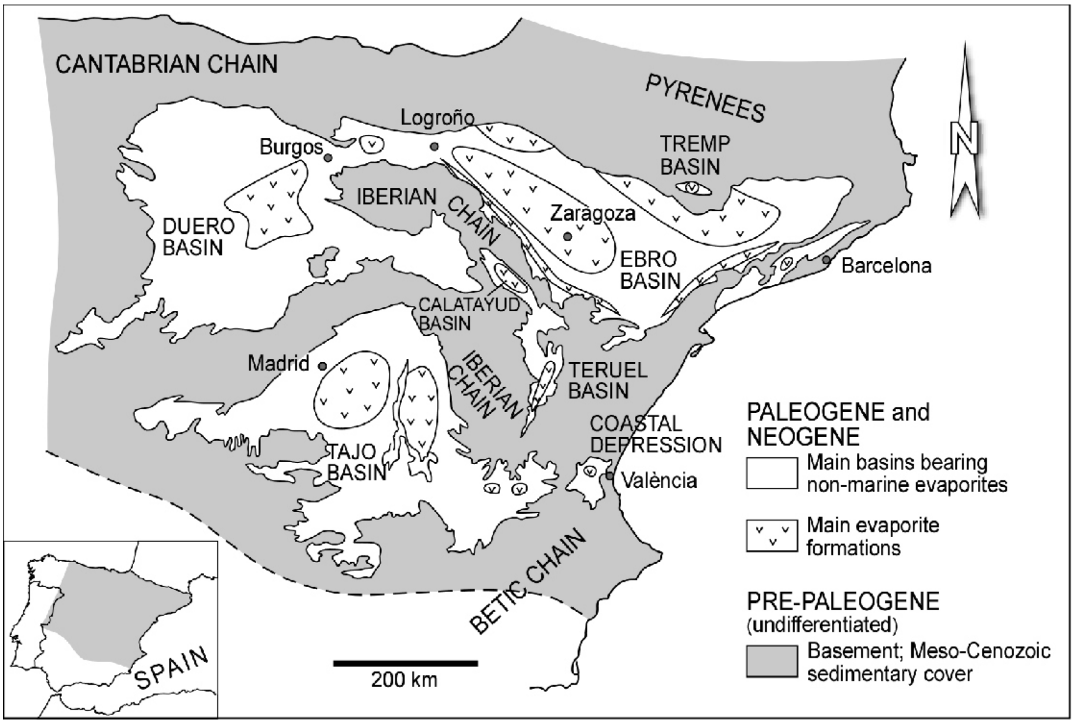

Fig. 2. Distribution of evaporite formations in the Paleogene and Neogene basins of Central and Northern Spain

Main occurrences of large nodules of secondary gypsum cited in this paper are located in the Calatayud, Tremp and Tajo basins, and in the Coastal Depression of València

central part of the Miocene Lower Unit; and an upper evaporite unit that is made up of laminated gypsum, located at the base of the Miocene Intermediate Unit (Fig. 3B). Along the southern boundary of the basin, the two evaporite units have gypsiferous outer rings mainly composed of massive, bioturbated, chert-bearing, primary gypsum facies. Near the village of Fuentes de Jiloca, these marginal facies include large nodules of secondary gypsum, which displace or replace the bioturbated gypsum (Fig. 4A, B).

The arrangement of these large nodules is commonly stratiform but may be vertical. In the latter case, the nodules form columns or walls up to 20 m high and several metres wide and cross-cut the bioturbated gypsum layers and the interbedded lutite beds (Fig. 5A, B). The stratiform arrangement of large nodules in the Fuentes de Jiloca area suggests repetitive sabkha episodes (Ortí and Rosell, 2000). However, given the size of the nodules and the height of the columns and given that no erosional surfaces are observed at the top of the nodules, the vertical arrangement could also have been caused by the circulation of deep, anhydritizing brines, which ascended through fractures.

TREMP BASIN (PALEOCENE UNIT)

The Tremp Basin (Fig. 2) is located in the allochthonous Central South Pyrenean structural unit (Seguret, 1972) and forms a wide syncline composed of Meso-Cenozoic strata. In this basin, the uppermost 200 m of the Tremp Formation (Garumn Facies of Paleocene age) constitute an evaporite unit which is intercalated within red lutites. Outcropping gypsum in this unit is secondary and derived from anhydrite hydration (García-Veigas, 1997).

In this unit, the common gypsum facies is laminated-to-banded and intercalates large nodules (from 0.5 m to >1 m across) of replacive/displacive gypsum (Fig. 6). A representative cycle of this unit in the Tremp Basin is shown in Figure 7A: the large nodules are mainly developed towards the top of the cycle where the thickness of the gypsum beds is maximal (Figs. 7A, B). The upper parts of the nodules, however, remain unaffected by any possible erosion/dissolution surface.

Although the cycle has been attributed to a sabkha setting by García-Veigas (1997), all these characteristics are compatible with the stratiform geometry of a post-depositional (burial) growth of large nodules of anhydrite.

COASTAL DEPRESSION OF VALÈNCIA (MIOCENE UNIT)

The Ninyerola Gypsum is a cyclic lacustrine unit (200 m thick) of Middle Aragonian (Early Miocene) age that is developed in the Coastal Depression of the city of València, Eastern Spain (Anadón and Alcalá, 2004; Fig. 8A). This depression is filled with Paleogene and Neogene and Quaternary deposits.

