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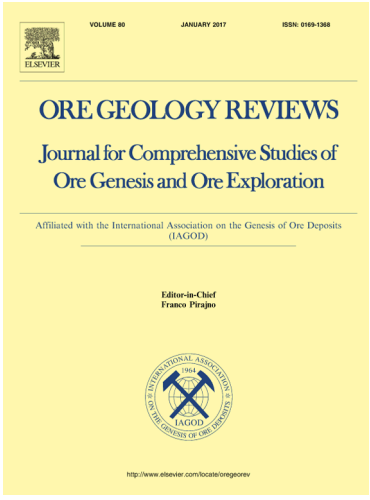

Re-assessing the European lithium resource potential – A review of *hard-rock* resources and metallogeny

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Re-assessing the European lithium resource potential – A review of *hard-rock* resources and metallogeny

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Graphical Abstract

Highlights:

- Lithium is not rare in Europe and is well represented in different orogenic settings;
- A pre-existing Li-rich source is required for Li-enrichment processes;
- Lithospheric thickening may reflect a favorable process for concentrating Li;
- Extensional geodynamical settings appear favorable for Li enrichment.

Keywords: lithium, metallogeny, pegmatite, Europe, resources

Abstract

Lithium, which is an excellent conductor of heat and electricity, became a strategic metal in the past decade due to its widespread use in electromobility and green technologies. The resulting significant increase in demand has revived European interest in lithium mining, leading several countries to assess their own resources/reserves in order to secure their supplies. In this context, we present for the first time a geographically-based and geological compilation of European lithium *hard-rock* occurrences and deposits with their corresponding features (e.g., deposit types, Li-bearing minerals, Li concentrations), as well as a systematic assessment of metallogenic processes related to lithium mineralization. It appears that lithium is well represented in various deposit types related to several orogenic cycles from Precambrian to Miocene ages. About thirty hard-rock deposits have been identified, mostly resulting from endogenous processes such as lithium-cesium-tantalum (LCT) pegmatites (e.g., Sepeda in Portugal, Aclare in Ireland, Läntta in Finland), rare-metal granites (RMG;

Beauvoir in France, Cinovec in the Czech Republic) and greisen (Cligga Head, Tregonning-Godolphin, Meldon in the UK and Montebras in France). Local exogenous processes may result in significant Li- enrichment, such as jadarite precipitation in the Jadar Basin (Serbia), but they are rarely related to economic lithium grades such as in Mn-(Fe) deposits, or in bauxite. We also identified major common parameters leading to Li enrichment: 1) a pre-existing Li-bearing source; 2) the presence of lithospheric thickening, which may be a favorable process for concentrating Li; 3) a regional or local extensional regime; and 4) the existence of fractures acting as channel ways for exogenous processes. Furthermore, we point out the heterogeneity of knowledge for several orogenic settings, such as the Mediterranean orogens, suggesting either a lack of exploration in this geographical area, or significant changes in the orogenic parameters.

1. Introduction

Over the past decade, lithium has become a strategic metal due to its physical and chemical properties, being the lightest solid element and an excellent conductor of heat and electricity. This makes it an excellent candidate for electromobility and green technologies, such as Li-ion batteries and other energy-storage devices (Armand and Tarascon, 2008; Tarascon, 2010; Ziemann et al., 2012; Manthiram et al., 2017). As a result, Li demand has increased significantly and a “lithium rush” is currently happening world wide (e.g., Roskill Information Services Ltd., 2016). In this context, the identification and assessment of lithium mineral resources and -reserves is a crucial step, as is understanding lithium metallogeny, a major subject for discovering new mineral resources.

Historically, two distinct deposit types are identified: 1) *brine* deposits in which the lithium grade is about 0.1% Li_2O; and 2) *hard-rock* deposits where the lithium grade generally varies from 0.6 to 1.0% Li_2O hosted by various Li-bearing minerals (Kesler et al., 2012; Mohr et al., 2012).

The brine-deposit type refers to relatively recent (mostly Quaternary), enclosed, tectonically active basins that contain Li-rich lacustrine evaporites. These are produced by high evaporation rates in an arid to hyper-arid climate and/or by various water inputs such as groundwater and spring water circulation (e.g., Ericksen and Salas, 1987; Bradley et al., 2013). In these deposits, the Li source and enrichment processes are specific to each brine. However, the most accepted model is the weathering of felsic rocks and/or local hydrothermal activity driven by a magmatic heat source through active channel pathways (Bradley et al., 2013; Hofstra et al., 2013). In North America, several deposits have been identified within the Basin and Range extensional province of the western United States, including the Clayton Valley and the Great Salt Lake (e.g., Bradley et al., 2013). In South America and more particularly on the Puna Plateau, brine deposits, referred to as *salars*, cover an area of about 400,000 km^2 from northern Argentina and northern Chile to western Bolivia (Ericksen and Salas,

1987) named the "lithium triangle". In this area, several lithium deposits have been identified and are operated by major mining companies including SCL/Chemetall and SQM/Tianqi Lithium Corp. In China, the Qinghai-Tibet plateau is a favorable setting for lithium deposits, as illustrated by the Qaidam (e.g., Shengsong, 1986; Yu et al., 2013) and Zabuye basins, from which some brines are exploited by governmental mining companies.

Hard-rock deposits comprise several styles of Li mineralization in magmatic and/or sedimentary rocks, related to both endogenous (magmatic) and exogenous (re-concentration by weathering, or supergene alteration and transport) processes. They can contain widespread varieties of Li-bearing minerals, such as Li-micas, Li-pyroxenes, Li-silicates, Li-phosphates, etc. (Table 1). Among them, hectorite (Li-bearing clay) from the Kings Valley, Nevada (e.g., Glanzman et al., 1978; Kesler et al., 2012) and jadarite (Li-bearing borosilicate) from Serbia (Stanley et al., 2007; Rio Tinto, 2017, Stojadinovic et al., 2017) represent potential world-class deposits, whereas spodumene-bearing lithium-cesium-tantalum (LCT) pegmatites in the Greenbushes (Australia) have been mined for decades by Talison Lithium Ltd. and others (e.g., Mudd and Jowitt, 2016).

Lithium production historically has been dominated by Australia (e.g., Greenbushes deposits), South America (*Salar de Atacama*) and China (Zabuye, Qaidam Lakes). Thus, of the 36.5 kt Li metal produced in 2016, 39% came from Australia, 32.8% from Chili, 15.6% from Argentina and 5.5% from China. Portugal, the first Li producing European country, represents only 1.3% of world production, especially for the ceramics and glass industry (BRGM, 2017). However, lithium exploration increased significantly (e.g., Roskill Information Services Ltd., 2016), leading other countries in the European Community (France, Austria, Czech Republic, Spain, Finland…) to assess their own mineral resources and –reserves, in order to evaluate their global competitiveness in the lithium industry.

Hereafter, we provide a key to understanding the geological context of lithium in Europe from a hard-rock perspective. To this end, we made a systematic, geographically- and geologically-based compilation of lithium occurrences, significant mineral showings and/or deposits, with their corresponding Li-deposit types, Li-bearing minerals and Li concentrations. For the first time, we present an overview and quantification of identified European Li deposit types and features, and their distribution in the different orogenic settings of Europe. A major effort was made to constrain metallogenetically the Li endowments in order to highlight potential processes (endogenous and exogenous) causing Li enrichment, and to introduce potential prospective regions. The resulting dataset may be used in future studies for constraining the possible relationships between Li-rich geothermal brines, surficial waters and Li-rich basement rocks.

2. Overview of hard-rock lithium deposit types in Europe

A compilation of lithium occurrences and -deposits was made by collecting information from various geological survey organizations, exploration and mining companies, and scientific research projects and related publications. This resulted in an up-to-date quantification of the European lithium potential, considering only hard-rock ore types, identifying 527 lithium occurrences, projects and deposits (provided as electronic supplementary material). This was almost five times more than the previous Mineral4EU-ProMine (http://minerals4eu.brgm-rec.fr/) inventory (Cassard et al., 2015).

In addition, mineral resource and -reserve and -production data were gathered from available published data by exploration and mining companies, such as technical and annual reports, from data repositories (e.g., https://sedar.com) and from governmental surveys. Note that the data from England, France and, locally, Germany are based on historical (before 1995), non-compliant CRIRSCO (Committee for Mineral Reserves International Reporting Standards) compliant estimates.

We emphasize that lithium deposits related to seawater, and geothermal- and oilfield brines are not considered in this study.

According to our compilation (and previous ones, e.g., Christmann et al., 2015), two distinct categories of lithium deposits and occurrences are found in Europe. They are: 1) Magmatic-related (Fig. 1A, B, C) deposits; and 2) Sedimentary/hydrothermal-related deposits (Fig. 1D).

