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Petrogenetic relationships between Variscan granitoids and Li-(F-P)-rich aplite-pegmatites in the Central Iberian Zone: Geological and geochemical constraints and implications for other regions from the European Variscides

E. Roda-Robles[a,*], C. Villaseca[b], A. Pesquera[a], P.P. Gil-Crespo[a], R. Vieira[c], A. Lima[c], I. Garate-Olave[a]

[a] Dpto. Mineralogía y Petrología, Universidad del País Vasco UPV/EHU, Barrio Sarriena s/n, 48940 Leioa, Spain
[b] Dpto. Petrología y Geoquímica, Universidad Complutense, IGEO (UCM, CSIC), 28040 Madrid, Spain
[c] Instituto de Ciências da Terra, Universidade do Porto/DGAOT, Rua do Campo Alegre 687, 4169-007 Porto, Portugal

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ABSTRACT

The Central Iberian Zone (CIZ) is characterised by a large volume of Variscan granitic intrusions, which can be grouped into five types: (1) two-mica peraluminous leucogranites (S_1); (2) P-rich highly peraluminous granites (S_2); (3) P-poor moderately peraluminous granites (S_3); (4) moderately to low peraluminous granites (S_4); and (5) I-type low peraluminous granites (I). Though not as abundant as granites, aplite-pegmatite rocks are nonetheless widespread in this region, occurring either as fields of aplite-pegmatite dykes or as leucogranitic cupolas. They are commonly enriched in Li-(F-P) minerals such as spodumene, petalite, micas, and phosphates of the amblygonite-montebrasite and triphylite-lithiophilite series. Many of the Li-rich bodies show an aplitic texture, frequently with the development of layered units. Coarse crystals are also common, but are mostly smaller than 12 cm long. They usually do not show internal fractionation, zoning or a quartz-core, and often have high values in Li_2O (> 1.3 wt%), and high Na, F and P contents. Evidence in support of a petrogenetic link among peraluminous granites and aplite-pegmatite bodies is provided by field relationships and geochemical affinities. The Li-(F-P)-mineralisation is closely related to the S_1 series in the northern CIZ realm, whereas in the southern portion of the CIZ the parental granites correspond to the S_2 series. The granites of the S_3 and S_4 series, and the I-type granite suite are not related to the Li mineralisation. The S_1 and S_2 granites are interpreted to derive mainly from the partial melting of highly peraluminous, Ca-poor and P-rich Neoproterozoic metasediments during the Variscan orogeny. The melts are presumed to evolve favoured by a high content in fluxing components, such as P, F, B, Li, and H_2O, which contributed to the lowering of viscosity, solidus temperature and polymerisation degree. This is in parallel to the increasing of the diffusion rates and mobility of the highly fractionated melts. The residual melts, enriched in incompatible elements such as Li and F, as well as B, Sn and other rare elements, tend to accumulate at the top of the granitic cupolas. Therefore, Li-rich dykes as observed in many aplite-pegmatite fields of the CIZ are arguably the result of the opening of the system, whereas Li-rich granitic cupolas form when the system remains closed. Lithium-rich rocks, comparable to those of the CIZ, are found in other parts of the European Variscan Belt also related to P-rich, Ca-poor, highly peraluminous S-type granites originated during the Variscan Orogeny. Accordingly, we postulate the existence of an extense Li-metallogenetic province including mainly the CIZ in Spain and Portugal, the Massif Central in France, the Bohemian Massif in the Czech Republic and Germany, the western Carpathians in the Slovak Republic, and the Cornwall region in the south west of England.

1. Introduction

Lithium has been recently defined as the "new gasoline" by some of the most important investment banks of the world. The use of this element in the production of batteries for electric cars has contributed to a substantial increase in its price, and this trend will continue as lithium is the lightest of the known metals with the best energy/weight ratio, which makes it perfect to feed batteries. The high demand has led to a "lithium rush", with many mining companies starting exploration campaigns in different countries worldwide. The largest reserves of Li

* Corresponding author.
E-mail address: encar.roda@ehu.es (E. Roda-Robles).

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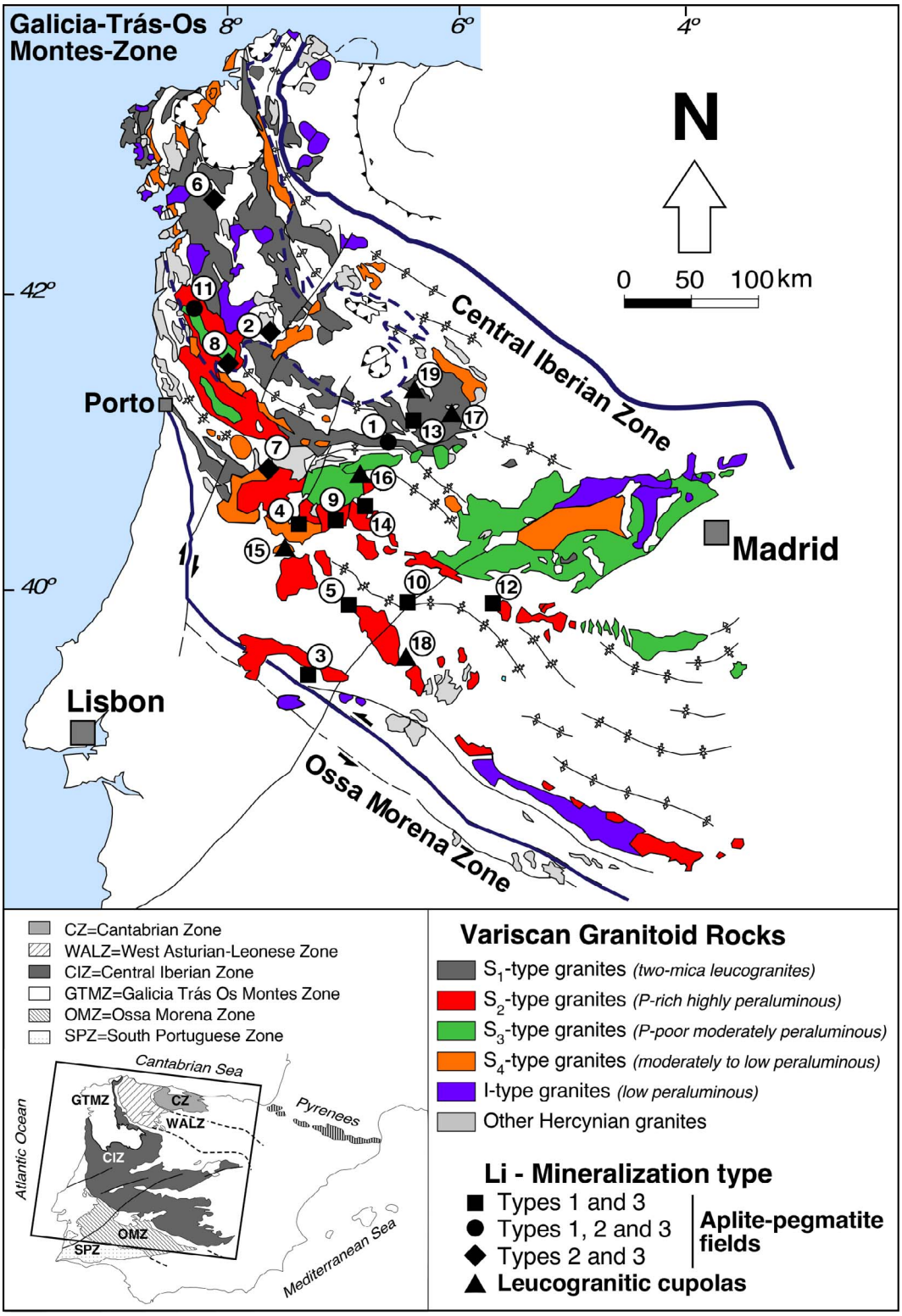

Fig. 1. Schematic geological map of the Central Iberian Zone (CIZ) and the Galicia-Trás-Os Montes Zone (GTMZ) (Spain and Portugal) (modified from Martínez-Catalán et al. (2004), with the permission of the SGE and authors) showing the distribution of the five Variscan granitic series proposed in the classification of Villaseca (2011), and of the Li-mineralization occurring in aplite-pegmatite fields and in leucogranitic cupolas. Numbers as in Table 1. Legend for the aplite-pegmatite fields, Type 1 = Li-mica rich aplite-pegmatites; Type 2 = spodumene and/or petalite-rich aplite-pegmatites; Type 3, simple aplite-pegmatites.

are found in low-grade evaporitic-type deposits, the most important located in the so-called Lithium Triangle, close to the borders between Bolivia, Chile and Argentina. According to the US Geological Survey, 58% of the world reserves occur in this area. However, this is not the only type of Li-deposit. Rare-element granitic pegmatites may also be enriched in this element, with concentrations that are high enough to make their mining economically profitable. A significant number of pegmatites are being mined or explored for Li at this moment (Jaskula, 2014, 2017; Kesler et al., 2012). The main Li-mineral in these pegmatites is spodumene, but other phases, such as petalite, lepidolite or

Table 1
Localities and main characteristics of Li-enriched aplite-pegmatite fields and leucogranitic cupolas in the Central Iberian Zone (Modified from Roda-Robles et al. (2016)).

N	Locality	Mineralization type (country rocks)	Mineral association*	Li-minerals**
1	**Fregeneda-Almendra** (Salamanca, Spain- Guarda, Portugal) Vieira et al. (2011)	Types 1, 2 and 3 Several dykes, some Li-rich (metapelites)	Qtz, Fsp, Li-Ms, Spd, Ptl, Cst, Mtb	**Spodumene** **Petalite** **Li-Muscovite** Montebrasite
2	**Barroso-Alvão** (Vila Real, Northern Portugal) Lima (2000), Martins et al. (2012)	Types 2 and 3 Several dykes, some Li-rich (metapelites)	Qtz, Fsp, Ms, Spd, Ptl, Cst, Nb-Ta oxides	**Spodumene** **Petalite** Lepidolite Montebrasite
3	**Tres Arroyos** (Badajoz, Spain) Garate-Olave et al. (2017)	Types 1 and 3 Aplite-pegmatite dykes, some Li-rich (metasediments)	Qtz, Fsp, Ms, Li-Ms, Mtb, Tpz, Cst, Nb-Ta oxides	**Li-Muscovite** Montebrasite
4	**Gonçalo** (Belmonte-Guarda, Portugal) Neiva and Ramos (2010)	Types 1 and 3 Li-rich aplite-pegmatite dykes (granite)	Qtz, Fsp, Ms, Lpd, Amb, Ptl, Tpz, Tur, Cst, Nb-Ta oxides + Zwd in country rock	**Lepidolite** **Amblygonite**
5	**Segura** (Central Portugal) Antunes et al. (2013)	Types 1 and 3 Li-rich aplite-pegmatite dykes (metapelites)	Qtz, Fsp, Lpd, Mtb, Tpz, Cst, Nb-Ta oxides	**Lepidolite** Montebrasite
6	**Lalin-Forcarei** (Galicia, Spain) Fuertes-Fuente and Martín-Izard (1998)	Types 2 and 3 Aplite-pegmatite dykes, some Li-rich (metasediments)	Fsp, Qtz, Spd, Ms, Mtb, Cst, Nb-Ta oxides	**Spodumene** Montebrasite
7	**Queiriga** (Alto Vouga, Portugal) Dias et al. (2013)	Types 2 and 3 Perigranitic pegmatites (andalusite schists)	Qtz, Fsp, Ptl, Lpd, Spd, Tpz, Brl, Cst, Nb-Ta oxides	**Petalite**, Lepidolite, Spodumene
8	**Seixoso-Vieiros** (Vila Real, Northern Portugal) Lima et al. (2009)	Types 2 and 3 Aplite-pegmatite dykes + leucogranitic cupola (metasediments)	Qtz, Fsp, Ptl, Mtb, Spd	**Petalite** Montebrasite, Spodumene
9	**Cabeço dos Poupos** (Sabugal, Portugal) Neiva et al. (2012)	Types 1 and 3 Li-rich aplite-pegmatite dykes (granite)	Qtz, Fsp, Ms, Li-Ms, Lpd, Zwd, Tur, Cst, Nb-Ta oxides, Ap, triplite	**Li-Muscovite**, Lepidolite Zinnwaldite
10	**Las Navas** (Cáceres, Spain) Gallego Garrido (1992)	Types 1 and 3 Li-rich aplite-pegmatite dykes, (metasediments)	Qtz, Fsp, Ms, Li-Ms, Mtb, Tpz, Spd, Cst, Nb-Ta oxides	**Li-Muscovite** Montebrasite Spodumene
11	**Serra de Arga** (Viana do Castelo, Portugal) Leal Gomes (1994)	Types 1, 2 and 3 Li-rich aplite-pegmatite dykes, (metasediments)	Qtz, Fsp, Ms, Ptl, Lpd, Spd, Cst, Nb-Ta oxides	**Petalite** **Lepidolite** Spodumene Montebrasite
12	**Belvís de Monroy** (Cáceres, Spain) Sanchez-Muñoz et al. (2017)	Types 1 and 3 Li-rich aplite-pegmatite dykes (metapelites)	Qtz, Fsp, Lpd, Mtb, Tpz,	**Lepidolite** Montebrasite
13	**Valderrodrigo** (Salamanca, Spain) Junta de Castilla y León (1986)	Types 1 and 3 Li-rich aplite-pegmatite dykes (michaschists)	Qtz, Fsp, Lpd, Tpz, Fe-Mn phosp	**Lepidolite** Lithiophilite
14	**La Canalita** (Salamanca, Spain) Llorens (2011)	Types 1 and 3Aplite-pegmatite dykes (metasediments)	Qtz, Fsp, Ms, Li-Ms, Amb, some Li-rich Tpz, Cst, Nb-Ta oxides	**Lepidolite** Amblygonite
15	**Argemela** (Guarda, Portugal)Charoy and Noronha (1996)	Microgranite + Quartz-Albite-dykes with montebrasite	Qtz, Fsp, Ms, Lpd, Mtb, Cst, Nb-Ta oxides	**Lepidolite** Montebrasite
16	**Castillejo de Dos Casas**(Salamanca, Spain) Martín-Izard et al. (1992), Roda-Robles et al. (2015)	Stockscheider over a leucogranitic cupola in contact to the metapelites	Qtz, Fsp, Li-Ms, Ptl, Cst, Mtb, Tpz, Fe-Mn phosp	**Li-Muscovite** Petalite Montebrasite Lithiophilite
17	**Golpejas**(Salamanca, Spain)Martín-Izard et al. (1992)	Leucogranitic cupola + Quartz-dykes with montebrasite (granite, metasediments)	Qtz, Fsp, Mtb, Cst, Nb-Ta oxides, Tpz	**Montebrasite**
18	**El Trasquilón** (Cáceres, Spain)Gallego Garrido (1992)	Leucogranitic cupola + Quartz-dykes with montebrasite	Qtz, Fsp, Mtb, Cst, Nb-Ta oxides, sulphides	**Montebrasite**
19	**Pinilla de Fermoselle (Zamora, Spain)**Roda-Robles et al. (2012)	Pegmatitic cupola over a granite, Li-rich in its upper part (granite, marbles)	Qtz, Ms, Fsp, Tur, Lpd, Bt, Zwd, Fe-Mn phosph	**Lepidolite** Ferrisicklerite Elbaite Zinnwaldite

*Abbreviations: Qtz-quartz; Fsp-feldspar; Brl-beryl; Lpd-lepidolite; Spd-spodumene; Ptl-petalite; Cst-cassiterite; Mtb-montebrasite; Amb-amblygonite; Ms-muscovite; Bt-Biotite; Tur-tourmaline; Tpz-Topaz; phosp-phosphates; Ap-Apatite; Zwd-zinnwaldite. **The most abundant Li-phases are presented in bold.
Aplite-pegmatite fields: Type 1 = Li-mica rich aplite-pegmatites; Type 2 = spodumene and/or petalite-rich aplite-pegmatites; Type 3: simple aplite-pegmatites.

amblygonite-montebrasite, may also be suitable to produce Li-carbonate, the most used compound for the manufacturing of batteries. Therefore, exploration for Li associated with pegmatites has also been increasing in an exponential way in recent years.

Lithium-enriched rocks are common in the Central Iberian Zone (CIZ) of the Iberian Massif (Spain and Portugal), in a ~75,000 km^2, NNW-SSE striking curved belt, (Fig. 1, Table 1). Even if Li may be enriched in some coarse-grained beryl-phosphate pegmatites, and in veins very rich in quartz and montebrasite; in the CIZ the Li-rich minerals occur mainly in aplite-pegmatite bodies grouped in pegmatite fields, sometimes spatially zoned around a larger granite intrusion. Less frequently, similar Li-enrichments may be observed in the marginal and/or apical parts of leucogranites (Roda-Robles et al., 2016). The textural and chemical features of these aplite-pegmatite bodies are not so commonly observed in other pegmatitic belts of the world according to the current bibliography on pegmatites. Field, geochemical and structural relationships, as well as the available geochronological data (Melleton and Gloaguen, 2015; Roda-Robles et al., 2009), indicate that

the origin of this Li-mineralisation is related to the major Variscan granitic magmatism occurring between 320 and 290 Ma. However, magmatism of different ages and geochemical signatures overlaps frequently in the same areas, making it difficult to decipher the petrogenetic relationships between the different types of Li-mineralisation and the different Variscan granitic series. The assessment of the linkage between this uncommon worldwide Li-mineralisation and the different granitic series in the CIZ may contribute not only to better understand the origin of the aplite-pegmatites and the last stages of crystallisation of the fractionated granitic systems, but may also advance the knowledge on the geology of the CIZ and of the whole European Variscan orogen. Last but not least, understanding the relationships between the Li-mineralisation and the different granitic series in the CIZ will facilitate the exploration campaigns for Li in this region.