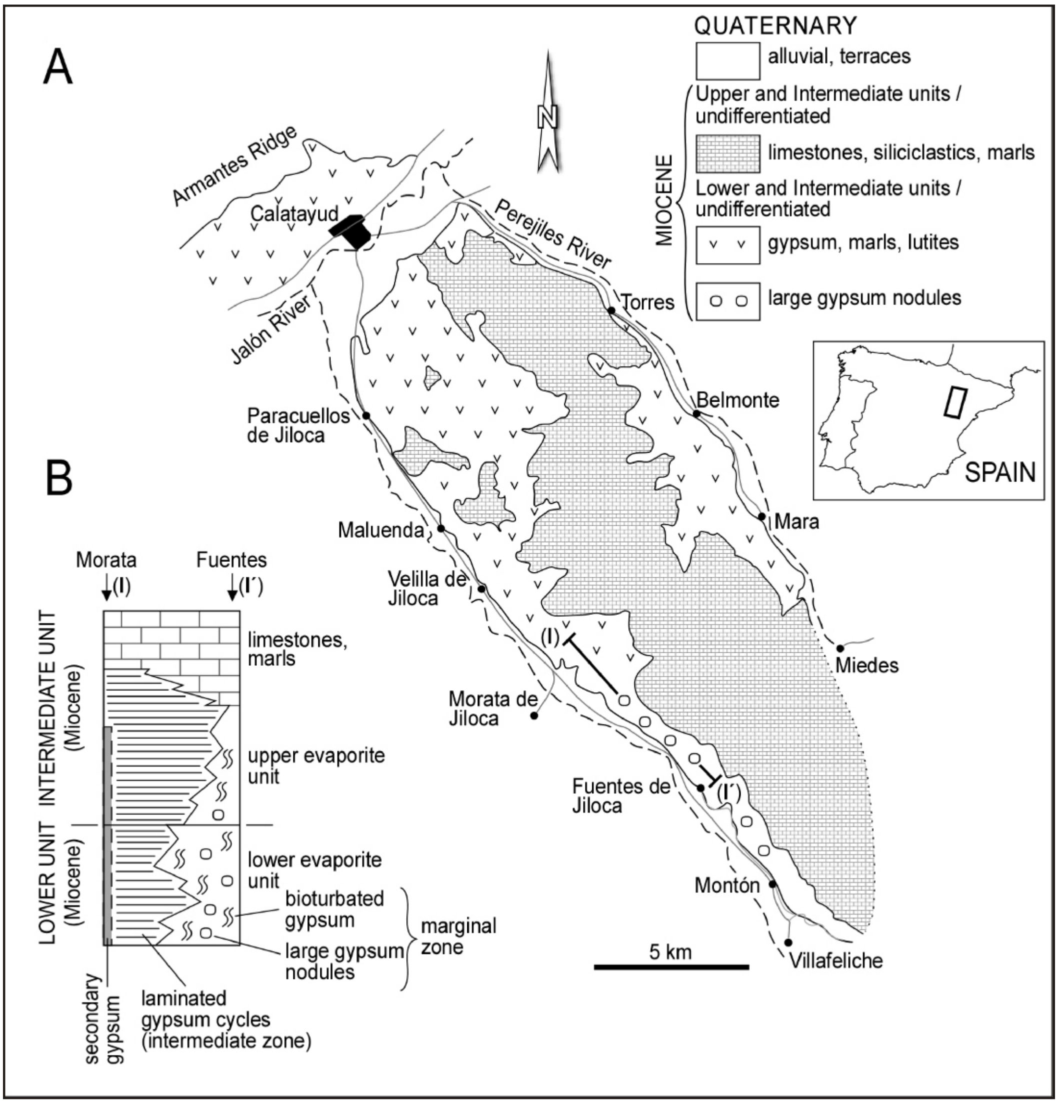

Fig. 3. Occurrence of large gypsum nodules in the Calatayud Basin (Miocene)

The general lithostratigraphic units of the Miocene (Lower, Intermediate and Upper units) are taken from Anadón and Alcalá (2004); the evaporite units, facies zones, and facies types are taken from Ortí and Rosell (2000); **A** – distribution of the (simplified) evaporitic facies between the town of Calatayud and the village of Villafeliche; the area of large gypsum nodules in the proximity of the village of Fuentes de Jiloca is indicated; **B** – distribution of large gypsum nodules in the lower and upper evaporite units along the section I–I' in the marginal area between Fuentes de Jiloca and Morata de Jiloca. Adapted from Ortí and Rosell (2000, fig. 2)

The best exposure of the Ninyerola Gypsum Unit is found in an anticline oriented NNE–SSW with a diapiric core formed by Upper Triassic gypsum (Keuper facies, which are rich in halite in the subsurface; Fig. 8B). This anticline is located between the villages of Montserrat and Picassent (València province). The Miocene succession on both flanks of the anticline displays some lithological differences, suggesting that initial diapirism of the Upper Triassic evaporites was coeval with the non-marine Miocene sedimentation (Ortí and Rosell, 2007).

The basal gypsum layers of the Miocene succession in contact with the Keuper facies are made up of secondary gypsum and display nodular facies; this gypsum has been exploited in quarries. The original bedding of these layers is severely disturbed by the presence of large nodules (up to 1.5 m across). In the overlying part of the succession, the gypsum remains primary (bioturbated gypsum facies). The structural complexity of this exposure prevents a precise observation of the nature, conformable or not, of the contact between the basal nodular gypsum facies of the Miocene succession and the overlying layers of primary gypsum.

