

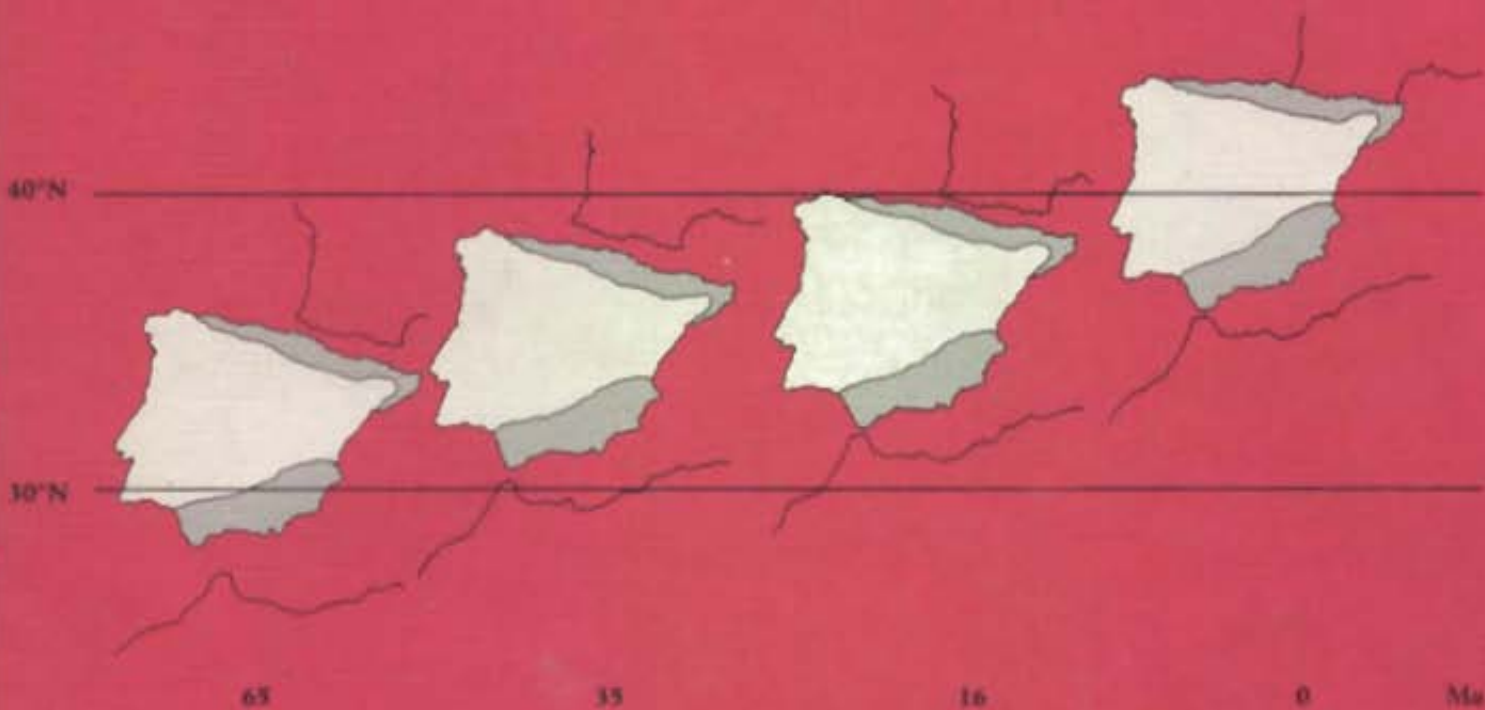
Tertiary basins of Spain

the stratigraphic record of crustal kinematics

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PETER F. FRIEND AND CRISTINO J. DABRIO

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G6 Mineral resources of the Tertiary deposits of Spain

M.A. GARCÍA DEL CURA, C.J. DABRIO AND S. ORDÓÑEZ

Abstract

Spain is the most self-sufficient country for minerals in the EU. A major proportion of these Spanish mineral resources has been obtained from Tertiary materials. The main materials exploited in Tertiary basins have been: brown coal and lignites, potassium salts, sodium salts (sulphates and chlorides), diatomite, sepiolite and other absorbent clays, bentonites, celestine, pumice, and also dimension (building-) stones and ceramics, portland cement and plaster raw materials.

Pb-Zn-Ag and gold *alunite* volcanogenic ores, related to Neogene volcanism, besides *Au*-placers have been mined from Roman times. Minor *Cu* and *Mn* occurrences are also reviewed.

The *brown coal* mines of Galician basins have provided all the significant production of Spain: more than 17 Mt. Low-quality Oligocene lignites in the eastern part of the Ebro Basin and Balearic Islands are less important from the economic point of view. Other occurrences are in the Guinzo de Limia, Guadix Baza, Granada (Arenas del Rey) and Alcoy basins.

The Spanish Tertiary basins (continental and marine) (Oligocene-Miocene) are filled by thick evaporites in which are obtained *potassium salts and sodium salts (sulphates and chlorides)*. The Montevives *celestine* mine is located in the evaporitic unit of the Granada basin (Miocene), and provides all of the Spanish celestine production. Spain is the world's third largest producer of celestine.

The Madrid basin and the minor Calatayud basin supply the whole of Spanish *sepiolite* production. The most important Spanish *attapulgite* production is obtained from the El Cuervo mine (Sevilla and Cádiz provinces). The genesis of the Cabo de Gata *bentonite* deposits is thought to be by hydrothermal alteration and halmyrolysis of Neogene volcanic rocks. The Madrid basin bentonites and 'pink clays' have been interpreted as an early diagenetic, even edaphic Mg-rich, attapulgite of illite clays. The most important areas of *ceramic clay* production in Spain are located in the Guadalquivir basin (Baillén area) and the Madrid basin (La Sagra-Alcalá de Henares).

The continental Neogene basin of the Hellin area supplies 90% of Spanish *diatomite* production.

Introduction

The estimated value of mineral production (oil and gas excluded) in 1990 in Spain was about \$4325 million, which represented over 1% of the country's Gross National Product. With a level of supply estimated at 35% of the domestic consumption of mineral raw materials, Spain is the most self-sufficient country in the EU (Mañana, 1992).

In 1990 the whole production of brown coal (20.9 Mt), potassium salts (0.78 Mt), sodium sulphate salts (0.71 Mt), bentonites (0.15 Mt), sepiolite-attapulgite (0.56 Mt), diatomite (92 kt), celestine (80 kt), and pumice was extracted from Tertiary rocks (ITGE, 1992). The Tertiary basins also provide a large proportion of lignites, natural stones, including commercial marbles, ceramic raw materials, cement raw materials (limestone, clays and gypsum), aggregates (crushed limestone and basalt) and gypsum for plaster manufacture.

This chapter presents a review of the most important mines, both active and historical, and occurrences located in – or related to – the sedimentary rocks of the Tertiary basins in Spain.

Metallic ores

The most important ore deposits are the *Zn-Pb-Ag* volcanogenic stockworks (network ore-bodies) mined in Neogene volcanic rocks in south-eastern Spain (Fig. 1). The origin and interpretation of these ore deposits are controversial, but in all cases the Neogene volcanism played an essential role in the ore genesis (Manteca & Ovejero, 1992). Silver has been mined since Roman times in La Unión-Cartagena district. Supergene enriched carbonates and sulphides in gossans were mined in the nineteenth century. The main deposits mined in this district are lead, silver, zinc, and minor baryte and iron ores. After the Spanish civil war, the production of the district rose to 90 Mt, producing lead (1.2 Mt), zinc (1.6 Mt) and silver (1.5 kt). Two *Pb-Zn-Ag* stockworks have been described recently in the Mazarrón district: one of these deposits contains 2.9% zinc, 1% lead, 28 g/t silver and 9 Mt ore, and the other contains 2.3% zinc, 0.7% lead, 20 g/t silver in a 5 Mt ore (Rodríguez, 1992).

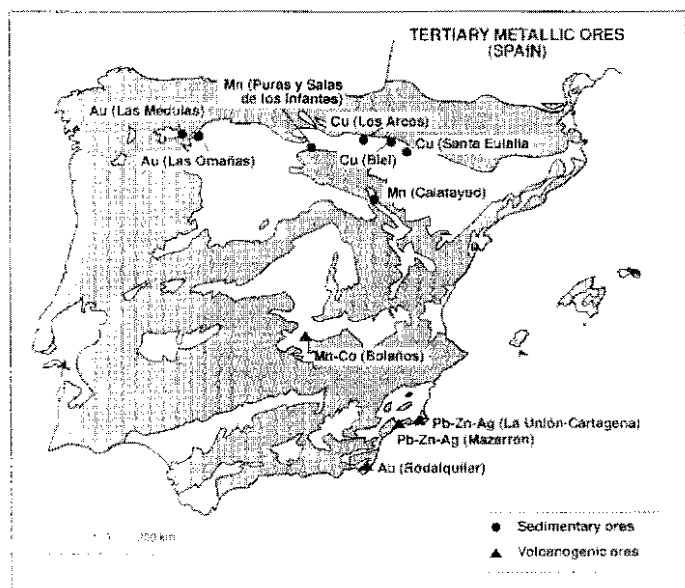


Fig. 1. Schematic location map of Tertiary metallic ores (see text).

The Rodalquilar *gold-alumite* field is related to calderas in the Cabo de Gata Neogene volcanic area (Fig. 1). Arribas Rosado (1992) described ore grades up to 10 g/t. Mined since 1970s, its production reached 85 kg in 1989.