In this paper a general description of the Li-bearing aplite-pegmatite dykes and leucogranitic cupolas, as well as of the different granite suites, is presented. We focus on the petrogenetic links among aplite-pegmatites and granites, based on field relationships and geochemical evidence. We also discuss the causes and mechanisms for the Li-enrichment in these rocks and their potential sources. Finally, a comparison of the Li-mineralisation and the associated granites in the CIZ with other equivalent occurrences in the European Variscan Belt is made, discussing the existence of an important Li-metallogenetic province in the European Variscides.

2. Geological setting

The CIZ represents the innermost region of the Variscan Iberian Belt and is characterised by a large volume of granitic intrusions (Fig. 1). The generation of such a huge volume of syn- to post-tectonic granitoids along the CIZ is an outstanding example of complex batholith construction in Western Europe (e.g., Bea et al., 1999; Capdevila et al., 1973; Dias et al., 2002; Villaseca et al., 1998b).

The CIZ is part of the northern Gondwana margin and is mainly composed of metasedimentary rocks, a thick Neoproterozoic to Early Paleozoic sequence, intruded by Cambro-Ordovician peraluminous metagranites, all of them variably metamorphosed by the Variscan orogeny. Early Ordovician to Early Carboniferous sediments lay unconformably over these metasedimentary and metaigneous sequences, defining an upper crustal suprastructure not involved in the main Variscan crustal recycling processes. Neoproterozoic to Early Cambrian metasediments outcrop widely in the southern CIZ (S-CIZ), where they define a thick succession of metamorphosed shales and sandstones with scarce interlayered volcanoclastic rocks (up to 11,000 m), known as the Schist-Greywacke Complex (SGC) (e.g., Rodríguez Alonso et al., 2004). Two units (Lower and Upper) have been distinguished, separated by the Cadomian unconformity but sharing similar geochemical features and a characteristic isotopic composition of low negative initial ƐNd signatures (Villaseca et al., 2014). The pre-Early Ordovician metasedimentary sequences of the northern CIZ (N-CIZ) (over 8000 m) are chemically different to the SGC of the southern parts (S-CIZ), being richer in LILE, REE and some HFSE (Th, U, Nb, Ta), and having lower P contents (Table 2) and higher negative initial ƐNd values (Villaseca et al., 2014). These chemical differences led to the consideration of a boundary separating the northern and southern CIZ domains based on different geological, geochemical and isotopic features (Orejana et al., 2015; Villaseca et al., 2014).

The CIZ has a superposed accreted terrane (the Galicia-Trás-Os-Montes Zone, GTMZ) assembled during the eo-Variscan collision at around 395 Ma (e.g., Ballévre et al., 2014). Other neighboring terranes (the West Asturian Leonese Zone, WALZ; and the Ossa Morena Zone, OMZ (Fig. 1)) were adjacent (WALZ) or definitively amalgamated during the late Paleozoic at the main Variscan collisional stages (at ca. 350–360 Ma, e.g., Martínez Catalán et al., 2014). The end of continental subduction under the CIZ was marked at ca. 345 Ma by the exhumation of eclogitised metabasites in the Spanish Central System (Villaseca et al., 2015). Massive production of granitoids took place during the period from 320 to 295 Ma (e.g., Dias et al., 1998; Fernández-Suárez et al., 2000; Gutiérrez-Alonso et al., 2011) and the batholiths are concentrated in the northern part of the CIZ – the most thickened Variscan domain (Simancas et al., 2013). Several authors have suggested the determinant role of crustal thickening in generating collisional magmatism in Iberia (Alcock et al., 2015; Bea, 2012; Merino Martínez et al., 2014; Simancas et al., 2013). The progressive thermal maturity of the thickened crust combined with the change from a compressional to a transtensional/transpressional regime that subsequently produces a complex pattern of strike-slip shear zones, resulted in extensive crustal melting (e.g., Bea et al., 1999; Dias et al., 2002).

Variscan granitoids intrude into Neoproterozoic to Palaeozoic sequences of metasedimentary and metaigneous rocks of variable metamorphic degrees, inducing thermal aureole metamorphism. The granites were mainly emplaced during post-collisional stages (*syn*-to post-D3, the last ductile deformation phase) (Dias et al., 2002). Four S-types (S_1, S_2, S_3 and S_4) and one I-type granite series have been summarised in the CIZ (Villaseca, 2011) and will be described below in the text. A remarkable feature of the CIZ Variscan magmatism is the lack of a spatial or temporal pattern distribution of granite series along the collisional belt during this long magmatic event of around 25 Ma. Nevertheless, geochronological data from leucogranite massifs (S_1 types of Villaseca, 2011, see section below) indicate that they peak at 312 to 316 Ma, whereas I-type granites from Galicia and northern Portugal are mostly post-tectonic (301–295 Ma) (e.g., Dias et al., 1998; Gutiérrez-Alonso et al., 2011). This post-collisional Variscan granite magmatism is accompanied by sporadic minor mafic inputs, which are variably contaminated and intermingled with the felsic magmas throughout this event (e.g., Dias et al., 1998; Orejana et al., 2009; Scarrow et al., 2009; Villaseca et al., 2011).

Besides the granitic rocks, pegmatites and aplite-pegmatites are also relatively common in the CIZ, mainly in the western part of both Spain and Portugal, occurring along a ~500 km-long and ~150 km wide NNW-SSE striking belt (Fig. 1). Most of the pegmatites in the CIZ belong to the Lithium-Cesium-Tantalum (LCT) family defined by Černý and Ercit (2005), and are commonly petrogenetically related to S-type peraluminous granitic magmas. In addition, in the central-eastern part of the CIZ, a few pegmatitic bodies belonging to the Niobium-Yttrium-Fluorine (NYF) family occur, usually filling miarolitic cavities inside I-type granites (González del Tánago et al., 2008). The LCT pegmatites usually appear hosted in the Neoproterozoic metasediments or within granitic rocks. Most of these pegmatites exhibit a poor internal differentiation. In this region, Li is frequently enriched in some of the most fractionated pegmatitic rocks (usually aplite-pegmatites), and in some quartz-rich veins (Roda-Robles et al., 2016). Their relationship to the different granitic series occurring in this region is not always clear, as granites of different age and composition occur commonly in the same area.

3. Data collection and analyses

Chemical values of the bulk composition of pegmatites are rarely provided in the literature due to the coarse grain size and the mineralogical and geochemical heterogeneity usually exhibited by these rocks, which makes it difficult to obtain representative data. However, the moderate thickness (mostly < 2 m) combined with the small grain size exhibited by the aplite-pegmatite dykes from the CIZ and the lack of an internal zoning allowed us to acquire 37 whole-rock data. In order to obtain representative analyses, only the most homogeneous bodies were chosen for sampling. In the case of the dykes, the thin ones were preferably sampled, taking at least 5 kg of rock across them. This material was crushed and homogenised before the analyses. These were made at the Activation Laboratories Ltd. (Actlabs, Canada), by X-ray fluorescence (XRF) for major elements and ICP-MS for trace elements. Li samples were fused with sodium peroxide. Fluorine was analysed at

Table 2
Whole-rock major (wt.%) and trace-element (ppm) mean composition of the three different aplite-pegmatite facies, including also data from leucogranitic cupolas in the first and third columns (Type 1 = Li-mica rich; Type 2 = petalite and/or spodumen rich; Type 3 = simple facies), the five granitic series, the shales from the N-CIZ and S-CIZ (NIBAS-Neoproterozoic Iberian Average Shale, from Ugidos et al. (2010)); plus the NASC (North American Shale Mean Composition, from Gromet et al. (1984)); the UCC (Upper Continental Crust, from Rudnick and Gao (2003)) and the BCC (Bulk Continental Crust, from Rudnick and Gao (2003)) for comparison. Besides own data, data from the next sources have been used to calculate the mean values in this table and in the different geochemical plots: Aplite-pegmatites, Gallego Garrido (1992), Martín Izard et al. (1992), Almeida (2003), Antunes et al. (2013), Neiva and Ramos (2010), Roda-Robles et al. (2012), Charoy and Noronha (1996), Leal Gomes (1994), Fuertes-Fuente and Martín-Izard (1998), Roza Llera (2014), Helal (1992). S_1 granites, López-Plaza and López-Moro (2003), Roza Llera (2014), Roda-Robles et al. (2012), Ortega (1998), López-Moro et al. (2014), López Plaza and Gonzalo (1993), Almeida (1994), Neiva et al. (2007). S_2 granites, Helal (1992), Gallego Garrido (1992), Ramírez and Menéndez (1999), González Menéndez (2002), Leal Gomes (1994), Antunes et al. (2008), Neiva et al. (2011, 2012), Merino Martínez et al. (2014), Chicharro et al. (2014). S_3 granites, Brandebourguer (1984), Casillas (1989), Andonaegui (1990); S_4 granites, Herreros (1998), Martins (1998), Martins et al. (2009). I granites, Brandebourguer (1984), Casillas (1989), Cuesta (1991), Mendes and Dias (2004), Pérez-Soba and Villaseca (2010).

n	Type 1 41	Type 2 25	Type 3 106	S1 162	S2 138	S3 228	S4 83	I 122	N-CIZ-SHALE 8 (F-Li-Cs = 57 samples)	S-CIZ-SHALE NIBAS + Li-F-Sn (13–4-12 samples)	NASC	UCC	BCC
(wt.%) SiO_2	68.82	71.80	72.39	71.52	72.00	72.00	71.04	71.56	59.60	59.47	64.80	66.62	60.60
Al_2O_3	17.84	16.73	16.03	14.83	14.81	14.36	14.29	14.28	19.26	19.19	16.90	15.40	15.90
Fe_2O_3(T)	0.62	0.39	0.58	1.79	1.86	2.35	2.76	2.64	7.93	7.39	6.29	5.60	7.45
MnO	0.07	0.04	0.03	0.02	0.03	0.05	0.08	0.05	0.10	0.04	0.06	0.10	0.10
MgO	0.08	0.06	0.04	0.45	0.46	0.57	0.72	0.60	2.40	2.71	2.86	2.48	4.70
CaO	0.41	0.46	0.40	0.77	0.74	1.47	1.74	1.77	0.69	0.29	3.63	3.59	6.40
Na_2O	4.80	5.24	5.40	3.19	3.37	3.32	3.52	3.44	1.31	1.85	1.14	3.27	3.10
K_2O	2.80	2.22	2.43	5.03	4.66	4.43	4.39	4.49	3.76	3.59	3.97	2.80	1.80
TiO_2	0.01	0.01	0.01	0.26	0.28	0.29	0.37	0.33	1.02	0.94	0.70	0.64	0.70
P_2O_5	0.90	0.75	0.75	0.34	0.42	0.17	0.14	0.10	0.17	0.21	0.13	0.15	0.10
Total	96.34	97.71	98.05	98.21	98.63	99.01	99.05	99.25	96.24	95.68	100.48	100.65	100.85
(ppm) F	9517	1329	2081	1510	1590	1081	804	931*	597	950	–	557	553
Li	3842	2819	313	165	207	48	95	75*	95	112	66	24	16
Ba	36.9	29.6	26.0	341	254	324	375	387	737	647	636	628	456
Rb	2031	737	529	302	349	237	221	212	173	126	125	82	49
Sr	164	137	80	94	79	92	99	98	132	80	142	320	320
Be	106	116	77	8.5	9.4	4.3	5.5	3.7	4.9	2.7	–	2.1	1.9
Cs	309	60	34	20	31	14	16	13	16	6.6	5.2	4.9	2
Sn	580	480	207	18	29	6.6	13	10	4.7	5.3	–	2.1	1.7
Pb	17	19	15	44	26	34	25	32	20	20	20	17	11
Zr	39	23	28	112	108	128	141	141	305	217	200	193	132
Hf	4.3	3.8	3.3	3.3	3.6	3.8	4.5	4.6	8.2	5.9	6.3	5.3	3.7
Nb	67	61	45	10	14	13	14	11	20	13	–	12	8
Ta	79	97	46	3.4	3.3	1.9	3.5	2.3	1.6	1.2	1.1	0.9	0.7
Y	2.3	1.1	2.7	9.6	12	27	31	36	39	36	27	21	19
A/CNK	1.53	1.45	1.31	1.23	1.24	1.11	1.04	1.04	2.57	2.57	1.32	1.03	0.85
ΣLREE	4.1	3.1	3.1	93	94	87	126	135	245	163	131	126	87
ΣHREE	0.9	0.5	0.8	7.1	7.9	14	18	19	23	20	–	13	12
$(La/Lu)_N$	7.9	9.2	4.5	32	18	8.5	7.1	7.9	10.6	7.4	7.1	10.4	6.9
K/Rb	11	27	42	139	122	158	170	182	182	233	264	283	305
Nb/Ta	0.9	0.6	1.0	2.9	4.3	6.9	4.0	5.0	13	11	–	13	11

*Average obtained from 20 data of a felsic (73.3–77.5 wt% SiO_2) I type granite (La Pedriza).

Actlabs by Fusion Specific Ion Electrode-ISE. In addition, 131 chemical analyses from the literature were compiled, including the major, minor and trace elements of bulk composition of aplite-pegmatite bodies and Li-rich leucogranitic cupolas in the CIZ.

In contrast to pegmatites, chemical data belonging to granites is abundant. Including our own data (over 500, published and unpublished) and others compiled from the literature (over 230), we have more than 700 analyses of granitic rocks from the CIZ, belonging to the four S-types (S_1, S_2, S_3 and S_4) and the one I-type granitic series. A summary of averaged chemical values of Li-rich aplite-pegmatites, granites and country wall-rocks is given in Table 2. We have considered it convenient not to include data belonging to the S_4 granitic suite in the plots of Figs. 4–6 and 9, and in the Supplementary Figs. 2 and 3 because this data significantly overlaps the compositional fields of the S_3 and/or I granitic types, which would mask the relationships among the other granitic suites and the Li-mineralisation. As it will be discussed in the text, S_3, S_4 and I suites do not show petrogenetical relationships to the studied Li-rich rocks.

Over 300 microprobe analyses of micas, 150 of lithium aluminosilicates, 80 of amblygonite-montebrasite and 50 of Li-bearing Fe-Mn-phosphates were obtained. In addition, 150 mica crystals were analysed for trace elements, including Li, by laser ablation (LA-ICP-MS). The technical details of these analyses are given in the Suplementary Text 1, and the data in the Suplementary Tables 1, 2, 3 and 4, and in the Suplementary Fig. 1.

4. General geology of the granitic rocks

Many granite-types have been defined in the CIZ according to their age of emplacement and chemical features (e.g., Capdevila et al., 1973; Ferreira et al., 1987), but here we use the nomenclature and synthesis of the five granite series of Villaseca (2011).

4.1. Two-mica leucogranite suite (S_1)

Two characteristics define the S_1 granite suite. First, it is the most felsic Variscan granitoid association, mostly comprising leucogranites (~55%) dominant over monzogranites (~45%), lacking granodiorites or more mafic sequences, and also characterised by the absence of mafic microgranular enclaves. Second, the S_1 suite comprises elongated batholiths that have been mostly emplaced synkinematically during D3 times (320 ± 6 Ma, in average) and are scarcer afterwards (e.g., Capdevila et al., 1973; Gomes et al., 2014; López-Moro et al., 2012; Teixeira et al., 2012). These batholiths represent the first important Variscan magmatic event in the CIZ, consequently, they were termed old Variscan granites (e.g., Oen, 1970). These granitic batholiths appear mostly confined to the nothwesternmost part of the CIZ (Fig. 1), being the southern branch of a large two-mica leucogranite belt in Western

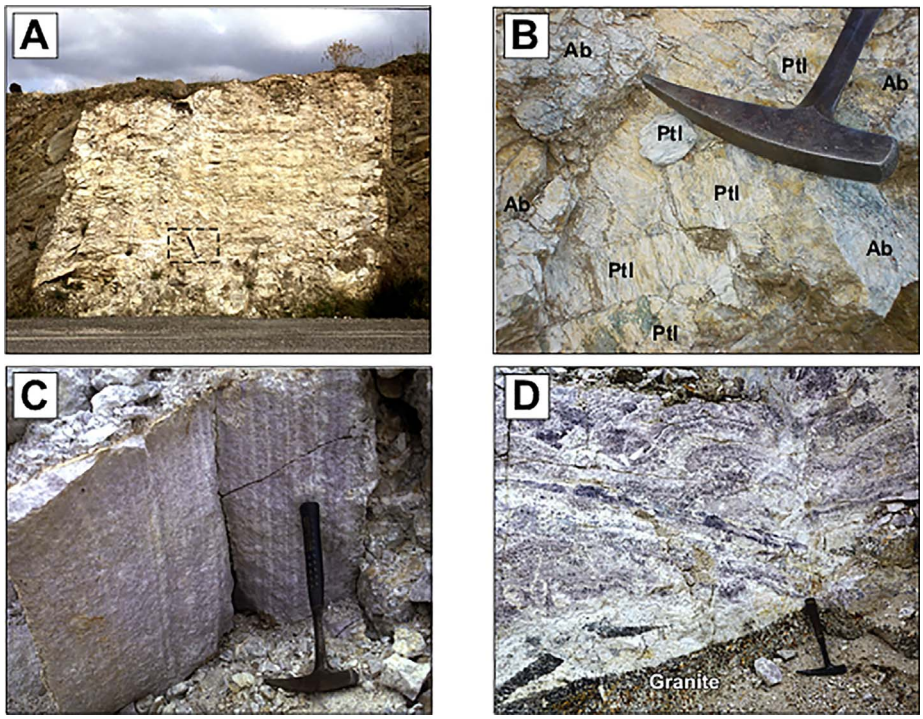

Fig. 2. (a) Sub-vertical homogenous simple aplite-pegmatite dyke (type 3), discordant to the hosting micaschists, from the Fregeneda-Almendra field (see hammer for scale); (b) mass of cone-shaped crystals of petalite randomly distributed together with albite crystals (type 2) from the Alberto open pit in the Fregeneda-Almendra field; (c) simple rhythmic layering with alternating lepidolite-rich and albite-rich bands in a sub-vertical aplite-pegmatite dyke (type 1) from the Feli open pit (Fregeneda-Almendra field); (d) complex rhythmic layering with lepidolite-rich, albite-rich and quartz-rich layers, alternating with different textures, from a subhorizontal dyke (type 1) hosted by a granite (Gonçalo field). (Abbreviations as in Table 1).

