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Origin of Late Cretaceous dolomites at the southern margin of the Central System, Madrid Province, Spain.

Origen de las dolomías del Cretácico Superior del borde sur del Sistema Central. Provincia de Madrid. España

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Abstract

This study focuses on the Late Cretaceous dolomitic units situated at the southern margin of the Central System in Spain. Dolomites were petrographically studied (transmited light, cathodoluminescence and scanning electron microscopy) and geochemically characterized (Mg/Ca ratio, $\delta^{13}C$, $\delta^{18}O$, $^{87}Sr/^{86}Sr$) to interpret the diagenetic environment in which they precipitated and to infer the possible mechanisms for dolomitization. Our data suggest that units of the lower and middle part of the section (Caballar, Castrojimeno and Burgo de Osma Fms.), although deposited in different sedimentary environments, were simultaneously dolomitized during the initial stages of burial, via the reflux of brines derived from the overlying Valle de Tabladillo Fm. This formation consists of interbedded evaporites and dolomicrites and/or collapsed breccias and was deposited in a coastal sabkha environment, where the unit was dolomitized during the initial stages of diagenesis.

Keywords: Dolomitization, petrography, geochemistry, stable isotope, strontium isotopes, Late Cretaceous, Central System, Spain

Resumen

Este trabajo ha consistido en la caracterización petrográfica y geoquímica de las unidades dolomíticas del Cretácico Superior que se encuentran en el borde Sur del Sistema Central, en la interpretación del ambiente diagenético de precipitación de las dolomías y en la definición del modelo de dolomitización para cada una de las unidades. El estudio petrográfico, realizado mediante microscopia convencional, catodoluminiscencia y microscopia electrónica, y el estudio geoquímico elemental (relación Mg/Ca) e isotópico ($\delta^{13}C$, $\delta^{18}O$ y $^{87}Sr/^{86}Sr$) de las dolomías sugieren que las unidades que se encuentran en la parte inferior y media de la serie (formaciones Caballar, Castrojimeno y Burgo de Osma), aunque se depositaron en ambientes sedimentarios distintos, se dolomitizaron a la vez, durante las primeras etapas del enterramiento, mediante el reflujo de salmueras procedentes de la unidad suprayacente, la Fm. Valle de Tabladillo. La Fm. Valle de Tabladillo, por su parte, está formada una alternancia de evaporitas y dolomías o bien por brechas de colapso y su dolomitización tuvo lugar durante las etapas más tempranas de la diagénesis de acuerdo con el modelo de "*sabkha*".

Palabras clave: Dolomitización, geoquímica, isótopos estables, isótopos de estroncio, Cretácico Superior, Sistema Central, España.

1. Introduction

The Late Cretaceous deposits examined in this study crop out along a NE-SW band bordering the southern margin of the Spanish Central System. In this area, sedimentation during the Late Cretaceous started with the deposition of siliciclastic facies and ended with sedimentation of carbonates, which are often dolomitized. The overall thickness of these deposits increases gradually from 15 metres in the SW to thicknesses close to 300 metres in the NE (Gil y García, 1996).

Although the carbonate units of the Late Cretaceous have been the subject of many stratigraphic and sedimentary studies (e.g. Alonso y Mas, 1982; Gil *et al.*, 1993; García *et al.*, 1996; Gil and García, 1996; Gil *et al.*, 2004), few investigations have explored the diagenetic processes that have affected these units (Mingarro and López de Azcona, 1974; Fernández Calvo, 1982; Fernández Calvo and Soriano Carrilli, 1984; Calvo *et al.*, 1993; Vegas, 1998) and there have been no such reports for the area examined here. In this study, we have characterized petrographically and geochemically the Late Cretaceous dolomitic units occurring at the border of the northern Madrid Province with the Guadalajara Province, and proposed a model for dolomitization of these units.

2. Stratigraphic context

The study area is located at the Barranco de las Cuevas (Madrid Province) and the Pontón de la Oliva (at the boundary between the Madrid and Guadalajara provinces), where Late Cretaceous deposits, which discordantly overlay the Variscan basement, reach a thickness of about 200 m (Fig. 1). The Late Cretaceous stratigraphic section starts with Cenomanian-Coniacian sandstones and lutites that correspond to the Utrillas, Castro de Fuentidueña and Segovia Formations (Alonso and Mas, 1982). Carbonate deposits of Coniacian-Campanian age, comprising four limestone and dolomite units, which are the focus of this study, overlie these formations (Alonso and Mas, 1982) (Figs. 1,2).

The first carbonate unit corresponds to the *Dolomías tableadas de Caballar Fm.*, which is characterized by well-stratified dolomites deposited in a tidal flat environment (Alonso and Mas, 1982). This unit has been assigned to the lower Coniacian (Alonso and Mas, 1982) and to the upper Turonian-lower Coniacian (García *et al*., 2004). In the lower part of this unit, mud-rich shallowing-upward sequences composed of dolomicrite predominate. Upwards, grain-rich sequences, formed of dolowackestones, dolopackstones and dolograinstones that exhibit traction structures increase in abundance. The top of the unit corresponds to a brecciation surface (Fig. 2).

The following unit, the *Calizas y dolomías de Castrojimeno Fm.*, was deposited during the upper Coniacian-Santonian (Alonso and Mas, 1982; García *et al*., 2004) and consists on two members (Fig. 2): The *lower member* comprises limestones, dolomitic limestones and marls that occur in shallowing-upward sequences deposited in an inner platform environment (Alonso and Mas, 1982).

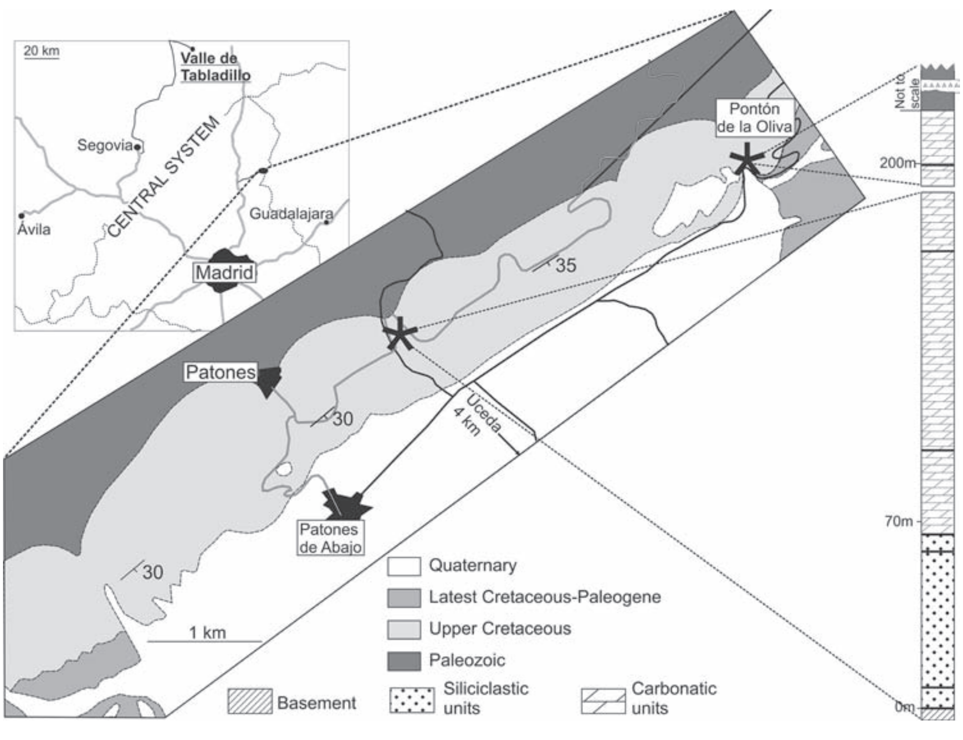

Fig. 1.- Location map, geological scheme and simplified stratigraphic section of the study area.

Fig. 1.- Situación geográfica, esquema geológico y columna estratigráfica simplificada de la zona de estudio.