2.1 Magmatic-related deposits

2.1.1 Rare-metal granites

Rare-metal granites (RMG; Černý et al., 2005) are felsic, peraluminous to peralkaline intrusive rocks that host magmatic disseminated mineralization. They occur as very small, mostly subsurface, granitic plugs, typically less than 1 km^3, such as the Beauvoir RMG in France (Raimbault et al., 1995; Fig. 1A). According to their geochemistry and geodynamic setting, three main types (Linnen and Cuney, 2005; Černý et al., 2005) are recognized:

1) Peralkaline RMG have very high contents of F, REE, Y, Zr, Nb, related to anorogenic settings; their Li content is relatively moderate (up to a few 1000 ppm) and is mainly illustrated by zinnwaldite and polylithionite occurrences (Tables 1, 2). This type is not documented in Europe;

2) Metaluminous to peraluminous, low- to intermediate-phosphorus RMG with high concentrations of Nb, Ta, Sn, that occur in both post- and an-orogenic geodynamic settings. The Li content again is moderate (up to a few 1000 ppm) and is mostly related to zinnwaldite. Examples include Cinovec (Fig. 1C), Podlesi (Fig. 2A), the Sejby and Homolka granites in the Czech Republic, the Chavence and Les Châtelliers granites in France (Table 2; Černý et al., 2005 and references therein);

3) Peraluminous, high-phosphorus RMG with strong enrichment in Ta, Sn, Li and F, occurring in a continental-collision setting. In this RMG type, Li concentrations can be high, from 0.5% to 1.0% Li_2O, and occurring as lepidolite, Li-rich muscovite and amblygonite-montebrasite series, such as at Beauvoir (Figs. 1A, 2B), Montebras (Fig. 2C) and the Richemont rhyolite dike in the French Massif Central, and Argemela in Portugal (Table 2; Černý et al., 2005 and references therein).

2.1.2 LCT Pegmatites

With the exception of some rare giant Precambrian occurrences, lithium-cesium-tantalum (LCT) pegmatites (e.g., London, 2008, 2018; Černý and Ercit, 2005) are relatively small-sized (a few m^3 to <1/2 km^3; Fig. 1B), coarse-grained and/or aplitic igneous rocks of granitic composition. Geochemically, Li-rich LCT pegmatites are similar to peraluminous high-phosphorus RMG. They are the result of crystallization of fluid-rich melts, enriched in various amounts of incompatible elements, such as Li, Ta, Sn, Rb, Be, Nb and Cs, and strongly depleted in REEs, close to chondritic values (London, 1995, 2005, 2008, 2018; Černý and Ercit, 2005; Černý et al., 2012; Linnen et al., 2012).

Pegmatites can form under various P/T conditions (Table 3) representing various classes (e.g., Černý, 1989, 1990; Černý et al., 2012). They are generally clustered in kilometer-size pegmatite fields (e.g., the Ambazac pegmatite field, Deveaud et al., 2013; Silva et al., 2018), and occur as dikes and/or sills (e.g., Emmes pegmatite, Finland, Eilu et al., 2012; Gonçalo pegmatite field, Portugal, Ramos et al., 1994) or lenticular bodies (e.g., Bohemian pegmatites, Melleton et al., 2012). Contacts with host-rock range from relatively sharp to progressive, depending on the nature of the host and the depth of emplacement. Host-rocks are mainly metasedimentary and/or metavolcanic rocks metamorphosed from lower greenschist to amphibolite facies (Černý, 1992) as well as granite intrusions (e.g., Gonçalo, Ambazac).

LCT pegmatites show heterogeneous textures and compositions, and are composed of variable amounts of quartz, plagioclase, potassium feldspar, micas, with various amounts of garnet, tourmaline, apatite and (usually) accessory Li-bearing minerals - locally rock-forming - such as spodumene, petalite, etc. Although not systematically observed, LCT pegmatites generally show layering and/or concentric zoning. Lithium-bearing minerals, including spodumene, petalite, the amblygonite-montebrasite group, lepidolite (Figs. 2D, E), eucryptite, elbaite and the lithiophilite-triphylite group are commonly found in pegmatite bodies, whereas cookeite and holmquistite occur mainly in the pegmatite aureole, or as secondary minerals (e.g., cookeite after petalite). Note that eucryptite may reflect the alteration of primary spodumene. Li_2O content varies as a function of the LCT pegmatite subtype and the Li-bearing minerals themselves (Table 3), ranging from 0.5 to 1.5%.

Mixed niobium–yttrium–fluorine (NYF)-LCT pegmatites are known from Norway (e.g., Birkeland, Frikstad, Skripeland) and Ukraine (Volodarsk-Volynsky). In Norway, pegmatite fields such as Evje-Iveland show a typical initial NYF chemical signature, but are depleted in REEs and F in

replacement areas. Moreover, the replacement zones show a "cleavelandite signature" as well as chemical and mineralogical LCT features, including beryl, columbite group minerals and tourmaline (Černý, 1991a, b). Lithium-ore tonnages or grades are not reported for these pegmatites.

2.1.3 Greisen

Greisen deposits (e.g., St Austell, UK; Cinovec, CZ) result from a high-temperature hydrothermal transformation of fractionated granitic intrusions (pegmatites, granites) with their upper part being a porous muscovite-quartz assemblage at the granite/host-rock contact. They can occur as multi-stage swarms crosscutting Sn-W quartz veins (e.g., Černý et al., 2005; Štemprok et al., 2005; Launay et al., 2018), or may form up to 100-m thick units with irregular to sheet-like bodies. Li is mainly hosted in micas, such as Li-rich muscovite, lepidolite, zinnwaldite, and amblygonite-montebrasite group minerals. Peraluminous RMG and metaluminous intrusions are favorable rock types for the development of such deposit types (Fig. 1C), whereas fractionated granites appear to be unrelated with significant Li endowment. Thus, the Li_2O content in the greisen around the world-class Panasqueira W deposit (Portugal) is only about 732 ppm (Bussink, 1984), whereas Li_2O values from the Erzgebirge province (e.g., Štemprok et al., 2005; Jarchovský, 2006) vary from 80 to 3100 ppm for greisen of the RMG Vykmanov and Schnöd granites.

Quartz, cassiterite, wolframite, micas, topaz, tourmaline, sericite and chlorite are common in greisen, showing vertical and horizontal zoning. Alteration is generally shown by kaolinization, tourmalinization, feldspathization (microclinization and/or albitization) and greisenization forming haloes around the granitic body. REE enrichment may occur and is indicated by precipitation of monazite, xenotime and other REE-rich minerals.

Note that this deposit type can be related to both magmatic and hydrothermal processes. Here, we consider a "granite-related" classification even though hot hydrothermal fluids are involved.

Co-products commonly consist of industrial minerals, such as feldspar, quartz and kaolin (e.g., Beauvoir, France); Sn and W are of first interest, Be, Ta, In, Sc, Rb representing potential byproducts.

2.1.4 Quartz-montebrasite hydrothermal veins

Several authors (e.g., Martín-Izard et al., 1992; Roda-Robles et al., 2016) reported the existence of quartz-montebrasite hydrothermal veins associated with leucogranitic cupolas in the central part of the Central Iberian Zone in Spain (e.g., Valdeflores, Barquilla, Golpejas, El Trasquilón) and Portugal (e.g., Argemela area and Massueime).

Hosted by granites or metasedimentary rock of the Schist-Metagreywacke Complex, these veins are generally <1 m thick and fill fracture sets. They contain a high proportion of quartz and few minerals such as K-feldspars and micas (Roda-Robles et al., 2016). Accessory minerals such as Nb-Ta oxides, cassiterite and sulfides are common, and Li-bearing minerals consist of the montebrasite-

amblygonite series (Table 1). Note that only few Li_2O values are reported for these occurrences (i.e., 0.45% Li_2O on average for the Argemela mine, PANNN, 2017) and, except for the Argemela mine, such deposits appear to be relatively uneconomic in view of their small size.

2.1.5 Tosudite mineralization related to gold deposits

An occurrence of Li-bearing tosudite in the the Châtelet gold deposit (France) was reported in several studies (Braux et al., 1993; Piantone et al., 1994); Li_2O values range from 142 to 920 ppm. These authors suggested that the Li is related to late hydrothermal fluid circulation, itself related to the RMG emplacement in the northern part of the French Massif Central.

2.2 Sedimentary/hydrothermal/supergene deposits

These types include deposits related to either sedimentary rocks affected (or not) by hydrothermal processes, or surficial rocks affected by supergene weathering.

2.2.1 Jadar deposit type

In 2004, Rio Sava (a subsidiary of the Rio Tinto mining corporation) discovered the Jadar Basin in Serbia, now considered as a "non-conventional" world-class lithium deposit through the occurrence of the mineral jadarite (Stanley et al., 2007), a lithium boron silicate (Table 1). The basin consists of a relatively large (>20 kilometers long) intramontane lacustrine (paleo)-evaporite basin composed of dolomite, marble, various siliciclastic sedimentary rocks, pyroclastic units and notable oil shales (Obradovic et al., 1997). The mineralization is hosted in a 400 to 500 m thick Miocene sedimentary unit dominated by calcareous claystone, siltstone, sandstone and clastic rocks, unconformably overlying a Cretaceous basement composed of various metasedimentary rocks, limestone, sandstone and granite, including Miocene intrusions (Fig. 1D).

The lithium and borate mineralization occurs as 1.5 to 35 m thick stratiform lenses of three gently dipping tabular zones covering a surface area of 3 by 2.5 km. The ore is composed of jadarite-bearing siltstone and mudstone with locally interbedded sodium borate lenses (i.e., ezcurrite, kernite, borax; Fig. 1D). Jadarite occurs as 1-10 mm white and rounded grains, nodules or concretions in the siltstone or mudstone matrix that contains various amounts of calcite, dolomite, K-feldspar, rutile, albite, pyrite, muscovite and ilmenite (Fig. 2F; Rio Tinto, 2017; Stanley et al., 2007). In 2017, the mineral resources were reported as 135.7 Mt of ore at a grade of 1.86% Li_2O and 15.4% B_2O_3 (Rio Tinto, 2017), representing a giant deposit of 2.524 Mt of Li_2O.