Europe that extends towards the Armorican and French Massif Central in France (see discussion below). Examples of well studied batholiths of this suite are: La Espenuca in Galicia (Ortega, 1998), Cabeceiras de Basto in northern Portugal (Almeida, 1994; Almeida et al., 1998) and Domo del Tormes leucogranites, in central Spain (López-Moro et al., 2012; López-Plaza and López-Moro, 2003).

This granitic suite has chemical similarities to those of the Himalayan leucogranites but they show marked higher P contents, in a wide range, the lowest contents being found in the northernmost La Espenuca massif, in Galicia. Some leucogranites of this suite show high $(La/Lu)_N$ ratios indicating a mid-to-lower crustal generation (e.g., Ortega and Gil Ibarguchi, 1990). They have been interpreted as melt fractions of pure crustal derivation (e.g., Cuesta and Gallastegui, 2004; Ortega, 1998), either from metasedimentary (Almeida et al., 1998) or peraluminous metaigneous sources (Dias et al., 2002; Ortega, 1998), or a combination of both (López-Moro et al., 2012).

4.2. P-rich highly peraluminous granite suite (S_2)

This second group has been referred to as mixed granitoids in the literature (e.g., Corretgé et al., 1977, 2004) for its intermediate character between anatectic two-mica leucogranites (crustal melts) and other less peraluminous granite series (locally hybridised with basic rocks), but all the Variscan granite suites of the CIZ geochemically overlap between them. This group shows a wider range of petrographic rocks than the S_1 suite, from biotite-tonalite (~1%), biotite-granodiorite (~5%), to biotite ± muscovite ± cordierite ± andalusite/sillimanite monzogranite (~80%), and two-mica ± cordierite ± andalusite/sillimanite leucogranite (~14%).

Most batholiths of this type are confined to the S-CIZ domain (Fig. 1), where the Neoproterozoic-Early Cambrian metasedimentary SGC is the epizonal wall rock. S_2 granitoids usually form inversely zoned plutonic complexes: Castelo Branco in central Portugal (Antunes et al., 2008); Jálama (Pesquera et al., 2018); Cabeza de Araya (Corretgé et al., 1985); Nisa-Alburquerque (Ramírez and Menéndez, 1999; Solá et al., 2009); and Belvís de Monroy (Merino et al., 2013). Most plutonic units have only restite or xenolith enclave types, being tonalitic microgranular enclaves circumscribed to areas close to the scarce tonalite bodies. This granite suite is emplaced in more epizonal levels (at conditions of 2–3 kbar: González-Menéndez et al., 2011; Merino Martínez et al., 2014) and is younger in age (mostly from 310 to 300 Ma) than S_1 granites (e.g., Antunes et al., 2008; Chicharro et al., 2014; Gutiérrez-Alonso et al., 2011; Merino Martínez et al., 2014).

A remarkable feature of this granitic group is the low CaO and high P_2O_5 content (Table 2), defining a marked perphosphorous trend. Moreover, it shows a notable coincidence in isotopic signatures (Sr, Nd, O, Hf) with the surrounding pelitic and greywackic sequences of the SGC (Antunes et al., 2008; Chicharro et al., 2014; Merino Martínez et al., 2014; Ramírez and Menéndez, 1999).

4.3. P-poor moderately peraluminous granite suite (S_3)

This suite also comprises a wide range of petrographic varieties, from tonalite to two-mica leucogranite, in similar proportions than in S_2 batholiths, but its lower peraluminosity degree leads to the absence of Al_2SiO_5 polymorphs in most S_3 granites. Moreover, minor basic massifs are coevally emplaced with them, giving rise to the common presence of mafic microgranular enclaves in many granite units. These granitoids outcrop dispersedly from northern Portugal to central Spain, being dominant in the Spanish Central System and the easternmost part of the Montes de Toledo batholith (Fig. 1). They are typical epizonal granite complexes giving rise to thermal aureoles, mostly late- to post-D3, from 308 to 299 Ma (e.g., Díaz Alvarado et al., 2013; Gutiérrez-Alonso et al., 2011; Merino Martínez et al., 2014; Orejana et al., 2012).

These granites follow large crystal fractionation processes, giving rise to leucogranites with typical REE normalised patterns of flat fractionation and large Eu negative anomalies (e.g., Sierra de Guadarrama batholith, Villaseca et al., 1998b). Their moderate peraluminosity, combined with similar isotopic (Sr, Nd, Pb, O, Hf) ratios to metaigneous lower crustal granulite xenoliths of central Spain, suggest that their crustal origin is mainly derived from felsic to intermediate metaigneous sources (e.g., Villaseca et al., 2012), in contrast with S_1 and S_2 granite suites of dominant metasedimentary derivation.

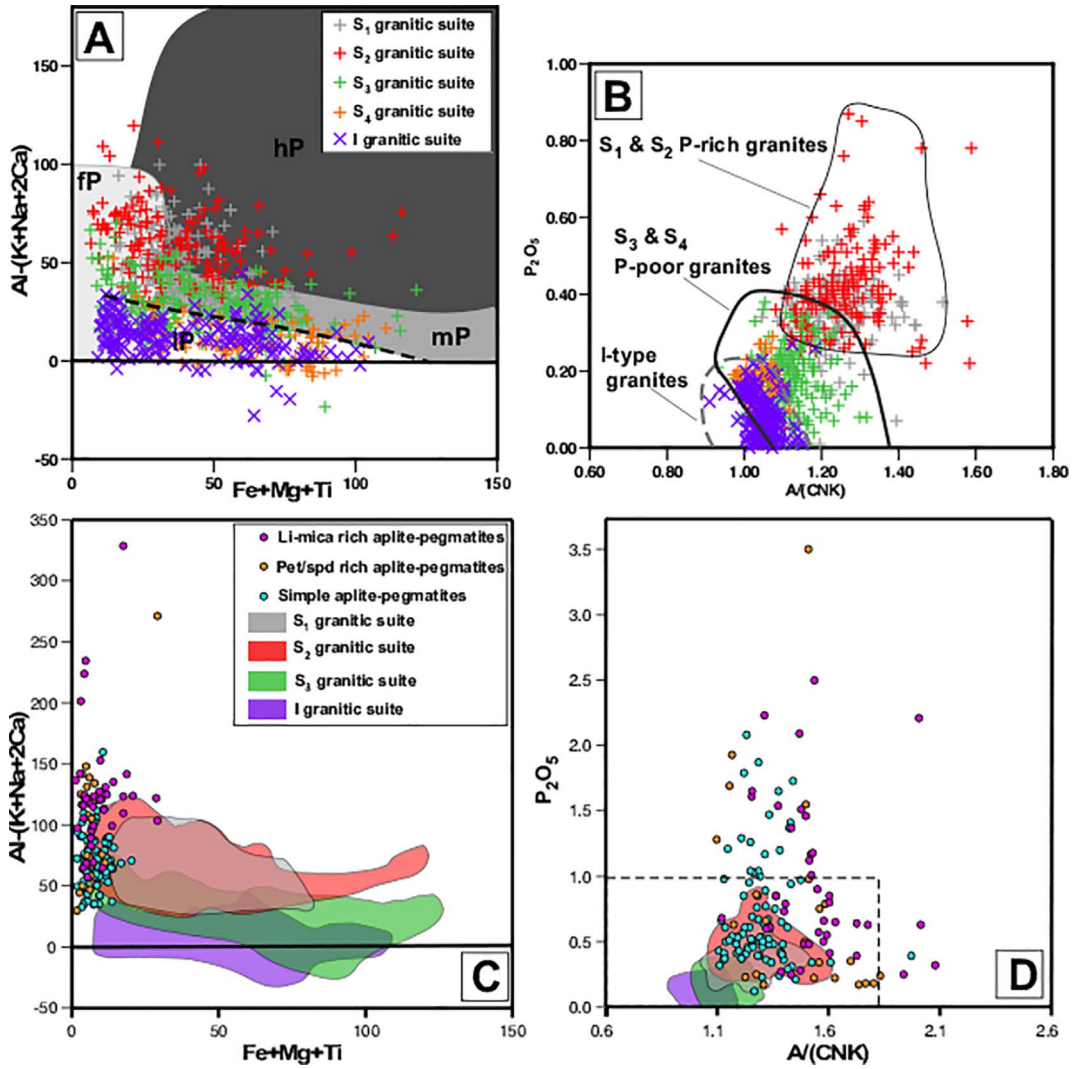

Fig. 3. (a) Composition of granites from the S_1, S_2, S_3, S_4 and I series in the A–B diagram of Debon and Le Fort (1983), where A = Al-(K + Na + 2Ca) and B = Fe + Mg + Ti, in mmol values, modified by Villaseca et al. (1998a), (hP = high peraluminous; mP = medium peraluminous; lP = low peraluminous; fP = felsic peraluminous); (b) A/CNK (molar Al_2O_3/(CaO + NaO + K_2O)) *vs* P_2O_5 (in wt.%) of the CIZ granitic rocks. The three discriminant fields are modified from Villaseca et al. (2008); (c) Composition of the three types of aplite-pegmatites and the leucogranitic cupolas (these included in *Li-mica rich* and *simple aplite-pegmatites* data), together with the compositional fields of the granites from the S_1, S_2, S_3 and I series in the A–B diagram; and (d) A/CNK (molar Al_2O_3/(CaO + NaO + K_2O)) *vs* P_2O_5 (in wt.%) for the three types of aplite-pegmatites and the leucogranitic cupolas. The dashed rectangle represents the limits of the diagram in the Fig. 3b for a reference. For data references see Table 2.

4.4. Moderately to low peraluminous granite suite (S_4)

This suite plots within the boundary between S- and I-type granites, comprising biotite-bearing granitoids without minerals of either peraluminous or metaluminous indication. Nevertheless, the presence of Ca-rich accessory minerals such as allanite or titanite, together with the evolution of biotite chemistries crossing the calc-alkaline to alumino-potassic fields defined by Nachit et al. (1985), seem to be diagnostic features of this granite suite. They are coeval to some minor basic intrusions, thus carrying a varied population of mafic microgranular enclaves.

S_4 granitoids appear from Galicia and northern Portugal to central Spain, comprising plutonic complexes of a wide age range, from early intrusions (*syn*-D3) (Vigo, Sameiro, Felgueiras, Lamego: Dias et al., 2000; Gallastegui, 2005), to late-to-postectonic plutons (Braga, Vila Pouca de Aguiar, Cota-Viseu, Navalosa: Dias et al., 1998, 2000; Herreros, 1998; Martins et al., 2009; Valle Aguado et al., 2005) (Fig. 1). Thus, they define a wide range of emplacement ages from 319–313 (*syn*-D3 intrusions) to 311–299 Ma (late to postkynematic intrusions).

S_4 monzogranites and subordinate granodiorites have been predominantly explained as crustal melts derived from metasedimentary (metagreywackes) and felsic metaigneous sources, mostly from lower crustal levels, and locally hybrised with mantle-derived basic melts (e.g., Dias et al., 2000; Martins et al., 2009).

4.5. Low peraluminous I-type granite suite (I)

This group comprises metaluminous to low peraluminous granites, with peraluminosity increasing with granite differentiation. This suite mostly consists of amphibole-bearing biotite-granodiorite (< 10%), biotite-monzogranite (65%), to biotite-leucogranite (25%). Some granite units show mafic microgranular enclaves.

The suite appears in the CIZ as a dispersed plutonic array outcropping in three main areas: Galicia-N Portugal (e.g., Caldas de Reis, Porriño, Gerês plutons); central Spain (Spanish Central System) (e.g., Villacastín, La Cabrera, La Pedriza plutons); and close to the boundary with the Ossa-Morena Zone (Villar del Rey and Los Pedroches plutons) (Fig. 1) (Villaseca et al., 2009). They intruded over a long period (mostly from 308 to 295 Ma) but some of these intrusions are the youngest of the Variscan CIZ magmatism (e.g., Dias et al., 1998; Fernández-Suárez et al., 2000; Gutiérrez-Alonso et al., 2011; Orejana et al., 2012).

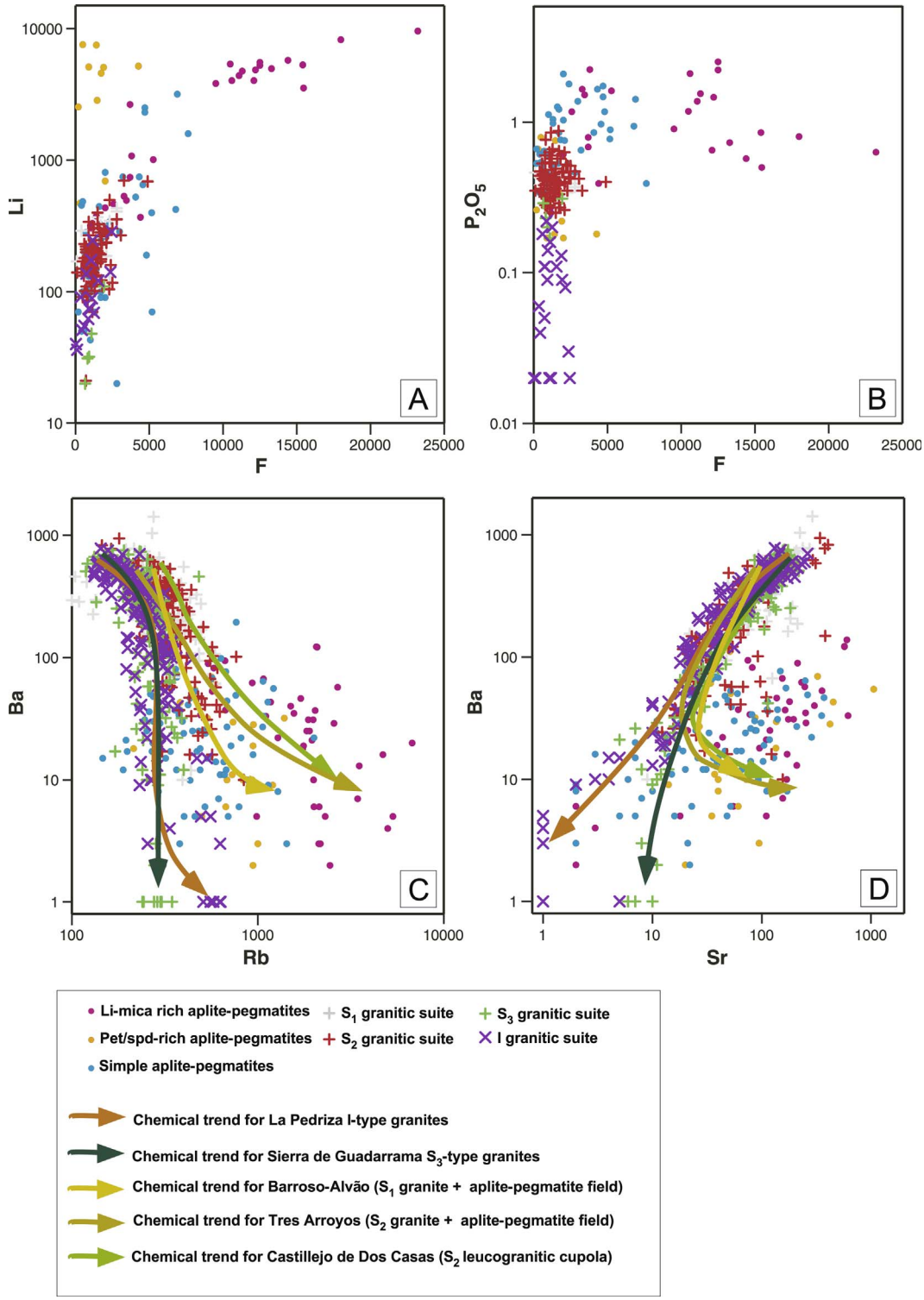

Fig. 4. Selected trace element (ppm) log–log variation diagrams for the CIZ granites, aplite-pegmatites and leucogranitic cupolas (these included in *Li-mica rich* and *simple aplite-pegmatites* data), (a) F *vs* Li; (b) F *vs* P_2O_5 (wt.%); (c) Rb *vs* Ba; and (d) Sr *vs* Ba. In c) and d) chemical trends for specific localities using crystal fractionation geochemical modelling, are shown, La Pedriza I-type pluton (Pérez-Soba and Villaseca, 2010); Sierra de Guadarrama S_3-type granites (Villaseca et al., 1998b); Barroso-Alvão S_1 granite + aplite-pegmatite field (Ortega et al., 2017); Tres Arroyos S_2 granite + aplite-pegmatite field (Garate-Olave et al., 2017); and, Castillejo de Dos Casas S_2 leucogranitic cupola (Roda-Robles et al., 2015). For data references see Table 2.