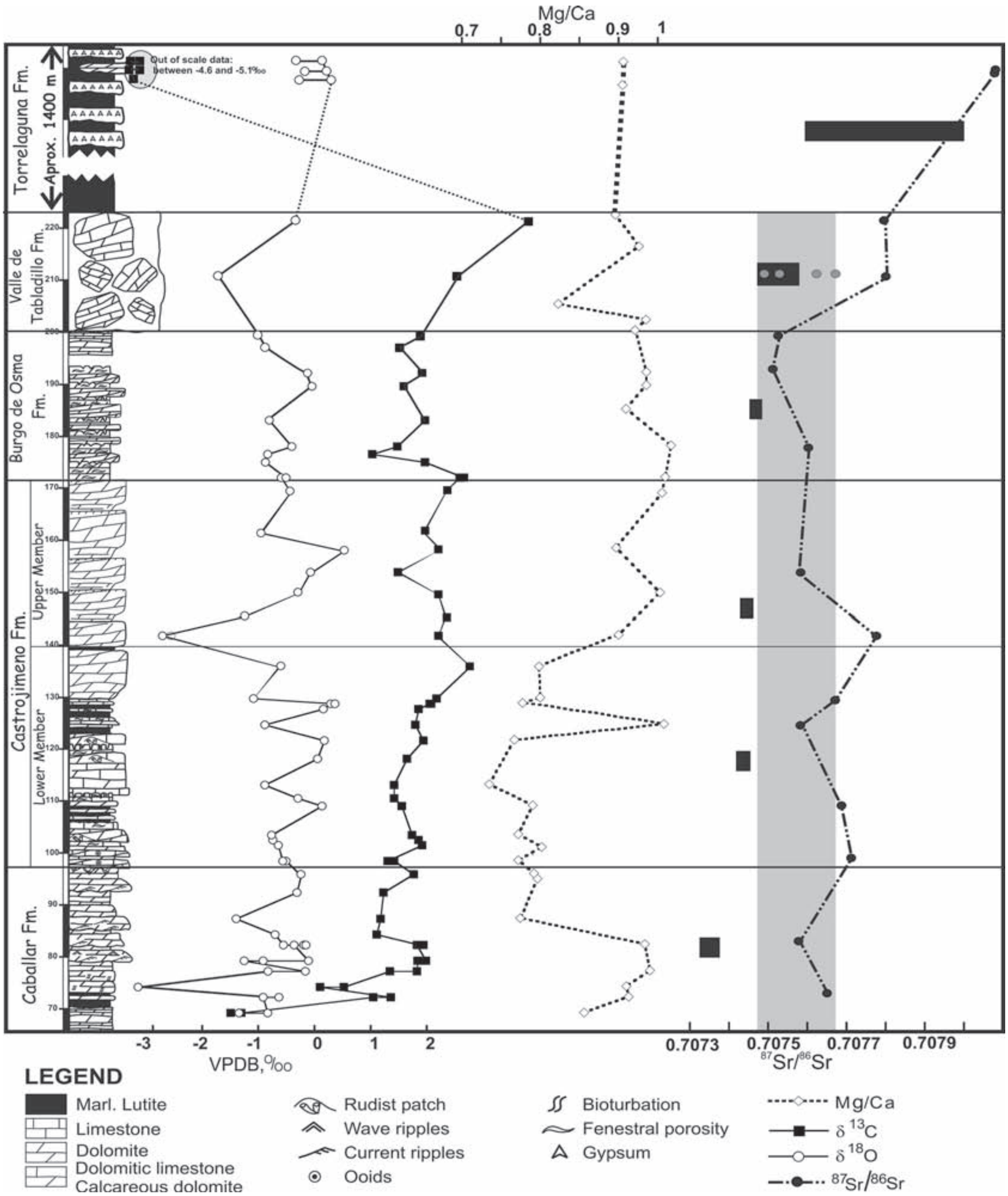

Fig. 2.- Stratigraphic section of the Late Cretaceous dolomitic units of the southern margin, Central System, Spain. Oxygen and carbon isotope compositions, and Mg/Ca and $^{87}Sr/^{86}Sr$ ratios for the dolomites are indicated to the right. Black rectangles to the right represent the range of $^{87}Sr/^{86}Sr$ ratios inferred for Late Cretaceous marine carbonates (based on McArthur *et al*., 1994). Grey circles and vertical bar indicate the individual analyses and range of $^{87}Sr/^{86}Sr$ ratios obtained for dolomites of the Valle del Tabladillo Fm. at the northern margin of the Spanish Central System (see Fig. 1 for location).

Fig. 2.- Columna estratigráfica de las unidades dolomíticas del Cretácico Superior del borde sur del Sistema Central. A la derecha se han representado los valores isotópicos de carbono y oxígeno, la relación Mg/Ca y la relación $^{87}Sr/^{86}Sr$ de las dolomías de las distintas unidades. Los rectángulos de color negro situados a la derecha del gráfico representan el rango de los valores de la relación $^{87}Sr/^{86}Sr$ que han sido inferidos para los carbonatos marinos del Cretácico Superior (basados en McArthur *et al*., 1994). Los círculos de color gris y la barra vertical de color gris representan, respectivamente, los valores de la relación $^{87}Sr/^{86}Sr$ obtenidos y su rango de variación en las dolomías de la Fm. Valle de Tabladillo en el borde Norte del Sistema Central (ver Fig. 1 para localización).

The lower part of these sequences is composed of limestones and dolomitic limestones of wackestone and packstone texture, within which accumulations of rudists in life position occur. The top of these sequences comprises marls, limestones and dolomitic limestones composed of mudstones and wackestones of benthic foraminifera and ostracods. These mudstones and wackestones usually show intense bioturbation, fenestral porosity and incipient nodulization. The *upper member* of the Castrojimeno Fm. is constituted of massive dolomites that locally exhibit large-scale cross-stratification and have been interpreted as subtidal bars (Alonso and Mas, 1982).

The next unit is the *Calizas de Burgo de Osma Fm.* of the upper Santonian-Campanian age (Alonso and Mas, 1982; García *et al*., 2004). It is formed of bioclastic dolomites and dolomudstones with fenestral porosity and locally with evaporite pseudomorphs that were deposited on a tidal flat (Alonso and Mas, 1982).

The dolomites of the *Burgo de Osma Fm.* at Pontón de la Oliva (Fig. 1), are overlain by highly porous breccias that could represent deposits of the *Valle del Tabladillo Fm.* In the northern sector of the Central System, where it was defined (Fig. 1), this formation comprises interbedded dolomites and evaporites, and breccias arising from the dissolution of these evaporites. This unit has been assigned to the lower Campanian (Alonso and Mas, 1982; García *et al*., 2004).

Over this carbonate succession, there is a unit essentially composed of clays, gypsum, marls and dolomites. This unit, which has been designated as the *Torrelaguna* Fm., has been assigned to the latest Cretaceous (Nodal and Águeda, 1976; Portero *et al*., 1990; Gil *et al*., 2004), and more specifically to the upper Campanian-Maastrichtian (García *et al*., 2004). Dolomite lithologies are scarce in this formation and, where present, show little lateral continuity. Dolomite of this unit could be analysed in the surroundings of Uceda, situated some 4 Km SE of the village of Patones (Fig. 1).

3. Methods

A total of 125 samples were used for the petrographic and geochemical study of sedimentary and diagenetic features of dolomites, utilizing standard petrographic techniques, cathodoluminescence, scanning electron microscopy (SEM), and elemental and isotopic analyses. For each sample, a polished 30 μm thin section and matching 150-200 μm thick section for microsampling was prepared. Cathodoluminescence (CL) examination was carried out using a Technosyn® cold cathodoluminescent unit operating at 14-17 kV with 350-450 μA beam current. Following examination with CL, all thin sections were stained with Alizarin Red S and potassium ferricyanide (Dickson, 1966).

Analyses for $\delta^{13}C$ and $\delta^{18}O$ values were performed in the Stable Isotope Laboratory at the University of Michigan. The strontium isotopes were performed in the Geochronology laboratory of the University Complutense of Madrid. In order to isolate dolomite from host calcite for isotopic analysis, sample powders were taken from thick sections using a microscope-mounted drilling system. A 1M solution of acetic acid and sodium acetate with a pH of 5 was prepared. Each powdered dolomite/calcite sample was placed into the solution for 48-72 hours, and the undissolved powder was then analyzed by XRD to be sure that all calcite was dissolved before isotopic analyses of residual dolomite.

For stable isotope analysis, all sample powders were roasted *in vacuo* for one hour at 200°C, to remove volatile organic contaminants, then reacted at 73°C in an automated carbonate reaction system (CarboKiel-I) coupled directly to the inlet of a Finnigan MAT 251 gas ratio mass spectrometer. Isotopic ratios were corrected for ^{17}O contribution and are reported in per mil notation relative to the VPDB standard. Values were calibrated utilizing NBS 19 as the primary standard, and analytical precision was monitored by daily analysis of NBS powdered carbonate standards. Measured precision was maintained at better than 0.1‰ for $\delta^{13}C$ and $\delta^{18}O$.