2.2.2 Mn-(Fe) deposits

Among the various types of host-rocks for Li-bearing minerals in Europe, the small-scale and discontinuous Mn-Fe-rich sedimentary units (e.g., Drosgol Mine in Scotland and Clews Gill in England), exploited in the 19th century for their Fe and Mn contents, are a favorable site for secondary

Li-oxides such as lithiophorite (Table 1). Stratigraphically, they can be subdivided into two distinct units: 1) a reddish siliciclastic host rock, mainly pelite, shale and/or sandstone, enhanced in Mn and Fe relative to the average shale composition; and 2) discontinuous Mn lenses or layers (coticules). The minerals include cryptomelane, goethite, hematite and other manganese oxides including Li-rich lithiophorite, chalcophanite and pyrolusite.

The Li_2O content in lithiophorite is relatively low (1.23 wt.% for Li_2O versus 55 wt.% for MnO), indicating an uneconomic lithium grade. However, its occurrence is relatively important in view of the sedimentary and European histories (cf. Section 3 hereafter).

2.2.3 Bauxite deposits

Similar to the Mn-Fe deposit type, bauxite deposits can contain various amounts of lithiophorite, cookeite (Table 1) and tosudite (Li-rich gibbsite; Nishyama et al., 1975). Lithiophorite is the most common Li-bearing manganese oxide mineral in karst bauxites. It occurs in several localities, such as the Halimba, Fenyöfö and Kincsesbanya deposits in Hungary, where aluminum and gallium in bauxite deposits were exploited from the 1950s to recently (Anderson, 2015).

Al and Mn are of first interest in this deposit type. The presence of lithiophorite and local cookeite reflect Li enrichment, though this is not systematically evaluated. However, values of up to 0.53% Li_2O within bauxite are known from China (Wang et al., 2013) and the USA (Tourtelot and Brenner-Tourtelot, 1977).

2.2.4 Other lithium deposit types

A few other deposit types show relatively minor anomalous lithium contents, including:
1) Mississippi-Valley type (MVT) deposits that include some lithiophorite (Usingen, Germany), and
2) Aalenian black shales from the Dauphinois region, Isère, France, where cookeite is disseminated in black shale and in tension gashes crosscutting the shales (Jullien and Goffé, 1993). For the latter occurrence, values (Henry et al., 1996) are in the range of 9 to 1 847 ppm Li_2O with an average of 441 ppm Li_2O (n=10). These lithium occurrences are symptomatic for local conditions allowing minor enrichment, implying that they are not economically significant regarding their Li content.

2.3 Lithium resources and reserves

In Europe, lithium resources and reserves have been estimated or evaluated according to a CRIRSCO (i.e., Committee for Mineral Reserves International Reporting Standards) reporting system, or based on historical evaluations, for only 35 sites (Table 4). Thirsteen occurrences in England, Ireland, France and Germany, such as Beauvoir, Montebras, St Austell or Aclare, were evaluated before systematic reporting was established, and their mineral-resource and -reserve estimates are mostly based on historic evaluations by geological survey organizations. Fifteen projects, including Jadar, are defined in the Australian JORC classification; one project (Zinnwald) was defined in the European PERC; one

project (Alberta I) is defined in the Canadian NI43-101 system; Four projects in Ukraine are defined in an unknown system (Table 4); and one project is defined in the United Nations Framework Classification (UNFC). This list reflects the active exploration of lithium in Europe.

However, most of these deposits report mineral resources and only five specify reserve values, indicating that a feasibility study was carried out. This generally includes a study of potential processing and metallurgical-treatment methods, resulting in an economically feasible recovery of lithium from the various Li-bearing minerals.

In order to compare the Li-content in known Li-deposits, the sum of ore production + ore reserves + ore resources has been converted into contained Li_2O (ore tonnage x ore grade) for each deposit. These deposits were then categorized and classified into categories A, B, C, D and E[1] according to their commodities and their reported mineral resources and -reserves (Table 4), following the system of the European ProMine database (Cassard et al., 2015). Note that we consider only the A, B, C and D categories as (potential) lithium deposits (Table 4), which corresponds to 28 deposits.

Among them, three deposits are identified as category A, including the Cinovec (Czech Republic), St Austell (UK) greisen and the Jadar deposit (Serbia). However, the historical estimate by the British Geological Survey for the St Austell deposit may be unrealistic, as it was based on extraction from an area of about 92.5 km^2 on the edges of protected landscape zones (British Geological Survey, 2016).

Lithium occurrences appear to be well distributed in Europe. However, the Iberian area and Finland regroup most of the identified lithium deposits (Table 4), indicating that these countries are relatively active in lithium exploration and suggesting that they have a strong Li-potential.

3. Lithium metallogeny in Europe throughout the Earth's evolution

In Europe, several orogenic events throughout geological history are associated with lithium mineralization. In this section, we assess and contextualize the lithium mineralization to orogenic features in order to establish—if possible—potential metallogenetic settings.

As a reminder, Europe's landmass results from a long geological history spanning 3.6 billion years, including the assemblage of numerous continental blocks. The European lithosphere can be broadly divided into two large regions: 1) The old East European Craton, partly covered by weakly deformed Phanerozoic and Meso- to Neo-Proterozoic rift and platform successions, mostly located in eastern and north-eastern Europe; and 2) A thinner, dominantly Phanerozoic, lithosphere, accreted to

[1] Category refers to: Category A ≥ 1,000,000 t Li_2O; 1,000,000 t ≥ Category B ≥ 100,000 t Li_2O; 100,000 t ≥ Category C ≥ 50,000 t Li_2O; 50,000 t ≥ Category D ≥ 5,000 t Li_2O; Category E < 5,000 t Li_2O, based on the sum of production + reserves + resources.

the East European Craton during Palaeozoic and younger orogenies, mostly in Western Europe (e.g., Gee et al., 2006; Artemieva and Thybo, 2013).

3.1 Hard-rock lithium mineralization in European Archean to Paleo-Proterozoic terranes

3.1.1 The Ukrainian Shield (3.5 to 1.9 Ga)

The Ukrainian Shield forms an assembly of Precambrian crystalline megablocks that is 900 km long and 60-150 km wide in the central part of the country (Fig. 3A). This area is fault-bounded by the younger Dnister-Fore Black Sea and the Dniprovsko-Donnetska metallogenic provinces, and was affected by three distinct magmatic events at *ca.* 3.2, 2.6 and 1.9 Ga (Vinogradov and Tugarinov, 1961), which may have been partly coeval with the Svecofennian magmatism (2.1 to 1.8 Ga).

Several Li-rich deposits and occurrences are reported, such as: 1) the spodumene- and petalite-subtype of lithium-cesium-tantalum (LCT) pegmatites that occur in the Dnester-Bug (Podolia) and Azov Megablocks (e.g., Krutaya Balka, Nadyia); and 2) zinnwaldite and lepidolite occurrences in mixed miarolitic niobium-yttrium-fluorine (NYF)-LCT pegmatites (e.g., Volodarsk-Volynsky) and rare-metal granites (RMG), such as the Perzhanskoe ore district) in the Northern Volyn Megablock (Kvasnista et al., 2016).

Although these lithium occurrences are known, only very little information is available. This makes their evaluation, regarding metallogenic settings and geological context from a European perspective, very difficult.

3.1.2 The Svecofennian orogenic belt (2.1 to 1.8 Ga)

The Svecofennian orogenic belt (Fig. 3) is part of the Columbia/Nuna supercontinent accretion that took place from 2.1 to 1.8 Ga (Zhao et al., 2002). It consists of magmatic-arc accretionary phases joining the juvenile Svecofennian arc terrane to the Archean Karelia Craton (Nironen, 1997) along the Luleå–Kuopio thrust zone (Fig. 3; Zhao et al., 2002).

Prior to the Svecofennian orogen, continental break-up of the Karelian Province led to the formation of an ocean basin and deposition of sedimentary units such as the 1.92 Ga Pohjanmaa schist belt. Initial accretion started at 1.91 Ga and ended at 1.87 Ga, followed by large-scale extension in a back-arc setting (1.87-1.84 Ga) shown by psammites/pelites and intruded by granites and mafic dikes (Korja et al., 2006). An oblique continent-continent collision occurred from 1.87 to 1.79 Ga, illustrated by the advancing accretion of retro-arc fold-and-thrust belts with alkaline bimodal magmatism (e.g., Lahtinen et al., 2009). Significant lithospheric thickening, local migmatization and formation of S-type granites in southern Finland and central Sweden occurred as well. Finally, gravitational collapse ended the orogen between 1.79 and 1.77 Ga. Two major amphibolite grade metamorphic events are recorded at 1.88-1.87 Ga (Lecomte et al., 2014) and 1.83 to 1.80 Ga (Eilu et al., 2012).