The NW Iberian Peninsula, NW of the Duero Basin and the El Bierzo Basin, was mined for *gold* in Roman times (Pérez García & Sánchez Palencia, 1985, 1992). More than 7×10^5 m³ of gold-bearing sediments have been removed in over 600 mines. The total amount of gold exploited by the Romans in the alluvial deposits was about 200 t (Porter & Alvarez Moran, 1992). Gold grades up to 300 mg Au/m³ have been described in the alluvial fan deposits of the Las Médulas Formation in the Bierzo Basin (see Chapter W9). Channel fill deposits contain gold flakes over 70 microns across (Pérez García & Sánchez Palencia, 1985). There are reports of gold grades up to 50 mg Au/m³ in the Omañas Formation in the northern Duero Basin.

Some *copper* occurrences have been described in the Oligocene conglomerates of the north Ebro basin. The main ore minerals are cuprite, calcosine, galena and native copper, all them related to granular cementation of conglomerates. Ore deposition is thought to be induced by the oxidising action of infiltrational water on the formation waters of the Oligocene sediments. The thickness of individual conglomerate layers reaches 8 m. The major occurrence is the Biel Mine (Zaragoza), with estimated reserves about 500 kt and copper contents around 1.7–2%.

Small *oxide-hydroxide manganese* ores have been mined in the Campos de Calatrava (Bolaños district) since the nineteenth century, (Crespo Zamorano, 1988 a and b). The ore deposits are related to Pliocene alluvial-fan sediments of the Guadiana Basin. Manganese ores occur as granular cement in conglomerates and also as manganiferous crusts. Both types may be related to low-temperature thermal waters derived from the Campos de Calatrava

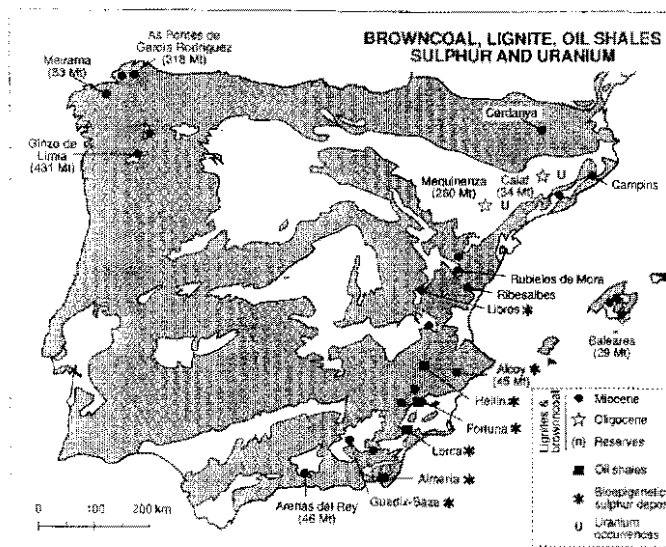


Fig. 2. Schematic location map of brown coal lignites and sulphur deposits, oil shales and uranium occurrences (see text).

vulcanism. Grade in these ores reaches 3.59% manganese, and 0.051% cobalt, with inferred resources of 15.6 Mt (Crespo Zamorano, 1988a and b). Occurrences of manganese oxides-hydroxides a granular cement ores have been cited in the western part of the Ebro and Calatayud basins.

Lignites, brown coal, oil shales and related sulphur and uranium

At present, the Tertiary basins of the Iberian Peninsula produce up to 17 Mt per year of brown coal, and some important quantities of lignite (Fig. 2). Spain was the second largest sulphur producing country in Europe during the first part of the present century and the Tertiary basins were the main suppliers of this production. In the 1970s native sulphur mining stopped as a consequence of sulphur recovery as a bi-product of oil and gas desulphuration.

The sulphur brimstone was obtained from biopigenetic sulphur deposits. The 1912 Spanish sulphur production was 14 kt, and at the same time Italy, the European leader, reached a production of 60 kt. The most important sulphur mines since Roman times are located in Coto Menor (Hellin basin). Up to eight sulphur seams, less than 1 m thick, have been exploited, with a 16% sulphur content. At the height of its activity this mine produced more than 50 kt per year. The rock age of the Hellin sulphur deposits is Late Miocene to Pliocene. Other biopigenetic sulphur deposits and occurrences have been described in the Baza, Fortuna, Lorca and Almería basins (Reyes *et al.*, 1992), the Libros basin (Anadón *et al.*, 1989), and Alcoy (Reyes, pers. commun.).

The increase of crude oil prices since 1971 has resulted in the increase of oil shale and coal exploration in the Tertiary basins (Reyes & Feixas, 1984; Reyes & Crespo, 1984). Laminated oil shale in the Neogene basins of SE Spain (Almería, Fortuna, Lorca

Table 1. Properties of oil shale deposits in the Cerdanya, Campins, Ribielos and Libros basins of eastern Spain (see Fig. 2 for the location of these basins)

Basin	Age	Thickness (m)	TOC (%)	Yield (l/t)
Cerdanya	Middle to Late Miocene	250	> 5	—
Campins	Upper Oligocene	150	11.5	50
Ribesalbes	Lower Miocene	100	1–15	87
Rubielos	Lower Miocene	250	—	30
Libros	Upper Miocene	10	1–2.6	20–70

Note:

TOC = total organic carbon.

Table 2. Uranium reserves in lignite seams of the Calaf, Sta. Maria de Queralt (village to the SSW of Calaf), and Mequinenza areas (see Fig. 2 for locations)

Area	Reserves ¹	Resources ¹
Calaf	15.3	26.3
Sta Maria de Queralt	13.6	26.3
Mequinenza – Fraga – Almaret	51.9	93.4

Note:

¹Reserves US \$15–30/lb U₃O₈

Hellin), located in a marine pre-evaporitic environment of Tortonian–Messinian age, have been described (Reyes *et al.*, 1984). On the other hand, Anadón *et al.* (1989) have described some oil shale occurrences in the Cerdanya, Campins, Ribesalbes, Rubielos and Libros basins (see Fig. 2). Some technical properties – TOC (total organic carbon), oil yield (l/t) and geological data (age, oil bearing shales unit thickness) – have been included in Table 1.

Radioactivity anomalies in the lignite seams of the Calaf and Mequinenza areas (Ebro basin) (Ramírez Ortega, 1966) led to the investigation of the uranium contents and distribution in these Tertiary basins. As a result of this exploration, the uranium resources of the lignite seams of both basins have been estimated as up to 80.8 kt of U (which sells at US\$15–30 per pound of U₃O₈) (Martín Deigado, 1975) (Table 2). At present, severe difficulties in extracting uranium from lignites, in addition to the availability of important Spanish high-quality uranium in proven quantity, make these Tertiary deposits uneconomic.

Fossil fuels (oil and gas excluded)

Brown coals of the Galicia basins

Several Tertiary basins occur along two dextral slip fault zones in NW Spain. Brown coals were formed in the terrigenous deposits of these basins, as a result of the evolution of the alluvial and limnic systems. The largest coal deposits recorded so far in the zone occur in the As Pontes de García Rodríguez and Meirama

basins (littoral basins) and the Xinzo da Limia basin (SE Galicia) (see Chapter W9). The age of the Tertiary coal-bearing sediments is not clear, because there is a controversial disparity in the dates interpreted from mammal vertebrate biozonation and from palynological biozonation (Chapter W9).

As Pontes de García Rodríguez basin This basin is located in the NW–SE-striking structural corridor that extends for 55 km, via Pedroso – As Pontes – Moinonoro (Santanach *et al.*, 1988). It is an elongate basin with 7 km maximal length and a width varying from 1.5 to 2.5 km (Bacelar *et al.*, 1991). As mentioned earlier, the As Pontes basin is a compressional basin partly controlled by a dextral strike-slip fault. The sedimentary fill was alternately overthrust by, and overlapping, the Hercynian basement during the successive deformation phases. The coal seams appear to have formed as a consequence of relative restriction and expansion between the limnic and alluvial systems in response to the varying subsidence of the deformation phases. Two depocentres developed in the basin: one located in the NW, the West Field, and another located in the SE, the East Field. The fill of the As Pontes basin has been divided into four sedimentary units represented in both coal fields. The thickness of individual seams varies from 0.5 to 25 m, and displays continuity along the basin, although there are lateral changes to terrigenous sediments in the marginal facies. In the basin, 19 coal seams have been identified. Three main lignite types have been distinguished: brown coal, xiloides and pyropisitic lignite, the last mainly formed by resins. The vitrinite reflectance varies from 0.1 to 0.4, the average being 0.3 (Martín Calvo, 1973). The average calorific value varies from 1600 to 2200 kcal/kg. The waste/lignite ratio average in the open pit mines is up to three. The 1979 reserves of the As Pontes basin were 318 Mt, and the 1989 lignite production in the basin was 12.6 Mt.

Meirama basin This basin is located in the NW–SE-striking structural corridor via Lendo–Meirama–Baimil (Santanach *et al.*, 1988). The surface area of the basin is about 1.5 km²; it has an elongate shape, with a long axis length of about 2.6 km, parallel to the main dextral strike-slip fault. The thickness of the basin fill is up to 250 m and it is longitudinally folded. The production in 1989 was estimated as up to 5 Mt, and the proved resources are nearly 80 Mt.

Xinzo da Limia basin This basin is located in the Vilalba–Maceda–Xinzo da Limia structural lineation, which trends NNE–SSW. The sediment fill of the basin is up to 250 m thick, and up to 130 m of this may be lignite seams. The quality of the lignite is brown coal, and the proven reserves may attain 431 Mt. The economic potential of the deposit was investigated recently by ENDESA (Baltuille, pers. commun.).