Dolomite powder for $^{87}Sr/^{86}Sr$ analyses was dissolved in 2 ml of a solution of 2,5N HCl, later evaporated to dryness at 80-100ºC. Samples were re-dissolved in 2,5 N HCl solution and Sr was pre-concentrated by standard methods of column chromatography. Following this, the Sr-concentrated samples were dissolved in 2 ml of phosphoric acid and Sr isotopic ratios were then determined with a VG SECTOR 54 five-collector mass spectrometer. Isotopic ratios were corrected for possible interferences from ^{87}Rb and normalized to the value $^{86}Sr/^{88}Sr = 0.1194$, to correct the isotopic fractionation effect. Analytical precision was monitored by analysis of the NBS-987 standard and measurement precision was maintained at better than $\pm$ 4-6x10^{-6}.

Elemental analyses for Ca, Mg, Sr, Mn, and Fe were performed on the electron microprobe of the University Complutense of Madrid, with an accelerating voltage of 15 kV and a spot size of 5 μm. Measured precision was maintained at better than ± 0.14% for Mg, ± 0.09% for Ca, ± 0.08% for Sr, ± 0.08% for Fe, and ± 0.06% for Mn, and the detection limits were 100 ppm for Mg, 250 ppm for Sr, 200 ppm for Mn, and 250 ppm for Fe.

4. Petrography and geochemistry of dolomites

4.1. Caballar Fm.

This unit is pervasively dolomitized, although most of the samples preserve their depositional texture. Mud-rich sequences of the lower part of the unit are formed by slightly ferroan dolomicrites and dolomicrosparites (Figs. 3A, 4A), which show a dull brown luminescence. In the grain-rich sequences of the upper part of the unit, both the micritic matrix and micritic components are composed of slightly ferroan dolomicrosparite displaying reddish luminescence (Fig. 3B, C). Intergranular porosity of grainstones is occupied by subidiotopic mosaics of dolomite that show red and brown zoning under CL and occlude pores smaller than 50 μm. Moldic porosity created after dissolution of most of the non-micritic carbonatic components is partially cemented by nonferroan rhombohedral dolomite, displaying zoned red-brown luminescence (Figs. 3B, C, 4B). Pores that are not totally filled by dolomite, are occluded by a mosaic of nonferroan to ferroan calcite (Figs. 3B, C, 4B).

The mean Mg/Ca ratio of dolomicrites and dolomicrosparites of the lower part of the Caballar Fm. is 0.91, close to the dolomite stoichiometric value (Fig. 2). However, dolomites forming the dolosparitic mosaics of the upper part of the unit are calcic, since their mean Mg/Ca ratio is 0.78 (Fig. 2). The overall oxygen isotope composition of dolomites of this unit is –0.1 to –3.1‰ and their carbon isotope contents range from –1.4 to +2‰; with the most negative values corresponding to dolomites at the base of the unit (Fig. 2). $^{87}Sr/^{86}Sr$ ratios are 0.707651 for dolomites of the lower part of the unit and 0.707573 for those of the upper part (Fig. 2).

4.2. Castrojimeno Fm.

4.2.1. Lower member

This member is partially dolomitized (Fig. 3D-G). The lower and upper parts of the member, comprising wackestones and packstones, are pervasively dolomitized. However, the patches of rudists appearing in the middle part, as well as the packstones and grainstones intercalated among these rudists, exhibit partial dolomitization; micritic matrix and micritic components are dolomitized but rudist skeletons, whose calcitic microstructure is usually well preserved, are not dolomitized (Fig. 3D). Similarly, mudstone and wackestone horizons that are intercalated with marls in the middle part of this member (Fig. 2) are scarcely dolomitized. In these horizons, dolomite appears as millimetre to submillimetre aggregates of nonferroan rhombohedral crystals, up to 50 μm in diameter (Fig. 3G), which have been partially to fully calcified.

In general, all dolomite present in this member is similar petrographically and geochemically to dolomite observed at the top of the Caballar Fm. (Figs. 2, 3B-F, 4B-C). Except for dolomite replacing matrix and micritic components, most dolomite forms subidiotopic to idiotopic mosaics of crystals of diameters up to 150-200 μm that show zoned red-brown luminescence and that cement primary and moldic porosity. Porefilling dolomite cement often has a rhombohedral habit and coarser crystal size than dolomite replacing the micritic matrix (Fig. 3E, F). As in the Caballar Fm., any porosity not entirely cemented by dolomite is occluded by zoned nonferroan-ferroan calcite cement, which exhibits zoned brown luminescence (Fig. 3E, F).

Occasionally, horizons interbedded with marls in the middle-upper part of the member are intensely dolomitized (Fig. 3H), with dolomite crystals exhibiting cloudy cores of reddish mottled luminescence and a transparent outer zone displaying brown luminescence. With SEM, the cores of the dolomite crystals appear rhombohedral and microporous, and are partially or fully calcified; the outer zones of the crystals, however, are scarcely porous and show euhedral terminations (Fig. 4D). At the boundary between the microporous core and the outer rim of the crystals, there is usually a band of up to 10-15 μm thick in which the dolomite is fully dissolved or calcified (Fig. 4D).

Chemically, most of the dolomites of the lower member of the Castrojimeno Fm. are calcic with a mean Mg/Ca ratio of 0.78 (Fig. 2). In the pervasively dolomitized horizons, dolomite yields a mean Mg/Ca ratio of 0.96 (Fig. 2). The oxygen isotope composition of the dolomites of this member ranges from –0.40 to –1.1‰ and the carbon isotope compositions range between +2.8 and +1.3‰ (Fig. 2). $^{87}Sr/^{86}Sr$ ratios yield values between 0.707575 and 0.707704, with the lowest value corresponding to dolomitic horizons interbedded with marls that showed the highest Mg/Ca ratio (Fig. 2).

4.2.2. Upper member

This unit is pervasively dolomitized, although in some samples, the depositional texture can be inferred through the abundance of moldic porosity. This dolomite is petrographically and chemically very similar to dolomite horizons observed within marls of the lower member (Figs. 2, 3H, 5A, 4D, E). It is formed by a subidiotopic mosaic of nonferroan dolomite crystals of up to 150 microns in diameter showing reddish luminescence. Dolomite crystals comprise a cloudy core displaying a rhombohedral habit, which is usually intensely calcified, and an outer clear rim displaying reddish luminescence that is better preserved (Fig. 5A, B). In this member, moldic porosity is partially cemented by euhedral crystals of dolomite that show zoned non-yellow-red-brown luminescence (Fig. 5A, B). SEM reveals that this dolomite is constituted of a microporous rhombohedral core that is partially or fully calcified, and an outer non-porous zone that commonly fills all inter-crystalline pore space among the crystal cores or partially fills these spaces. When partially filling porosity dolomite crystals develop euhedral terminations (Fig. 4E). At the boundary between the core and the outer rim there is usually a transitional band in which the dolomite is completely dissolved or calcified (Fig. 4E).

The mean Mg/Ca ratio of dolomites of the upper member is 0.94 (Fig. 2). Oxygen isotope compositions range between –0.1 and –2.8‰ and carbon isotope compositions range between +2.7 and +0.6‰ (Fig. 2). $^{87}Sr/^{86}Sr$ ratios range between 0.70757 and 0.707772, with the highest value corresponding to dolomite with the most negative oxygen isotope value (Fig. 2).

4.3. Burgo de Osma Fm.

This unit is pervasively dolomitized, although the depositional texture may be generally inferred by the presence of abundant fenestral and moldic pores. Petrographically, dolomite of this formation is identical to that observed in the upper member of the Castrojimeno Fm. Dolomite occurs as subidiotopic mosaics of crystals, which have microporous rhombohedral cores. These cores, which are usually partially or fully calcified, are overlain by a clear rim that fills inter-crystalline porosity (Figs. 4F, 4G, 5C), which is usually better preserved, shows red-brown or intense red luminescence and often occupies all the space existing among the microporous cores (Fig. 5C, D).

This dolomite is also chemically indistinguishable from the underlying unit (Fig. 2). Their mean Mg/Ca ratio is 0.94, oxygen isotope compositions range between +0.05 and –0.9‰ and carbon isotope compositions between +2.2 and +1.5‰ (Fig. 2). $^{87}Sr/^{86}Sr$ ratios range from 0.707517 to 0.707603 (Fig. 2).