Assuming that this contact is conformable, the large nodules of the precursor anhydrite can be interpreted as having formed in a sabkha (very early diagenetic) setting, as has been tentatively proposed by Ortí and Rosell (2007). However, assuming that the boundary is irregular or oblique to bedding, a different diagenetic scenario could also be considered. Thus, the circulation of ascending brines associated with the diapirism of the Upper Triassic (Keuper) evaporites during the

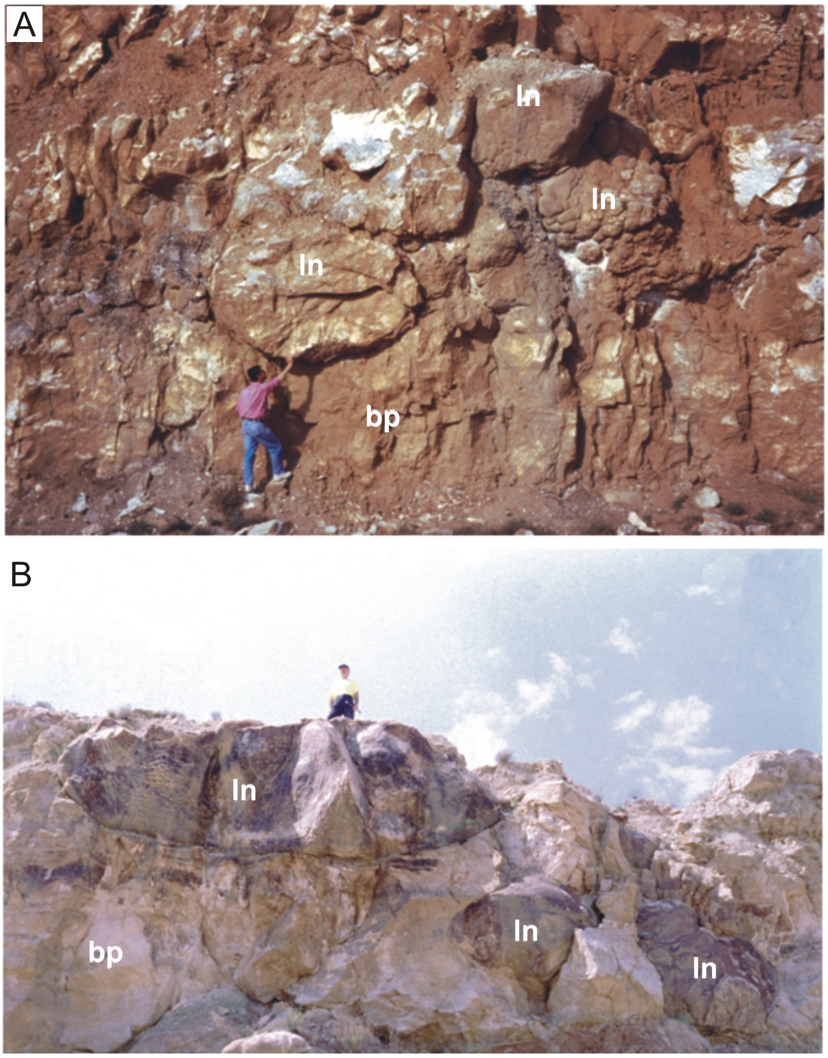

Fig. 4. Large gypsum nodules of alabastrine secondary gypsum (ln) in the lower evaporite unit (Miocene) of the Calatayud Basin; the host bioturbated gypsum of the large nodules is preserved as primary (bp); Yesera del Pilar quarry front near Fuentes de Jiloca

A – group of large nodules; **B** – individual, replacive, large nodule of about 8 m across; other large nodules are present

Miocene could be considered. In this case, the ascending brines, rich in sodium chloride, would have caused the nodular anhydritization of the gypsum beds in contact with the Keuper deposits on both sides of the anticline core (Fig. 8C). This alternative interpretation is favoured by (1) the large size of the nodular structures and by (2) the fact that only the Miocene succession in direct contact with the diapiric materials was transformed into nodular anhydrite (the bioturbated facies has been preserved as primary gypsum in other outcrops of the Ninyerola Gypsum Unit in the region).

TAJO BASIN (PALEOGENE AND MIOCENE UNITS)

The Tajo (Madrid) Basin is an intracratonic Paleogene and Neogene basin located in Central Spain (Fig. 2). This basin was filled with a sedimentary succession over 2000 m-thick mainly composed of lacustrine and alluvial materials. In the central part of the basin, the main evaporitic deposits consist of a thick (about 600 m) continental unit known as the Saline Unit or the Miocene Lower Unit of Early to Mid Miocene age (Ramblian to early Aragonian; Calvo *et al*., 1996). The unit is formed by layers of highly-soluble minerals (glauberite, thenardite and halite) in association with gypsum/anhydrite and lutite beds (Ortí *et al*., 1979; Ordóñez *et al*., 1991). Some gypsum deposits are also present in the so-called Miocene Intermediate Unit (of middle Aragonian to Vallesian age). Besides these two major units that occupy the basin centre, some older (Paleogene) units, which are mainly composed of Ca-sulphate facies (gypsum/anhydrite), crop out in the eastern and northern parts of the basin.

In various evaporite units of this basin, some occurrences of large, displacive to replacive nodules of secondary gypsum have been cited. These occurrences are preferentially located towards the basin margins (Ortí *et al*., 1992). One of them is present in the gypsum quarries near the village of Añover de Tajo (Fig. 9) to the south of the basin, where large nodules and irregular masses (>2 m across) displace mechanically and also replace the laminated gypsum at the base of the Miocene Intermediate Unit (Fig. 10).

Another occurrence of Miocene age is found near the village of Leganiel located to the east of the basin and close to the overthrust of the Altomira Chain on the Paleogene and Neogene deposits of the basin. At this locality, large nodules (up to 1 m across) replace a primary selenitic gypsum facies of lacustrine origin (Fig. 11). This facies is considered as an independent marginal gypsum unit that developed coevally with the upper part of the Saline Unit but remained geographically separated from it (Rodriguez-Aranda *et al*., 1995). In this locality, the large gypsum nodules also replace the beds made up of bioturbated gypsum facies that form the base of the Miocene Intermediate Unit.

Additional occurrences are also present in the Paleogene gypsum deposits to the north of the basin in the quarries near the villages of Aleas and Torrelaguna (Fig. 9). In the Aleas quarries, large nodules (>1 m across) of replacive, secondary gypsum may occupy significant parts of the exploitation fronts (Fig. 12).