Most CIZ I-type granites define a felsic low peraluminous suite with typical fractionation trends towards high Th, Nb, Y-HREE contents giving rise to rare-metal-rich leucogranites (e.g., Pérez-Soba and Villaseca, 2010; Villaseca et al., 1998b). They have been interpreted as crustal derived magmas from deep *meta*-igneous sources, being common the presence of inherited orthogneissic Cambro-Ordovician zircons (e.g., Orejana et al., 2012).

5. General geology of the Li-(F-P)-rich rocks

Different rock types enriched in Li-(F-P) are distinguished in the CIZ, including aplite-pegmatite fields, leucogranitic cupolas, beryl-

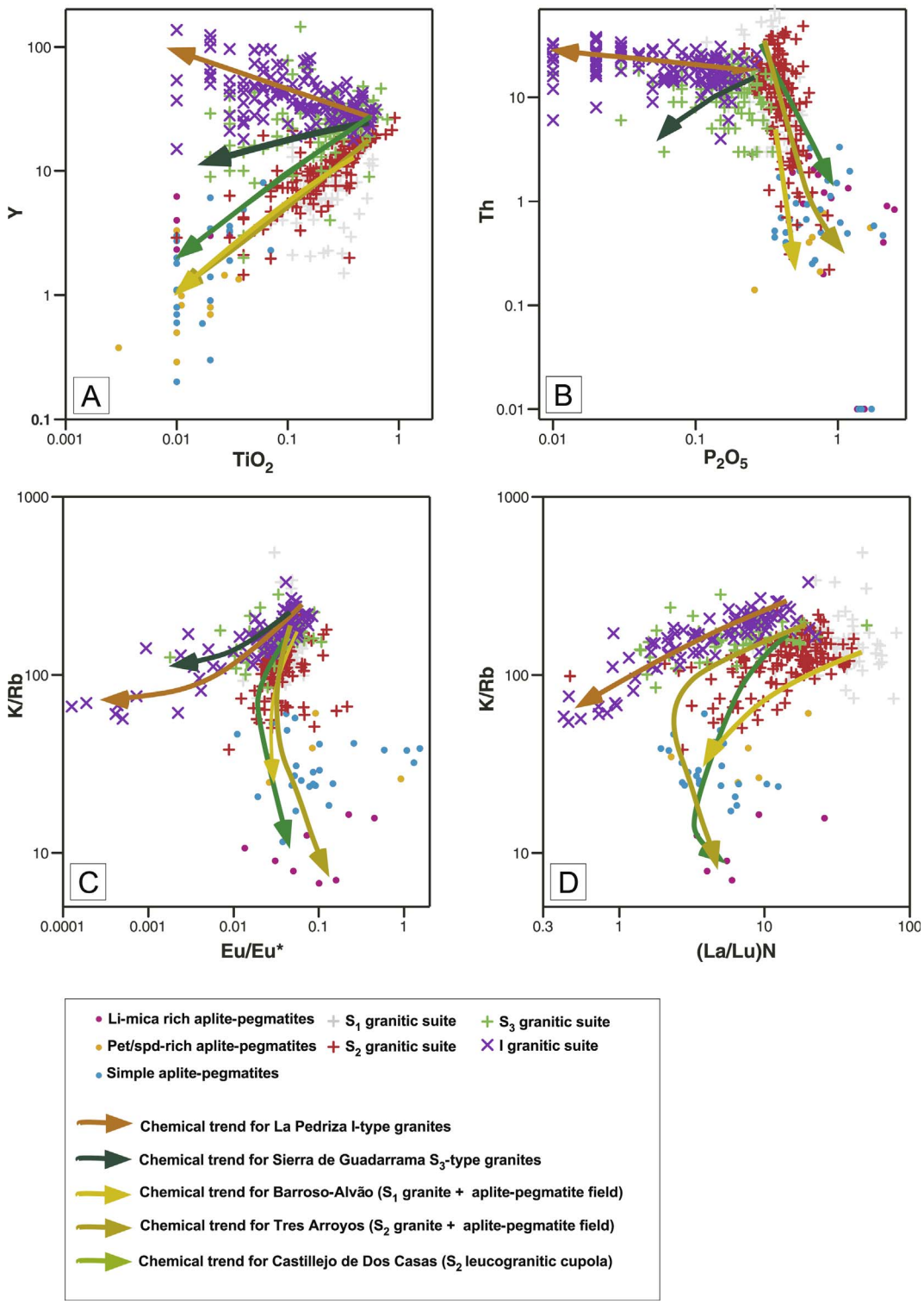

Fig. 5. Log-log plots of (a) TiO_2 (wt.%) *vs* Y (ppm); (b) P_2O_5 (wt.%) *vs* Th (ppm); (c) Eu/Eu* *vs* the K/Rb ratio; and (d) $(La/Lu)_N$ *vs* the K/Rb ratio for the CIZ granites, aplite-pegmatites and leucogranitic cupolas (these included in *Li-mica rich* and *simple aplite-pegmatites* data), showing chemical trends calculated by using crystal fractionation modelling for specific localities, La Pedriza I-type pluton (Pérez-Soba and Villaseca, 2010); Sierra de Guadarrama S_3-type granites (Villaseca et al., 1998b); Barroso-Alvão S_1 granite + aplite-pegmatite field (Ortega et al., 2017); Tres Arroyos S_2 granite + aplite-pegmatite field (Garate-Olave et al., 2017); and, Castillejo de Dos Casas S_2 leucogranitic cupola (Roda-Robles et al., 2015). For data references see Table 2. Symbols as in Fig. 4.

phosphate pegmatites and quartz-montebrasite-rich dykes (Roda-Robles et al., 2016). In this paper we will just focus on the first and second types, which are the most widespread and present intriguing mineralogical, textural and chemical features that are not commonly observed in other orogenic regions. For more detail on the beryl-phosphate pegmatites and the quartz-amblygonite-rich dykes occurring in the CIZ see Roda-Robles et al. (2016).

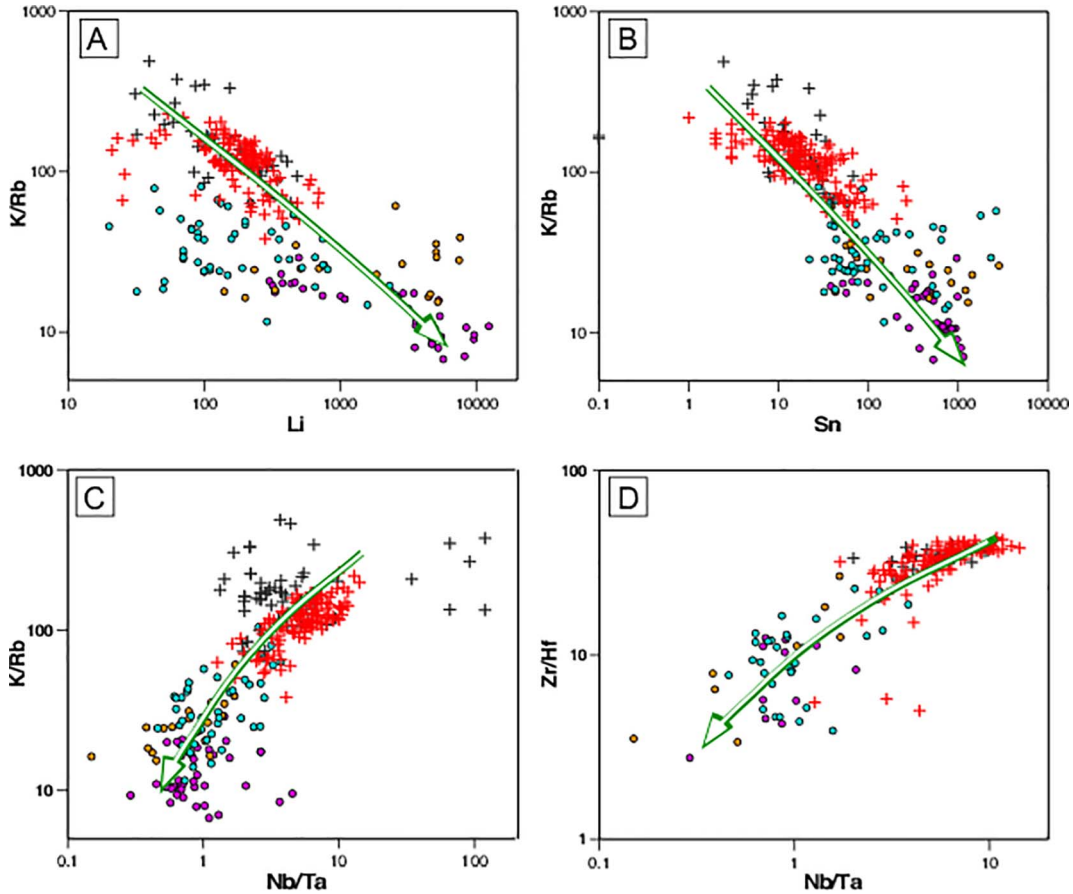

Fig. 6. Log-Log plot of the K/Rb ratio *vs* the trace elements (ppm) (a) Li; (b) Sn; and plot of the ratio Nb/Ta *vs* the ratios (c) K/Rb; and (d) Zr/Hf, for the S_1 and S_2 granitic series, the aplite-pegmatites and the leucogranitic cupolas (these included in *Li-mica rich* and *simple aplite-pegmatites* data) from the CIZ, showing their chemical trends. For data references see Table 2. Symbols as in Fig. 4. Green arrow, chemical trend for S_1, S_2 and the aplite-pegmatites.

5.1. Aplite-pegmatite fields

Lithium-rich aplite-pegmatites occurring in fields of up to a few hundred dykes are widespread along the CIZ and in the south of the GTMZ (Fig. 1, Table 1). These dyke-like bodies intrude with variable dips (from subhorizontal to subvertical, Fig. 2a) into both granitic and metasedimentary rocks. Their grain size may be very homogeneous (either aplitic or pegmatitic) or clearly heterogeneous (aplitic and pegmatitic facies coexisting in the same body). In all cases crystals with lengths over 12 cm are quite exceptional. These big crystals, usually feldspars and less frequently, petalite and spodumene, are often wedge-shaped (Fig. 2b), frequently growing perpendicularly to the contacts with the host-rocks. Another relatively common textural feature of the aplite-pegmatites from the CIZ is the development of a more or less complex layering parallel to the contacts with the host-rocks throughout the pegmatite (e.g. Fregeneda-Almendra, Gonçalo, Las Navas, and Tres Arroyos fields) (Fig. 2c, d). It is remarkable that other textural features commonly observed in granitic pegmatites, such as the graphic intergrowths, the development of a quartz core, or an internal zoning, are very rarely observed inside these bodies (Fig. 2a). In some fields the aplite-pegmatites show a regional zoning characterised by an enrichment of rare-metals with increasing distance from the parental granite (e.g., Fregeneda-Almendra and Tres Arroyos fields) (Gallego Garrido, 1992; London et al., 1999; Roda-Robles et al., 2016).

Quartz, albite and K-feldspar are the predominant minerals in most of the dykes of the aplite-pegmatite fields; whereas spodumene, petalite, Li-muscovite and/or lepidolite, may be abundant in the Li-richest dykes. The Li-F-richest bodies are rich in Li-micas (up to 35% mica in volume). In the remaining dykes, micas (mainly muscovite) are usually scarce comprising less than 5 vol%. Chemical composition of micas reflects the progressive enrichment in Li and F, showing continuous trends for specific aplite-pegmatite fields from granites, through the barren aplite-pegmatites up to the most differentiated dykes, which are enriched in lepidolite (e.g., Fregeneda-Almendra, Barroso-Alvão, and Tres Arroyos) (Supplementary Fig. 1 and Table 1).

Petalite and spodumene only occur in some of the dykes of the aplite-pegmatite fields (Table 1). The two phases may coexist in the same dyke, but more often only one of the two lithium aluminosilicates is present. Locally either petalite or spodumene may be totally or partially replaced by sub-vitreous, greasy aggregates of late eucryptite and/or cookeite. The concentration of these lithium aluminosilicates may be very high in some zones of the dykes, constituting up to ~50 vol% of spodumene and/or petalite. The chemical composition of spodumene, petalite and eucryptite is given in Supplementary Table 2.

The most common accessory mineral phase in all the aplite-pegmatites is montebrasite, which moreover is the only Li-bearing phase in some of the less evolved bodies of the aplite-pegmatite fields. Although less abundant, Fe-Mn ± Li phosphates are also common accessory phases in the aplite-pegmatites, mainly in the dykes with intermediate degrees of fractionation (e.g., Fregeneda-Almendra, Barroso-Alvão, Tres Arroyos, Lalín-Forcarei). Members of the Li-rich triphylite-lithiophilite series are usually replaced by ferrisicklerite-sicklerite and heterosite-purpurite. As micas, phosphates also reflect the fractionation degree of their hosting pegmatite. In the case of montebrasite, the F/OH ratio increases towards amblygonite in the most fractionated bodies (Supplementary Table 3), whereas in the Fe-Mn-phosphates there is a decrease in the Fe/(Fe + Mn) ratio parallel to the increase in Li and F of the hosting aplite-pegmatite, with sicklerite crystals observed in the

Table 3
Whole-rock major (wt.%) and trace-element (ppm) representative compositions of the three different aplite-pegmatite facies from the aplite-pegmatite fields and from the leucogranitic cupolas from the CIZ. *Locality (numbers as in Fig. 1 and Table 1): F-A = Fregeneda-Almendra; TA = Tres Arroyos; CDC = Castillejo de Dos Casas; PF = Pinilla de Fermoselle; BA = Barroso-Alvao; JAL = Jálama. **Rock: AP fields = aplite-pegmatite fields; Lg cupola = Leucogranitic cupola. ***Facies: Type 1 = Li-mica rich; type 2 = spodumene and/or petalite-rich; type 3 = simple facies. n.d. = not determined. For information about analytical methods see text.