Lignites of the Ebro Basin

Several coal sequences developed in Oligocene times in the Tertiary Ebro basin. Despite the fact that the individual seams of coal are thin and subeconomic, mining has taken place since the nineteenth century in the Calaf and Mequinenza areas. In both

areas, the organic biomarkers identified by Gorch *et al.* (1992) show that the original organic matter derived mainly from higher plants and bacteria.

Calaf area The coal-bearing stratigraphic section formed in the Segarra lacustrine system and consists of limestones, marls, terrigenous and even evaporitic sediments of Early Oligocene age (Gorch *et al.*, 1992). The paludine-lacustrine, coal-bearing intervals consist mainly of limestone and grey mudstone beds that interfinger with lenticular channel-fill sandstones showing cross-bedding and ripple laminations. The grey massive mudstones are interbedded with coal seams ranging from a few centimetres to 0.8 m in thickness. Organo-sulphur compounds (Gorch *et al.*, 1992) are mainly responsible for the high sulphur content of the lignites. The coals consist mainly of lignite to sub-bituminous coal with calorific values that vary from 6.400 to 7.042 kcal/kg (Cabrera & Sáez, 1987), a sulphur content that varies from 2.98 to 8.36% and ash content ranging from 16.46% to 24.18%. The reserves in the Calaf area are up to 34 Mt (IGME, 1985). From the petrological point of view, the coals are vitrinite-rich with minor amounts of exinite and inertite. The vitrinite reflectance ranges from 0.4 to 0.7 (Martín Calvo, 1973). This author pointed out the relationship between the uranium content and the organic matter content, and that uranium is preferentially associated with humines and humic acid fractions. Local values of up to 0.180% U_3O_8 content have been recorded, although average values are nearer to 0.020% U_3O_8 .

Mequinenza area The coal-bearing stratigraphic interval formed in the Los Monegros lacustrine system and consists of limestones, marls, and evaporitic and terrigenous sediments of the Late Oligocene. The immediate coal-bearing strata of the Los Monegros lacustrine system consist mainly of pale brown to grey micritic limestones, grey mudstones and minor sandstone lenses and sheets. Thin lenticular coal seams interfinger with the lower and middle part of the unit. Individual seams are generally thin with sharp and well-defined boundaries. The average thickness is less than 0.3 m, and varies from a few centimetres to 0.9 m. The coal deposits consist mainly of an ash-rich lignite, with 30–53% average ash content, a high sulphur content, ranging from 1.54 to 11.88%, and calorific values ranging from 3.500 to 5.500 kcal/kg (Cabrera & Sáez, 1987). The proven reserves are 260 Mt (IGME, 1985).

Table 2 presents data on the uranium resources in both the Calaf and the Mequinenza coal areas.

Other Tertiary coal basins

The only mines active at present and not already discussed are in Balearic Basins. These mines produce up to 14 kt/year of lignite. The identified reserves of lignite in these basins are up to 71.4 Mt, and the proven reserves are 29 Mt (Fig. 2).

In the Balearic Islands, lacustrine deposits have been described, of Early Oligocene, Ludian-Sannoisian age (Colom, 1983) and of Middle Eocene age (Ramos-Guerrero *et al.*, 1989). Only the deepest lacustrine facies of the central part of Mallorca contains lignite seams (Sta María, Binisalem, Alaró, Inca, Lloseta and Selva).

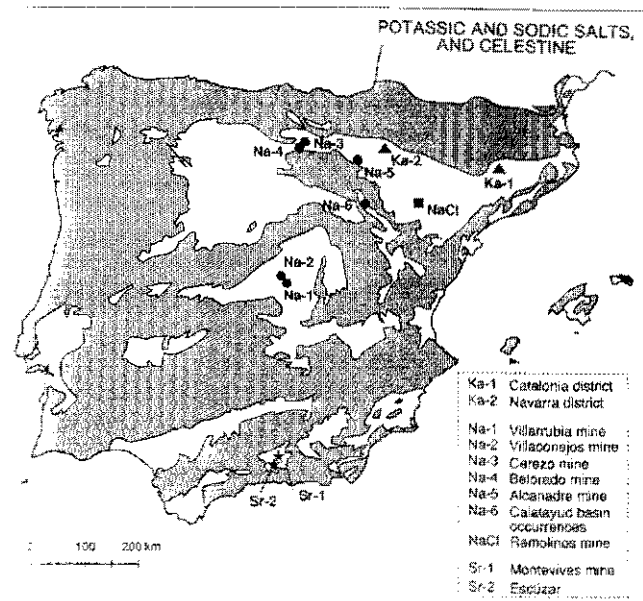


Fig. 3. Schematic location map of potassic salts, sodic salts and celestine mines (see text).

Recently, Ramos-Guerrero *et al.* (1989) have described the Binisalem member, which consists mainly of an alternation of limestone (bioclastic wackestone) and lignites. The coals consist mainly of a humic matrix that contains exinic macerals, such as resinite and cutinite (Ramos Guerrero *et al.*, 1989) with calorific values up to 4.6 kcal/kg, 1.7% sulphur content and 18.5% ash content (Colom, 1983).

Lignite occurrences in Tertiary basins have been cited in the Cerdanya, Alcoy, Guadix-Baza and Granada basins (Arenas del Rey). In the Prats-Alp area of the Neogene Cerdanya basin some lignite beds have been identified in unit C of Anadón *et al.* (1989) (Fig. 2). The lignite-bearing sediments of the Alcoy, Guadix-Baza and Granada basins have been reported as Late Miocene – Pliocene. Lignites from these basins are mainly soft brown coals.

In the Neogene Madrid and Duero basins, in the lower evaporitic units, and also in the limnic shallow marly sediments, minor organic matter occurrences (lignites and/or oil shales) have been cited.

Potassic and sodic salts, and celestine

Spanish Tertiary basins, both continental and marine, contain thick evaporitic deposits from most ages of the Tertiary. Potassium salts, sodium salts (sulphate and minor chloride), celestine and gypsum are obtained from these Tertiary basins. In this section we focus on potassic and sodic salts and celestine. (Fig. 3).

Potassic salts

The Spanish production of potassic salts has decreased from 860 kt of K_2O in 1981 to 585 kt in 1991. All the production is concentrated in two mineral districts: Catalonia and Navarra.

Table 3. *Estimated mineral reserves of potassic salts in the Catalonia and Navarra districts*

District	Amount of resource (kt)		
	Proven	Probable	Possible
Catalonia	10470	9185	16645
Navarra	2000	8000	12000

Source: From ITGE (1992).

Estimated mineral resources and reserves in these areas are presented in Table 3.

Salinity increased dramatically in the Sub-Pyrenean basin during the Late Eocene–Early Oligocene (Ludian); most probably it was this salinity crisis that was responsible for the deposition of potassic salts in the basin. Deep drill holes reach potassic salts in the central part of the Ebro basin below a thick cover of Oligocene–Neogene sediments. Pueyo (1975) and Rosell (1983) interpreted the deposition of salt as related to marine brine with a low content of magnesium sulphate. The basin underwent active nappe (overthrust) tectonics after the deposition of the salts and, according to Rosell (1983), the Catalonia district remained in the autochthonous zone, whereas the Navarrese district was transported by thrusting. As a result, erosion removed the seams of potassic salts in the cores of the anticlines of the Navarrese district, while the anticlinal cores were preserved in the Catalonia district under a thick cover of Oligocene sediments.

Catalonia district The structure of the Catalan district is an E–W-trending syncline, with salt thicknesses ranging from 150 to 500 m (Pueyo, 1975) as a consequence of local tectonics. In the Cardona diapir the thickness of potassic salts reaches 2000 m. The main, and best-known mines (Cardona, Balsareny and Suria) are located in this district.

The stratigraphic section includes, from bottom to top (Fig. 4):

1. Marine marls (Igualeda marls).
2. 4 to 5 m, laminated anhydrite.
3. 130 to 200 m, massive halite (sal de muro).
4. 5 to 20 m, lower potassic unit, consisting of decimetre sequences of terrigenous clay, carbonates, sulphates, halite and silvite. This unit is divided into two by a thick layer of massive halite.
5. 40 to 80 m, upper potassic unit (carnallitic unit) including several seams (three or four in the Suria mine, and more in other localities). It is interesting to note that the content of halite (low-grade potassic salt) in this unit increases when the seams are thinner and less numerous.
6. 85 to 120 m, grey mudstones, or transition unit. Some intercalations of halite have been described in the lower part of this unit.
7. 500 m (or more), lacustrine reddish deposits (top).