OTHER OCCURRENCES

In the Carpentras Basin of SE France (Paleogene), the Mazan gypsum quarry exhibits a selenite facies of primary gypsum which was deposited in a lacustrine environment. In the

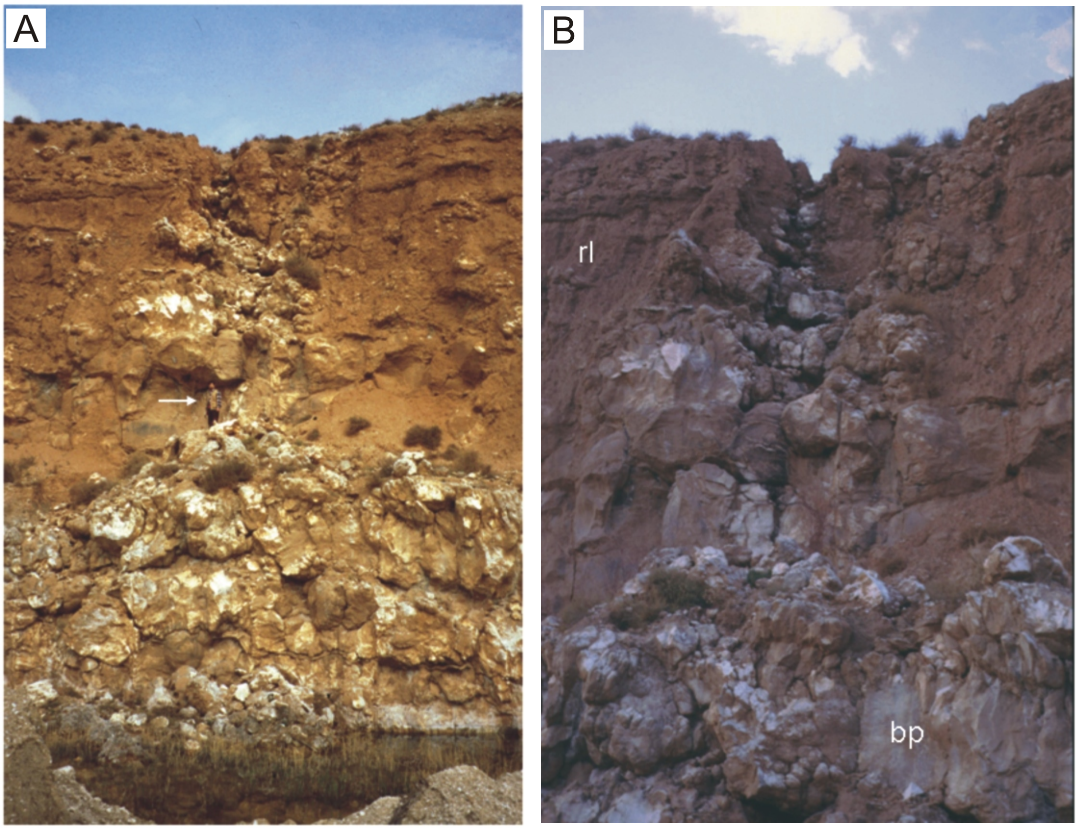

Fig. 5. Occurrence of large nodules of alabastrine secondary gypsum in the lower evaporite unit (Miocene) of the Calatayud Basin; the host bioturbated gypsum of the large nodules is preserved as primary; Yesera del Pilar quarry front near Fuentes de Jiloca

A – stratiform (lower part of the picture) and vertical (upper part) arrangement of the large nodules; the host-rock of the large nodules is primary bioturbated gypsum in the stratiform arrangement and red lutites in the vertical arrangement; the lutite beds in the upper part of the picture are cut or deformed by large nodules; note the human scale (arrow); **B** – close-up of the former picture; bp – bioturbated gypsum, rl – red lutites; the picture is taken at a time different from that of the picture in (A)

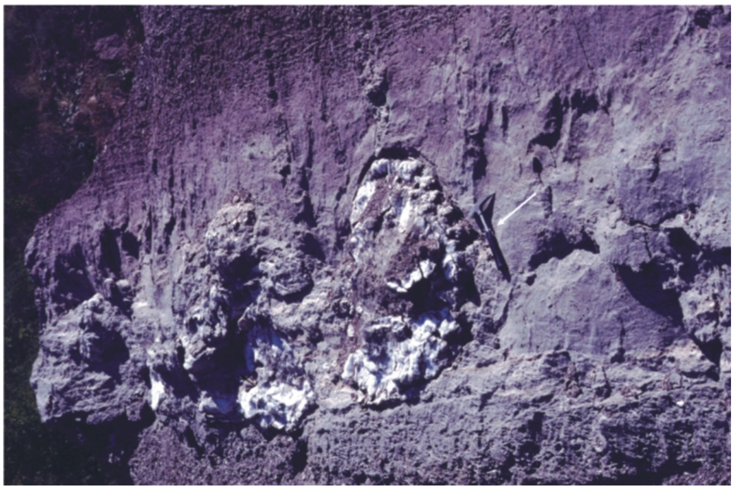

Fig. 6. Occurrence of large nodules of alabastrine secondary gypsum in the evaporitic unit of the Tremp Formation (Garumn facies; Paleocene) in the Tremp Basin (South Pyrenees)

Hammer for scale (arrow)

quarry fronts, the presence of secondary gypsum features in vertical columns or walls that are several metres in height and a few metres in width was cited by Truc (1983) and by Ortí (1989*a*). These columns cut and replace the selenitic layers. In the central part of the columns, large nodules (>0.5 m across) of alabastrine secondary gypsum are surrounded by irregular masses of megacrystalline secondary gypsum (Fig. 13). According to the former authors, the selenitic gypsum was replaced along vertical fractures by large anhydrite nodules (central part of the columns) and by massive anhydrite (external parts of the columns). During the exhumation of the deposit, the large nodules and the massive anhydrite were replaced, respectively, by the alabastrine and the megacrystalline textural varieties of secondary gypsum. This anhydritization was attributed to the ascending circulation of chloride-rich brines in burial conditions by Truc (1983).