Locality*	F-A (1)	TA (3)	TA (3)	TA (3)	CDC(16)	CDC(16)	PF (19)	F-A (1)	BA (2)	BA (2)	BA (2)	TA (3)	TA (3)	TA (3)	TA (3)	TA (3)	JAL	JAL	JAL	CDC(16)
Rock**	AP fields	AP fields	AP fields	AP fields	Lg cupola	Lg cupola	Lg cupola	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	Lg cupola
Facies***	Type 1							Type 2				Type 3								
SiO_2	70.14	68.88	71.13	67.17	61.85	70.86	61.29	73.31	72.07	68.82	72.36	68.86	68.02	69.15	70.64	70.50	73.67	73.90	74.18	74.77
Al_2O_3	16.22	17.54	16.64	18.50	23.75	17.90	23.59	16.72	15.96	18.79	16.05	16.50	19.56	18.52	16.29	18.48	15.06	15.10	14.73	14.27
Fe_2O_3(T)	0.25	0.70	0.23	0.06	0.31	0.29	0.25	0.54	0.74	1.03	0.41	0.30	0.79	0.93	0.41	0.34	0.80	0.68	2.64	0.97
MnO	0.07	0.07	0.08	0.09	0.15	0.12	0.19	0.04	0.08	0.07	0.06	0.02	0.02	0.02	0.03	0.02	0.05	0.03	0.03	0.06
MgO	0.73	0.01	0.02	0.02	0.03	0.05	0.04	0.13	0.03	0.65	0.04	0.05	0.05	0.03	0.03	0.04	0.10	0.04	0.18	0.05
CaO	1.71	0.17	0.35	0.31	0.36	0.24	0.09	0.99	0.27	0.12	0.45	0.64	0.35	0.24	0.39	0.35	0.37	0.28	0.54	0.40
Na_2O	5.51	5.50	4.27	4.04	4.25	5.12	3.07	4.66	3.75	3.76	6.17	5.24	9.31	8.91	5.85	9.14	4.59	4.25	3.45	5.00
K_2O	2.12	1.90	2.80	4.00	3.83	2.63	6.43	1.00	5.03	3.23	2.49	3.34	0.75	0.66	2.79	0.55	3.04	3.98	4.28	3.15
TiO_2	0.01	0.01	0.01	0.01	0.01	0.01	0.01	0.02	0.02	0.01	0.01	0.01	0.00	0.00	0.01	0.00	0.02	0.01	0.12	0.03
P_2O_5	0.68	2.50	0.57	0.80	0.63	1.18	0.25	0.79	0.66	0.26	0.63	2.08	0.54	0.53	1.79	0.61	0.64	0.76	0.54	0.51
F	0.37	1.25	1.44	1.80	2.32	1.05	n.d.	0.05	0.03	0.02	< 0.01	0.20	0.05	0.02	0.24	0.04	0.07	0.18	0.23	0.06
LOI	2.90	2.11	2.60	3.88	3.25	1.99	3.44	0.96	1.08	5.15	1.02	2.02	1.18	1.10	0.92	0.82	0.84	0.30	1.29	0.54
Total	100.71	100.64	100.14	100.67	100.75	101.44	98.65	99.20	99.72	101.91	99.69	99.26	100.62	100.11	99.39	100.89	99.25	99.51	102.21	99.81
B	54	n.d.	n.d.	n.d.	107	63	n.d.	13	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	127	400	14	1010
Li	2660	5260	5730	8230	9600	5400	8450	7570	470	2550	510	810	100	70	290	50	281	258	450	200
Be	203	10	11	7	36	66	n.d.	135	136	213	198	5	7	6	7	8	n.d.	n.d.	12	7
Sc	< 1	< 1	< 1	< 1	< 1	< 1	n.d.	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	2.85	0.8	4	3
V	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	7.02	< 5	5	< 5
Cr	40	< 20	< 20	< 20	40	60	30	130	30	30	< 20	< 20	< 20	< 20	90	< 20	n.d.	n.d.	n.d.	110
Co	< 1	< 1	32	33	< 1	7	< 1	< 1	< 1	4	< 1	26	< 1	< 1	< 1	< 1	1.07	< 0.5	1	< 1
Ni	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	< 20	n.d.	n.d.	n.d.	< 20
Cu	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	< 10	n.d.	n.d.	n.d.	< 10
Zn	110	140	150	80	70	80	300	60	70	60	40	< 30	< 30	< 30	< 30	< 30	n.d.	n.d.	80	90
Ga	38	50	50	65	40	32	70	20	26	16	27	45	51	41	45	44	24.5	23.2	24	23
Ge	9.9	7.4	9	10	29.7	11.9	13.1	4.7	5.1	2.3	9.7	5	4.3	5.4	7.4	4.6	n.d.	n.d.	n.d.	4.9
As	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	14	< 5	< 5	< 5	< 5	< 5	< 5	n.d.	n.d.	n.d.	9
Rb	1070	1990	3450	4710	3520	1740	5000	214	1200	440	832	1130	260	188	2000	247	556	681	534	534
Sr	154	169	156	171	210	95	3	135	14	52	72	113	114	29	285	139	35	48.8	87	36
Y	0.2	0.2	0.2	0.2	2	6.2	0.5	0.8	0.7	2	0.5	0.8	0.9	0.6	0.2	0.5	6.1	1.8	9	3.3
Zr	20	28	24	36	115	35	112	8	20	15	19	19	107	11	13	53	36	19	55	16
Nb	56	80.7	71.5	93	67	63	84.9	19	37	44	76	48	186	21	40	207	17.1	16.4	15	11
Mo	2	< 2	< 2	< 2	< 2	3	3	< 2	< 2	< 2	< 2	< 2	< 2	< 2	5	< 2	n.d.	n.d.	n.d.	< 2
Ag	0.5	< 0.5	< 0.5	< 0.5	< 0.5	0.9	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	n.d.	n.d.	n.d.	< 0.5
In	1.2	1.2	1.7	3.6	3.7	0.8	0.2	0.3	0.2	0.1	0.3	1.1	0.1	0.2	0.6	0.2	n.d.	n.d.	n.d.	0.2
Sn	350	370	530	1150	975	208	284	71	57	39	95	311	40	42	150	43	39	53	45	46
Sb	< 0.2	1.1	0.6	0.4	< 0.2	0.8	0.8	< 0.2	0.3	0.3	0.3	0.5	0.4	0.4	< 0.2	0.4	n.d.	n.d.	n.d.	< 0.2
Cs	236	136	341	260	722	272	877	27	57	58	73	24	9.4	6.3	62	14.5	51	70.1	49	48
Ba	67	10	7	16	13	18	4	18	9	50	10	43	36	12	22	31	66	44.2	67	15
La	0.44	0.31	0.19	0.23	0.69	2.14	0.22	0.67	0.46	5.42	0.38	0.24	0.63	0.27	0.4	0.3	3.13	0.6	8.04	2.53
Ce	0.78	0.36	0.24	0.26	0.96	4.92	0.45	1.24	0.77	6.75	0.61	0.34	0.89	0.37	0.7	0.4	6.08	1.2	18.7	2.62
Pr	0.11	0.04	0.02	0.03	0.11	0.73	0.05	0.13	0.09	1.44	0.08	0.05	0.12	0.06	0.08	0.05	1	0.13	2.29	0.53
Nd	0.39	0.17	0.1	0.1	0.47	3.29	0.18	0.5	0.36	5.73	0.36	0.3	0.63	0.2	0.23	0.25	3.04	0.4	8.76	1.97
Sm	0.03	0.07	0.03	0.02	0.13	1.04	0.11	0.11	0.09	1.24	0.13	0.09	0.16	0.08	0.04	0.06	0.85	0.13	2.2	0.43
Eu	0.038	0.011	0.01	0.015	0.015	0.264	0.007	0.033	0.009	0.328	0.012	0.05	0.051	0.03	0.005	0.03	0.07	0.025	0.213	0.057
Gd	0.08	0.06	0.03	0.04	0.17	1.19	0.22	0.13	0.12	0.86	0.15	0.12	0.18	0.1	0.04	0.08	1.01	0.13	1.53	0.37
Tb	< 0.01	< 0.01	< 0.01	< 0.01	0.05	0.2	0.06	0.02	0.02	0.11	0.02	0.02	0.03	0.02	< 0.01	0.01	0.1	0.04	0.3	0.08
Dy	0.05	0.05	0.04	0.03	0.23	1.2	0.14	0.14	0.11	0.55	0.08	0.13	0.15	0.09	0.04	0.08	0.93	0.25	1.74	0.47
Ho	< 0.01	< 0.01	< 0.01	< 0.01	0.04	0.23	0.01	0.03	0.02	0.09	0.01	0.02	0.03	0.01	< 0.01	0.01	0.17	0.04	0.27	0.09
Er	0.04	0.03	0.02	0.02	0.1	0.5	0.02	0.08	0.06	0.23	0.04	0.06	0.08	0.04	0.03	0.03	0.24	0.17	0.73	0.28

(continued on next page)

Table 3 (*continued*)

Locality*	F-A (1)	TA (3)	TA (3)	TA (3)	CDC(16)	CDC(16)	PF (19)	F-A (1)	BA (2)	BA (2)	BA (2)	TA (3)	TA (3)	TA (3)	TA (3)	TA (3)	JAL	JAL	JAL	CDC(16)
Rock**	AP fields	AP fields	AP fields	AP fields	Lg cupola	Lg cupola	Lg cupola	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	AP fields	Lg cupola
Facies***	Type 1							Type 2				Type 3								
Tm	0.005	0.007	< 0.005	< 0.005	0.023	0.074	< 0.005	0.009	0.011	0.036	0.006	0.011	0.013	0.01	< 0.005	0.006	0.07	0.03	0.105	0.052
Yb	0.03	0.06	0.01	0.03	0.15	0.51	0.03	0.06	0.1	0.22	0.04	0.06	0.09	0.06	< 0.01	0.04	0.44	0.28	0.74	0.36
Lu	0.005	0.008	< 0.002	0.004	0.013	0.065	0.005	0.009	0.021	0.028	0.006	0.007	0.013	0.008	< 0.002	0.005	0.06	0.04	0.105	0.053
Hf	1.8	2.7	2.2	3.2	9.3	2.9	13.5	0.3	1.1	1.2	2.4	2.2	13.3	1	1.1	7.6	1.61	1.4	2	0.7
Ta	82	91	64.8	72	95	70	41.3	11	26	25	200	52	194	26	55	269	6.3	5.8	4.25	5.61
W	5.8	34.6	301	249	45	29	13.1	< 0.5	2.6	0.7	1.8	123	8.5	2.3	6.2	9.7	7	4	6.6	4.3
Tl	6.5	8.06	15.3	20.5	21.2	8.4	27.6	1.2	7.3	2.8	5.0	6.5	0.9	0.73	10.1	1.05	n.d.	n.d.	2.83	3.4
Pb	30	6	< 5	8	8	21	7	9	19	< 5	16	14	31	14	12	35	16	6	15	12
Bi	0.5	< 0.1	0.1	< 0.1	< 0.1	< 0.1	0.1	0.2	< 0.1	< 0.1	< 0.1	0.8	0.2	< 0.1	0.2	< 0.1	n.d.	n.d.	n.d.	< 0.1
Th	2.03	0.83	0.95	1.21	2.71	1.34	2.84	0.77	0.45	0.14	0.4	0.47	1.96	0.59	0.58	0.93	3.25	0.5	6.45	0.96
U	8.6	29	7.72	11.7	8.1	4.5	3.79	2.3	16	5.11	11.5	7.6	5.8	4.78	12.9	7.07	n.d.	n.d.	8.48	2.6
A/(CNK)	1.12	1.54	1.56	1.60	2.01	1.53	1.94	1.59	1.32	1.89	1.17	1.23	1.17	1.17	1.22	1.14	1.31	1.28	1.30	1.15
K/Rb	16.4	7.9	6.7	7.1	9.0	12.5	10.7	38.8	34.8	60.9	24.8	24.5	23.9	29.1	11.6	18.5	45.4	48.5	66.5	49.0
Nb/Ta	0.7	0.9	1.1	1.3	0.7	0.9	2.1	1.7	1.4	1.7	0.4	0.9	1.0	0.8	0.7	0.8	2.7	2.8	3.6	2.0
Zr/Hf	11.1	10.4	10.9	11.3	12.4	12.1	8.3	26.7	18.2	12.5	7.9	8.6	8.0	11.0	11.8	7.0	22.2	13.6	27.5	22.9
$(La/Lu)_N$	9.13	4.02	–	5.97	5.51	3.42	4.57	7.73	2.27	20.09	6.57	3.56	5.03	3.50	–	6.44	5.41	1.56	7.95	4.95
Eu/Eu*	0.22	0.05	0.10	0.16	0.03	0.07	0.01	0.08	0.03	0.09	0.03	0.15	0.09	0.10	0.04	0.13	0.02	0.06	0.34	0.04
ΣLREE	1.64	0.91	0.56	0.61	2.25	11.39	0.96	2.52	1.68	19.14	1.48	0.97	2.31	0.92	1.37	1.00	13.10	2.33	41.73	7.55
ΣHREE	0.21	0.208	–	0.12	0.66	3.47	0.415	0.42	0.41	1.89	0.32	0.38	0.51	0.30	–	0.24	2.68	0.87	3.99	1.53

most fractionated Li-mica-rich dykes (e.g., Fregeneda-Almendra, Roda-Robles et al., 2010) (Supplementary Table 4). Other common minor and/or accessory minerals in the aplite-pegmatites are topaz, cassiterite and Nb-Ta oxides.

As the degree of enrichment in Li ± F in all the dykes belonging to the same field may change drastically, we distinguish three types: (Type 1) Li-mica bearing dykes that are Li-rich and the richest in F; (Type 2) Petalite and/or spodumene bearing dykes, which are also Li-rich but much poorer in F; and (Type 3) simple aplite-pegmatites which show low Li and F contents, usually with no Li-rich silicates and just accessory crystals of montebrasite. These three types of aplite-pegmatites are not equally represented along the CIZ (Table 1, Fig. 1): in the central parts of the CIZ, the three categories of aplite-pegmatites may be present (e.g., Fregeneda-Almendra), whereas in the south only the first and third group appear (e.g., Tres Arroyos). In the northern areas only the second and the third type are usually well represented (e.g., Barroso-Alvão, Lalín-Forcarei) (Table 1, Fig. 1).

5.2. Leucogranitic cupolas

Lithium-enriched leucogranitic cupolas are less abundant than the aplite-pegmatite fields and occur predominantly in the central part of the CIZ (Table 1, Fig. 1). In this case Li is enriched in the apical or marginal units of highly fractionated leucogranites that are typically fine grained and enriched in B, Li and F (Charoy and Noronha, 1996; Martín-Izard et al., 1992, Roda-Robles et al., 1998; Roda-Robles et al., 2012; Roda-Robles et al., 2015). The Li-rich cupolas may be very heterogeneous and show different mineral associations. Stockscheiders are observed at the top of some of the cupolas, where wedge-shaped feldspar crystals growing perpendicular to the contact with the hosting rocks (comb texture) are relatively common, as well as layered, subhorizontal units with fine-grained lepidolite-rich bands alternating rhythmically with feldspar-rich bands (e.g., Castillejo de Dos Casas: Roda-Robles et al., 2015). In other cases a complete gradual transition from granitic to highly evolved pegmatitic facies may be observed (e.g., Pinilla de Fermoselle: Roda-Robles et al., 2012). More homogeneous microgranitic cupolas enriched in Li, F and P, with a mineral association similar to that of the Li-rich aplite-pegmatites also occur in the CIZ (e.g., Argemela: Charoy and Noronha, 1996). In general, the contact between the granitic and the Li-enriched units is not well defined, with a gradual transition between them, which makes it difficult to establish the limit between both rock units (Roda-Robles et al., 2012; Roda-Robles et al., 2015).

Main minerals include quartz, albite, K-feldspar, muscovite and Li-mica. These micas exhibit quite similar compositions to those of the micas from the aplite-pegmatite fields, i.e., from muscovite in the granite and barren facies, through trilithionite, and up to polylithionite in the most fractionated units (Supplementary Fig. 1) (e.g. Castillejo de Dos Casas, Pinilla de Fermoselle) (Supplementary Table 1). Fe-Li-rich micas are also present in some leucogranite cupolas with compositions of biotite in the granitic facies, through zinnwaldite in the barren pegmatitic units, and polylithionite in the most fractionated ones occuring frequently at the top of the cupolas (Supplementary Fig. 1) (Roda-Robles et al., 2012).

Lithium-bearing phosphates are common accessory minerals in the leucogranitic cupolas. Fluorine-rich members of the amblygonite-montebrasite series (up to 7 wt% F) and Li-Fe-Mn-rich phosphates of the triphylite-lithiophilite series, (or their replacement products ferrisicklerite-sicklerite and heterosite-purpurite), are common in these cupolas. As in the aplite-pegmatites, there is a gradual decrease in the Fe/(Fe + Mn) ratio as the fractionation degree increases, with lithiophilite and sicklerite only associated with the Li-richest facies (e.g., Pinilla de Fermoselle, Castillejo de Dos Casas). Other common accessory minerals include topaz, cassiterite and Nb-Ta oxides.

6. Chemistry of the aplite-pegmatites and granites of the CIZ

The Li-rich pegmatites are peraluminous rocks (Tables 2 and 3, Fig. 3, and Supplementary Fig. 2a), with A/CNK values in the range 1.12–2.61 for the Li-mica-rich bodies (type 1); 1.10–1.89 for the spodumene and/or petalite bodies (type 2); and 1.11–1.97 for the simple aplite-pegmatite dykes (type 3) (Tables 2 and 3). The SiO_2 contents for the aplite-pegmatites are highly variable, from 54.79 to 84.25 wt% SiO_2, although most of the analyses are in the range 66.53 to 75.82 wt% (Tables 2 and 3, Supplementary Figs. 2 and 3). All the aplite-pegmatites are Na-rich, with the highest values belonging to the simple aplite-pegmatite bodies (up to 10.37 wt% Na_2O) (Supplementary Fig. 2d). The K contents are fairly low, the Li-mica bearing aplite-pegmatites showing the highest contents (< 8.85 wt% K_2O) (Table 2, Supplementary Fig. 2e). Phosphorous, Fe and Ca are minor elements in these rocks (Tables 2 and 3, Supplementary Fig. 2b, 3g, b, c), whereas the Mg, Ti and Mn contents are much lower (Tables 2 and 3; Supplementary Fig. 2f).

There are also important differences among the three types of aplite-pegmatites in the trace element contents. The Li-mica bearing dykes are the Li- and F-richest bodies (Tables 2 and 3, Fig. 4a, b). The spodumene- and/or petalite-bearing dykes are also Li-rich but poorer in F, whereas the simple aplite-pegmatites show low Li and F contents (Fig. 4a, b). The highest mean values for incompatible elements such as Rb, Nb, Cs, Be, Hf and Sn are found in the Li-mica bearing aplite-pegmatites; intermediate mean values in the spodumene and/or petalite bearing dykes; and the lowest contents in the simple aplite-pegmatites (Tables 2 and 3, Figs. 4a, b, c, 5, 6 and Supplementary Fig. 3a). Other elements usually behaving as compatible in granitic melts (such as Ba and Sr), show a more complex distribution with enrichment of both in some of the Li-mica richest aplite-pegmatites of type 1 when compared to the aplite-pegmatites from types 2 and 3 (Tables 2 and 3; Fig. 4c, d and Supplementary Fig. 3).

In contrast with the pegmatites, available chemical data of granite rocks is abundant. For simplification data of S_4 granites are not included in the Figs. 3c, d, 4, 5, 6, and 9 in order to not obscure the chemical trends of the S_1 to S_3, and I-type suites. The data of S_4 granites plot inbetween or overlap the compositional fields of the S_3 and/or I granitic types (Fig. 3a, b; Table 2). The S_4 granite suite is not related to the Li-mineralisation anyway.

The S_1, S_2, S_3 and S_4 suites correspond to granitic rocks with decreasing alumina saturation values (mean A/CNK values 1.23, 1.24, 1.11 and 1.04 respectively, Table 2). The A-B plot (Debon and Lefort, 1983), modified by Villaseca et al., 1998a), discriminates the highly peraluminous S_1 and S_2 granitic series from the less peraluminous S_3 and S_4 suites, and also from the slightly peraluminous I-type granites (Fig. 3a). It is remarkable that most of the data from the S_1 and S_2 series overlaps in this diagram, except for a few Al-richer samples of S_2, which overlap the more peraluminous compositional field of the CIZ aplite-pegmatites. In Harker diagrams S3 and I-type granites show a near overlap of decreasing Al_2O_3, Fe_2O_3, CaO, MgO (not shown), TiO_2, and P_2O_5 values, and slightly increasing Na_2O and K_2O contents (Supplementary Fig. 2). In contrast, S_1 and S_2 series, which overlap widely in most of the chemical plots, show narrow compositional ranges, with high P_2O_5 contents for many of the samples, defining a perphosphorous character for most of the S_1 and S_2 granites (Table 2, Fig. 3b, d, and Supplementary Fig. 2g). S_2 granites define better than S_1 this P enrichment (Bea et al., 1992; Villaseca et al., 2008) overlapping with the aplite-pegmatite compositional field (Fig. 3b, d, and Supplementary Fig. 2g).