Fig. 3.- (opposite page) A. General appearance of dolomicrites from the lower part of the Caballar Fm. (Q) Quartz. B. Stained sample showing dolomites from the upper part of the Caballar Fm. Dolomite (D) replaces the micritic matrix and partially fills moldic porosity, which, in turn, is occluded by zoned nonferroan-ferroan calcite cement (CC). C. Transmitted light (left) and cathodoluminescence (CL) (right) micrographs showing dolomites from the upper part of the Caballar Fm. Dolomite crystals (D) that replaces the micritic matrix shows mottled, orange luminescence. The dolomite cementing porosity is rhombohedral and exhibits zoned red-brown luminescence (blue arrow). Calcite, which occludes porosity, shows zoned dull brown luminescence (CC). D. Stained sample showing a rudist fragment (R) from the lower member of the Castrojimeno Fm. Note that dolomitization (D) affects micritic matrix and micrite filling the growth-framework and boring pores, it yet does not affect the rudist skeleton. E. Stained sample showing dolomites of the lower member of the Castrojimeno Fm. Dolomite (D) replaces the micritic matrix and partially fills the moldic porosity, which is occluded by zoned nonferroan-ferroan calcite cement (CC). Note the similarity between these dolomites and those of the upper part of the Caballar Fm. (Fig. 3B). F. Transmitted light (left) and CL (right) images of dolomites from the lower member of the Castrojimeno Fm. The dolomite (D) replacing the micritic matrix shows mottled, orange luminescence. The dolomite cementing porosity is rhombohedral and shows zoned red-brown luminescence (blue arrow). Calcite (CC) occluding porosity exhibits zoned brownish luminescence. Note the similarity between these dolomites and those of the upper part of the Caballar Fm. (Fig. 3C). G. Stained sample showing the partially dolomitized limestone, which is interbedded with marls in the middle part of the lower member of the Castrojimeno Fm. (Fig. 2). Dolomite (D) replaces the micritic matrix and partially fills intraskeletal porosity. H. Detailed view of the dolomite layers, which are occasionally interbedded with marls in the middle part of the lower member of the Castrojimeno Fm. (Fig. 2). Dolomite crystals (D) have a cloudy rhombohedral core, which is commonly calcified, and a clear and well-preserved outer rim (blue arrows). Porosity is occluded by nonferroan calcite (CC).

Fig. 3.- (página opuesta) A. Aspecto general de las dolomicritas de la parte inferior de la Fm. Caballar. Q: Cuarzo. B. Aspecto de las dolomías de la parte media y superior de la Fm. Caballar (la muestra está teñida). La dolomita (D) remplaza a la matriz formando mosaicos dolomicroesparíticos y cementa parcialmente la porosidad móldica que está ocluida por un cemento de calcita (CC) zonada no ferrosa-ferrosa posterior (de color rosa y azulado, respectivamente). C. Fotografías realizadas con microscopia de luz transmitida (izquierda) y catodoluminiscencia (derecha) de una muestra de la parte superior de la Fm. Caballar. La dolomita (D) que remplaza a la matriz micrítica presenta una luminiscencia moteada anaranjada. La dolomita que cementa la porosidad, de mayor tamaño de cristal, tiene un hábito romboédrico y una luminiscencia zonada de color rojo y marrón (flecha azul). La calcita (CC) que ocluye la porosidad presenta una luminiscencia zonada mate de color marrón. D. Detalle de un fragmento de rudista (R) del miembro inferior de la Fm. Castrojimeno. Obsérvese que la dolomitización (D) afecta a la matriz, al relleno de la porosidad intraesquelética y al relleno de las perforaciones que afectan al rudista (áreas no teñidas, flechas azules) pero no afecta al esqueleto calcítico del rudista (teñido de rosa) cuya microestructura se encuentra bien conservada. E. Aspecto de las dolomías del miembro inferior de la Fm. Castrojimeno. La dolomita (D), no teñida, remplaza a la matriz y cementa parcialmente la porosidad móldica que está ocluida por un cemento de calcita (CC) zonada no ferrosa-ferrosa. Obsérvese la similitud entre estas dolomías y las de la parte superior de la Fm. Caballar (Fig. 3B). F. Fotografías realizadas con microscopia de luz transmitida (izquierda) y catodoluminiscencia (derecha) de una muestra del miembro inferior de la Fm. Castrojimeno. La dolomita (D) que remplaza a la matriz micrítica presenta una luminiscencia moteada anaranjada. La dolomita que cementa la porosidad, de mayor tamaño de cristal, tiene un hábito romboédrico y una luminiscencia zonada de color rojo y marrón (flecha azul). La calcita (CC) que ocluye la porosidad presenta una luminiscencia zonada mate de color marrón. Obsérvese la similitud entre estas dolomías y las de la parte superior de la Fm. Caballar (Fig. 3C). G. Aspecto de las calizas, muy poco dolomitizadas, que se encuentran intercaladas entre los niveles margosos de la parte media del miembro inferior de la Fm. Castrojimeno (Fig. 2). La dolomita (D) remplaza a la matriz micrítica y cementa parcialmente la porosidad intraesquelética. H. Detalle de los niveles dolomíticos que ocasionalmente se encuentran intercalados entre las margas de la parte media del miembro inferior de la Fm. Castrojimeno (Fig. 2) y que suelen estar parcial o intensamente calcitizados. En este caso los cristales de dolomita (D) presentan un núcleo de aspecto sucio y hábito romboédrico, que suele estar parcial o totalmente calcitizado y una parte externa limpia, bien preservada (flechas azules). La porosidad está ocluida por un cemento de calcita no ferrosa (CC).

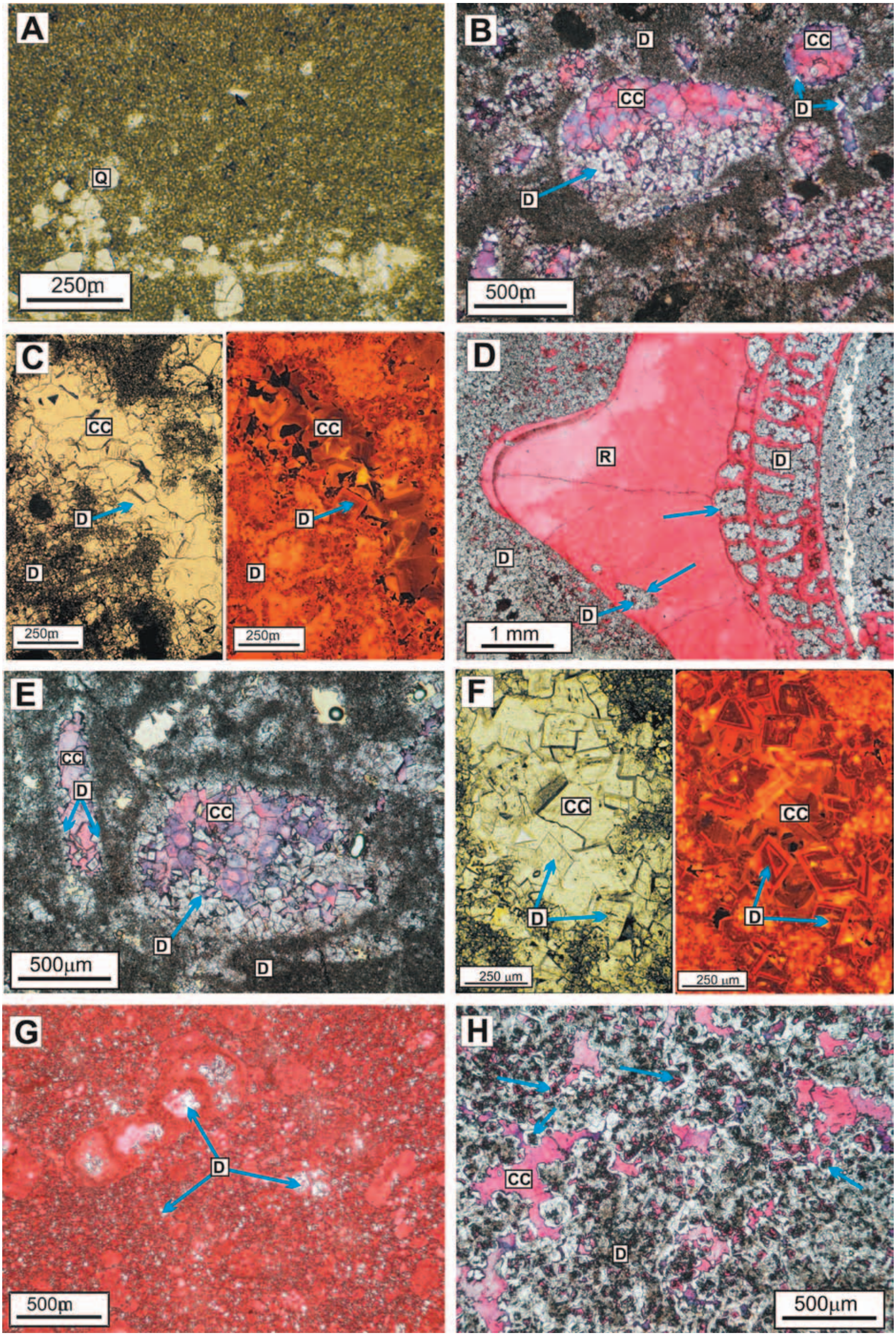
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Fig. 4.- (opposite page) Backscattered electron microscopy images. A. Detail of dolomicrites from the lower part of the Caballar Fm. Note the rhombohedral shape of some of the crystals (blue arrows). B. Dolomites from the upper part of the Caballar Fm. The intraclast located to the right of the image is comprised of dolomicrite (DM). Dolomite crystals replacing the micritic matrix (D) are anhedral or subeuhedral and larger than those that replace the intraclast. The dolomite crystals filling porosity (blue arrow) are the largest, euhedral in shape, and display a zonation similar to that observed under CL (Fig. 3C). Porosity is occluded by calcite (CC). C. Detailed view of dolomites from the lower member of the Castrojimeno Fm. The dolomite crystals replacing the micritic matrix (D) are anhedral to subeuhedral; crystals cementing porosity (blue arrow) are larger, euhedral and display a zonation similar to that observed under CL (Fig. 3F). Porosity is occluded by calcite (CC). Note the similarity between these dolomites and those from the upper part of the Caballar Fm. (Fig. 4B). D. Detailed image of dolomites, which are occasionally interbedded with marls in the middle part of the lower member of the Castrojimeno Fm. (Fig. 2). Dolomite crystals (D) have microporous and rhombohedral cores, which are commonly calcified (green arrows), and clear and well-preserved outer zones (blue arrows). Note the band of calcite (red arrows) between the core and the rim of the dolomite crystals. E. Detailed image of dolomites from the upper member of the Castrojimeno Fm. Note their similarity to those shown in the previous image. The dolomite crystals are constituted of microporous and rhombohedral cores, which can be partially or fully calcified (green arrows), and limpid outer rims, which commonly occlude intercrystal porosity. Note that between the core and the rim, there is also a band of calcite (red arrows). F. Dolomite of the Burgo de Osma Fm. This dolomite is also comprised of microporous and rhombohedral cores, which can be partially or fully dissolved (white arrows), and limpid outer rims, which occlude porosity. Between the core and the rim of the dolomite crystals there is a band of dissolved (green arrow) or calcified (red arrow) dolomite. G. Detailed view of dolomite (D) of the Burgo de Osma Fm. cementing moldic porosity. Note that between the microporous and rhombohedral cores and the limpid rims of the crystals there is a band of calcite (red arrow). Porosity is also occluded by calcite (CC). H. Detail of the dolomitic breccias of the Valle de Tabladillo Fm. at the southern margin of the Central System. Dolomite (D) consists of small anhedral inclusions, which are embedded in the calcite crystals (CC). I. Detailed image of the dolomite (D) of the Torrelaguna Fm. The dolomite contains gypsum pseudomorphs (white arrows), which are partially filled by dolomite crystals (blue arrow).