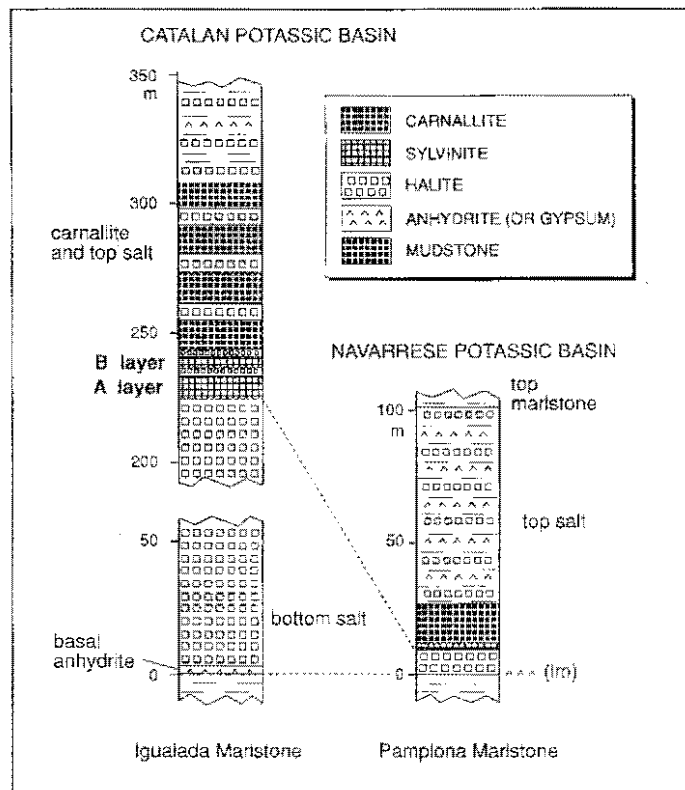


Fig. 4. Stratigraphic section correlation between Catalan and Navarrese potassic basins. After Rosell (1983).

Navarrese district

As in the former case, the potassic salts are associated with marine marls (Pamplona marls). The only mine (Subiza shaft) is located in a synclinal basin (the so-called Pamplona potassic basin or Sierra del Perdón potassic basin), but a research programme has only recently been initiated.

A general stratigraphic succession includes:

1. Deep marine marls (bottom).
2. 0.6 to 1 m, laminated anhydrite unit.
3. Almost 10 m, lower banded, massive halite unit (sal de muro).
4. 2 m, lower potassic unit, formed by 18 silvinit–halite couplets; some of them are used as markers.
5. 12 m, carnallitic unit, made up of eight halite–carnallite couplets.
6. Top halite unit, made up of sequences of decimetre- to metre-thick red mudstones and centimetre-thick halite layers.
7. Up to 50 m, top marls, mudstones with some anhydrite layers.

It should be noted that this stratigraphic section is very similar to the one in the Catalonia district (Fig. 4). The only difference is the total thickness.

Table 4. Reserves of sodium salts in Spanish mining districts

Company	Raw material	Plant location	Basin	Extractive method	Proved reserve (Mt)
Crimidesa	Glauberite	Cerezo de Río Tirón	Ebro Trench	a	19
Foret	Thenardite	Villarrubia	Madrid basin	b	15.5
Minera S. Marta	Glauberite	Belorado	Ebro Trench	a	63
Sulquisa	Glauberite	Villaconejos	Madrid basin	a	57

Notes:

(a) Open-pit solution; (b) underground mine.

Source: From ITGE (1991)

Sodium salts

For many years, Spain has been the European leader in natural sodium sulphate production, and all of it comes from Tertiary continental basins. The 1990 Spanish natural sodium sulphate production, in terms of Na_2SO_4 content, amounted to 714 kt, 240 kt from thenardite only in the Madrid basin, and the remainder from glauberite in both the Madrid and Ebro basins, particularly the Tertiary Trench or Bureba Corridor in the western part of the Ebro basin (ITGE, 1992). Table 4 presents some data on the reserves of the mining districts.

There are two active mines in the Madrid basin (Fig. 5), one located near Villaconejos–Colmenar de Oreja, and another located near the eighteenth-century mine in Villarrubia de Santiago. There are many no-longer-active mines in the Madrid basin, but, in the past, the most common method of recovering salts was to use saline springs and wells (e.g. Espartinas, Carabaña, Carcaballana, Loeches), and some of these were even being exploited in Roman times.

The mine located near Villaconejos–Colmenar de Oreja (the Fátima mine), is an open-pit dissolution mine. The mineral glauberite is preferentially dissolved in pools located over a bed of glauberite–anhydrite that is 27 m thick. The brine is recovered by pumping from wells and sent to an evaporation plant where top-quality sodium sulphate precipitates. The source brines of the evaporation plant are introduced into pools and then recirculated (Ordóñez *et al.*, 1982).

The mine located near Villarrubia de Santiago ('El Castellar') is a pillar and room underground mine that works a 5–8 m thick thenardite bed with minor glauberite. The mineral extracted is processed in an evaporation plant to obtain the commercial sodium sulphate (Ordóñez *et al.*, 1982).

The Neogene stratigraphic record of the Madrid basin is summarised by Megias *et al.* (1983) and by Calvo *et al.* (Chapter C2). The sodium sulphate mines are located in the Lower Unit. The sedimentological interpretation of the Lower Unit or Saline Unit has been discussed by Ordóñez *et al.* (1992) and by Ordóñez & García del Cura (1995). The mineralogical and petrological features of the economic saline deposits have been described by García del Cura (1979), García del Cura *et al.* (1979) and Ortí *et al.* (1979). The

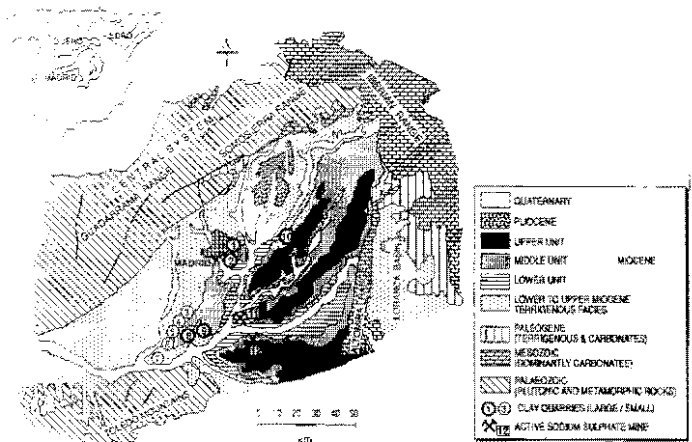


Fig. 5. Location map of main active mines in Madrid basin: 1, Vicálvaro sepiolite mine; 2, Almodóvar sepiolite mine; 3–6, Yepes–Cabañas de La Sagra mines (bentonites and sepiolite); 7, La Sagra ceramic raw clay mines; 8 and 9, Esquivias special clay mines; 10 Alcalá ceramic raw clay mines; 11, Villaconejos ('Fátima mine') sodium sulphate mine (glauberite); and 12, Villarrubia ('El Castellar') sodium sulphate mine (glauberite and thenardite).

only paleontological data obtained from the Saline Unit are the flora found in an exploration drill hole near the Villaconejos mine. This flora indicates a Late Oligocene – Early Miocene age for the Saline Unit (Alvarez Ramis *et al.*, 1989).

The 'El Castellar' mine is placed in the Upper Saline Subunit. The sedimentology and facies distribution of this Upper Saline Subunit are poorly understood, and are, at present, under review by the authors of this chapter. However, it is possible to point out some distinctive features of this Upper Saline Subunit, one being the presence of thenardite as the main mineral, and another being the local and restricted character of this Subunit. In this Upper Subunit it is possible to identify at least six repetitions of the following sedimentary dm to m sequence: 1. reddish mudstone containing interstitial halite; 2. muddy terrigenous sediment \pm halite \pm glauberite; 3. massive thenardite \pm glauberite. The thenardite \pm glauberite bed mined in Villarrubia is up to 8 m thick (Fig. 6). The massive thenardite mined there displays a typical blue colour and is thought to be a secondary mineral from mirabilite. This soft

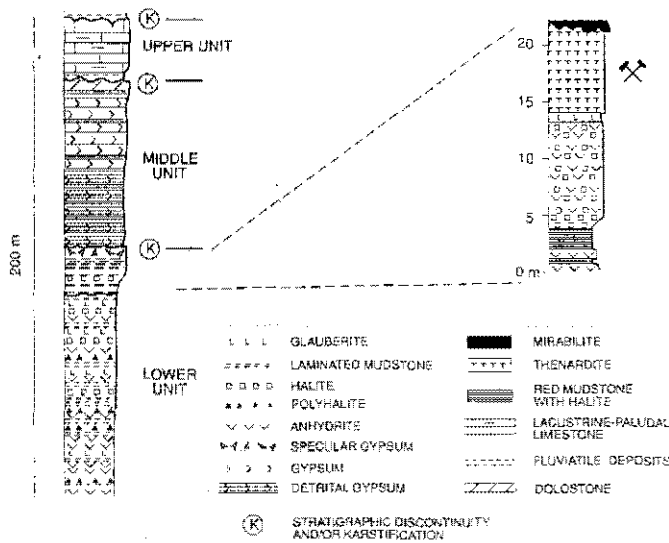


Fig. 6. Location of Villarrubia thenardite ± glauberite mine. Stratigraphic section of central part of Madrid basin and a detailed section of thenardite seam mined there.

and wet crystalline mirabilite precursor of thenardite formerly contained some precipitated idiomorphic crystals of glauberite that are now enclosed in massive thenardite.

The glauberite mine of Villaconejos exploits the lower Saline Subunit (Fig. 7). This Subunit is thought to have formed in a perennial saline lake surrounded by a wide mudflat. As pointed out by Ordóñez *et al.* (1983) and Utrilla *et al.* (1992), brines of this saline lake were derived mainly from the weathering of Upper Cretaceous–Paleocene marine evaporites. Saline mineral associations in this stratigraphic interval are reported in Fig. 7. Glauberite occurs as a massive bed of idiomorphic glauberite crystals, with minor magnesite marls interbedded with anhydrite and micritic magnesite. Glauberite cements and nodular anhydrite have recently been interpreted to result from the early diagenetic glauberitisation of primary anhydrite in the muddy lacustrine belt during episodes of low lake water levels (Ordóñez & García del Cura, 1993).