Overall SiO_2, Na_2O and K_2O are uncorrelated in S_1 and S_2 granites as for the aplite-pegmatites, but in general Na_2O values are lower and K_2O values are higher in any granite type than in the three aplite-pegmatite varieties (Table 2, Supplementary Fig. 2d, e). The S_3 and I-suite granites are geochemically different from S_1 and S_2, with slightly lower Al_2O_3 and higher Fe_2O_3, TiO_2, but markedly higher CaO contents (Table 2, Supplementary Fig. 2c). Thus, the A/CNK-P_2O_5 plot discriminates among the different granitic series and these from the aplite-pegmatites (Fig. 3b, d). The lowest A/CNK and P_2O_5 values correspond to the I-type, S_3 and S_4 granitic series (Fig. 3b, d). Aplite-pegmatite data shows a wider scatter, generally with higher A/CNK and/or P_2O_5 values than the five granitic series (Fig. 3d).

Rubidium, Sr and Ba also show significant variations for the different granitic and pegmatitic series. The S_3, S_4 (not shown) and I-type series are again quite similar in chemical composition, showing a positive correlation of Rb and negative of Ba and Sr (not shown) versus SiO_2 contents (Supplementary Fig. 3). The LILE contents for the S_1 and S_2 series are quite similar among them, but different to the other suites. In the plots SiO_2 versus Ba (Supplementary Fig. 3b), and Ba versus Rb and Sr (Fig. 4c, d) for the data of S_1 and S_2 granites show wider scatter, as the aplite-pegmatites data. The Ba of pegmatites is on average lower than that of these granites (Fig. 4c, d).

The contrasting chemical evolution of Th, Y (HREE) in I- and S_3-type granites has been previously described by Villaseca et al. (1998b) and Pérez-Soba et al. (2014). These evolution trends series are even more different for the highly peraluminous S_2 and S_1 series. In the TiO_2 versus Y plot the S_3 and I series appear quite scattered (Fig. 5a). By tendency the Y content of the I-type suite increases slightly with decreasing the TiO_2 content. S_3 granites show a slightly positive Y-Ti correlation but this is better defined in the more peraluminous granites. S_1 and S_2 series show a good positive correlation, with a significant decrease in the Y content as the TiO_2 decreases from the granitic to the different types of aplite-pegmatites, the lowest Y values belonging to some Li-rich bodies. Differences for S_1, S_2 and aplite-pegmatites regarding the S_3 and I series are also observed for the plots P_2O_5 versus Th, K/Rb versus Eu/Eu*, and K/Rb versus $(La/Lu)_N$ (Fig. 5b, c, d).

Contents in incompatible elements such as F, Li, Ta, Cs, Rb and Sn are higher in S_1 and S_2 than in S_3, S_4 (not shown) and I-type granites (Figs. 4 and 6; Supplementary Fig. 3; Table 2). Some S_2 granites (e.g., Pedroso de Acim, Belvís de Monroy, Logrosán) show the highest values in those incompatible elements.

7. Discussion

7.1. Geochemical affinity among pegmatites and granites in the CIZ

The most abundant Li-bearing pegmatites in the CIZ, that is, those occurring in the aplite-pegmatite fields, show textural, mineralogical and geochemical features that even when present in other parts of the European Variscan Belt, are not usually observed in such abundance in other pegmatitic regions of the world (for additional information, see Suplementary Text 2). This makes it especially interesting to determine which granites are genetically related to pegmatites, and to understand the processes that gave rise to the development of this volumetrically important Li-mineralisation occurring in the CIZ. Fractionation of peraluminous granitic melts is the most widely accepted mechanism to explain the extreme differentiation degree shown by many rare-element pegmatites worldwide. However, other theories such as crystallisation of anatectic melts (e.g., Müller et al., 2017; Novak et al., 2013; Roda-Robles, 1993; Shaw et al., 2016; Shmakin, 1983; Simmons et al., 2016; Simmons and Webber, 2008; Stewart, 1978) or the influx of rare metal-rich fluids from the devolatilisation of the lower crust (e.g., Christiansen et al., 1988; Cuney and Barbey, 2014; Marignac and Cuney, 1999; Simons et al., 2016) have also been proposed for the origin of the rare-element rich granites and pegmatites.

The fractionation of granitic magmas is the most plausible mechanism to explain the extreme differentiation degree shown by the Li-rich aplite-pegmatites of the CIZ taking into account: (i) the continuous chemical variation from the parental granite, through the barren, intermediate and Li-(F-P)-richest bodies observed in some of the pegmatite fields (e.g., Fregeneda-Almendra, Tres Arroyos, Barroso-Alvão, (Garate-Olave et al., 2017; Ortega et al., 2017; Roda Robles et al., 1999;

Vieira et al., 2011); (ii) the gradual transition from granitic to highly fractionated pegmatitic facies observed in some granitic cupolas (e.g. Pinilla de Fermoselle) (Roda et al., 2005; Roda-Robles et al., 2012); and (iii) the whole chemical composition of the studied aplite-pegmatites, which is significantly different from the minimum melt fractions observed in anatectic terrains. Although some of the aplite-pegmatite fields are temporally and spatially related to granites of the S_2 series in the S-CIZ domain, in some cases with a zonal distribution around the parental granite (e.g., Tres Arroyos district close to the Nisa-Alburquerque batholith), field relationships between granites and pegmatites are not always easy to establish. Moreover, the studied Li-rich aplite-pegmatite fields are never located around S_3, S_4 or I-type granite batholiths (Fig. 1).

Different binary diagrams have been used to discriminate between those granitic series that could be potential parental granites of the Li-rich pegmatites and those that could not be genetically related to them (Figs. 3–6; Supplementary Figs. 2 and 3). One representative sample of aplite-pegmatite fields from the S-CIZ (Tres Arroyos) and another one from the N-CIZ (Barroso-Alvão), together with one from a leucogranitic cupola (Castillejo de Dos Casas), are highlighted in some of the plots showing continuous trends from the granites belonging to S_1 or S_2 to the Li-F-richest aplite-pegmatites. In addition, the chemical trends for specific S_3 (Sierra de Guadarrama) and I-type (La Pedriza) granitic series are also shown. There is a very strong depletion in CaO, an increase in P_2O_5, F, and peraluminosity in S_1, S_2 granites and the aplite-pegmatites when compared to S_3, S_4 and I granitic series. Rubidium, Sr and Ba also show significant variations for the different granitic and pegmatitic series, with an increase in Rb and shorter and poorly defined decrease in Ba and Sr in the most felsic leucogranites of the S_1 and S_2 series, with some of the data overlapping the field of the aplite-pegmatites (Fig. 4c, d; Supplementary Fig. 3). Other incompatible trace elements, including Sn, Cs, Nb and Ta, as well as the K/Rb, Nb/Ta and Zr/Hf ratios show continuous trends from the S_2 granitic series through the different facies of aplite-pegmatites (Fig. 6). In other graphs aplite-pegmatites correlate with the data of the S_1 and S_2 granitic suites, and the data plot far from the S_3, S_4 (not shown) and I series (Fig. 5). Accordingly, there is no geochemical affinity among the granites belonging to the S_3, S_4 and I series and the aplite-pegmatites from the CIZ. These findings indicate that the S_1 and the S_2 series, occurring mainly in the N-CIZ and in the S-CIZ respectively (Fig. 1), are the most likely candidates for the five granitic series in the CIZ (being the parental granites of the Li-rich aplite-pegmatites).

Aplite-pegmatites from N-CIZ are very similar to those occurring to the south. The main chemical difference among the aplite-pegmatites from N-CIZ and S-CIZ is the degree of enrichment in F and P attained by the most fractionated pegmatites, which is usually lower in the aplite-pegmatite fields of the northern realm. In the central parts of the CIZ, the F content in the most fractionated aplite-pegmatites, is intermediate. Thus, a minor chemical difference may exist between Li-rich pegmatites from north to south of the CIZ that is also correlated with decreasing P contents shown by the S_1 batholiths (e.g., La Espenuca granites from NW Spain have markedly lower P contents than equivalent S_1 types from Portugal or the Domo del Tormes areas; Ortega, 1998).

There are also some significant chemical and geochronological differences between S_1 and S_2 granites. While S_1 corresponds to syn- to late-D3 granites, the S_2 are younger, late- to post-tectonic ones (see section above). Moreover, S_1 forms commonly sheet-like, *para*-authochtonous and relatively small bodies, whereas the S_2 constitutes larger, allochthonous batholiths often exhibiting a reverse zoning, with some leucocratic marginal facies highly enriched in incompatible elements such as F, B, P and Li (e.g., Alburquerque, Belvís de Monroy, Jálama plutons). Thus, even if geochemically the S_1 and S_2 series show important similarities, they represent different granitic events. However, both series are likely related to the most important Li-mineralisation in the CIZ. The available geochronological data on the Li-rich aplite-pegmatites supports a diachronous character of this mineralisation in the CIZ. Two different events of formation of rare-element pegmatites have been recognised in this region by U-Pb dating of columbite-group minerals with older ages in N-CIZ (310 ± 5 Ma) and younger ages in S-CIZ (301 ± 3 Ma) (Melleton et al., 2011). The Li-(F-P)-rich aplite-pegmatites from the Fregeneda-Almendra field, in the centre of the CIZ, have been dated by the step-heating $^{40}Ar/^{39}Ar$ method on micas yielding ages in the range 295.1 ± 4.2–296.4 ± 3.5 Ma (Roda-Robles et al., 2009, Vieira, 2010).

Summing up, the parental granitic magmas of the aplite-pegmatites in the CIZ are Ca-poor, P ± F-rich, strongly peraluminous melts. The Li-mineralisation occurring in aplite-pegmatite fields has a diachronous character, with an increasing age from S-CIZ to N-CIZ. There is also a chemical polarity in the aplite-pegmatites, probably reflecting the minor chemical differences existing between S_1 and S_2 series. The most rare-element ernriched aplite-pegmatites located in S-CIZ (e.g., Tres Arroyos, Las Navas) are P- and F-richer than those in N-CIZ (e.g., Barroso-Alvão, Lalín-Forcarei), with intermediate values in the central parts of the CIZ (e.g. Fregeneda-Almendra, Gonçalo).

7.2. From metasediments to Li-(P-F)-rich melts

The granitic pegmatites belonging to the LCT family (Černý and Ercit, 2005) usually occur in orogenic belts. They are commonly considered to be the result of the crystallisation of granitic melts originated in a thickened continental crust as a consequence of the heating generated by mechanical and/or radiogenic decay processes (Tkachev, 2011). Granites related to Li-rich pegmatites are usually S-type and peraluminous. Metasedimentary rocks (mainly greywackes and shales) are the main source for S-type peraluminous granites (e.g. Puziewicz and Johannes, 1988; Patiño Douce and Johnston, 1991; Sylvester, 1998). Marine shales are potential sources for alkalis, such as Li, Na, K, Rb and Cs, and alkaline earths, such as Be, (mainly in micas), as well as for fluxing components, such as F, B and P (London, 2008). Metasedimentary rocks from the CIZ have been proposed frequently as a source for the S_1 and S_2 granites, the most peraluminous granitoids in this region (Bea et al., 1999, 2003; Merino Martínez et al., 2014), although some peraluminous orthogneissic sources have been also proposed (e.g., Ortega, 1998). The peraluminosity of the granitic melts is probably not the only condition necessary for the formation of these rare-element pegmatites. The main chemical differences between the potential parental granites for the Li-rich aplite-pegmatites in the CIZ (S_1 and S_2) and those that are not related to them (S_3, S_4 and I series), are the higher peraluminosity and concentrations of P, F and Li, together with lower Ca values (Table 2). The cause of these differences is very likely the geochemistry of the protoliths. The S_3 and S_4 peraluminous granitic series are assumed to be derived from a lower crustal metaigneous protholith (Martins et al., 2009; Villaseca et al., 2012), whereas S_1 and S_2 were derived mainly from metasedimentary sources (e.g., Almeida et al., 1998; Antunes et al., 2008; Merino Martínez et al., 2014).

The study area is characterised by a thick (8–11 km) Neoproterozoic to Early Cambrian (e.g., Ribeiro et al., 1990; Rodríguez-Alonso et al., 2004) metasedimentary sequence of alternating pelitic and psammitic beds. Even if some authors have suggested a geochemical homogeneity in the pelitic metasedimentary rocks of the CIZ (Ugidos et al., 2010), according to Villaseca et al. (2014; and references therein) the Neoproterozoic metasedimentary units from the N-CIZ have different geochemical signatures (Table 2). The N-CIZ domain has foreland features showing more mature and recycled continental sedimentary sources, whereas the S-CIZ domain (the so-called Schist-Greywacke Complex, SGC series) was closer to the Cadomian active margin of Gondwana continent, being comparatively richer in juvenile (mantle-derived) components. The N-CIZ materials partly overline the materials belonging to the SGC, and could wedge themselves beneath them in the central parts of the CIZ due to flat subduction during

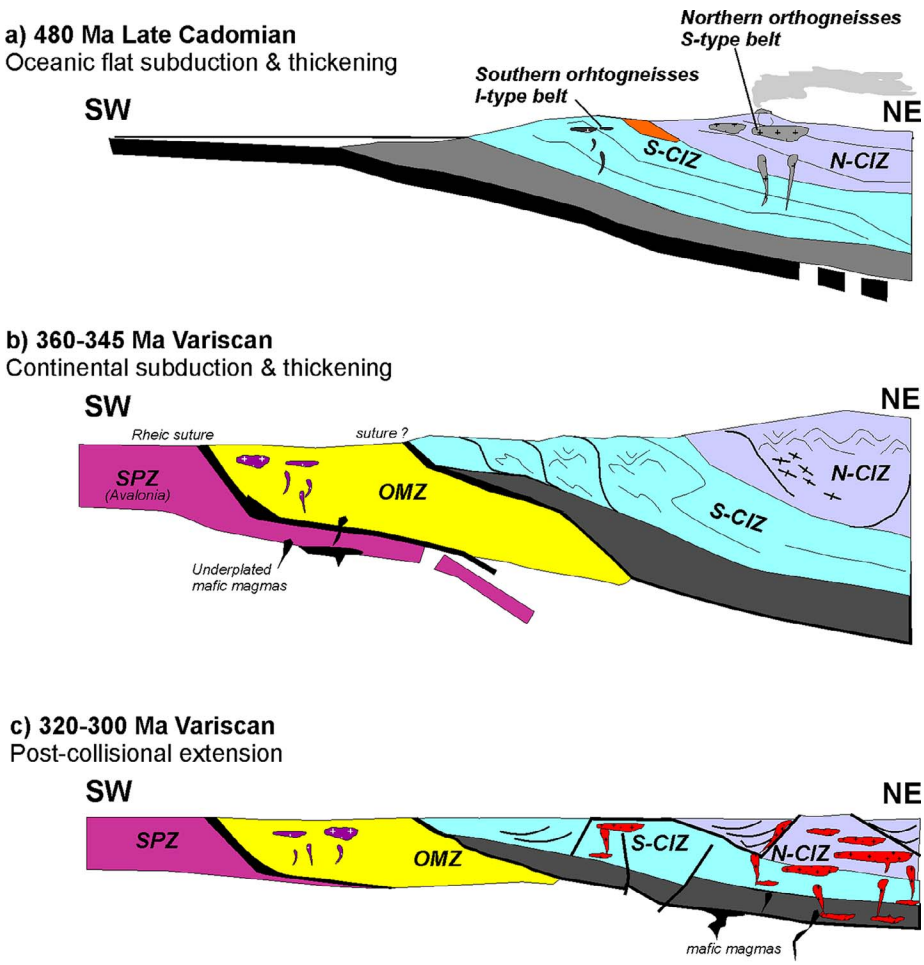

Fig. 7. Schematic diagrams illustrating the tectonic evolution of the Central Iberian terranes during the Paleozoic. Reconstructions for, (a) flat subduction and continental thickening (collision event) during late Cadomian times (Cambrian-Early Ordovician), modified from Villaseca et al. (2016); (b) continental collision during the Variscan Orogeny (Early Carboniferous) and calc-alkaline 350–335 Ma plutonism in the OMZ (modified from Simancas et al., 2013); and (c) after thickening late-orogenic collapse and massive peraluminous granite plutonism in the CIZ by radiogenic heat production. SPZ = South-Portuguese Zone, OMZ = Ossa Morena Zone, S-CIZ = southern Central Iberian Zone, N-CIZ = northern Central Iberian Zone. See text for explanation.

Cambrian–Ordovician late-Cadomian stages (Villaseca et al., 2014, 2016) (Fig. 7a). The metasedimentary sequences from the northern CIZ are slightly richer in LILE, LREE and in some HFSE elements (and poorer in Li, F and P_2O_5) than the materials from the S-CIZ (Villaseca et al., 2014) (Table 2). Nevertheless, when both metasedimentary sequences are compared to other averaged shale compositions, such as the NASC (Average North American shale: Gromet et al., 1984), and with the values for the upper continental crust (UCC) and bulk continental crust (BCC) (Rudnick and Gao, 2003), the metasediments from the CIZ show a higher peraluminosity, significantly lower Ca-(Sr)-contents, and higher P, F and Li values (Table 2).