Within this geological framework, lithium mineralization took place in metasedimentary and metavolcanic units along major fault and shear zones. They are dated as relatively late, between 1.8 to 1.79 Ga (Fig. 3, Table 5; e.g., Alviola et al., 2001), post-dating local migmatization. They include the *ca.* 1.88-1.86 Ga Vaasa Migmatite Complex on the margins of the Evijärvi belt (Suikkanen et al., 2014), and the *ca.* 1.84-1.82 Ga late-orogenic migmatizing microcline granites in southwestern Finland (Kurhila et al., 2005), and appear coeval with the regional amphibolite-grade metamorphism. The mineralization occurs as LCT pegmatite fields, such as the Kaustinene and Somero-Tammela fields (Fig. 3). The Kaustinene one occurs in the 1.92 Ga Pohjanmaa schist belt comprising the Länttä, Syväjärvi and Outovesi deposits, which are albite-spodumene pegmatites owned by Keliber Oy. In the Somero-Tammela region (Fig. 3), the petalite/spodumene Luolamaki, Hirvikallio and Kietyömaäki LCT pegmatites, owned by Nortec Minerals Corp., are hosted in the Håme belt that consists of metavolcanic rock intercalated with metagreywacke and metapelite. In these pegmatites, petalite was formed first and later converted to spodumene, suggesting a temperature decrease at constant pressure during crystallization that involved rapid cooling of the terranes (Eilu et al., 2012). Triphylite is reported from several LCT pegmatites, mainly in Sweden.

3.1.3 The Sveconorwegian orogenic belt (1140 to 850 Ma)

Part of the Grenvillian orogeny, the Sveconorwegian orogenic belt is related to the collision between Fennoscandia and an undetermined major plate (likely Amazonia), which contributed to the Rodinia supercontinent assemby (e.g., Li et al., 2008). The orogen spans from 1140 to 850 Ma, amalgamating Mesoproterozoic (1750-1500 Ma) lithotectonic units separated by major shear zones (Bingen et al., 2008a, b); according to these authors, the orogen can be divided into several tectonic phases. Among these, the Arendal phase (1140-1080 Ma) marks the collision between the Idefjorden and Telemarkia terranes (Fig. 3). This initial phase was related to closure of an oceanic basin, and subsequent accretion of a volcanic arc and a high-grade metamorphic event (*ca.* 1140-1125 Ma). The Adger phase (1050-980 Ma) corresponded to oblique continent-continent collision, and underthrusting and burial/exhumation of the Idefjorden Terrane (Fig. 3). This phase was contemporaneous with crustal thickening of the Telemarkia Terrane, when widespread syn-collisional magmatism was followed by high-grade metamorphism. Finally, the Dalane phase (970-900 Ma) corresponded to gravitational collapse, associated with post-collisional magmatism, and formation of a gneiss dome and core complex (930-920 Ma) with low-pressure/high-temperature metamorphism.

In this context, lithium mineralization occurred within polymetamorphic Paleoproterozoic amphibolite gneiss, gabbroic amphibolite and metadiorite, mainly within the Idefjorden and Telemark terranes. In the latter, the Evje-Iveland pegmatite field, recognized as the largest one (Fig. 3; e.g., Birkeland and Frikstad), comprises over 400 pegmatite bodies. These were dated at *ca.* 909±14 Ma (Scherer et al., 2001; Table 5), appear to be unrelated to granites, but are coeval with late regional

partial melting and crustal collapse. Among these NYF pegmatites, several indicate a late-magmatic event shown by a REE-depleted replacement zone consisting of "cleavelandite", amazonite, quartz and muscovite, suggesting overprinting of a LCT magma onto pre-existing NYF pegmatite bodies (Černý, 1991a,b). Lepidolite and zinnwaldite are reported from these pegmatites, which are considered as mixed NYF-LCT pegmatites, although the Li enrichment is related to the replacement zones.

3.2 Hard-rock lithium mineralization in European Neoproterozoic to Neogene terranes

3.2.1 The Cadomian orogenic belt (620-540 Ma)

In Europe, the Cadomian orogeny is characterized by a continental magmatic arc, which occurred during the Ediacaran along the rim of the West African Craton and resulted in opening of the Rheic Ocean between the Avalonia and Armorica microplates, respectively associated with the Laurentia and Gondwana supercontinents. This took place from Cambrian to Ordovician (Fig. 4; e.g., Linnemann et al., 2008; Nance et al., 2012).

A notable relic of this event is the occurrence of discontinuous Cambrian-Early Ordovician Mn-(Fe) rich metasedimentary rocks in Scotland, Wales, the Lake District of England, Belgium and Germany (Fig. 4; Kroner and Romer, 2013). Here, Li-bearing minerals such as lithiophorite can occur. Such occurrences are restricted to the Avalonian Shelf, constrained by the Rheic suture in the south, and were formed by weathering of the Cadomian continental magmatic arc at the edge of the peri-Gondwana plate in an extensional regime (Romer et al., 2011). They were formed during the first stage of the orogeny (*ca.* 590-570 Ma) and are not stratigraphically correlative, but can be found along the Avalonia Shelf from Nova Scotia through the Government Point Formation of the sedimentary Goldenville Group (Canada; White, 2008) to Poland. Kroner and Romer (2013) suggested that coeval and similar deposits may be found in the southern part of the Ossa Morena Zone (Spain).

3.2.2 The Caledonian orogenic belt (475-380 Ma)

The Caledonian orogeny was a series of tectonic events related to the closure of the Iapetus Ocean (McKerrow et al., 2000), reflecting Ordovician-Silurian oblique interactions between the Laurentian (Scotland), Avalonia (Ireland) and Baltica terranes. The initial Grampian phase (475-460 Ma) consisted, at the north end of the Iapetus Ocean, of collision between the Laurentian continental margin and an intra-ocean island arc complex. This resulted in the emplacement of S-type granites in the NW highlands of Scotland and was followed by oblique subduction under the Laurentian (north), Avalonian (south) and Baltica (east) terranes (Fig. 5A). In the Late Silurian (425 Ma), the Iapetus Ocean was closed and continents collided with the Laurentian Terrane along the Iapetus Suture Zone (Fig. 5A). Widespread calc-alkaline magmatism occurred from *ca.* 425 to 380 Ma as a post-subduction event (Miles et al., 2016), related to orogen-wide sinistral transtension induced by subsequent episodes of lithospheric extension during the Early Devonian (Brown et al. 2008).

In this Caledonian context, LCT pegmatites are known from Scotland and Ireland. In Scotland, the Glenbuchat pegmatite lies in the northern part of the Iapetus Suture Zone, hosted by Dalradian metasedimentary rocks of the Grampian Terrane. It consists of lepidolite and elbaite rich pegmatite (Fig. 5B; Jackson, 1982). The Dalradian Inzie Head gneiss and Grampian granite are associated with the *ca.* 470 Ma Grampian migmatization (Johnson et al., 2001).

In Ireland, the *ca.* 412 Ma Leinster LCT pegmatite field (Table 5; Barros, 2017) that includes the spodumene Aclare and Molyisha pegmatites, shows a relatively late time of formation (Fig. 5B). The pegmatite field is hosted by the *ca.* 417-405 Ma poly-phase Tullow Lowlands pluton (Fritschle, 2016) along the East Carlow Deformation Zone and includes up to 60 wt.% spodumene (Luecke, 1981). Thus, their emplacement may be related to a transtensional regime in this late orogenic process.

3.2.3 The Variscan orogenic belt (400-250 Ma)

The European Variscan orogen extends from southern Iberia to northeastern Bohemia, forming a 3000 km long and 700-800 km wide belt. It results of Late Paleozoic convergence and collision of the Gondwana (south) and Laurasia-Baltica (north) megacontinents along the Variscan Front (Fig. 6), involving several intermediate microcontinents and closures of oceanic domains (e.g., Matte, 1986, 1991).

The earliest continental collision started locally in the Early Devonian (385-380 Ma) with migmatization and related anatexis of continental crust, as well as exhumation of Late Silurian rocks along a regional deformation event. In the Middle-Late Devonian (360-350 Ma), arc and back-arc magmatism occurred in the northern Gondwana margins and Central Armorican Domain, attesting of southward subduction and subsequent closure of the Rheic Ocean (Fig. 6; Faure et al., 2005). This event was associated with a variable pressure-temperature metamorphic and deformation event. Late Visean synorogenic extension related to a synorogenic collapse of the inner zones occurred along NW-SE stretching and 333 to 326 Ma migmatization (Faure et al., 2005). Finally, post-orogenic collapse took place around 300 Ma. It was coeval with N-S extension, development of intramontane coal basins and *ca.* 306 Ma local migmatization (Faure et al., 2005). These events appear to have been diachronous throughout the Variscan orogeny.

Considerable amounts of granitic intrusions and several districts of RMG/greisen and LCT pegmatite deposits illustrate the Variscan orogeny. At the scale of the belt, such deposit types are relatively late in the orogeny, coeval with crustal extension together with regional partial melting and melt emplacement. Thus, in the Bohemian Massif, the easternmost part of the European Variscan belt (Fig. 6), greisen and RMG are common in the Saxothuringian and Teplá-Barrandian zones consisting of Neoproterozoic basement (e.g., Matte et al., 1991). The Moldanubian area contains mainly LCT pegmatites (e.g., Cháb et al. 2010, Ackerman et al., 2017), which appear to be spatially related to migmatitic domes and shear zones. According to Melleton et al. (2012), two ages of pegmatite

emplacement were identified, including an independent orogenic stage in the Bohemian Massif with LP-HT regional metamorphism related to significant reheating and anatexis; they note that the emplacement of LCT pegmatite here is the oldest known magmatic event of the Variscan orogeny.