The Cerezo de Río Tirón, Santa Marta and Belorado glauberite mines are located in the Tertiary Trench, 'La Bureba Corridor', that connects the Ebro and Duero basins. Glauberite mines are located in the Cerezo Evaporite Formation of uncertain Late Miocene age, and are restricted to the central part of a small basin. The Evaporite Formation displays sharp lateral facies changes to non-economic detrital sediments through saline mudstones with nodular anhydrite. A perennial saline model has been proposed to explain the general features of the Evaporite Formation (Menduiña *et al.*, 1984). The detrital sediments laterally associated with the saline deposits have been interpreted as the distal part of a prograding lacustrine deltaic system, and this model has been used in the exploration of the Santa Marta mine, which started production in 1989.

The recovery method used in Cerezo de Río Tirón and Belorado is similar to that described in the Villaconejos mine, that is to say, an

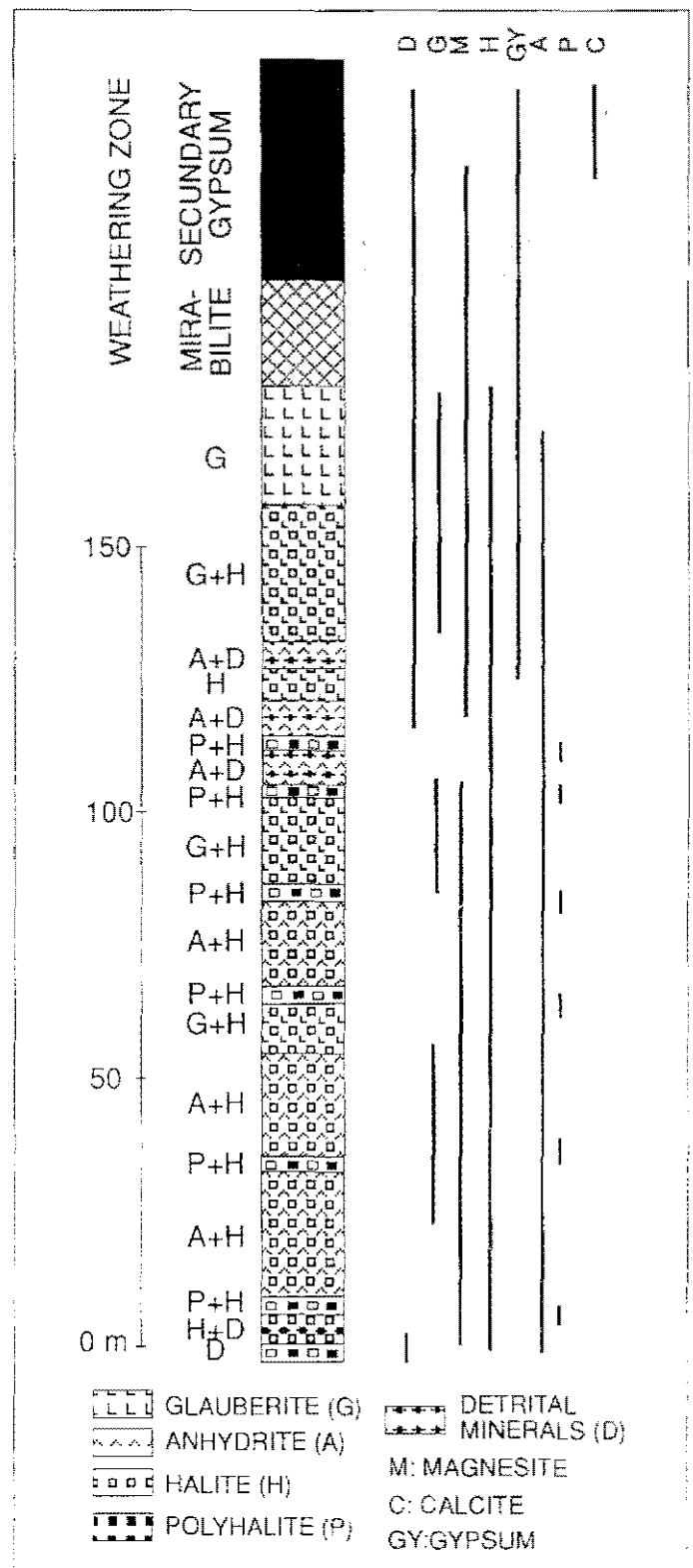


Fig. 7. Schematic mineralogical section of an exploration drill hole located close to the Villaconejos glauberite mine.

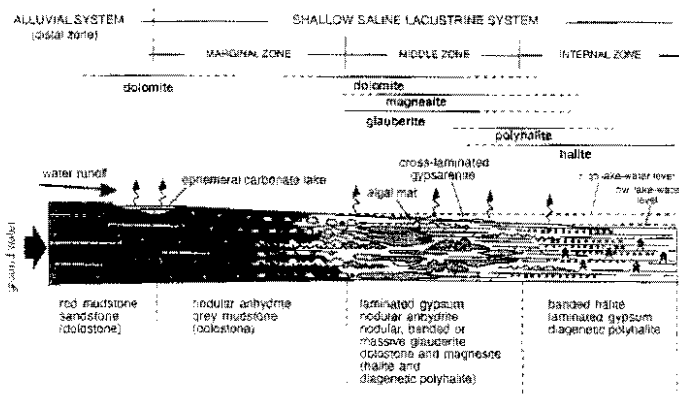


Fig. 8. Paleoenvironmental and sedimentological model of Alcanadre non-active mine. Adapted from Salvany & Orti (1992).

open pit solution mine cut down to a glauberite seam; the resulting brine is the raw material for top-quality commercial sodium sulphate.

The Cerezo Evaporite Formation stratigraphic interval contains alternations of metre-thick seams of microcrystalline glauberite and non-economic muddy anhydrite. The number of glauberite seams is up to eight, but the recovery methods, and the commercial productivity of the seams, are conditioned by the present-day topography and by the impoverishment of the upper seams by infiltration of meteoric waters.

Some no-longer-active underground mines with glauberite have been described from the western Ebro basin. These glauberite deposits are located in the Lerin Formation of Late Oligocene–Early Miocene age (Salvany, 1984; Salvany & Orti, 1992, 1994). These authors have recently proposed a paleogeographical and sedimentological model to explain the main features of the Alcanadre mine zone (Fig. 8). The thickness of the overburden and the thinness of the glauberite beds result in high values of the stripping ratio and the submarginal economic character of these occurrences.

Some glauberite occurrences in the central part of the Ebro basin and below the halite beds of La Real mine (Remolinos–Zaragoza) have been described (Fernández Nieto & Galán, 1979; García Veigas *et al.*, 1991). Some outcrops of glauberite beds have also been reported in the Calatayud basin (IGME, 1980; Sánchez Moral *et al.*, 1993).

Halite rock salt is present in both continental and marine Tertiary basins of the Iberian Peninsula. The 1990 Spanish production of sodium chloride was 3.3 Mt, 19.8% of which was obtained as a byproduct of Tertiary potassic salt recovery, and 3.2% was obtained from the Tertiary Ebro basin. A high proportion of the remainder was obtained from the natural evaporation of marine brines, and a smaller proportion from Triassic rock salt deposits. The rock salt deposits of Remolinos and Torres de Berrellen in the Zaragoza Formation (Lower Miocene) of the central part of the Ebro basin have been mined for a long time. Proved reserves amount to more than 16 Mt NaCl. The halite deposit is up to 100 m thick, and consists of decimetre layers of halite with minor nodular

anhydrite (Orti & Pucyo, 1977). The lateral continuity of the deposit is over 30 km along the Ebro river valley (Orti, 1990).

Celestine

Spanish celestine production has risen from about 19 kt in 1980 to more than 80 kt in 1990. Spain is the third country in the world ranking of production of this mineral. Celestine is produced from the Montevides mine, about 12 km southwest of Granada (Granada Province). The Montevides ore grade is 80% SrSO_4 , with proven reserves of 3 Mt. The regularity of the ore body in this case enables the deposit to be competitively concentrated by the flotation method.

The Montevides mine is located in a small hill in the central part of the Tertiary Granada basin. The host sedimentary materials are of Middle to Late Miocene age. Recently, Rubio Navas (1990) has pointed out the near domical structure of the ore body and the tectonic character of the celestine ore body outcrop.

The celestine ore is strata bound, up to 40 m thick, and it dips 20° to 50° towards the northwest. The celestine mineralisation displays a mm-scale, clear–dark banded texture similar to the primary stromatolite textures of the host limestone and marls. The rich ore displays a massive structure and earthy and/or microcrystalline textures with obliteration of primary structures. The ore body is well stratified in layers ranging from 20 to 50 cm thick. In addition to celestine, the mineral paragenesis of the Montevides ore deposits includes calcite, dolomite, quartz, gypsum, strocianite, Fe–Mn oxides and hydroxides, phyllosilicates, etc.

The Escúzar village celestine occurrence is an E–W-striking outcrop with more than 10 km lateral continuity – the Escúzar Celestine Belt of Martín *et al.* (1984). The host rock of the celestine ore is also a stromatolitic limestone, and the thickness of the deposit is roughly 20 m. The average celestine content is lower than at Montevides, and rarely rises to 55%. A significant feature of this occurrence is the presence of a karstic surface at the top of the deposit, with dolines and other karstic depressions filled by low-grade brecciated ore and stromatolite carbonates. Escúzar has estimated reserves of about 1 Mt.