Chappell and White (1992) attributed low-Ca contents in some strongly peraluminous granites from the Lachland Fold Belt to the sedimentary sources, where this element (together with Na) became lost during the formation of clay from feldspar. In the study area some authors have attributed the origin of some chemical differences between sediments to their early Paleozoic continental weathering on the stable parts of northern Godwana (Avigad et al., 2005; Noblet and Lefort, 1990; Romer and Kroner, 2015). As a consequence, the resulting sediments would be Ca, Na and Sr depleted, reflecting the alteration of feldspars (Mingram, 1998), and enriched in Rb, Sn, W and other incompatible elements such as Li, Be, Cs, Nb and Ta. These elements would be incorporated into the clay minerals resulting from the weathering of the feldspars (Romer and Kroner, 2015). The redeposition of those weathered sediments during the opening of *peri*-Gondwanan oceanic basins would give rise locally to important volumes of fluxing- and rare-element-rich sediments (Romer and Kroner, 2015). In addition, phosphorite levels of Ediacaran-Early Cambrian ages are relatively abundant in the metasedimentary sequence of the S-CIZ (Álvaro et al., 2016), with P_2O_5 values in the range 10.25–27.63 wt% (Álvaro et al., 2016) and also some high F contents (e.g. 1.71 wt%) reported in phosphorites from the Valdelacasa anticline (Gabaldón López et al., 1987). Although the phosphorites are presumed to be very difficult to melt during the anatectic processes, their occurrence intercalated in the pelitic series suggests a high-P-(F) sedimentary environment. Based on these considerations, the Neoproterozoic-Early Cambrian metapelites from the CIZ, which are Al, P, F and Li-richer and Ca-poorer when compared with other averaged shales and with the UCC and BCC, are considered to be a previously enriched metasedimentary protholith for the S_1 and S_2 granites (Table 2). The geochemical signature of these metasediments could be modified locally by weathering of the sediments (Romer and Kroner, 2015). However, taking into account the significant thickness of the CIZ metasedimentary sequences (up to 11 km), the enriched composition of these sediments would most probably have a primary origin.

The S_2 granite suite is mostly confined to the S-CIZ domain (Villaseca et al., 2008), where the metasedimentary SGC is an epizonal wall rock. These granites show a notable coincidence in isotopic signatures (Sr, Nd, O, Hf) with the surrounding pelitic and greywackic sequences of the SGC (Antunes et al., 2008; Ramírez and Menéndez, 1999) and also show the same populations of inherited zircons (e.g., Chicharro et al., 2014; Merino Martínez et al., 2014), suggesting that the SGC metapelitic material may be a suitable protholith for the S_2 granitic suite. To the N-CIZ the S_2 is not well represented. There, the earlier S_1 suite is volumetrically dominant. These granites have been interpreted as melt fractions of pure crustal derivation (e.g., Cuesta and Gallastegui, 2004; Ortega, 1998), either from metasedimentary (Almeida et al., 1998), metaigneous (Dias et al., 2002; Ortega, 1998), or mixed sources (López Moro et al., 2012). Granites from the S_1 having a metaigneous protholith occur mainly in the northwestermost areas of

the N-CIZ (e.g., La Espenuca granite: Ortega, 1998). In this case, granites are P-poorer and are not related to any Li-mineralisation. Considering that the N-CIZ metasediments partly overline the sequences belonging to the SGC (Villaseca et al., 2014, 2016), we speculate that metasedimentary rocks from the SGC under the N-CIZ could have melted together with metasediments from the N-CIZ in southern parts of the N-CIZ (Fig. 7). The proportions of these materials would decrease northwards; therefore decreasing the concentrations of F and P in the eventually generated granitic melts. High concentrations in these two elements, together with other fluxes (such as B, Li and H_2O), may be crucially important in explaining the occurrence of the Li-rich mineralisation in the CIZ.

The low Ca concentration in the S_1 and S_2 granitic melts would prevent significant crystallisation of apatite, which would favour the concentration of P and F in the residual melts. Water, together with F, seem to be the most important fluxing agents in natural aluminosilicate melts, with a strong influence in the chemical and physical properties of magmas (e.g., Bartels et al., 2013; Dingwell et al., 1993, 1996; Holtz et al., 2001; Maneta, 2015; Nabelek et al., 2010). Boron, P and Li also act as fluxes (Bartels et al., 2013, 2015); Holtz et al., 1993, 1995; Maneta et al., 2015). Moreover, the H_2O solubility increases with increasing F and B content in the melt (Holtz et al., 1993) and with peraluminosity (Acosta-Vigil et al., 2003; Dingwell et al., 1997; Holtz et al., 2001). For the *P-T* regimes estimated for the emplacement of the S_2 granites, close to 800 °C and ~2.5 kbar (Merino Martínez et al., 2014), and taking into account the ASI value (mean of 1.27), the H_2O content would probably be > 6 wt% according to the experimental data of Acosta-Vigil et al. (2003). The occurrence of Li-mineralisation in the apical parts of leucogranitic cupolas in the CIZ, with an upward zoning (e.g. Pinilla de Fermoselle, Castillejo de Dos Casas), supports a model of in situ upward movement of the residual melt and volatiles (Roda et al., 2005; Roda-Robles et al., 2012). Moreover, the distribution of the rare-element aplite-pegmatite fields of the CIZ, with the Li-F-richest dykes occurring frequently farthest from the parental granite, could reflect a previous vertical chemical zonation of the melt within the source magmatic chamber (e.g., Bea et al., 1994; London 1990, 1992; Roda et al., 2005; Roda-Robles et al., 2012, 2016; Simmons et al., 1987). Therefore, it seems feasible that at the top of some magma chambers, H_2O and flux-rich portions of the granitic melts accumulated. The enrichment in Li, F, P, B and H_2O in those melts would notably reduce their viscosity (Dingwell et al., 1996), and would also lower the liquidus temperature, controlling the ability of melts to segregate from the source area and to migrate through the crust (Holtz et al., 2001). Most of the aplite-pegmatite dykes in the CIZ are filling fractures in the country rock, either granitic or metamorphic. Consequently, the melt could be expelled along fissures and the flux- and volatile-richest melt (which originates at the top of the magma chamber) would move first and arrive further due to its lower viscosity and solidus temperature. The dykes of the aplite-pegmatite fields in the CIZ are notoriously poor in OH– and H_2O-bearing minerals, as micas are very scarce (unless they are Li-rich micas, in which F is more abundant than OH in the mica structure, Roda-Robles et al., 2016). The main minerals in these dykes are feldspars, quartz and lithium aluminosilicates. If the granitic melt underwent a vertical fractionation prior to the opening of the system, the amount of H_2O in the apical portion would be high, probably close to saturation levels. Therefore, the low proportion of OH– or H_2O-bearing minerals in these bodies is likely the result of exsolution of a H_2O-rich fluid phase once the pegmatitic melt intruded the fracture where it later solidified. The decrease in P and T during the ascent of these melts, along with the crystallisation of Li ± F-rich minerals from the very beginning of crystallisation, would deplete the capability of dissolution of water in the melt, prompting the exsolution of a fluid phase. The intense tourmalinisation developed frequently in the mica-schist hosting the aplite-pegmatites (e.g., Fregeneda-Almendra, Barroso-Alvão, Tres Arroyos and Belvís de Monroy fields) attests for metasomatic processes involving a H_2O-B-rich fluid exsolved from the pegmatitic-melt injected into the fractures. The loss of H_2O-rich fluids, as well as the fast cooling in contact with the country rocks, would lead to an important undercooling of the melt, which could be related to the aplitic character of an important volume of the dykes (Fenn, 1977; Nabelek et al., 2010). It may also be related to the development of rhythmic banding (London, 2008; Nabelek et al., 2010; Webber et al., 1997, 1999), and other directional textures such as feldspar and lithium aluminosilicate comb-crystals, observed frequently in some of the aplite-pegmatites from the CIZ. The lack of pockets inside these dykes, relatively common in other pegmatites intruded at shallow levels, may also be related to this undercooling.

The chemical, paragenetic and often textural similarities of the Li-mineralisation between the dykes of the aplite-pegmatite fields and the highly fractionated leucogranitic cupolas suggest that in this case the magmatic system should remain closed until the last stages of pegmatite crystallisation (Roda et al., 2005; Roda-Robles et al., 2012, 2016). Thus, high crystal fractionation rates in metasedimentary-derived parental melts (S_1 and S_2 types) seems to be the main magmatic process involved in the origin of the uncommon CIZ Li-rich rocks (Figs. 5 and 6).

7.3. Distribution and significance of Li-(F-P)-rich aplite-pegmatites and granites within the European Variscan Belt

Although less common, Li-bearing aplite-pegmatites showing similar petrographic, mineralogical and chemical characteristics to those occurring in aplite-pegmatite fields of the CIZ are also present in other parts of the central and eastern regions of the European Variscan Belt (Fig. 8). Such is the case for the Chedeville Li-rich body in the Monts d'Ambazac pegmatite field (Deveaud et al., 2015; Raimbault, 1998) and the Richemont dyke (Raimbault and Burnol, 1998) both in the Massif Central, France. Moreover, Li-rich granitic rocks comparable to the leucogranitic cupolas of the CIZ are documented in a number of localities, e.g., the Saxothuringian Zone of the Bohemian Massif, Czech Republic (Breiter, 2002; Breiter et al., 1997, 2005; Klomínský et al., 2010) and Germany (Förster et al., 1999); the Moldanubian Zone of the Bohemian Massif, in South Bohemia and Northern Austria (Breiter and Scharbert, 1995; Klomínsky et al., 2010); the Gemeric superunit in the Slovak Republic (Breiter et al., 2015; Petrík et al., 2011; Uher and Broska, 1996); and the Montebras granite (Cuney et al., 2002; Raimbault et al., 1995); and the Beauvoir granitic cupola (Cuney and Barbey, 2014) in the French Massif Central. In these cases similar mineral associations, sometimes including stockscheider comb-crystals and layered textures, are observed usually in relation to P-rich peraluminous granitic cupolas. Lithium-B-rich fractionated granites also occur to the north of the Rheic suture within the Rhenohercynian Zone, belonging to the Late-Variscan Cornubian Batholith (SW England). Layered aplite-pegmatite dykes, some of them enriched in F ± Li, occur in this region as well and are related to these granites, for example, the Megiliggar sheet complex (e.g., Stone, 1969; Fig. 8).

In this discussion many of the Li-rich pegmatites occurring in the Moldanubian Domain of the Variscan Bohemian Massif are not included because the "style" of pegmatite is very different from that of the aplite-pegmatites occurring in the CIZ. Those from the Moldanubian Domain show textural features typical for common pegmatites, having a coarse grain size and a well-developed internal zoning, including a prominent quartz core (Melleton et al., 2012). These pegmatites have been interpreted as the result of anatexis (Melleton et al., 2012).

It is difficult to extrapolate the classification established for the different granitic series from the CIZ to those that appear in the rest of the Variscan belt. However, according to the bibliographic data, granites with a chemical signature similar to the S_1 and/or S_2 series seem to be abundant mainly in the French Massif Central, Bohemian Massif and Rhenohercynian Zone (Figs. 8 and 9). Phosphorous-enriched and Ca-poor highly peraluminous granites from these regions plot in the same areas of the S_1 and S_2 granites (Fig. 9). For example, in the French Massif Central the "Muscovite-bearing Peraluminous Granitoids"

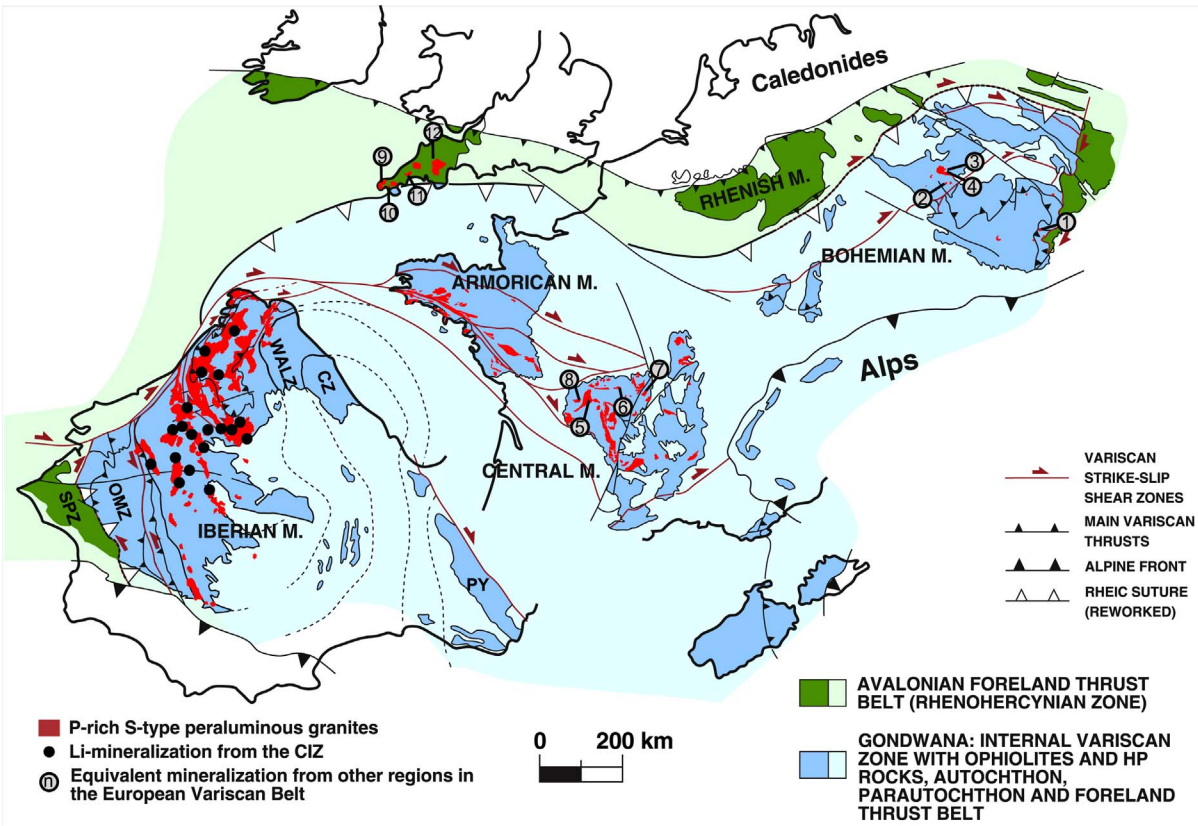

Fig. 8. Schematic geological map of the European Variscan Belt (modified from Martínez Catalán et al. (2014)) showing the distribution of the P-rich, Ca-poor peraluminous Variscan granitic series (Lameyre, 1987; Augier et al., 2015; Klomínský et al., 2010), and of the Li-mineralization similar to that from the aplite-pegmatites and leucogranitic cupolas in the CIZ, (1) Homolka (Breiter and Scharbert, 1995); (2) Krizovy Kámen (Klomínsky et al., 2010); (3) Podlesí (Breiter et al., 1997, 2005); (4) Perninkstock (Klomínsky et al., 2010); (5) Monts d'Ambazac (Raimbault, 1998, Deveaud et al., 2015); (6) Montebras (Raimbault et al., 1995); (7) Beauvoir granite (Cuney and Barbey, 2014); (8) Richemont (Raimbault and Burnol, 1998); (9) Porth Ledden (Müller et al., 2006; Shail et al., 2014); (10) Megiliggar Rocks (Stone, 1969, 1975; Simons and Shail, 2014; Simons et al., 2016); (11) Saint Austell (Manning et al., 1996; Thompson et al., 2014); (12) Meldon (Scrivener and McVicar Wright, 2014).

(MPG), e.g., Saint Sylvester, Millevaches and La Marche (Barbarin, 1999; Moyen et al., 2017), are classified as syn- to late D3, muscovite- or muscovite + biotite (two mica) leucogranites (e.g., Downes and Duthou, 1988; Williamson et al., 1996). These are related to shear zones that could be correlated to the S_1 granites from the CIZ. In the Saxothurigian Zone of the Bohemian Massif the P-rich peraluminous granites, comparable to those of the S_2 suite in the CIZ, are also abundant in the Fichtelgebirge-Erzgebirge Batholith. They belong to the series of "Younger Intrusive Complex" (YIC) described by Rajpoot and Klomínský (1994) and Klomínský et al. (2010) as the groups 3, 4 and 5 (G3, G4 and G5). They consist of moderately to strongly differentiated, post-orogenic, stanniferous, highly peraluminous granites, some of them Li-mica and topaz-bearing, e.g., in the Fichtelgebirge, Slavkovský les Mts. (Krudum massif) and the western and central part of the Krusné Hory/Erzgebirge Mts. (Nejdek-Eibenstock pluton, Blatná massif, and Podlesí stock), and small stocks of Geyer, Ehrenfriedersdorf, Satzung, and Pobershau (Breiter et al., 1999; René, 2003) (Fig. 8). In the case of the Moldanubian Zone of the Bohemian Massif, slightly mineralised small post-orogenic granitoids from the Moldanubian Composite Batholith, including Nebelstein, Hirschenschlag-Kozi Hora and Homolka granites (Klomínsky et al., 2010) (Fig. 8), have a highly peraluminous, P-rich character (Breiter and Scharbert, 1995) that could be compared to those of the S_2 series in the CIZ. In the Western Carpathians, chemically similar granites have been described in the Gemeric superunit (Petrík et al., 2011; Uher and Broska, 1996). These granites are Sn-F-rich and crop out as several small bodies. To the west of the Gemeric unit some of these granites show Li-enrichment (Dianiška et al., 2002). Granites from the Cornubian batholith have also been compared to and correlated with those from the S_1 and S_2 series of the CIZ (Simons et al., 2016), some of them associated with Sn, F and/or Li enrichments, as is the case of the Land's End pluton (Müller et al., 2006; Shail et al., 2014); the Tregonning granite (Bevins et al., 2010; Floyd et al., 1993; Stone, 1969, 1975); the Dartmoor granite (Scrivener and McVicar Wright, 2014); and the Saint Austell granite (Manning et al., 1996; Shail et al., 2014) (Fig. 8).