In France, the northwestern part of the Massif Central is a favorable area for rare-element magmatic bodies (Marignac and Cuney, 1999). This province can be divided into three distinct deposit types (Table 5): 1) rare-metal granite such as the 317±6 Ma Beauvoir and the 314±4 Ma Montebras (Aubert, 1969; Cuney et al., 1992, 2002); 2) rare-metal rhyolite represented by the 313±3 Ma Richemont rhyolite (Raimbault and Burnol, 1998); and 3) LCT pegmatites such as the Mont d'Ambazac rare-element pegmatite field (e.g., Raimbault et al., 1995; Deveaud et al., 2013). The latter includes the 309±5 Ma lepidolite-subtype LCT Chédeville pegmatite, which postdates the 324±4 Ma host granite (Hollinger et al., 1986) and appears to be sub-synchronous with local partial melting (315±4 and 316±4 Ma; Gébelin et al., 2009) and with shearing (La Marche shear zone: 316±5 to 312±2 Ma, Gébelin et al., 2007, 2009). This east-west La Marche fault system, located in the northern part of the Limousin, appears to have been a key-control on magmatic activity (Cuney et al., 2002).

The Galicia-Trás-os-Montes Zone (GTOMZ) and the Central Iberian Zone (CIZ) in the Iberian Variscan belt host widespread LCT pegmatite fields. At least five main mineralized pegmatite fields are recognized in the former from north to south: Forcarei-Lalín, Serra de Arga, Barroso- Alvão, La Fregeneda-Almendra and Gonçalo-Guarda. In the CIZ, the 326±3 Ma Argemela granite is the only RMG known from Iberia (Charoy and Noronha 1991; 1996). The average age of intrusion of LCT pegmatites in the GTOMZ is 310±5 Ma, whereas in the CIZ and in the southern GTOMZ the ages are younger: 301±3 Ma (Melleton et al., 2011), 295.1±4.1 Ma and 296.4±4.1 Ma (Roda-Robles et al., 2009; Vieira, 2010). Moreover, late quartz-montebrasite hydrothermal veins are reported from several areas in the CIZ (e.g., Roda-Robles et al., 2016).

Finally, in the Austroalpine unit of the Eastern Alps, Permian LCT spodumene-bearing pegmatites are known (e.g., Thöni and Miller, 2000; Ilickovic et al., 2017). These pegmatites appear to be coeval with lithospheric extension, causing crustal basaltic underplating, HT and LP metamorphism, as well as intense magmatic activity (Schuster and Stüwe, 2008).

3.2.4 The Mediterranean and circum-Mediterranean orogens (Mesozoic-2.5 Ma)

Several styles of lithium mineralization are contemporaneous with the circum-Mediterranean and Mediterranean Tethys mountain belts, such as the Carpathians Mountains or the Egean Domain, which resulted from oceanic closure and collision of the European continental foreland (Bohemian Massif) with the African promontory of the Adriatic microplate (Fig. 7). Rifting of the Alpine Tethys and its subsequent subduction underneath the Adriatic margin, followed by continent-continent collision, promoted widespread magmatic activity through time, as well as the development of related orogens such as the Carpathians Mountains.

The initial continental rifting of the Alpine Tethys and its related magmatism occurred from the Middle to Late Triassic in the eastern part of the Mediterranean domain (Bertotti et al., 1999; Schmid et al., 2008). In the central Alpine-Carpathian-Dinaridic orogenic system, *ca.* 242 Ma Li-phosphate pegmatites occur in the Brissago area (Switzerland; Vignola et al., 2008); these authors suggested that the pegmatites formed from partial melting of the Early Permian Ivrea gabbro.

This tectono-magmatic event was followed by development of the Adriatic passive margin in the Middle Jurassic (Bertotti et al., 1999; Schmid et al., 2008) in an extensional tectonic regime, and later by the subduction of the Tethys oceanic lithosphere beneath the Adriatic margin from Cretaceous to Late Paleogene. This crustal shortening led to the final consumption of the Neotethys Ocean associated with widespread calk-alkaline magmatism in the Carpathian arc (Fig. 7; Schmid et al., 2008) and formation of the Apennines in Italy. Meanwhile, Jurassic to Cretaceous bauxite deposits with lithiophorite are reported from Hungary and Greece (Fig. 7), suggesting a tropical climate during this period.

In the external domain of the Dauphinois zone (French Alps), an Eocene greenschist metamorphic event led to the formation of cookeite-bearing formations and -tension gashes within Aalenian black shales (Fig. 7). According to Jullien and Goffé (1993), the Li was sourced from the metasediments that themselves resulted from erosion of the continental crust.

Within the Central Alps (i.e., Penninic Zone), a kilometer-scale east-west extensional area occurs with several Oligocene-Miocene LCT pegmatites in the Vigezzo, Bodengo and Codera areas. They are Tertiary (Fig. 7; 30 to 20 Ma, Guastoni et al., 2014; Romer et al., 1996) and show a beryl-phosphate affinity with elbaite and columbite as potential accessory minerals (Guastoni et al., 2014, 2016).

Finally, extensional collapse and back-arc extension promoted development of the Miocene Pannonian and the Jadar basins within the Alpine-Carpathian-Dinarides domain along several late Oligocene–Miocene detachment zones (Jolivet et al., 2009; Menant et al., 2018; Stojadinovic et al., 2017) and in response to rapid slab roll-back (Simić et al., 2017; Stojadinovic et al., 2017). Basin formation was accompanied by calc-alkaline magmatism with a paroxysm of silicic volcanism during the early and middle Miocene (Kovács et al., 2007). Rapid exhumation of metamorphic rocks caused episodic migmatization as well as related magmatism (Bergell intrusion; Beltrando et al., 2010).

Within the northern part of the Apennines, the *ca.* 6.7-6.9 Ma (Ferrara and Tonarini, 1985) LCT pegmatites from Elba Island are famous for their gem-quality elbaites.

4. Interpretation and discussion

Considering all lithium occurrences in Europe, one of the first observations regarding their distribution is their apparent clustering (Figs. 3, 4, 5, 6, 7). This clustering defines pegmatite and/or RMG fields with similar ages of emplacement, suggesting a relatively coeval magmatic activity related to common endogenous processes. Furthermore, the Li-rich sedimentary basins such as Jadar reflect late sedimentary/hydrothermal Li re-concentration through exogenous processes.

Endogenous processes related to lithium mineralization

We have identified several Li-magmatic events through time as illustrated by RMG, greisen and LCT pegmatites (Table 5; Figs. 3, 4, 5, 6, 7, 8), ranging from Paleoproterozoic to Miocene. These events occurred during times of collisional orogeny, including the Svecofennian (2.1-1.8 Ga; Fig. 3), Sveconorwegian (1140-850 Ma; Fig. 3), Caledonian (490-390 Ma; Fig. 5), Variscan (400-250 Ma; Fig. 6) and Mediterranean (Mesozoic-2.5 Ma; Fig. 7) orogenies. These events were mainly related to supercontinent formation, as observed elsewhere by Bradley (2011). Accordingly, in the Svecofennian orogen, LCT pegmatite ages are 1.8-1.79 Ga This suggests relatively late emplacement in the orogenic cycle that postdated arc accretion and the first regional metamorphism, and might be related to crustal thickening as well as to a late amphibolite-facies metamorphic event.

During the Sveconorwegian orogen, emplacement of Li-rich pegmatites (910-906 Ma) appears coeval with the late Dalane phase (970-900 Ma), corresponding to gravitational collapse and post-collisional magmatism, as well as the formation of a gneiss dome and core complex related to low pressure/high temperature metamorphism. The Scottish and *ca.* 412 Ma LCT pegmatites from Ireland, which are part of the Caledonian orogenic belt, appear coeval with crustal thickening and post-subduction magmatism.

In the Variscan belt, RMG (Beauvoir, Montebras and Richemon, France; Argemela, Portugal; St Austell, UK), greisen (Cligga Head, Tregonning-Godolphin, Meldon, UK; Dlha Dolina, Slovakia; Montebras, France; Krasno-Konik, Krupka, Czech Republic) and various LCT pegmatites (Table 5, Fig. 6) are widely distributed (Fig. 8). Their ages suggest mostly emplacement during the Late Carboniferous (Table 5) reflecting highly fractionated magmatic events throughout the European core related to a post-collisional stage ending the Variscan orogeny *sensu stricto* (Fig. 6, Table 5; e.g., Bonin, 1998; Chen et al., 1993; Cuney et al., 2002; Melleton et al., 2012; Neace et al., 2016). Moreover, from the internal to the external orogenic domains Li-magmatism appears to be diachronous, indicating southward prograding Li-rich magmatic activity traversing the entire belt. In the internal zones (France, Germany, Czech Republic and NW Iberia) RMG, greisen and LCT pegmatites were mostly emplaced between 320 and 307 Ma corresponding to the Bavarian phase (330-315 Ma; Finger et al., 2007) in the Bohemian Massif and to synorogenic collapse and NW-NE stretching in the French Massif Central (320-310 Ma). In the external zones (UK, parts of Spain and

Portugal), similar deposits tended to be emplaced around 305 and 301 Ma (excluding the *ca.* 326 Ma Argemela granite) suggesting late-orogenic magmatism (Melleton et al., 2015). Finally, deposits belonging to the Gemeric unit in the Western Carpathians of Slovakia, as well as the Austroalpine pegmatites of the Eastern Alps that form the extreme margins of the belt, indicate Permian ages coeval with regional partial melting (Finger et al., 2003, Petrik et al., 2014; Ilickovic et al., 2017).