Both the Montevides and Escúzar celestine deposits have been interpreted as the results of early diagenetic cementation of stromatolitic carbonates when dessication processes followed the fall in sea-level (Martín *et al.*, 1984). In this model, the strontium is derived from marine connate waters.

Other celestine occurrences have been cited in the Duero Tertiary basin (Ordóñez *et al.*, 1980), and Ebro basin (Rubio Navas, 1990).

Special clays (sepiolite, attapulgite and bentonites), ceramic clays and diatomites

Sepiolite, attapulgite and bentonite deposits are commonly associated in the Tertiary clayey formations, and in consequence these minerals are mined together or in the same areas. The official statistical data of these special clays show some contradictory interpretations because the uses of the special clays are similar

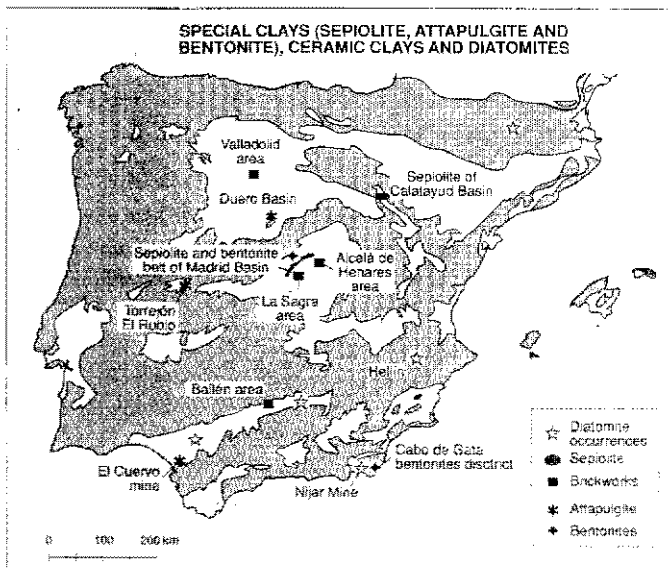


Fig. 9. Schematic location map of special clays (sepiolite, attapulgite and bentonite), ceramic clays and diatomites (see text).

and generally connected with the physical properties of absorption, exchange capacity, etc. As a consequence, we also include in this section the diatomite deposits because these physical properties are also their most characteristic feature.

Sepiolite

The 515 kt produced in Spain in 1990 were obtained entirely from the Tertiary basins: 453 kt from the Madrid basin, 61 kt from the Calatayud basin and 1 kt from the Duero basin. The sepiolite resources in the Madrid basin reach 100 Mt, and the estimated production capacity is more than 1 Mt/year. Sepiolite is also present in the attapulgite deposit of Lebrija (El Cuervo mine Guadalquivir basin), with a sepiolite grade of up to 5% (Fig. 9).

In the Madrid basin, sepiolite is quarried at Vicálvaro, Vallecas, Parla and San Blas, to the south of the city of Madrid (Fig. 5), and Yuncillos (prov. Toledo) (Galán & Castillo, 1984; Ordóñez *et al.*, 1992). The sepiolite seams occur at the top of fining-upward sequences of arkosic sandstones. The economic sepiolite seams occur at an intermediate position between the lacustrine deposits of the Middle Unit of the Madrid basin and arkosic sediments derived from granitoid and high-grade metamorphic source areas that outcrop in the northwestern part of the Madrid basin (Fig. 10).

The classic sepiolite deposits of Vallecas–Vicálvaro were exploited three centuries ago to obtain the light rough stone of Madrid buildings, and special refractory clays. The areal extent of these deposits is almost 6.6 km², and they consist of two exploitable layers of sepiolite with sharp lateral changes of facies to the neighbouring abundant arkosic sandstones. The thickness of the upper layer, the richest in sepiolite, is up to 10 m, and it is separated from the lower sepiolite layer by more than 15 m of non-economic muddy sandstones. The lower sepiolite layer is 1 to 5 m thick. A

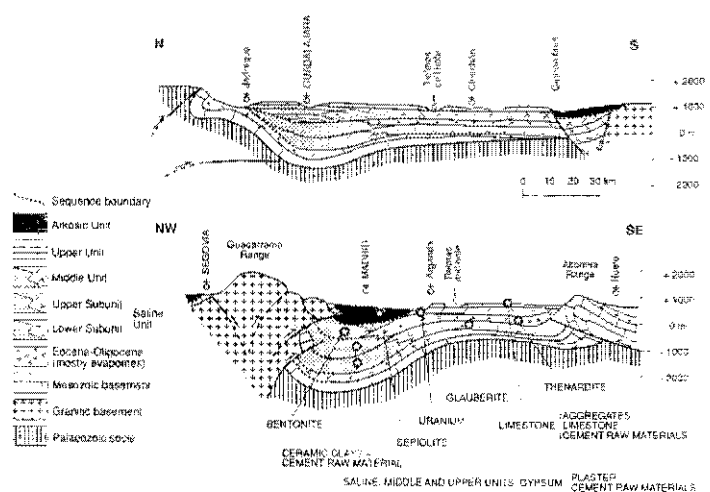


Fig. 10. Schematic cross-section of the Neogene Madrid basin (adapted from Megias *et al.*, 1983) showing stratigraphic location of main mined mineral deposits: glauberite, thenardite, sepiolite, bentonite, ceramic clays, Portland cement raw materials, dimension stone (Colmenar limestone) aggregates (crushed limestones), lime raw materials and uranium occurrences.

classic mammal site (Cerro Almódovar site) of Middle to Late Aragonian age is located in the muddy marls at the top of the lower sepiolite layer. The sepiolite grade varies between 65% and more than 95%, being accompanied by smectites, quartz, illite, feldspars and carbonates. This is the world's most important known deposit of sepiolite (Galán & Castillo, 1984).

The Yuncillos sepiolite deposits extend over 3 km² and there are two sepiolite-containing layers. The lower sepiolite layer is richer than the upper layer and its thickness is up to 3 m. The upper layer contains smectite and nodular chert. The location is close to Vallecas and Vicálvaro. In Paracuellos del Jarama (east of the city of Madrid) there are important reserves of sepiolite, probably subeconomic because of the thick cover and its location in an urban area. The seams of the Paracuellos sepiolite deposits are interbedded with burrowed muddy arkosic sandstones and, in places, with reworked sepiolite and paleosols (Alonso *et al.*, 1986). The genesis of the Paracuellos sepiolite deposits has been reported as edaphic to paludal in a distal alluvial fan environment (Calvo *et al.*, 1986). Some uranium vanadates (yuyamunite) have been formed in the edaphic environment, associated with dolocretes and with vertebrate remains in the marls (Arribas, 1963). In our opinion, it is possible to define a *sepiolite belt* along the Madrid arkosic trench, elongated from NW to SE from Yuncillos to Paracuellos del Jarama, with many sepiolite occurrences as at north La Sagra, north Esquivias, Parla, North Getafe, etc.

In the Calatayud basin, sepiolite marls were exploited a few years ago in the Isabel mine (Orera). The annual production of this mine, as we have pointed out before, was up to 61 kt in 1990 (ITGE, 1992). The sepiolite deposits of the Calatayud basin are interpreted as lacustrine deposits in a brackish shallow lacustrine environment. Arauzo *et al.* (1989) describe the mineralogical paragenesis: dolomite, illite, smectite and quartz, with minor amounts of calcite,