A province of P-rich rare-metal granites was defined for the central zone of the European Variscan Belt by Breiter and Scharbert (1995), including the Czech-Austrian-Bavarian Moldanubicum and the French Massif Central. This metallogenetic province could be extended to the Saxothuringian and Cornwall regions, as well as to the CIZ, based on new geochemical data. The origin of many of these P-rich granites is assumed to be mainly related to the dehydration melting of metasediments at crustal levels (Gerdes et al., 2000; Moyen et al., 2017; Petrík and Kohút, 1997; Vozárová et al., 2009). As in the case of the metasediments from the CIZ, P-(F, Li) and Al-rich and Ca-poor shales have been also described in the Saxo-Thuringian zone of the Bohemian Massif (Romer and Hahne, 2010). The existence of marginal oceanic basins at the northern *peri*-Gondwana margin, where thick mature pelitic sequences were deposited at Neoproterozoic times, was crucial in generating enriched metasedimentary protoliths later involved in the collisional Variscan crustal recycling that generated large peraluminous granite batholiths.

Finally, there are also some late Variscan Li-rich albite granites to the east of the Saxo-Thuringian Zone (Cínovec-Zinnwald, in the Erzgebirge) (Breiter, 2012; Breiter et al., 1999), coeval to the described S-type, P-rich granites, but related to highly fractionated A-type granites with a slightly peraluminous F-rich P-poor composition (Stemprok et al., 2003). According to Romer et al. (2014), metasedimentary rocks

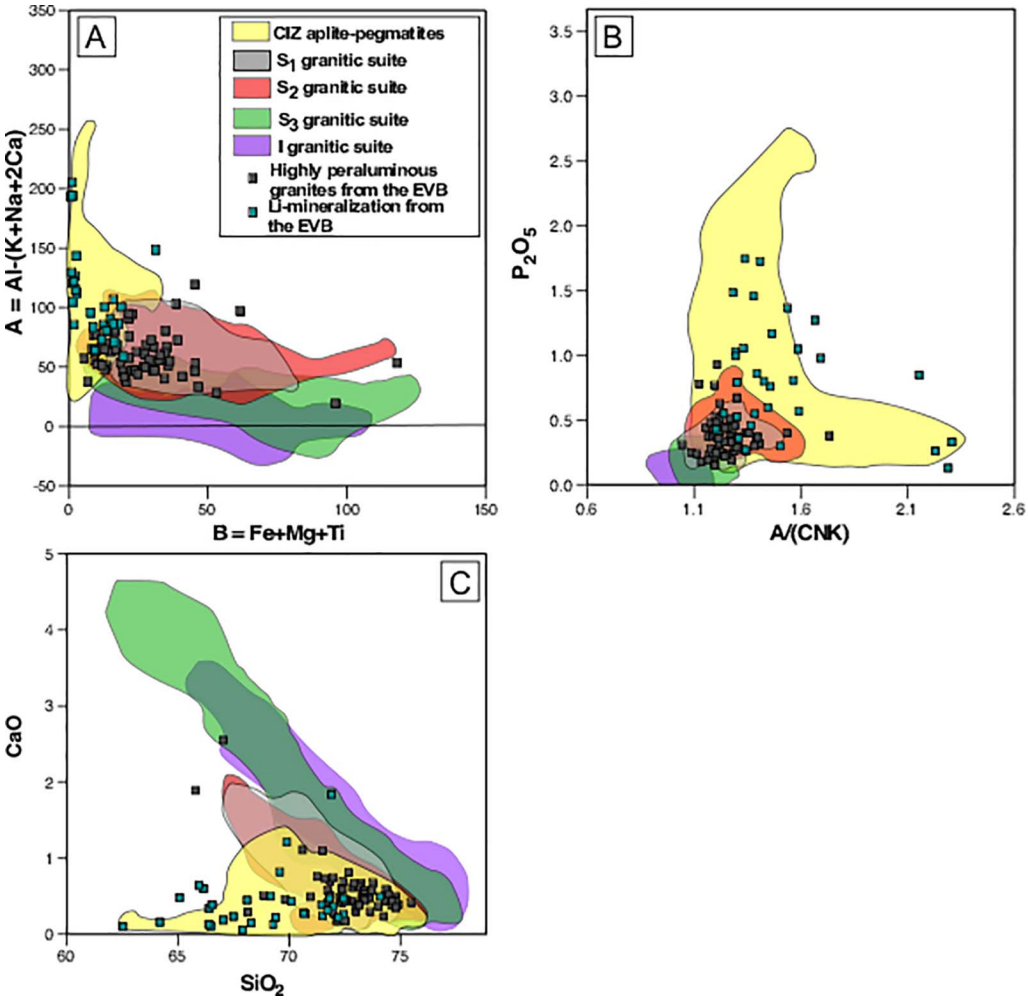

Fig. 9. Composition of the P-rich Variscan granites and of the Li-mineralization belonging to the European Variscan Belt (EVB) in the French Massif Central, the Bohemian Massif an the Cornubian Batholith, together with the compositional fields of the aplite-pegmatites, the leucogranitic cupolas and the S_1, S_2, S_3 and I granitic series of the CIZ (a) in the A–B diagram; (b) in the A/CNK (molar $Al_2O_3/(CaO + NaO + K_2O)$) *vs* P_2O_5 (in wt.%); and (c) in the SiO_2 *vs* CaO (in wt.%) values. Chemical data of European Variscan granites and Li-rich mineralization from: Müller et al. (2006), Simons et al. (2016), Williamson et al. (1996), Downes et al. (1997), Léger et al. (1990), Cuney and Barbey (2014), Raimbault et al. (1995), Raimbault (1998); Breiter et al. (1991, 2005), Klomínsky et al. (2010), Breiter and Scharbert (1995), Petrik et al. (2011) and Förster et al. (1999). We have considered as Li-mineralization rocks with Li contents > 1000 ppm.

enriched in F, Li, Rb, Cs, Sn and W could be in the source of these granites, probably involving higher *meta*-graywacke than metapelite proportion (Breiter, 2012). Again, the involvement of enriched metasedimentary sources is crucial for the development of Li-rich highly fractionated granite magmas.

7.4. Open questions to the proposed model

Although the close spatial association of Li-(F-P)-rich pegmatites with P-rich highly peraluminous granite complexes suggests a direct link via fractional crystallisation from the neighbouring granite batholiths, some questions remain open and will be the topic of future studies.

Over past decades research on pegmatites has yielded a wealth of information, particularly on rare-element pegmatites. Numerous geological, petrographic, mineralogical and geochemical studies have been carried out in order to document the characteristics and petrogenesis of such systems. However, isotopic studies to establish the age, affiliation and evolution of pegmatites are relatively scarce. Actually, only a small number of geochronological data on the Li-mineralisation associated with aplite-pegmatite fields and/or leucogranitic cupolas from the CIZ, as well as from the rest of the European Variscan Belt, are available. The different types of pegmatites occurring in the Fregeneda-Almendra field were dated by the $^{40}Ar/^{39}Ar$ step-heating method on micas (Roda-Robles et al., 2009; Vieira, 2010), giving ages in the range 295.1 ± 4.2 to 296.4 ± 3.5 Ma. In addition, Melleton and Gloaguen (2015) dated some Li-mineralisation from the Galicia-Trás-Os-Montes Zone and the CIZ (U-Pb ages on columbite and tantalite) with results of 310 ± 5 Ma and 301 ± 3 Ma, respectively. With the same method these authors also dated some Li-occurrences from the French Massif Central in the North Limousin area, obtaining ages of 317 ± 6 Ma, 314 ± 4 Ma and 309 ± 5 Ma for the Beauvoir, Montebras and Chèdeville rare-elements magmatic bodies, respectively. This scarcity of geochronological data on Li-mineralisation is probably due, on the one hand, to the difficulty to find mineral phases suitable for the application of the geochronological methods in highly evolved pegmatites and, on the other hand, to significant subsolidus processes that variably alter the mineral paragenesis and may lead to inaccurate results. In contrast, there is more geochronological data on potential parental granites. In the case of the pegmatites from the Fregeneda-Almendra field, the age of the closest granite (Meda-Penedono-Lumbrales Complex of the S_1 series) was estimated in 311.2 ± 3.7 Ma ($^{40}Ar/^{39}Ar$ step-heating micas; Roda-Robles et al., 2009; Vieira, 2010); i.e. ~15 Ma older than the aplite-pegmatites in this field. A similar situation was recognised for the St Sylvestre granite in the proximity to the Chèdeville pegmatite in the French Central Massif, with an age of 324 ± 4 Ma (U–Pb on zircon and monazite; Hollinger et al. (1986), but again the spatially related pegmatites are approximately 15 Ma older. This age difference is too large to consider that those pegmatites could be petrogenetically linked to the closest outcropping granite. In the case of the Fregeneda-Almendra field, the origin of the Li-rich aplite-pegmatites is likely to be related to a non-outcropping late-orogenic granite. This granite probably belongs to the S_2 series, which is common in this region (Roda-Robles et al., 2009; Vieira, 2010). We do not possess enough data to interpret the relationships between the Chèdeville pegmatite and the St Sylvestre granite. Any case, it is evident that the understanding of the

relationships between granites and pegmatites in the CIZ and in the European Variscan Belt is limited due to the lack of geochronological data on Li-mineralisation, but also on the potential parental granites.

Another controversial issue to establish linkages between granites and pegmatites is the interpretation of the geochemical data. For example, chemical trends similar to those observed for the Li-aplite-pegmatites and granites from the CIZ (Supplementary Fig. 2d, f) reported by Simons et al. (2016) for the granites of the Cornubian batholith, are interpreted in a different way. They conclude that the most fractionated topaz-rich G5 granites (equivalent to the most fractionated S_2 facies and/or to some aplite-pegmatites in the CIZ) could not be derived from any of the other four types of peraluminous granites in the region, and propose for them "an origin through melting of the biotite-rich restite remaining after extraction of G3 (biotite) granite magmas. Such melting would have been induced by fluids containing a high abundance of F, Li, P and HFSE derived from granulite facies dehydration melting of the lower crust during emplacement of mantle-derived melts"; as it had been previously proposed for the origin of some rare metal-rich fractionated granites in the French Massif Central (Cuney and Barbey, 2014). However, this scenario fails in the case of leucogranitic cupolas enriched in Li-(F-P) because there is an evident genetic link between the granite below and the enriched cupola, with a gradual chemical, mineralogical and textural transition (e.g., Pinilla de Fermoselle (Roda-Robles et al., 2012), Castillejo de Dos Casas (Roda-Robles et al., 2015), and Podlesí (Breiter et al., 2005)). Moreover, and even if some isotopic data makes it more difficult to correlate some Li-rich aplite-pegmatites with their nearby granite (e.g., Antunes et al., 2013), a good number of aplite-pegmatite fields and leucogranitic cupolas in the CIZ have been studied in detail and in most of cases an origin by fractionation of granitic melts has been proposed, e.g., Argemela (Charoy and Noronha, 1996); Las Navas (Gallego Garrido, 1992); and Gonçalo (Neiva and Ramos, 2010). In addition, geochemical modelling has taken place on some representative aplite-pegmatite fields, e.g., Tres Arroyos, (Garate-Olave et al., 2017); Barroso-Alvão, (Ortega et al., 2017); Fregeneda-Almendra, (Vieira, 2010), and leucogranitic cupolas, e.g., Pinilla de Fermoselle, (Roda-Robles et al., 2012); and Castillejo de Dos Casas, (Roda-Robles et al., 2015), (Figs. 4–6). In all these cases modelling has revealed that fractional crystallisation of S_1 and S_2 granitic melts is the most plausible petrogenetic process for the Li-mineralisation.

In contrast, the hypothesis of a metasomatic imprint by Li-P-F-B-rich fluids expelled from the granulitic lower crust, induced either by its delamination (Cuney and Barbey, 2014) or by interaction with underplated mafic magma (e.g., Simons et al., 2016), is not straight forward to apply to the CIZ. The P-Li-richest metasedimentary sequences outcropping in the southern realm (the SGC materials), show low metamorphic degrees, no evidence of having underwent any melt extraction process (partial melting), and are located far from granulite-facies terranes. Thus, the hypothetic Li-P-F-B-rich fluids expelled from a granulitic lower crust should cross several km and be pervasive through a huge volume of rocks to metasomatise significantly the protoliths of the Li-mineralisation, which is difficult to explain.

The formation of the Li-rich aplite-pegmatites of the CIZ by anatexis is unlikely for the following reasons. These pegmatites do not occur in migmatitic terrains, but intrude into low degree metamorphic areas. In addition, their bulk composition, highly enriched in Li-(F-P-B) is very different from the minimum melt fractions observed in anatectic terrains (e.g., López-Plaza and Gonzalo, 1993). Thus, even if low degrees of partial melting of previously enriched metasediments may have formed melts with relatively high Li content, the important volumes of Li-rich material observed in the aplite-pegmatite fields and their spatial association with highly fractionated plutonic bodies make it difficult to assume that the pegmatitic melts could have been formed directly by extremely low degrees of partial melting.

Therefore, in the Variscan Iberian belt at least, the chemical suitability of metasedimentary protoliths seems to be an original feature, and the Li-rich aplite-pegmatites and leucogranitic cupolas appear to be most probably derived by differentiation of granitic melts originated from these initially enriched metasedimentary sources.

Nevertheless, there is still much to learn to fully understand the petrogenetic uniqueness of the European Variscan Li-(F-P) metalogenetic province. This research has raised many questions in need of further investigation including geochemistry (major, minor and trace elements and isotopic geochemistry) and geochronology of the Li-rich aplite-pegmatites, granites and metasediments in both the CIZ and the rest of the European Variscan Belt.

8. Summary and conclusions

Based on field observations, mineralogical, textural and compositional characteristics of the different types of Li-mineralisation and granitic rocks from the CIZ, together with the bibliographic data on Li-mineralisation and associated granites from the central and eastern European Variscan belt, the following conclusions can be drawn:

1) The majority of Li-rich rocks from the CIZ are represented by aplite-pegmatite fields and fractionated leucogranitic cupolas. Their textures (mainly aplitic, often layered and exhibiting occasionally coarse-grained comb textures), the lack of internal zoning and quartz core, and the bulk chemical composition (high Li $>$ 1.3 wt% Li_2O, Na, F and P contents) are uncommon for most pegmatite provinces worldwide considering the available literature.
2) Based on the degree of Li $\pm$ F-enrichment, the aplite-pegmatites are classified into three types: (1) Li-mica bearing dykes that are Li-rich and the richest in F; (2) Petalite and/or spodumene bearing dykes, which are also Li-rich but much poorer in F; and (3) Simple aplite-pegmatites which show low Li and F contents, usually with no Li-rich silicates and just accessory montebrasite. The three types are not always present in all the pegmatite fields. The F-rich aplite-pegmatites (type 1) are more common south of CIZ and the F-poor ones (type 2) in the N-CIZ.
3) The main Li-rich minerals occurring in the aplite-pegmatite fields and leucogranitic cupolas of the CIZ are Li-mica, spodumene and/or petalite, with montebrasite and ferrisicklerite as main accessory minerals.
4) Field relationships and geochemical features suggest that granites belonging to the S_1 series (two mica leucogranite suite) in the N-CIZ and S_2 series (P-rich highly peraluminous granite suite) in the S-CIZ represent the parental magmas of the aplite-pegmatites and of the Li-mineralisation associated with leucogranitic cupolas. The P-poor, moderately peraluminous granite suite (S_3), the moderately to low peraluminous granite suite (S_4), and the low peraluminous I-type granite suite (I) are not related to these types of mineralisation.
5) Pelitic and metagreywackic Neoproterozoic to Early Cambrian metasediments of the CIZ are considered to be the main sources of the perphosphorous-peraluminous granite batholiths (S_1 and S_2 series) related to these Li-rich rocks. The strong enrichment in P-(F, Li) and the highly peraluminous (Ca-Sr-poor) character of these metasediments suggest a specific compositional prerequisite for the formation of abundant P-rich, highly peraluminous granites, uncommon in other collisional scenarios (Caledonian, Tasmanian or Himalayan).
6) Magmatic fractionation processes, enhanced by the effects of primary high contents in fluxing components such as P, F, B, Li and H_2O, are interpreted to be the main cause for the Li-enrichment in the studied aplite-pegmatites. Such components result in the lowering of the viscosity, solidus temperature and polymerisation degree of the melts, favouring their mobility and internal diffusion.
7) Lithium-rich rocks of similar composition and similar textures to those from the aplite-pegmatite dykes and/or from the leucogranitic cupolas in the CIZ, are found in other parts of the European Variscan Belt. Accordingly, an extensive Li-metallogenetic province, related to P-rich, Ca-poor, highly peraluminous S-type granites originated

by the melting of highly peraluminous, P-rich, Ca-poor metasediments during the Variscan orogeny could be defined.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.oregeorev.2018.02.027.

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