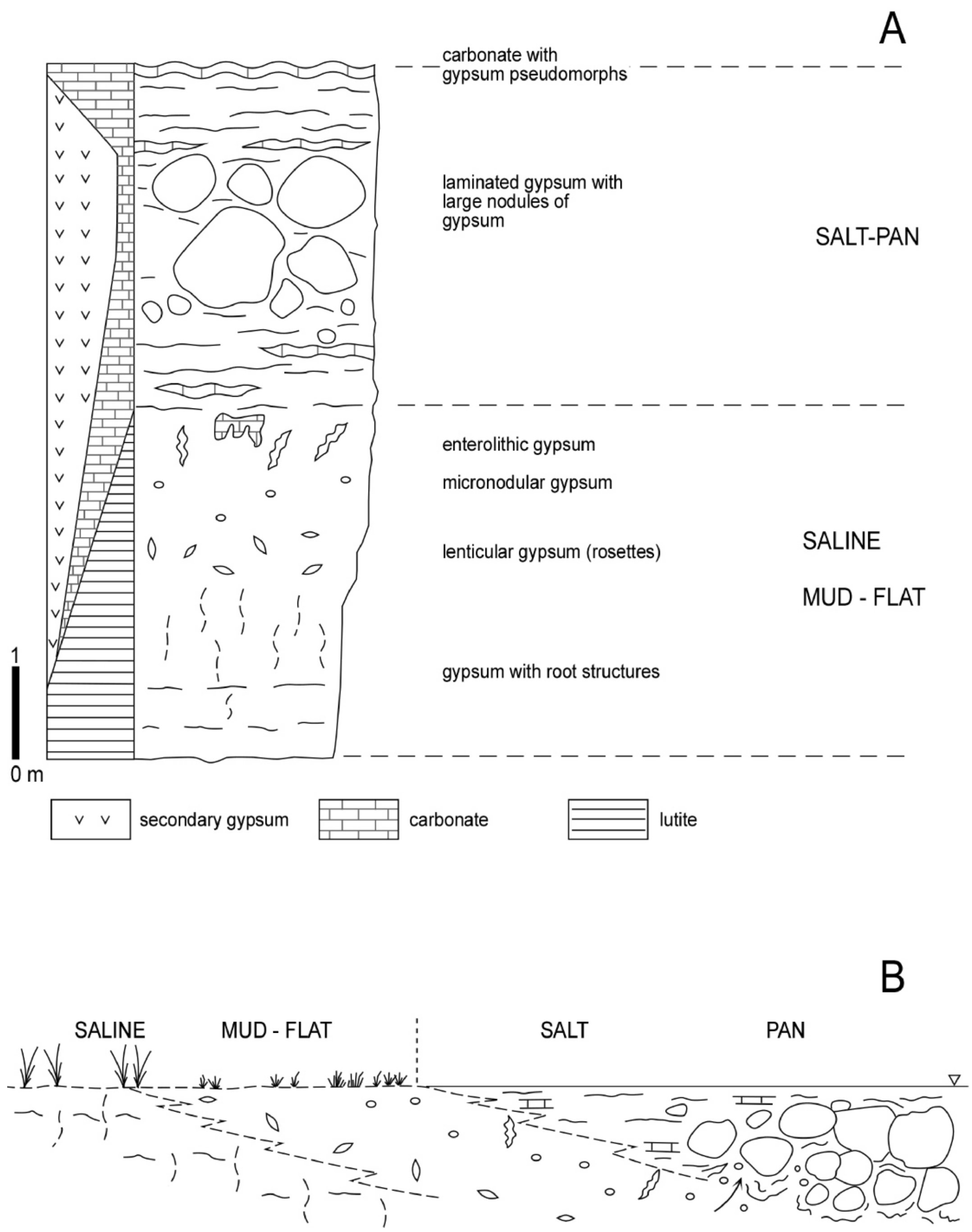

Fig. 7. Occurrence of large gypsum nodules of secondary gypsum in the Tremp Formation; adapted and simplified from García-Veigas (1997, fig. 9.2)

A – evaporitic, depositional cycle bearing large nodules; **B** – interpretative subenvironments of the depositional cycle according to García-Veigas (1997); the suggestion that the growth of the large nodules of (precursor) anhydrite postdated the sedimentation of the whole cycle (i.e., occurred during burial diagenesis) is cited in the text (see section "Other occurrences")

In the Betic Chain (SE of Spain), Salvany and Ortí (1990) reported the presence of large nodules in the Neogene succession (Mid–Late Miocene to Pliocene) of the Campo Coy Basin (Murcia province), where a cyclic, non-marine gypsum unit up to 45 m-thick crops out along the southern margin of the basin. Both primary and secondary gypsum facies are present in this unit, including secondary gypsum nodules up to 1 m across. Salvany and Ortí (1990) concluded that the primary gypsum facies in the depositional cycles were partly transformed into nodular anhydrite in a sabkha setting during early diagenesis. Subsequently, large nodules of anhydrite developed preferentially at the top of the cycles, displacing/replacing the already formed nodular (sabkha) anhydrite and deforming the bedding.