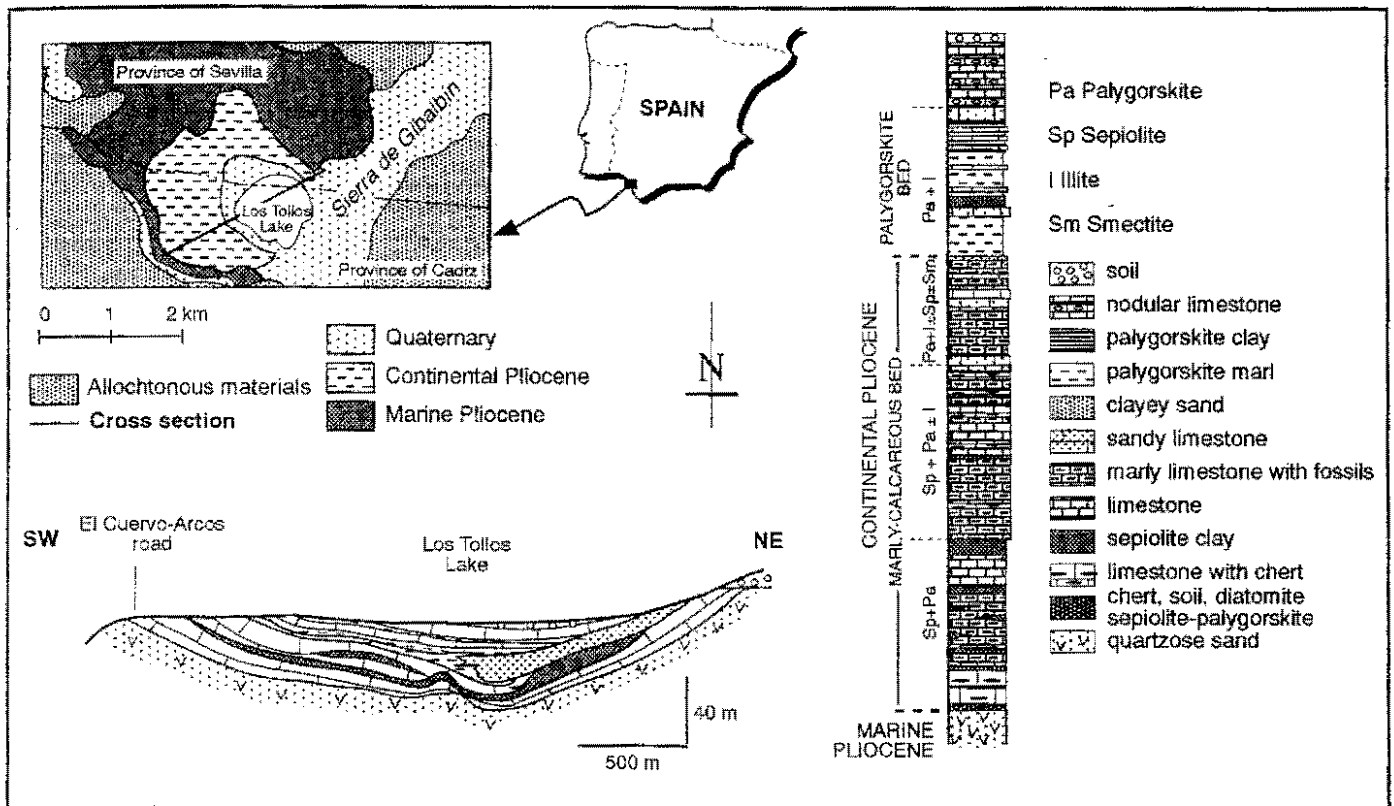


Fig. 11. Geological location of El Cuervo attapulgite mine, cross-section and detailed stratigraphic section. After Galán & Ferrero (1982).

feldspars, chlorite and kaolinite in the Mara area, and sepiolite alternating with smectites in the Orera area. Early diagenetic vadose to edaphic processes of carbonate-siliceous (due to diatoms and/or detrital clays) sediments may explain the genesis of the sepiolite marls. Other occurrences of sepiolite marls south of Guadalajara (Madrid basin) and Cuestas Facies (Duero basin) may be interpreted in a similar way.

Attapulgite

The 1990 Spanish attapulgite production was 54 kt. The most important production comes from the El Cuervo attapulgite mine (Sevilla and Cádiz provinces) and only 4 kt was from the Torrejón el Rubio attapulgite deposit (Fig. 9).

The El Cuervo mine is located in a Pliocene to (?)Quaternary lacustrine deposit (Galán & Ferrero, 1982) (Fig. 11). Two main units are distinguished: a lower marly layer and an upper palygorskitic (attapulgitic) layer. The thickness of the attapulgite layer varies from 3 to 30 m with an attapulgite grade of between 35 and 75% and with minor sepiolite (0–30%) and calcite. It includes some fossiliferous limestone beds interbedded in the attapulgite seam. The estimated resources are up to 30 Mt.

The Torrejón el Rubio deposit is located in the small Tertiary fault basin of Torrejón el Rubio (Galán & Castillo, 1984). The source area and the basement of the basin are deeply weathered slates. The lowest basin fill is composed of weathered slate material

and this layer is overlain by a terrigenous-muddy bed with a thickness that varies from 6 to 50 m. On top of this bed (0.5–4 m) an attapulgite grade of up to 70% can be measured. Paragenesis of the attapulgite deposit is, in addition to attapulgite, palygorskite ± illite ± smectite (saponites) ± chlorites ± sepiolite ± quartz ± feldspars and dolomite. The genesis of the attapulgite deposits appears to be a result of a magnesium-rich weathering process that affected clayey, probably illite-rich, sediments (Fig. 12).

Some attapulgite occurrences have been described in the lacustrine sequences of the Tertiary Duero basin: García del Cura & López Aguayo (1974); Pozo & Leguey (1985); in Sacramenia: Martín Pozas *et al.* (1983); and in Bercimuel: Suárez *et al.* (1989) and Suárez *et al.* (1991). Other less-important attapulgite occurrences have been described in which the mineral forms a cement of the proximal detrital facies of the Madrid and Duero basins (Ordóñez *et al.*, 1977; Megías *et al.*, 1982; Leguey *et al.*, 1984a and b). The paragenesis of these attapulgite occurrences includes palygorskite ± cristobalite ± dolomite, interpreted as late diagenetic cement of the arkosic sandstones.

Bentonites

The 1990 Spanish production of Ca-bentonite was 151 kt, and this came from two districts: Cabo de Gata (Almería province) and Villaluenga (Toledo province).

The Cabo de Gata district deposits are associated with the

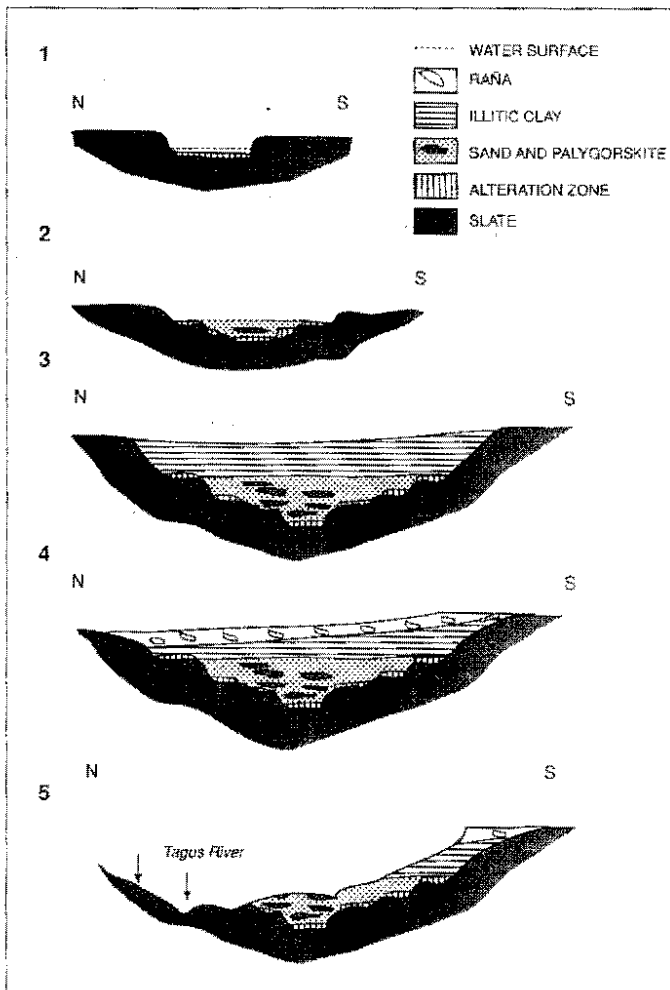


Fig. 12. Schematic interpretation of Torrejón el Rubio attapulgite deposit from the 1, weathering of substrate; 2, clayey sand sedimentation and early diagenetic (vadose) magnesium-rich attapulgite, probably related to low sedimentation; 3, silting up sedimentation; 4, alluvial fan ('raña') deposits; 5, fluvial network trenching. After Galán & Castillo (1984).

Neogene calc-alkaline volcanism of SE Spain, with an age of the volcanism varying between 8 and 17 Ma.

The main bentonite mine is in the La Serrata-Los Trancos deposit, and the production of this mine is up to 100 kt/year. The proven reserves are estimated as 3.5 Mt (J. Teodoro, pers. commun.). According to Doval (1992) the average purity of the smectites is up to 98%. The genesis of these bentonite deposits is low temperature (about 40–70 °C) hydrothermal alteration of volcanic rocks (Leone *et al.*, 1983): about 70 °C in the Sierra de Gata and about 40 °C in the Serrata de Nijar. The mineralogical paragenesis of the Serrata de Nijar deposits has been described by Caballero *et al.* (1983) as formed mainly of smectites, the only phyllosilicate present in the fine fractions belonging to the montmorillonite-beidellite-nontronite series, along with jarosite, pyrolusite and alpha-trydimite (probably neoformed) and quartz, plagioclase and potassium feldspars; inherited minerals from the parent rocks; the

parent rocks are rhyolites, dacites and trachytes. The exchange cation capacity is $\text{Na}^+ > \text{Mg}^{2+} > \text{Ca}^{2+} > \text{K}^+$, and the chemical composition of the bentonite displays wide variations even within the same deposit. The most important bentonite deposits at Serrata de Nijar are closely related to N50E striking fissures and commonly display vein-fill features.

The bentonites of the South Cabo de Gata zone consist of smectite, jarosite, zeolite and trydimite, as neoformed minerals, and of plagioclase, potassium feldspar, amphiboles and micas as minerals inherited from the parent materials (Caballero *et al.*, 1985).

In addition to this volcanogenic interpretation of these bentonite deposits, F. Ferrero (1984) has pointed out the probable influence of halmyrolytic processes in the genesis of some of the blanket bentonite deposits. The geochemical data seem to exclude a marine origin for the alteration solution, and favour the idea that the solutions resulted from a system of meteoric waters heated by a geothermal cycle (Caballero *et al.*, 1985).

The Villaluenga district (Toledo province) is located in the Middle Unit of the Neogene Madrid basin (Middle Aragonian). It occurs in a transitional zone between the outer marginal facies and inner chemical facies of the lacustrine sequences. These bentonite deposits usually occur in laterally continuous levels 0.4–2 m thick. Commonly, the bentonites are intercalated in the lacustrine sequences with distal deltaic micaceous sands and dolomitic carbonates, some chert nodules and other mudstones. The mineralogy of the bentonites is essentially Mg-rich smectites, mostly stevensite and saponite (Galán *et al.*, 1986; Pozo *et al.*, 1991), and their potential as backfilling and sealing material in high-level radioactive waste disposal is very interesting (Cuevas, 1992; Cuevas *et al.*, 1993). The reserves of the Madrid basin are probably up to 10 Mt, located in a closed stratigraphic position from Cerro del Aguila (Villaluenga) to the southern part of the city of Madrid, markedly parallel to the sepiolite belt. The reserves of the mined deposits at Cerro del Aguila (Villaluenga) are up to 0.7 Mt.