Thus, it appears that the emplacement of RMG, greisen and LCT pegmatites was relatively late in the orogenic cycle and may have been coeval with continent-continent collision, commonly postdating arc accretion. It could also be related to crustal thickening (Alviola et al., 2001), a favorable setting for crustal peraluminous melt (Cuney et al., 1992; 2002) through wall-rock assimilation, the unmixing of restite, and/or internal fluid circulation related to convective fractionation (Lehmann, 1994; Martin and De Vito, 2005).

Importantly, it also appears that the reported greisen were developed from RMG, suggesting that most of the greisen associated with fractionated S-type peraluminous granite may not show significant Li contents. The formation model involves early exsolution of an F-CO_2-H_2O-rich aqueous phase from the granitic magma, along with fluid/rock interactions leading to dissolution/precipitation and re-concentration of incompatible elements, such as Li, F, Sn, W, etc., within the greisen (Heinrich, 1990). Here: 1) miarolitic cavities are common and reflect volatile saturation; and 2) a decrease in rock volume and increase of porosity are reported. In the 321.5±3 Ma Cinovec deposit, dissolution of the protolithionite—formed during magmatic intrusion—and precipitation of zinnwaldite during greisenization have led led to remobilization of Li into its final host mineral (Johan and Johan, 2005).

4.2 Exogenous processes related to lithium occurrences

Several Li-occurrences in Europe reflect a concentration of lithium in sedimentary rocks through various exogenous processes, such as hydrothermal circulation and/or erosion and transport. Thus, distinct occurrence types can be distinguished.

4.2.1 Jadar deposit type

The Jadar deposit type is exclusively Neogene (Oligocene to Pliocene), based on available data. The existence of several other isolated intramontane lacustrine evaporite basins is suggested in Serbia (Fig. 8) as well as in Bosnia, such as the Valjevo-Mionica or Lopare basins from where jadarite was reported. Interestingly, the subsurface of these basins includes LCT pegmatite and Cretaceous to Miocene granitic intrusions, suggesting local lithium enrichment in the basement (Stojadinovic et al., 2017). These basins were formed during the late stage of the Dinadiric orogeny (Fig. 7), coeval with the extensional collapse and back-arc extension due to Carpathian slab retreat (Simić et al., 2017).

Jadarite precipitation is poorly constrained. Some authors suggested that interaction between clastic sedimentary rocks and the surrounding brine—possibly involving hydrothermal devitrification

and hydration of andesitic-dacitic pyroclastic material or alteration of clay minerals—may contribute to its precipitation (Stanley et al., 2007; Stojadinovic et al., 2017).

4.2.2 Mn-(Fe) deposits

Two distinct periods of Mn-(Fe) precipitation in Europe are described here. A first group of deposits in Scotland, Wales (e.g., Drosgol Mine), England (Clews Gill), Belgium (Ottré, Beez) and Germany (Harz) is Cambrian to Early Ordovician in age (Fig. 8; Waldron et al., 2011; Romer et al., 2011), representing a notable relic of the Cadomian orogeny (650-550 Ma; Fig. 4). They are exclusively located in the Avalonian plate, constrained by the Rheic Suture, and are formed from weathering of the Cadomian magmatic arc of the Gondwana plate, as suggested by Nd and Sr isotopes (Romer et al., 2011). Interestingly, in Europe, the Cadomian orogeny was subsequently reworked during the Caledonian and Variscan orogenies (Zelazniewicz et al., 1997; Melleton et al., 2012).

The second group is mainly found in Hungary, where the Eplény and Urkut Mn deposits are Jurassic in age (Polgari et al., 2005; Figs. 7, 8). These deposits are associated with marine sedimentary rocks mainly composed of bioclastic limestone and black shale.

The Li-bearing mineral lithiophorite, as most Mn- and Fe-oxides, was formed from secondary fluid circulation in the host rock (Nicholson and Anderton, 1989). Romer et al. (2011) suggested that the lithium component derived from chemical weathering of continental crust and was originally concentrated in siliciclastic or carbonate rocks. Thus, late hydrothermal fluid circulation may have remobilized—and still remobilizes—the Li_2O content via dissolution/precipitation processes , thus helping the precipitation of Li-bearing oxides under oxidizing conditions. Moreover, several authors pointed out their distribution along regional faults that are favorable sites for fluid circulation, such as the Candwr Fault in Wales (Cotterell et al., 2009) and the Red Gill Fault in the UK (Clark, 1963).

4.2.3 Bauxite deposits

Li-bearing minerals in bauxite deposits are Cretaceous in Hungary (D'argenio and Mindszenty, 1986) and Jurassic to Cretaceous in Greece where they are located along the northern shores of the Mediterranean Sea (Bardossy, 1982).

In Hungary, these deposits are stratiform where bedrock is a non-uniform karst carbonate rock, in which the bauxite horizons are relatively large. The Halimba mining district is one of the largest, with bauxite thickness varying from 1 to 40 m. Here, the Li-bearing mineral lithiophorite originated from secondary fluid circulation through the host rock (Bardossy, 1982) forming Mn-rich layers in epi- and supergene crusts. Cookeite is also reported from these deposits (Bardossy, 1982).

4.3 Discussion of rare-metal magma formation

There are currently two distinct models of rare-metal magma formation. The first one involves the escape of late-stage melts ending the crystallization of huge highly fractionated felsic magma chambers (e.g., Jahns and Burnham, 1969; London, 1992). One of the major arguments for this model is the regional zoning of pegmatite bodies in the margins of supposed parent granites (e.g., Cameron et al., 1949; Černý et al., 2005). An example is the Fregeneda-Almendra pegmatite field where crystal-fractionation modelling and geochronology support a magmatic origin (Vieira 2011; Roda-Robles et al., 2016). Moreover, the widespread presence of pegmatites and RMG in granites (e.g., London, 1992; Černý et al., 2005) suggests a granite-related origin (e.g. Roda-Robles et al., 2016).

However, some aspects disagree with this model, suggesting a second model that involves low-grade partial melting of crustal sequences (e.g., Norton, 1973; Zasedatelev, 1977; Stewart, 1978; Melleton et al., 2011; Müller et al., 2015; Bongiolo et al., 2016). In southern Ireland, geochemical evidence points to the absence of a relationship between the LCT-spodumene Aclare pegmatite field and the surrounding Tullow Lowlands and Blackstairs plutons that are part of the Leinster Granite. This absence concerns their capacity of generating a residual pegmatite melt (Barros et al., 2016). These authors suggested a separate partial-melting event for both units, although the intrusion of the granitic unit may have triggered anatexis of the surrounding sedimentary rocks.

In the French Massif Central, the Monts d'Ambazac pegmatite field has δ^7Li mica values that are consistent with a crustal metasedimentary source. It is also coeval with evidence of a partial-melting event, excluding the influence of magmatic fractionation from the nearby St Sylvestre granite in the formation of pegmatites. It also demonstrates that strong Li enrichment in pegmatite is not related to a fractionation process (Deveaud et al., 2015). In the Bohemian Massif and in Austria, LCT pegmatite fields are also supposed to be formed by partial melting processes (Melleton et al., 2012).

Thus, a model of partial melting during anatexis of sedimentary rock (evaporites, cookeite-bearing metapelite, Li-rich metasedimentary rock, or Li-rich Ordovician orthogneiss, etc.) may be applied (Fig. 10), involving coeval emplacement for S-type granite (e.g., Kontak et al., 2002) and nearby LCT pegmatite, but not promoting a "parental" relationship. Moreover, micas, garnet and staurolite, which are widespread in metasedimentary rock, are seen as a potential source for lithophile elements such as Li (London, 2005, 2018). For instance, in the Brissago-Valle di Ponte area (Switzerland-Italy), poorly fractionated LCT phosphate pegmatites are suspected to be derived from local partial melting of kinzigites during high-temperature metamorphism (Vignola et al., 2008). At the European scale, various Li-bearing minerals are reported from pegmatites, including Li-micas, lepidolite, spodumene and petalite, involving variations in the fluxing content (F, B and P) of the magmatic fluid (Roda-Robles et al., 2010), as well as varying P/T conditions (e.g., spodumene versus petalite; London, 1986, 1990). These variations are also seen in the Scandinavian orogenies, where zonation of Li-bearing

minerals is highlighted: 1) the Svecofennian LCT pegmatites are associated with Li-silicate and Li-phosphate minerals; but 2) the Sveconorwegian LCT pegmatites show a Li-phyllosilicate affinity.

4.4 From source to sink

As suggested above, several parameters may control the lithium mineralization locations in Europe. The observed clustering of endogenous lithium deposits such as LCT pegmatites, RMG and/or greisen may involve a crustal anomaly (>20 ppm; Rudnick and Gao, 2004), or a Li "pre-concentration" related to paleoenvironmental sedimentation conditions (e.g., type of basin, host rock, climate) and/or post-deposition processes (weathering, basin-fluid circulation in the crust), more generally preserved along a paleo passive-margin (Fig. 9; Romer and Kroner, 2015).