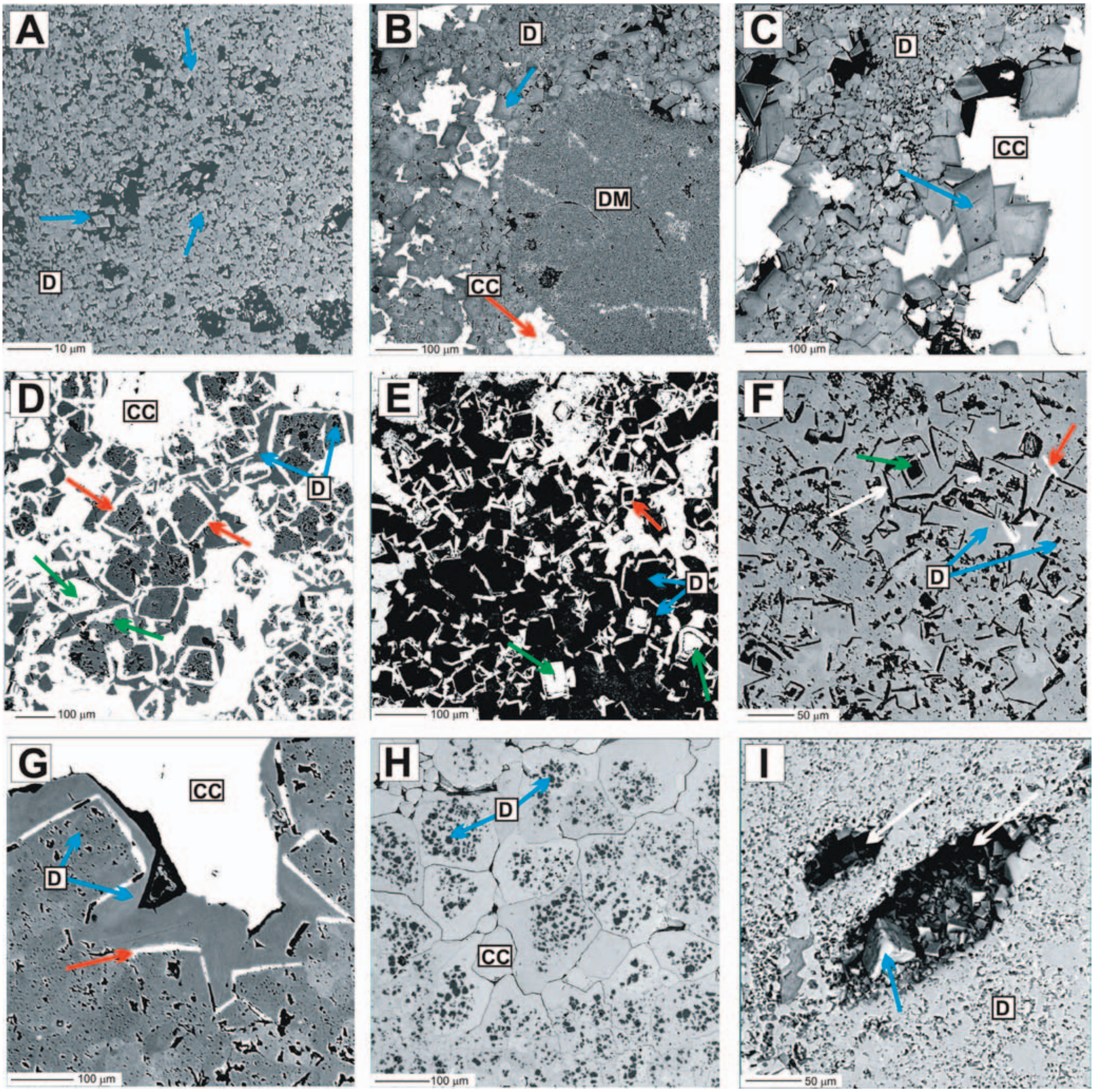
Fig. 4.- (página opuesta) Fotografías tomadas mediante el microscopio electrónico de barrido en el modo de electrones retrodispersados. A. Aspecto de las dolomías de la base de la Fm. Caballar. Obsérvese el hábito romboédrico de parte de los cristales (flechas azules). B. Dolomías de la parte superior de la Fm. Caballar. El intraclasto situado a la derecha de la imagen está formado por un mosaico dolomicrítico (DM). Los cristales de dolomita que remplazan a la matriz micrítica (D) son anhédricos y subeuhédricos y de mayor tamaño que los del intraclasto. Los cristales de dolomita que cementan la porosidad (flecha azul) son de mayor tamaño, euhédricos y presentan un zonado de crecimiento similar al observado con catodoluminiscencia (Fig. 3C). La porosidad está ocluida por calcita (CC). C. Aspecto de las dolomías del miembro inferior de la Fm. Castrojimeno. Los cristales de dolomita que remplazan a la matriz micrítica (D) son anhédricos o subeuhédricos y los que cementan la porosidad (flecha azul) son euhédricos, de mayor tamaño y presentan un zonado similar al observado con catodoluminiscencia (Fig. 3F). La porosidad está ocluida por calcita (CC). Obsérvese su similitud con las dolomías de la parte superior de la Fm. Caballar (Fig. 4B). D. Aspecto de las dolomías que se encuentran en algunos de los niveles intercalados entre las margas de la parte media del miembro inferior de la Fm. Castrojimeno (Fig. 2) y que suelen estar intensamente calcitizadas (CC). En este caso los cristales de dolomita (D) presentan un núcleo microporoso de hábito romboédrico, que suele estar parcial o totalmente calcitizado (flechas verdes), y una parte externa limpia, bien preservada y con terminaciones generalmente euhédricas. Obsérvese que entre el núcleo y la parte externa de los cristales hay una franja que se encuentra totalmente calcitizada (flechas rojas). E. Aspecto de las dolomías del miembro superior de la Fm. Castrojimeno. Como las dolomías de la imagen anterior, los cristales de dolomita están formados por un núcleo de hábito romboédrico, comúnmente, que puede estar parcial o totalmente calcitizado (flechas verdes) y una parte externa que suele ocluir la porosidad intercristalina. También en este caso entre el núcleo y la parte externa de los cristales hay una franja que se encuentra totalmente calcitizada (flechas rojas). F. Aspecto general de las dolomías (D) de la Fm. Burgo de Osma que también están formadas por un núcleo microporoso de hábito romboédrico, que puede estar parcial o totalmente disuelto (flechas blancas) y una parte externa de aspecto limpio que ocluye la porosidad. Entre el núcleo y la parte externa de los cristales hay una franja en la que la dolomita está disuelta (flecha verde) o calcitizada (flecha roja). G. Detalle de las dolomías de la Fm. Burgo de Osma que en este caso están cementando porosidad móldica. Obsérvese el núcleo microporoso y romboédrico de los cristales de dolomita, la parte externa limpia y la franja calcitizada entre ambas (flecha roja). La porosidad está ocluida por calcita (CC). H. Aspecto de las brechas dolomíticas de la Fm. Valle de Tabladillo en el borde Sur del Sistema Central. La dolomita (D) se encuentra en forma de pequeñas inclusiones anhédricas que se encuentran en el interior de los cristales de calcita (CC). I. Aspecto de las dolomías (D) de la Fm. Torrelaguna que contienen pseudomorfos de evaporitas, yeso probablemente (flechas blancas). Los pseudomorfos están parcialmente cementada por cristales de dolomita (flecha azul).