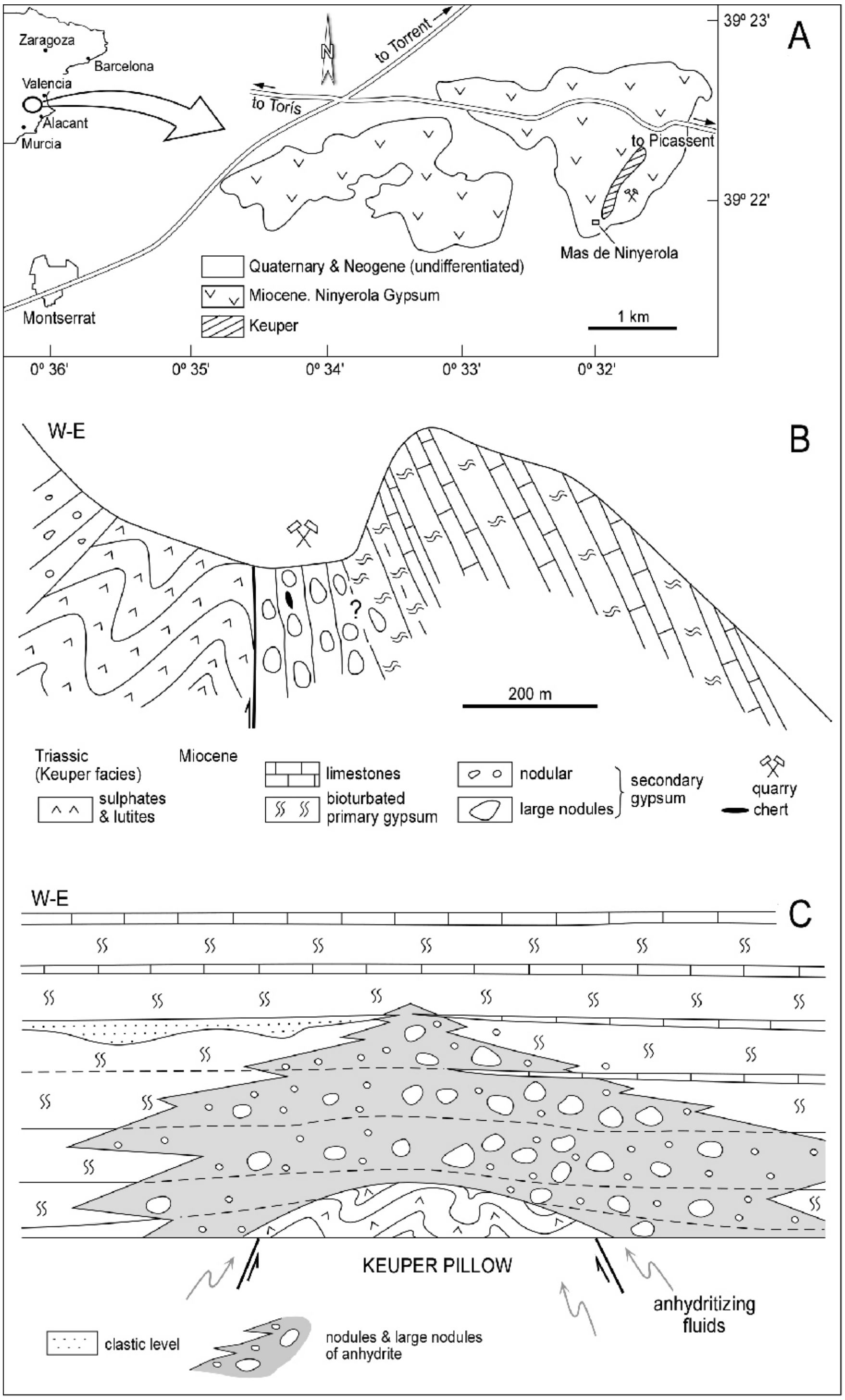

Fig. 8. Occurrence of large nodules of secondary gypsum in the Ninyerola Gypsum Unit (Miocene; western zone of the Neogene Coastal Depression of València)

A – location map of the Ninyerola Gypsum Unit. The area shown is the SE corner of the Geological Map 721 (Cheste) of Spain on a scale of 1:50 000 (after Ortí and Rosell, 2007, fig. 1); **B** – geological section across the diapiric anticline in the Ninyerola Zone between the villages of Montserrat and Picassent. At the base of the Miocene succession the gypsum is totally secondary and displays nodular facies and large nodules; adapted from Ortí and Rosell (2007, fig. 3A); **C** – interpretative scheme of the gypsum-to-anhydrite transformation involving large anhydrite nodules. This transformation would have been caused by ascending fluids acting on the primary (bioturbated) gypsum facies of the Ninyerola Gypsum Unit. The chloride-rich, ascending fluids would be linked to initial, synsedimentary diapirism (pillow stage?) of the Keuper materials. The scheme assumes that in (B) the boundary between the nodules/large nodules (anhydritized facies) and the preserved primary gypsum is not depositional

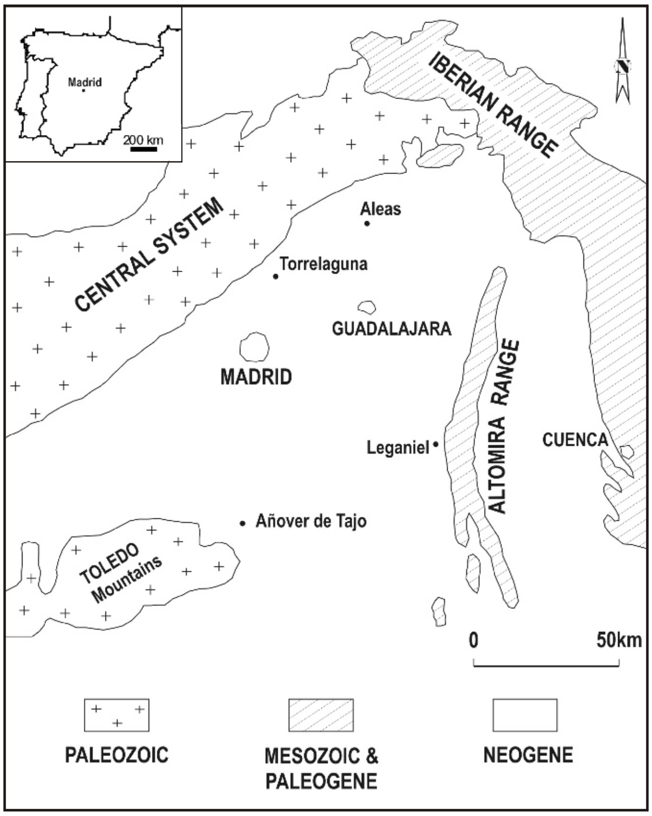

Fig. 9. Situation of the localities near the occurrences of large gypsum nodules in the Miocene evaporite units of the Tajo (Madrid) Basin cited in this paper