In any case, this involves the existence of a primary Li-source that can be of magmatic origin, such as erosional material from a continental magmatic arc and related Mn(-Fe) deposits and lithiophorite occurrences. Another possibility is a sedimentary origin, such as the Schist-Metagreywacke Complex in the Galicia-Trás-Os-Montes Zone (Roda-Robles et al., 2016), or the Pohjanmaa schist belt in Finland and related LCT pegmatites (Eilu et al., 2012). In that respect, significant lithospheric thickening may be favorable for concentrating Li in a specific location (Eilu et al., 2012). The occurrence of several types of Li mineralization in the French Massif Central (Fig. 6; LCT pegmatite, greisen, RMG, Li-bearing tosudite) forming clusters may suggest a possible common Li-source (Cuney and Barbey, 2014). Moreover, a recent isotopic δ^7Li study of pegmatites (Deveaud, 2015; Deveaud et al., 2015), suggested that beryl-columbite and lepidolite-petalite LCT pegmatites from the Monts d'Ambazac show a distinct crustal contribution, indicating that Li-rich sources may contribute to "secondary" lithium deposits if suitable processes are involved.

The timing of fluid circulation appears to be another important feature for Li-concentration, whether related to a regional or to a local extensional regime in an orogenic cycle (Figs. 9, 10; e.g., Eilu et al., 2012; Jolivet et al., 2009; Melleton et al., 2015; Stojadinovic et al., 2017; Menant et al., 2018). Sedimentary/hydrothermal lithium deposits are thus mainly related to regional extension (rifting or back-arc extension; Kroner and Romer, 2013; Simić et al., 2017; Stojadinovic et al., 2017), whereas magmatic-related lithium deposits are associated with local decompression and/or transtension strike-slip deformation in late continent-continent orogenic cycles, leading to the formation of a volatile-rich melt (Fig. 10). According to Kontak et al. (2002), this melt may cause over-pressuring and/or hydro-fracturing, resulting in the formation of dilatant zones and related fracture sets. Remarkably, sedimentary rocks enriched in Li during an extensional regime (e.g., Jadar Basin) may be a favorable Li source during a subsequent magmatic event. Unfortunately, a lack of data makes it impossible to confirm this hypothesis.

Finally, the distribution of Li-occurrences is strongly influenced by the location and geometry of fracture sets (Figs. 9, 10; Deveaud et al., 2012, Deveaud, 2015; Silva et al., 2018). High permeability

fractured zones seem to act as favorable channels for: 1) The emplacement of LCT pegmatite or RMG from evolved magma; or 2) Hydrothermal fluid circulation through sedimentary successions (Jadar Basin, lithiophorite occurrences in Aalenian black shales) leading to secondary Li-bearing mineral (lithiophorite, cookeite) precipitation. A recent geostatistical study showed that most pegmatites occur less than 500 m from a fault system (Deveaud et al, 2013).

4.5 Grade assessment of and tonnage estimation

4.5.1 Data quality

When regarding the available dataset, several problems are obvious. The differences in knowledge and definition of mineral resources and reserves vary between historical data (St Austell, Beauvoir), JORC (Jadar project) and NI43-101 (Alberta I project). Historical data estimates predate the CRIRSCO system (before 1995; www.crirsco.com). They are based on drilling campaigns managed by geological surveys and related subsidiaries. Such data may lead to under- or over-estimates. Figure 11 shows that projects and occurrences are distributed homogeneously despite their reference system (historical data in italics *versus* CRIRSCO system in bold). Note that mineral resources from the St Austell deposits appear to be strongly anomalous from the main population (Fig. 11, Table 4); they are unrealistic for environmental and societal reasons, and probably not economically viable regarding ore grades. For these reasons, the St Austell data are not used in the following sections. The JORC system, however, may exclude some commercially sensitive information, such as mineral reserves if mineral resources (Table 4), which is not allowed with the NI43-101 report. However, these points do not affect the mineral resources comparison on which our estimates are based.

As mentioned above, thirsteen projects in England (Meldon), Ireland (Aclare), France (Beauvoir, Montebras), Germany (Altenberg) and Ukraine were evaluated before setting up a "reporting system", as such mineral resource and -reserve estimates refer to historical evaluations. These data represent only 5% of the entire dataset (Table 4). Fifteen projects were defined in the Australian JORC system (e.g., Wolfsberg, Austria); one project (Zinnwald, Germany) in the European PERC; one in the Canadian NI43-101 system (e.g., Alberta I project which comprises Presqueria deposit, Spain) and one project is defined in the United Nation Framework Classification (UNFC; Alijo deposit).

However, we think that, despite the apparent discrepancies between reference systems, our dataset is a good starting point for estimating the European Li hard-rock potential (Figs. 11, 12).

Moreover, Li-bearing minerals in lithiophorite and Li-chlorite occurrences in sedimentary deposits—bauxite, Mn-(Fe), MVT and Li-bearing clay deposits—were not systematically described, as Li was not of first interest at the time. This was the case in Spain, France and Turkey, and may hide significant local Li grades, as in China (Wang et al., 2013) and USA (Tourtelot and Brenner-Tourtelot, 1977). Available data from bauxite deposits in China show that the average Li grade is very low

(2045 ppm Li_2O; Wang et al, 2013). The same is true for Li-bearing shales in the Dauphinois area, France, where the average Li content is 949.34 ppm Li_2O (Henry et al., 1996). Accordingly, these occurrences can hardly be considered as potential Li deposits regarding their ore grade.

Based on the available dataset, 8,839,750 t of Li_2O (Table 4) are presently reported in Europe from various deposit types and related to various Li-bearing minerals.

Furthermore, our study does not cover lithium in seawater, whose average content has been estimated at 0.17 ppm (Fasel and Tran, 2005; Yaksic and Tilton, 2009), nor that potrentially contained in oilfields (e.g., Pechelbronn in France) and geothermal brines (e.g., the South Crofty deposit in the UK; Cornish Lithium/Strongbow). Such potential lithium sources are difficult to quantify due to fluid mixing, dilution and/or movement (Houston et al., 2011), but research is ongoing (e.g., Eramet-IFPEN), as are potential resource estimates from such sources.

4.5.2 Range of ore grade and tonnage

At the deposit scale, metric tons of ore and average grades (Fig. 11, Table 4) of European hard-rock lithium deposits are relatively competitive compared to the world-class LCT pegmatites from the Greenbushes in Australia and Whabouchi in Canada, which are representative examples of such Li-deposits. In detail, the Proterozoic Ukrainian pegmatites show similar grades and tonnages, whereas Variscan pegmatites host lower tonnages. Thus, as pointed out before, significant differences in Li content are seen among the various orogens. Interestingly, the Svecofennian, Sveconorwegian and Variscan orogenies, which involved supercontinent accretion, continent-continent collision and notable late lithospheric thickening, resulted in richer lithium deposits than the Cadomian, Caledonian and Alpine orogenies. This suggests that processes involving extensive crustal anatexis from lithospheric thickening lead to significant Li enrichment (Černý, 1991a, b). For the Apine orogen, the dearth in Li deposits could be related to a present-day deep erosional level, as indicated by the presence of few LCT pegmatites emplaced at the highest structural levels, highlighted by miarolitic (Guastoni et al., 2014, 2016) features (MI class of Černý and Ercit, 2005).

It also appears that greisen and RMG containing mainly Li-micas (lepidolite, zinnwaldite, Li-muscovite) and Li-phosphates, may contain relatively higher tonnages, but lower grades, than the LCT pegmatites, which contain mainly spodumene- and petalite-dominated Li-bearing minerals (Figs. 11, 12). This is, first, a function of deposit size: pegmatites are narrow and well constrained whereas greisen and RMG can form kilometer-scale cupolas and may have deep roots (e.g., Beauvoir). Second, the type of Li-bearing mineral, within which the Li_2O content may vary significantly (Table 1), is another major parameter, illustrated by spodumene *versus* zinnwaldite.

Finally, in addition to the above points, ore grade can be controlled by several other parameters. These include the geochemistry of fluxing fluids (F, B and P), different crystallization parameters and

P-T conditions (spodumene *versus* petalite; Černý and Ferguson, 1972), and variable degrees of fractionation (Li-phosphate occurrences against amblygonite-montebrasite or triphylite-sicklerite-ferrisicklerite series; Černý, 1991b). All these may affect the number and type of Li-bearing minerals, as well as their mineral size and relative abundance, and therefore the overall Li content.

4.6 Perspectives

As emphasized by this study, lithium in hard-rock deposits is not rare in Europe and well distributed within Proterozoic to Cenozoic orogens (Fig. 8). The Variscan orogeny (Fig. 6) shows the most important Li-content (more than 60% of the identified deposits in Table 4) in various deposit types (greisen, RMG, pegmatite). The oldest orogens mainly contain LCT pegmatites (Figs. 3, 5) that tend to cluster, potentially because of successive orogenic reworking. However, only very few studies report lithium occurrences related to young Mediterranean orogens, suggesting either a lack of exploration or a significant difference between the Variscan and Alpine orogenies.