4.4. Valle de Tabladillo Fm.

This unit, examined at the Pontón de la Oliva sector (Fig. 1), is composed of breccias formed by metre-sized blocks, which are often embedded in a calcareous to marly reddish matrix, and comprise intensely or completely calcified dolomite. When dolomite relics persist, they occur as small anhedral inclusions up to 20-30 μm (Fig. 4H) within nonferroan calcite crystals that show zoned non-yellow-brown luminescence (Figs. 4H; 5E).

The mean Mg/Ca ratio recorded for these dolomites is 0.9 (Fig. 2). Oxygen and carbon isotope compositions are +0.3 to –1.6‰ and +2.5 to +3.5‰, respectively (Fig. 2). $^{87}Sr/^{86}Sr$ ratios range between 0.707789 and 0.707796 (Fig. 2).

We have also examined dolomites of the Valle de Tabladillo Fm. in its type locality in the northern margin of the Central System (Fig. 1). In this zone, the unit, deposited in a sabkha coastal setting (Alonso and Mas, 1982), is formed by alternating centimetre to metre horizons of dolomicrite and alabastrine gypsum, which preserve abundant chicken-wire structures. In this area, dolomite displays oxygen isotope compositions ranging between +1.1 and +1.6‰, and carbon isotope compositions between



+2.5 and +3.1‰. That is, the isotope composition of dolomites of the Valle de Tabladillo Fm. at the northern margin of the Central System is similar in carbon but more positive in oxygen than dolomitic breccias examined at the southern margin. The $^{87}Sr/^{86}Sr$ ratios of dolomites of the northern border range from 0.707486 to 0.707675. In this case, the lowest $^{87}Sr/^{86}Sr$ ratios were recorded in dolomite with negligible clay and quartz contents, whereas the highest ratios were obtained in two samples that contained abundant clay and quartz. The $^{87}Sr/^{86}Sr$ ratios are always lower than those recorded in the dolomitic breccias of the southern margin (Fig. 2).

4.5. Torrelaguna Fm.

The dolomite of this unit, interpreted here to have formed as dolocretes, comprises reddish luminescent dolomicrites, which are commonly nodulized, brecciated, and contain abundant lenticular pseudomorphs of evaporites, probably gypsum. These pseudomorphs are now cemented by nonferroan and non- to bright luminescent calcite (Figs. 4I; 5F).

The mean Mg/Ca ratio of dolomites of the Torrelaguna Fm. is 0.86 (Fig. 2). Oxygen and carbon isotope compositions range from –0.4 to +0.3‰ and from –4.6 to –5.1‰, re-

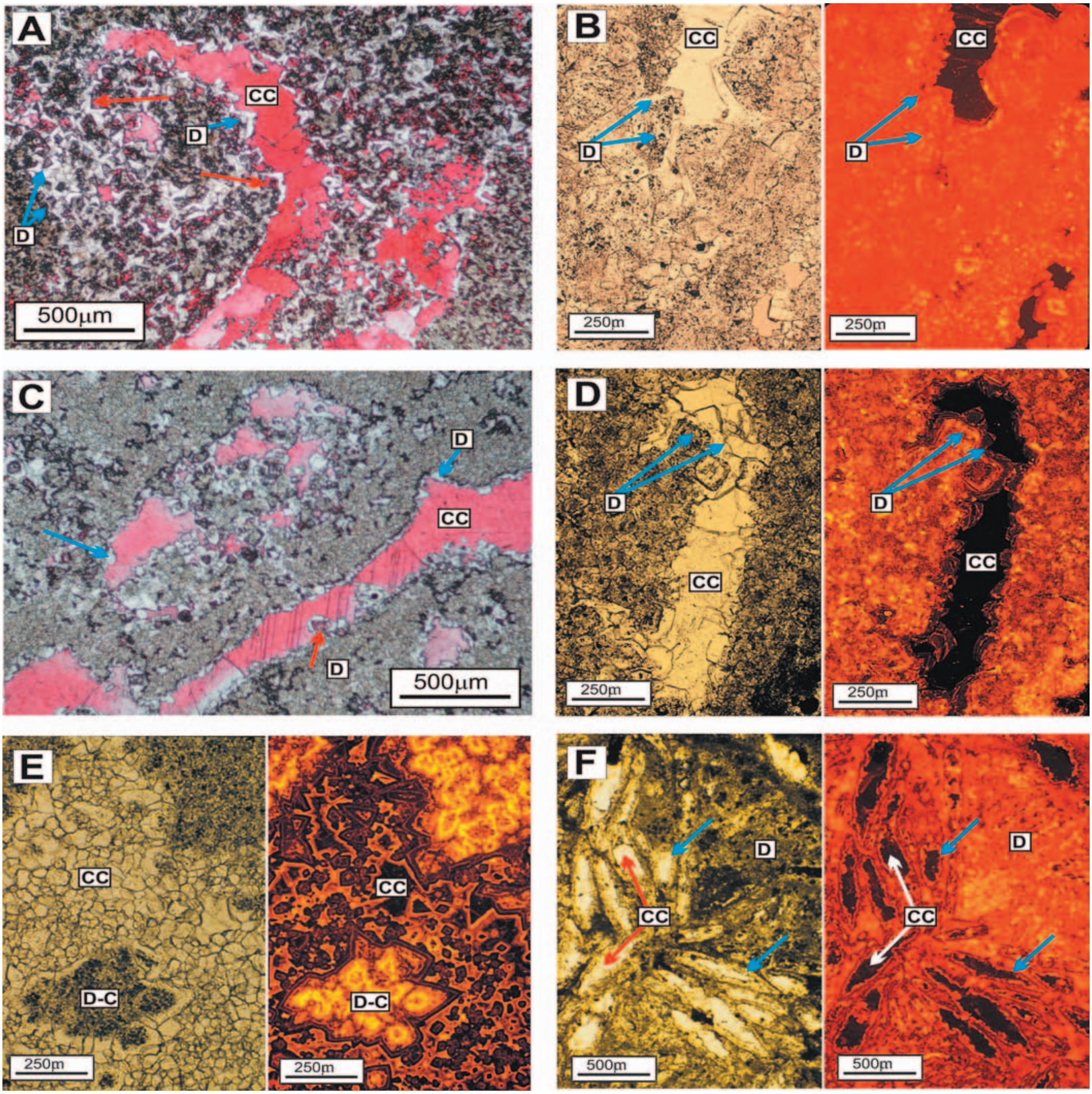


Fig. 5.- A. Stained sample showing dolomite from the upper member of the Castrojimeno Fm. Dolomite crystals (D) replace the micritic matrix and partially fill the moldic porosity. Dolomite crystals have cloudy cores, which can be partially or fully calcified (red arrows), and limpid outer rims. Porosity is occluded by nonferroan calcite (CC). B. Transmitted light (left) and CL (right) micrographs of dolomites from the upper member of the Castrojimeno Fm. Dolomite (D) replacing the micritic matrix is cloudy and shows reddish-orange luminescence. Dolomite crystals cementing porosity (blue arrows) have cloudy and rhombohedral cores, showing reddish-orange luminescence, and limpid outer rims, displaying red luminescence. Porosity is occluded by nonluminescent calcite (CC). C. Stained sample of dolomite from the Burgo de Osma Fm. Dolomite (D) replaces the micritic matrix and partially cements moldic porosity, which is occluded by nonferroan calcite (CC). The dolomite replacing the matrix is cloudy. Dolomite crystals cementing pores (blue arrows) are constituted of cloudy and rhombohedral cores, which may be partially or fully calcified (red arrow), and limpid outer rims. Note the similarity between this dolomite and that of the upper member of the Castrojimeno Fm. (Fig. 3H). D. Transmitted light (left) and CL (right) micrographs of dolomites from the Burgo de Osma Fm. The dolomite (D) replacing the micritic matrix is cloudy and shows a mottled, orange CL. Dolomite crystals cementing porosity (blue arrows) have cloudy and rhombohedral cores, which show zoned orange-red-brownish luminescence, and limpid outer zones, which exhibit dark brown luminescence. Porosity is occluded by nonluminescent calcite (CC). E. Transmitted light (left) and CL (right) micrographs of breccias from the Valle de Tabladillo Fm. at the southern boundary of the Central System. The cloudy areas, which show mottled and zoned yellowish-orange luminescence, are composed of intensively calcified dolomite (D-C). The more limpid areas, which produce zoned nonluminescent-brownish-orange

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spectively (Fig. 2). $^{87}Sr/^{86}Sr$ ratios range between 0.7078072 and 0.7078091. These values are considerably higher than those of dolomite from the underlying units (Fig. 2).