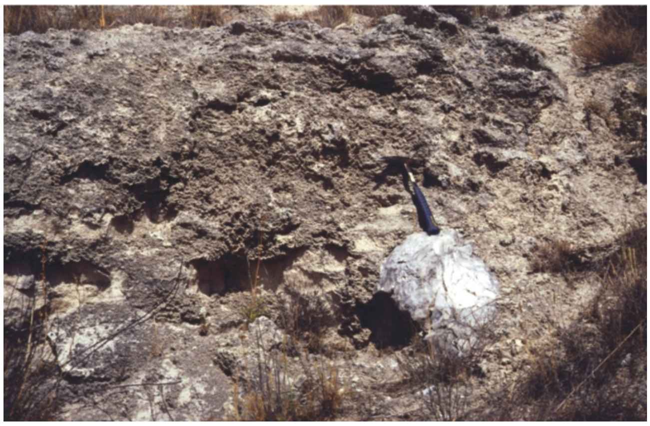

Fig. 11. Large nodules of megacrystalline secondary gypsum replacing (primary) selenite facies

Top of the Miocene Lower Unit (Saline Unit) near the village of Leganiel (eastern part of the Tajo Basin); diameter of the complete nodules is close to 60 cm

As in the case of the Tremp Basin (see above), we cannot rule out the possibility that the occurrences in the Campo Coy Basin derived from ascending, anhydritizing brines in the proximity of the basin margin (in contrast to an origin related to a sabkha setting). In the two cases, the preferential presence of the large nodules at the top of the cycles could also be linked to the availability of larger volumes of gypsum (presence of thicker gypsum layers towards the top of the cycles) to be converted into anhydrite during burial.

ENVIRONMENT OF FORMATION

The foregoing summary of occurrences suggests an origin of the large (precursor) anhydrite nodules that is not consistent with a sabkha setting. As regards a possible origin due to deep burial diagenesis, the coexistence of the large nodules with un-

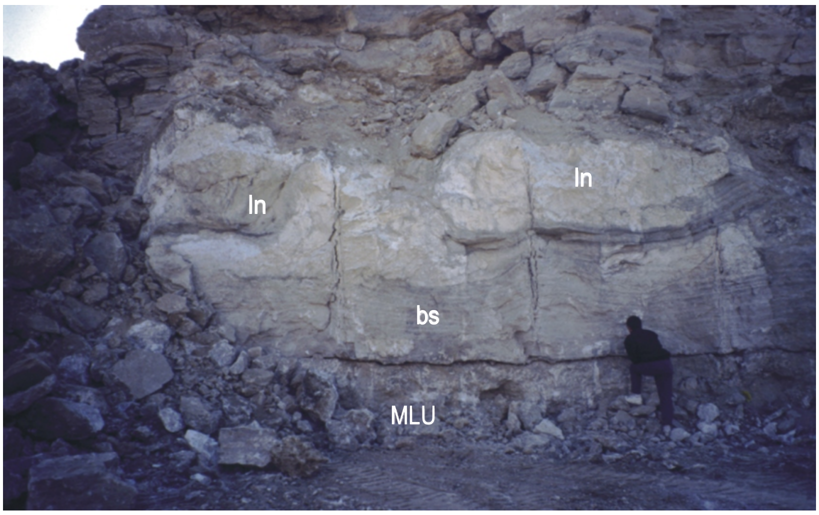

Fig. 10. Occurrence of large nodules of alabastrine secondary gypsum (ln) at the base of the Miocene Intermediate Unit of the Tajo (Madrid) Basin in a gypsum quarry front near the village of Añover de Tajo

The gypsum host-rock of the large nodules is laminated to banded secondary gypsum (bs); the lowermost part of the front corresponds to the top of the Miocene Lower Unit (MLU – Saline Unit); person for scale

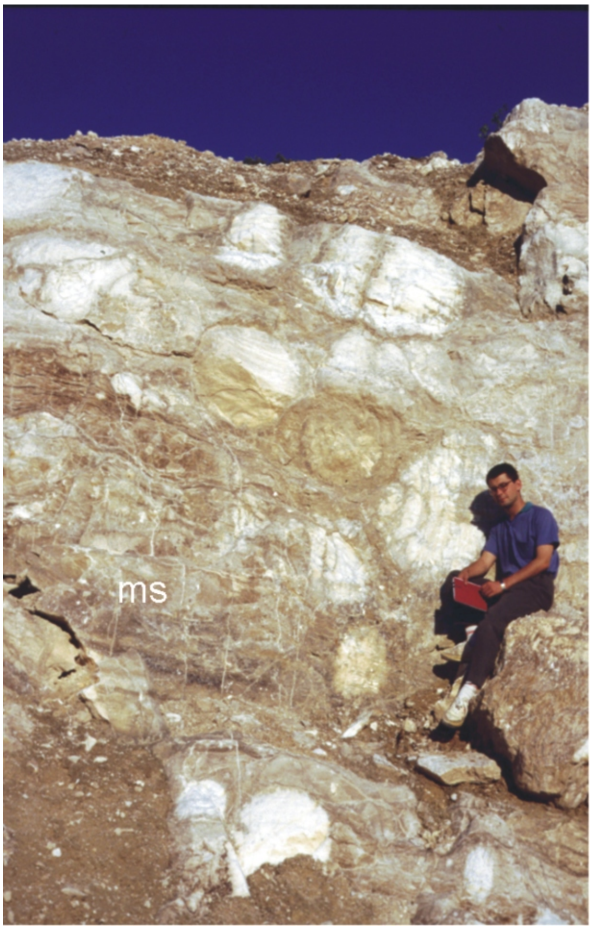

Fig. 12. Occurrence of large, replacive gypsum nodules of alabastrine secondary gypsum in the quarries near Aleas (northern part of the Tajo Basin)