Some 'pink clays' formed by interstratified kerolite, most probably stevensite with interlayered kerolite related to the bentonite deposits, are found (Martín de Vidales *et al.*, 1989, 1991). These 'pink clays' have been used as an oil discolorant. Pink clay layers are associated with dessication levels developed over bentonitic clay beds (green colour). The possible resources of pink clays in the Madrid basin are up to 2 Mt.

Ceramic clays

Almost all the marginal and basal formations of the continental Tertiary basins of the Iberian Peninsula contain clay sediments that are being used as ceramic raw materials. In this facies there are large volumes of material that are potential sources of ceramic clays. They have a variable mineralogical composition, but illite is generally the most abundant phyllosilicate mineral. Commercial interest in these clay-rich sediments depends on the physical properties of the crude materials: plasticity, drying capacity without shrinkage, extrusionability, firing temperature, and, after firing, degree of efflorescence, permeability and compressive strength,

etc. However, the distance from the location of the clay-rich sediment deposits to the potential consumption centres may be the most important economic factor, because haulage costs may quickly make the price prohibitive.

Recently, González Díaz (1992) has reviewed Spanish ceramic clays. The most important ceramic-clay production areas are located in the Guadalquivir basin (Bailén) and in Madrid basin (La Sagra-Alcalá de Henares) (Fig. 9).

The ceramic raw materials mined in the Bailén area are of Miocene age, and have an average mineralogical composition of 20% quartz, 12% feldspars, 25% calcite and 54% phyllosilicate, mainly illite and smectite (González *et al.*, 1986).

The ceramic raw materials exploited in La Sagra (Toledo province) and Alcalá de Henares (Madrid province) are located in the Lower Unit of the Neogene Madrid basin (Lower Aragonian). The average content of illitic clays is up to 50% (Menduiña, 1988; García *et al.*, 1990). Resources amount to 500 Mt. These clay-rich sediments are used to add a top-quality silicate component to Portland cement (García Calleja *et al.*, 1991). The potteries production at La Sagra reaches 2–5 Mt per year.

Clays from the basal Unit (Tierra de Campos) of the Duero basin in the area near Valladolid are also exploited for use as ceramic raw materials.

Diatomites

Spanish diatomite production during 1990 was 107 kt, lower than the 1986 production. Spanish diatomite deposits occur in Neogene formations in the southern part of Spain in the Guadalquivir Basin (Porcuna and Martos), the Prebetic intramontane basins and the Internal Betics intramontane Late Miocene basins (Sorbas, Vera, and Nijar in Almería and Murcia provinces) (Calvo, 1984; Regueiro *et al.*, 1993).

The Prebetic area supplies more than 90% of the total Spanish diatomite production. The Hellin diatomite district extends over more than 100 km², and the diatomite occurs here in several basins that were either separated or episodically interconnected: the Cenajo Basin, Camarillas Basin, Calderones Basin, Elche de la Sierra Basin and Hijar Basin (see Table 5). These five intramontane basins outcrop along the Mundo and Segura river valleys. A schematic stratigraphic column for these basins has been proposed recently by Elizaga & Calvo (1988) (Fig. 13). Sedimentological and isotope data have been reviewed recently by Bellanca *et al.* (1989). The diatomaceous sediments are close to Facies E just above a 30–50 m thick megaslump. These megaslumps are associated with volcanism and/or a probably tectonic reactivation of the basins. The K/Ar age of the volcanism measured in the Monegrillo is up to 5 Ma (Bellón *et al.*, 1981). The megaslump-level offers the best marker in prospecting for diatomite in the district (Regueiro *et al.*, 1993).

Ten per cent of Spanish diatomite production is from the Nijar Basin (Almería); these diatomaceous sediments are located in mm-thick laminated shales formed by couplets of dark organic-rich oil shale and clear diatomite-rich laminae. The age of the deposits

Table 5. Reserves and resources of diatomite deposits in the Hellin district

	Cenajo	Camarillas	Calderones	Elche	Hijar
r I	7.5	—	—	—	—
r II	18.7	39	—	—	—
r III	—	—	—	—	5
R I	152	—	—	—	—
R II	133	12	—	337	—
R III	8.8	—	34	—	—

Notes:

Reserves (r) and identified resources (R) in Mt for the different basins. I SiO₂ < 30%; II 30% < SiO₂ < 70%; III SiO₂ > 70%.

Source: Adapted from Regueiro *et al.* (1993).

spans the interval from Late Tortonian to Early Messinian (Reyes *et al.*, 1984).

Diatomaceous sediment occurrences, sometimes mines, have been cited in the Guadalquivir Basin, for example at Sanlúcar de Barrameda, Porcuna, and Martos. Earlier in this chapter we have cited the diatomaceous sediments in the El Cuervo attapulgitic deposit. Other occurrences have been cited in the Madrid basin (Calvo *et al.*, 1988), and in the Cerdanya basin. In this last occurrence, dated as Middle–Late Miocene, Anadón *et al.* (1989) have described a diatomaceous mudstone facies with fine lamination.

Dimension (building) stone

Spain plays a very important role in the world stone industry. The natural stone production of Spain accounts for 13.2% of the world total (Lombardero & Regueiro, 1992).

Some Tertiary limestones of the Inner Prebetic Units have great purity and soundness, are easily polished and may be considered as 'commercial marbles' or marbles of type C, according to the Marble Institute of America (MIA). The quarries are grouped mainly in three areas: Coto Pinoso and Peña de Zafra in Alicante province and Sierra de la Puerta in Murcia province. The most famous quarries are located in Coto Pinoso, where the 'Crema Marfil' marble is obtained; the 1989 production of blocks was up to 10⁵ m³, and the proven reserves are 7 × 10³ m³. From a petrographic point of view the Coto Pinoso limestone may be classified as a biosparite–biomicrite (Llopis & López Jimeno, 1991).

The 'Colmenar de Oreja Limestone' has been obtained from the Upper Unit of the Madrid basin (Fig. 10), and it has been used since the eighteenth century in the construction of buildings and monuments in Madrid (Dapena *et al.*, 1988). According to the ASTM (American Society for Testing of Materials) Standard C–568–79, Colmenar limestone is among the most suitable for use for the outside of buildings (Dapena *et al.*, 1988). Colmenar limestones are paludine–lacustrine biosparites with less than 2.45% porosity. At present, Colmenar limestones and other similar limestones of the Upper Tertiary Unit of Madrid are intensively used as crushed

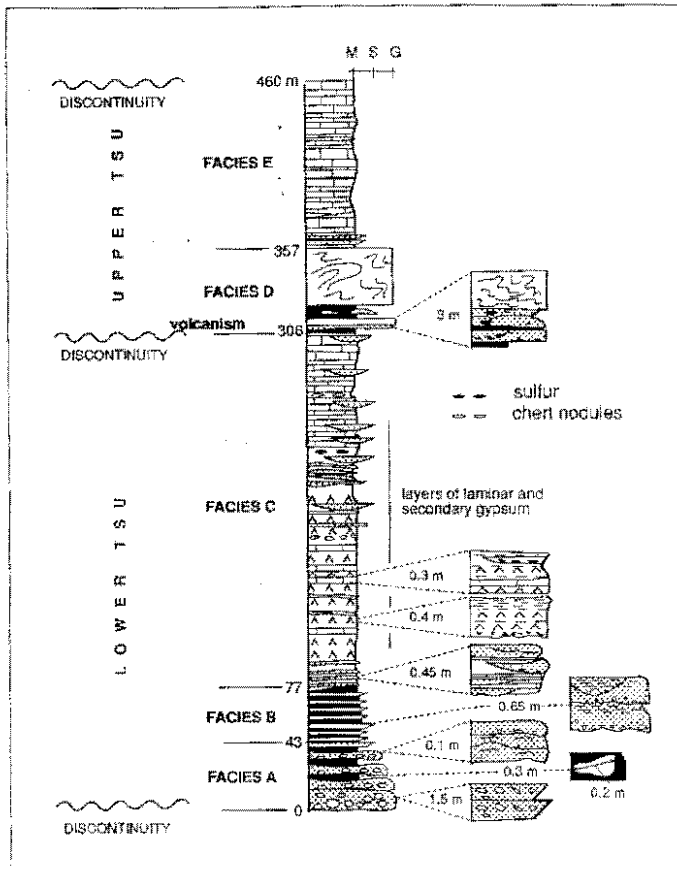


Fig. 13. Schematic stratigraphic section of Cenajo basin: A, conglomeratic basal beds; B, sandy layers in laminated oil shales; C, this subunit consists of alternations of laminated detrital carbonate and gypsum; D, megaslump subunit probably related to eruptive volcanism (5.7 ± 0.3 Ma. after Bellón *et al.*, 1981); E, diatomaceous sediments. After Elizaga & Calvo (1988). 'TSU' refers to Tecto sedimentary unit (chapter G1).

aggregate raw materials, cement raw materials, and lime raw materials (> 98% CaCO_3) and fillers (García Calleja, 1991; García Calleja *et al.*, 1991; García del Cura *et al.*, 1993).

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