5. Discussion: Origin of the Late Cretaceous dolomites at the southern margin of the Central System.

5.1. Torrelaguna Fm.

It is likely that dolomite of the Torrelaguna Fm. precipitated during early stages of diagenesis in an evaporitic setting, as suggested by their relatively high oxygen isotope compositions and the presence of gypsum pseudomorphs. The carbon isotope composition of this dolomite, which is substantially more negative than that of the underlying units, is typical of meteoric and pedogenic carbonates that incorporate soil-derived, isotopically light carbon (Lohmann 1987). The $^{87}Sr/^{86}Sr$ ratio of this dolomite is also consistent with precipitation from meteoric waters (e.g. Faure, 1986; Tucker and Wright, 1990) since the $^{87}Sr/^{86}Sr$ ratio obtained in these dolomites is much higher than the $^{87}Sr/^{86}Sr$ ratios inferred for carbonates precipitated from seawater during the latest Cretaceous (McArthur *et al*., 1994; Veizer *et al*., 1999).

5.2. Valle de Tabladillo Fm.

Dolomite of the Valle de Tabladillo Fm. of the northern margin of the Central System, which lack signs of subsequent recrystallization, was probably precipitated during or just after sedimentation via sabkha dolomitization. Dolomitization in a sabka environment with high evaporation rates is supported by the interbedded nature of this dolomite with evaporite horizons and by carbon and oxygen isotope values, which are relatively high (+2.5‰ - +3.1‰ and +1.07‰ - +1.6‰, respectively) (e.g. Tucker and Wright, 1990; Warren, 2001). Moreover, the $^{87}Sr/^{86}Sr$ ratios obtained for this dolomite correspond to the ratio inferred for carbonates precipitated from seawater during the Campanian (McArthur *et al*., 1994; Veizer *et al*., 1999; Fig. 2), when this unit was deposited. The observation that the $^{87}Sr/^{86}Sr$ ratios are higher in samples containing higher clay and quartz contents is probably attributable to a greater supply of continental sediments and waters, which have higher $^{87}Sr/^{86}Sr$ ratios than seawater (e.g. Faure, 1986; Tucker y Wright, 1990).

The higher $^{87}Sr/^{86}Sr$ ratios of dolomite in the southern Central System compared to those of the northern border (Fig. 2), probably reflects their subsequent recrystallization. Recrystallization could have occurred during sedimentation of the Torrelaguna Fm. (Fig. 5E), as suggested by the fact that the breccia dolomites have higher $^{87}Sr/^{86}Sr$ ratios than those of underlying units and that these values are intermediate with respect to the ratios obtained for the Valle de Tabladillo Fm. at the northern margin of the Central System and for the Torrelaguna Fm. (Fig. 2).

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luminescence, are composed of calcite (CC). F. Transmitted light (left) and CL micrographs (right) of dolomites from the Torrelaguna Fm. The micritic matrix shows mottled reddish-orange luminescence (D). The dolomite also partially cements gypsum pseudomorphs (blue arrows). Pseudomorphs are occluded by nonluminescent calcite (CC).

Fig. 5.- (página opuesta) A. Aspecto de las dolomías del miembro superior de la Fm. Castrojimeno. Los cristales de dolomita (D) remplazan a la matriz y rellenan parcialmente la porosidad móldica. Estos cristales presentan, en general, un núcleo romboédrico de aspecto sucio que se encuentra parcial o totalmente calcitizado (flechas rojas) y una parte exterior de aspecto claro. La porosidad está ocluida por un cemento de calcita no ferrosa (CC). B. Fotografías realizadas con microscopia de luz transmitida (izquierda) y catodoluminiscencia (derecha) de una muestra del miembro superior de la Fm. Castrojimeno. La dolomita (D) que remplaza a la matriz presenta un aspecto sucio y una luminiscencia de color rojo anaranjado. Los cristales de dolomita que rellenan la porosidad (flechas azules) tienen un núcleo de aspecto sucio, hábito romboédrico y luminiscencia de color rojo anaranjado y una parte externa, más limpia, de luminiscencia de color rojo. La porosidad está ocluida por calcita no luminiscente (CC). C. Aspecto de las dolomías de la Fm. Burgo de Osma. La dolomita (D) remplaza a la matriz y rellena parcialmente la porosidad móldica que, a su vez, está ocluida por un cemento de calcita no ferrosa (CC). La dolomita que remplaza a la matriz presenta un aspecto sucio. Los cristales de dolomita que cementan los poros (flechas azules) tienen un núcleo, parcial o totalmente calcitizado, de aspecto sucio y hábito romboédrico (flecha roja) y una parte externa más limpia. Obsérvese la similitud de estas dolomías con las del miembro superior de la Fm. Castrojimeno y con las de la figura 3H. D. Fotografías realizadas con microscopia de luz transmitida y catodoluminiscencia de una muestra de la Fm. Burgo de Osma. La dolomita (D) que remplaza a la matriz presenta un aspecto sucio y una luminiscencia moteada anaranjada. Los cristales de dolomita que rellenan la porosidad (flechas azules) tienen un núcleo de aspecto sucio, hábito romboédrico y luminiscencia zonada de color anaranjado, rojo y marrón y una parte externa, más limpia, de luminiscencia de color marrón oscuro. La porosidad está ocluida por calcita no luminiscente (CC). E. Fotografías realizadas con microscopia de luz transmitida y catodoluminiscencia de las brechas de la Fm. Valle de Tabladillo del borde Sur del Sistema Central. Se pueden apreciar zonas de aspecto más sucio y de luminiscencia moteada y zonada de color amarillo y naranja que están formadas por dolomías intensamente calcitizadas (D-M). Las zonas más claras y de luminiscencia zonada no luminiscente, marrón y anaranjada están formadas por calcita (CC). F. Fotografías realizadas con microscopia de luz transmitida y catodoluminiscencia de las dolomías de la Fm. Torrelaguna. La matriz dolomicrítica tiene una luminiscencia moteada de color rojo anaranjado (D). La dolomita también rellena parcialmente los pseudomorfos de las evaporitas, yeso probablemente (flechas azules). Los pseudomorfos están cementados posteriormente por calcita no luminiscente (CC) que ocluye la porosidad