Host-rock of the nodules is massive secondary gypsum (ms)

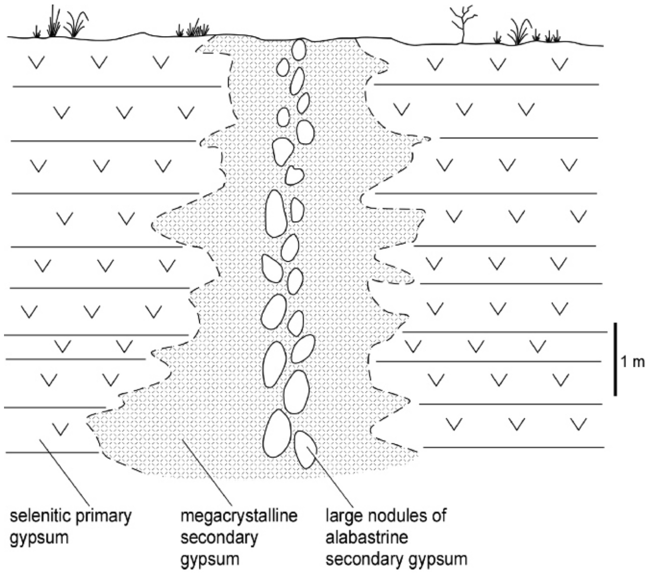

Fig. 13. Occurrence of large nodules of alabastrine secondary gypsum in a columnar arrangement in the Mazan gypsum quarry, Carpentras Basin, SE France (Paleogene); adapted from Ortí (1989*a*, fig. 17c)

affected primary gypsum facies in a number of Paleogene (Carpentras Basin) and Miocene units (Calatayud Basin, Tajo Basin, Coastal Depression of València) indicates that their growth was not related to mineral transformations in a deep burial setting.

In contrast, the growth of large replacive/displacive anhydrite nodules in the basins studied seems to be related to the existence of palaeohydraulic systems connecting the bounding mountain chains and the basins. In these systems, groundwater of deep, regional aquifers nourished in the chains would have discharged into the basin margins. This type of circulation has been documented in the Quaternary hydrogeologic systems of the Iberian margin in the Ebro Basin by Sánchez Navarro *et al*. (1999). Similarly, in Paleogene–Miocene times waters ascending from deep would have been discharged into the margins of the basins under study (Tajo and Tremp basins; Coastal Depression of València) recycling Mesozoic (Ca-sulphates, Na-chloride) evaporites. Presumably, these ascending, highly-saline and relatively warm waters became anhydritizing flows.

In the Calatayud Basin, particularly in the Fuentes de Jiloca Zone, two additional factors could have favoured the gypsum-to-anhydrite conversion into large nodules during burial. One factor is the lateral facies change in the lower evaporite unit from the highly-saline central zone, rich in Na-sulphate minerals, to the marginal zone of Fuentes de Jiloca, dominated by primary gypsum facies that are characteristic of low ionic concentration waters. Compactional brines from the central zone of this unit probably circulated laterally and also influenced the gypsum-to-anhydrite conversion in the marginal zones. The other factor, and the more important one, is that solutes coming from older Paleogene and Neogene evaporite units in this basin could have been added to the chemically recycled Triassic solutes in the deep regional aquifers. In fact, Ortí and Rosell (2000) reported a Na sulphate-chloride spring that currently discharges water at 24°C at the village of Paracuellos de Jiloca (Fig. 3A). According to borehole data, a thick halite unit (of Miocene age?) at a depth of between 170 and 537 m exists in this zone. This suggests that, during the Miocene, deep brines at relatively high T (probably >25°C) ascending from older Paleogene and Neogen evaporite units also contributed to the replacive or displacive growth of large nodules of anhydrite along the outer zones of the Miocene evaporite units close to the basin margins.

In the case of the Tajo Basin, some of the occurrences that directly overlie the highly-soluble materials of the Miocene Lower Unit (Saline Unit) suggest that compactional brines also favoured the growth of large anhydrite nodules in the marginal zones. Moreover, the influence of chloride-rich brines ascending through particular zones of the gypsum units under a structural control was considered in the Ninyerola Gypsum Unit and in the Carpentras Basin.

Although the depth at which the large anhydrite nodules developed during progressive burial cannot be precisely established, it is clear that the process occurred before the gypsum units became buried deeply enough to undergo a total gypsum-to-anhydrite conversion; thus, the process would have occurred during shallow-to-moderate burial conditions.

As stated above, the evaporite units bearing the large nodules are located mainly at the basin margins. Similar structures, however, could also develop in particular zones of the units with a different palaeogeographic or structural position, as in the diapiric anticline of the Ninyerola Gypsum Unit in the Coastal Depression of València. Besides the Iberian examples, the growth of large anhydrite nodules could also have occurred in other geological domains, particularly in evaporitic basins that recycled older evaporites similar to those described in this work.

CONCLUDING REMARKS

1. The textural characteristics of the gypsum that makes up the large nodules in the Iberian Paleogene and Neogene basins indicate that this diagenetic facies originated as anhydrite.

2. This mode of anhydrite formation is clearly different (large size and vertical arrangement of the nodules; no clear relationship of the nodules with depositional cycles) from that of the sabkha anhydrite. This mode bears no relation to the anhydritization that pervasively affects the gypsum units in a deep burial setting.

3. The growth of these anhydrite nodules occurred during shallow-to-moderate burial. In general, this growth affected (displaced/replaced) the host-gypsum sediments prior to their complete lithification.

4. The large anhydrite nodules developed preferentially in the gypsum units located in the basin margins and in the gypsiferous outer rings of the central evaporite units. However, they could also form in particular zones of the gypsum units that are associated with structural elements such as deep faults and diapirs.

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