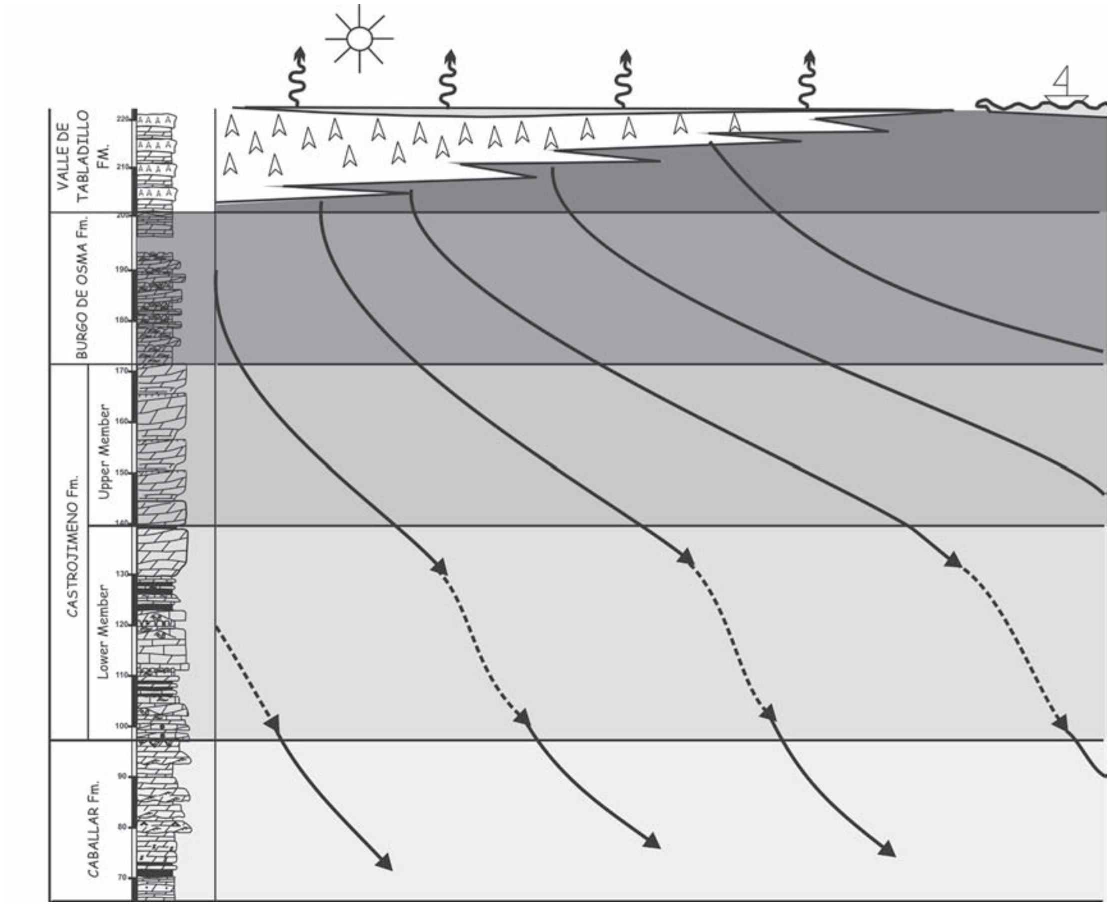

Fig. 6.- Diagram showing the brine reflux dolomitization model proposed for the Late Cretaceous carbonate units at the south margin of the Spanish Central System. Arrows indicate the descending movement of brines formed during the deposition of the evaporative Valle de Tabladillo Fm. Dashed arrows indicate partially dolomitized stratigraphic levels containing marls, which may have acted locally as barriers of low-permeability for dolomitizing fluids.

Fig. 6.- Esquema representativo que resume el modelo de dolomitización por reflujo de salmueras propuesto para las unidades del Cretácico Superior en el borde Sur del Sistema Central. Las flechas indicarían el movimiento descendente de estas salmueras desde la Fm. evaporítica Valle de Tabladillo que se depositó y dolomitizó en un ambiente de *sabkha* costera. Las flechas de trazo discontinuo se han representado en los tramos que se encuentran parcialmente dolomitizados y que contienen los tramos margosos que pudieron actuar como barreras poco permeables para el paso de los fluidos dolomitizantes.

5.3. Caballar, Castrojimeno and Burgo de Osma Fms.

Petrographic and geochemical analysis of the dolomite within the Caballar, Castrojimeno and Burgo de Osma Formations at the southern margin of the Central System, suggests that dolomitization occurred after sedimentation, during the initial stages of burial of these units. Because most of the dolomites comprises dolosparitic mosaics, which in addition to replacing matrix and allochems also infill moldic porosity, dolomite precipitation must have occurred after the dissolution of the depositional components whose onset probably occurred once burial had commenced. In contrast to the Caballar and Burgo de Osma Fms., which were deposited in a tidal flat environment where a process of synsedimentary dolomitization could have taken place, the Castrojimeno Fm. was deposited in a platform setting, which is not compatible with this type of dolomitization. Moreover, the shallower facies of the lower member of the Castrojimeno Fm., which consist of mudstone and wackestone with incipient nodulization and fenestral porosity, are those that most probable would have undergone synsedimentary dolomitization. These facies, however, show the least degree of dolomitization. This contrasts with the upper member of the Castrojimeno Fm., which is totally dolomitized even though this sequence was deposited in an open platform setting. In addition, petrographic and geochemical data are identical for dolomite of the upper member of the Castrojimeno Fm., comprised of subtidal platform bars, and for the Burgo de Osma Fm., deposited on a tidal flat (Figs. 2, 4A-D). This suggests that dolomite in both units precipitated from similar fluids and under a similar diagenetic environment.

The fact that the middle part of the lower member of the Castrojimeno Fm. is scarcely dolomitized is probably due to the presence of marls, which may have acted as low-permeability barriers or aquicludes for dolomitizing fluids. This could also have been responsible for the calcic and non-stoichiometric dolomite of the lower member, since these aquicludes could have limited the supply of Mg necessary for complete dolomitization of this unit. It is possible that Mg necessary for dolomitization was derived from the micritic matrix, typically formed by high-magnesium calcite (HMC) and aragonite, and from HMC components, since dolomitization generally affects the micritic matrix and micritic components and not the rudist shells (Fig. 3D), which are composed of low-magnesium calcite.

In the uppermost Caballar Fm., the grain-rich sequences are largely dolomite, which is petrographically and geochemically identical to dolomite of the overlying lower member of the Castrojimeno Fm. (Fig. 3B-E), suggesting that dolomitization probably occurred at the same time and from the same fluids. The basal mud-rich sequences of the Caballar Fm. are the only lithologies of the entire stratigraphic section that are formed of dolomicrite and dolomicrosparite, supporting an early syndepositional dolomitization. Nevertheless, the geochemistry of this dolomicrite is similar to that of dolomites present in other units, such that one cannot preclude subsequent recrystallization, which led to the growth of the dolomite crystals and the re-equilibration of their element and isotope compositions.

Finally, another finding that suggests that the Caballar, Castrojimeno and Burgo de Osma Fms. were concurrently dolomitized from the same fluids and under the same diagenetic conditions is the similarity of both oxygen and carbon isotope compositions and $^{87}Sr/^{86}Sr$ ratios of the dolomite appearing in each of the three units (Fig. 2). Significantly, the $^{87}Sr/^{86}Sr$ ratios in dolomite of these units are not in accordance with the marine $^{87}Sr/^{86}Sr$ ratios inferred for the Late Cretaceous (McArthur *et al*., 1994; Veizer *et al*., 1999; Fig. 2). In effect, the $^{87}Sr/^{86}Sr$ ratios inferred for carbonates precipitated from seawater during the time interval when these units were deposited are lower than the expected marine $^{87}Sr/^{86}Sr$ ratios and differ especially from those of the basal formations (Fig. 2).

Regarding the model for dolomitization of the Caballar, Castrojimeno and Burgo de Osma Fms., it is likely that dolomitization occurred via a reflux of evaporative brines. This model is consistent with the petrographic data, as the younger uppermost units are the most intensely dolomitized, whereas, horizons interbedded with marly lithologies, which could have acted as aquicludes, are only partially dolomitized or non-dolomitized. This model is also supported by isotope data, because carbon and oxygen isotopic values are relatively positive, as would be expected from reflux dolomitization by evaporative bines (e.g. Tucker and Wright, 1990; Warren, 2001). The observation that in some samples the more negative oxygen isotopic compositions is accompanied by higher $^{87}Sr/^{86}Sr$ ratios may indicate some mixing with meteoric waters which typically have more negative oxygen isotopic values and higher $^{87}Sr/^{86}Sr$ ratios than seawater (e.g. Faure, 1986; Tucker and Wright, 1990). Such mixed-water reflux dolomitization models, where dense saline waters mix with meteoric water, have been proposed for the dolomitization of the Messinian coral reefs in the south of the Iberian Peninsula (Meyers, *et al*., 1997; Warren, 2001).

Dolomitization of these units by brine reflux may have taken place during deposition of the evaporative Valle de Tabladillo or Torrelaguna Formations. The $^{87}Sr/^{86}Sr$ ratios obtained for the dolomites of the Caballar, Castrojimeno and Burgo de Osma Fms. are similar to those recorded for the dolomites of the Valle de Tabladillo Fm. at the northern margin of the Central System, but differ considerably from the carbonates of the Torrelaguna Fm. Thus, it is likely that dolomitization of these units was produced by the reflux of brines from the Valle de Tabladillo Fm. (Fig. 6).

6. Conclusions

The detailed pretrographic and geochemical study of the dolomitic units of the Late Cretaceous in the southern margin of the spanish Central System suggests that even though Caballar, Castrojimeno and Burgo de Osma Formations were deposited in different sedimentary environments, dolomitization likely occurred at the same time, during the first stages of burial diagenesis via reflux of evaporative brines or mixed-waters. The source of these brines were likely related to the depositional environment of the overlying Valle de Tabladillo Formation, which is constituted of interbedded evaporites and dolomicrites that were deposited and dolomitized in a coastal sabkha environment.

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