As for jadarite occurrences, greenfield exploration in Balkan countries such as Serbia and Bosnia may identify latent deposits related to lacustrine evaporite basins. Currently, this area is relatively underexplored and several exploration and mining companies showed recent interest in acquiring permits, such as the Australian firm South East Asia Resources, recently renamed Jadar Lithium.

Regarding Li-production, LCT pegmatites that generally have high lithium grades and low tonnages could be rapidly in production as Li-extraction processes for spodumene are operational. Greisen and RMG, which have low Li grades and high tonnages, will take somewhat longer to reach production as the extraction processes of Li-bearing micas must be demonstrated at deposit scale.

5. Conclusions

This review of Li hard-rock lithium metallogeny in Europe demonstrates that a wide range of deposit types, including endogenous (LCT pegmatites, RMG, greisen) and exogenous (Jadar, bauxite) processes, is involved. The lithium is contained in various Li-bearing minerals, such as spodumene, lepidolite and zinnwaldite, which are related to different orogenies through time. A favorable geodynamic setting for endogenous magmatic lithium accumulation comprises a late orogenic process, commonly postdating arc accretion, coeval with continent-continent collision, and related to local crustal thickening. A post-orogenic extensional setting is favorable for exogenous processes that can concentrate lithium into a deposit.

At present, 27 potential hard-rock deposits have been identified in Europe. The sum of such Li resources is estimated at 8,839,750 t of Li_2O. Their production may secure, in part, European lithium requirements in the near future.

Our inventory also reflects the heterogeneity in knowledge regarding lithium occurrences. This is due to a relative lack of interest in lithium until recently and suggests that new targets might be defined in the foreseeable future through active ongoing exploration.

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8. Table captions

Table 1: Main Li-bearing minerals encountered in Europe, their corresponding chemical formula, Li content (Li_2O and Li metal) and physical characteristics.

Table 2: RMG classification according to Linnen and Cuney (2005) with European examples.

Table 3: Pegmatite classification according to Černý and Ercit (2005) and Černý et al. (2012) and corresponding P/T conditions. LCT pegmatites in red show a significant lithium potential and those in orange a moderate lithium potential, with corresponding examples if known. LCT = lithium-cesium-tantalum; NYF = niobium–yttrium–fluorine.

Table 4: Li projects in Europe and their past production and estimated Li metal resources/reserves. NA refers to data not available. (data were collected from exploration and mining companies; Lulzac and Apolinarski, 1986; Smolin and Beaudry, 2015; British Geological Survey, 2016)

Table 5: Location and dating of several pegmatites, RMG and greisen deposits in Europe

9. Figure captions

Figure 1. Cross sections of various lithium deposits. Lithium-bearing units are identified in red in each section. A) Beauvoir RMG (France) and related stockwork (modified from Cuney and Autran, 1987). B) Sepeda pegmatite in Portugal (modified from Dakota Minerals, 2017); pink area represents barren pegmatite. C) Cinovec deposit in Czech Republic (modified from Breiter et al., 2017). D) Jadar Basin in Serbia and location of the jadarite layers (modified from Rio Tinto, 2017). Color should be used

Figure 2. Photographs of various styles of lithium mineralization. A) Quartz-feldspar and zinnwaldite mineralization (Podlesi, Czech Republic). B) Greisen and related La Bosse stockwork (Beauvoir, Massif Central, France). C) Montebras stocksheider hosted in RMG (Massif Central, France). D) Phenocrysts of petalite (yellowish-greenish minerals) surrounded by purplish lepidolite in quartz-potassic feldspar-albite matrix (Chédeville pegmatite, Massif Central, France). E) Alternating lepidolite-rich and aplite-rich layers in a horizontal pegmatite (Chédeville pegmatite, Massif Central, France). Scale represents 7 cm. Abbreviations: Lpd: lepidolite; Ptl: petalite; Znw: Zinnwaldite; Qz+Fsp: quartz+feldspar. F) Jadarite mineralization (Jadar Basin, Serbia) in mudstone (courtesy of Matevž Novak, Geological Survey of Slovenia) Color should be used

Figure 3. A) Schematic map of the East European Craton and distribution of the main shields (modified after Roberts and Slagstad, 2015). The red box highlights the studied area. B) Simplified geological map of the Fennoscandian Shield (modified after Koistinen et al., 2001 and Bergh et al., 2015) showing distribution of the Svecofennian and the Sveconorwegian orogens, and distribution of LCT and mixed NYF-LCT pegmatites. Red circles and surrounded red names refer to LCT pegmatite fields; blue circle and name refer to mixed NYF-LCT pegmatite field; *n* = number of identified pegmatite bodies. Abbreviations: B: Bamble Terrane; ES: Eastern Segment; I: Idefjorden Terrane; T: Telemark Terrane; SFDZ: Sveconorwegian Frontal Deformation Zone. Color should be used

Figure 4. Simplified geological map of the crustal block involved in the Caledonian orogeny along the Rheic Suture and location of contemporaneous Mn-(Fe) rich deposits (modified after Linnemann et al., 2007; Garfunkel, 2015). Note that Caledonian relics within Gondwana were reworked during the Variscan and Alpine orogenies making reconstruction of their respective contacts difficult. Color should be used

Figure 5. A) Schematic map of the Mid-Devonian paleo-continental reconstruction (modified after Woodcock et al., 2007). Red box highlights studied area. B) Simplified geological map of Ireland and northern Britain with distribution of LCT pegmatites related to the Caledonian orogeny (modified after Miles et al., 2016). Red circle and surrounded red name refer to Leinster LCT pegmatite fields from which nine LCT pegmatites are identified. Color should be used

Figure 6. Simplified geological map of the Variscan orogeny in Europe, location of various lithium-bearing deposits and a selection of ages (modified after Murphy et al., 2010; Martínez Catalán, 1990). Note that the Variscan orogeny was subsequently reworked along the Alpine Front by the Alpine orogeny. Additionally, 260 Li-rich bodies including LCT pegmatites, greisen and the Argemela RMG are identified in Portugal and Spain, 50 Li-rich occurrences are identified in the Bohemian Massif and 26 LCT pegmatites are identified in South Austria. Abbreviations: AM: Armorican Massif; AVZ: Arveno-Vosgina Zones; BM: Bohemian Massif; CAZ: Central Armorican Zone; CIZ: Central Iberian Zone; CZ: Cantabrian Zone; GTMZ: Galicia-Tras-os-Montes Zone; MZ: Moldanubian Zone; NAZ: North Armorican Zone; OS: Ossa-Morena Zone; RM: Rhenish Massif; SAZ: South Armorican Zone; SPZ: South Portugese Zone; SZ: Saxothuringian Zone; TBZ: Tepla-Barradian Zone; WALZ: West Asturian-Leonese Zone. Color should be used

Figure 7. Simplified geological map of the Alpine-Mediterranean area and location of lithium-bearing deposits (modified after Tomljenovic, 2002; Bousquet et al., 2012). Brissago and Elba Islands

pegmatite fields are highlighted by a red circle; black circle is Dauphinois region with cookeites; *n* suggests number of identified pegmatite bodies. Abbreviations: BM: Bohemian Massif; EA: Eastern Alps; EC: East Carpathians; IWC: Internal West Carpathians; NCA: Northern Calcareous Alps; SC: South Carpathians. Color should be used

Figure 8. Simplified geotectonic map of Europe (modified after Artemieva et al., 2006; Charles et al., 2013) and distribution of various Li-bearing occurrences. Color should be used

Figure 9. Location map of the Li deposits (Table 4) in Europe (modified after Artemieva et al., 2006; Charles et al., 2013). Categories refer to: A ≥ 1,000,000 t Li_2O; 1,000,000 t ≥ Category B ≥ 100,000 t Li_2O; 100,000 t ≥ Category C ≥ 50,000 t Li_2O; 50,000 t ≥ Category D ≥ 5,000 t Li_2O; Category E <5,000 t Li_2O and past production and mineral resources. Color should be used

Figure 10. Geological sections of favorable lithium setting. Continental collision (A; modified from Menant et al., 2018) shifting to post-collision setting (B; modified from Menant et al., 2018) represents favorable context for Li-hard-rock formation such as LCT pegmatites, RMG and greisen along orogenic collapse (C). Superscripts on C correspond to European examples: [1] = Chédeville, Mina Feli, or Gonçalo; [2] = Richemont; [3] = Beauvoir, Argemela and Montebras; [4] = Barroso-Alvao, Läntta and Aclare. Continental subduction (D; modified from Menant et al., 2018) shifting to back-arc setting (E; modified from Menant et al., 2018) represents favorable context for sedimentary/hydrothermal Li deposits such as Jadar (F; modified from Stojadinovic et al., 2016). *LCT* pegmatite can also occur in such a context. Color should be used

Figure 11. Average lithium grade (wt.% Li_2O) *versus* metric tons of ore (Mt) for the 28 identified Li deposits in Europe regarding their deposit type. Deposits and projects based on historical estimates are written in italics and those based on the CRIRSCO system in bold. The Whabouchi (Canada; blue triangle) and Greenbushes (Australia; green triangle) pegmatite deposits are mentioned here in order to compare these world-class deposits to European grades and tonnages. Color should be used

Figure 12. Grade (wt.% Li_2O) *versus* deposit types (A) and deposit tonnage (Mt) *versus* deposit types (B) considering resource estimates. The boxes indicate the median (black line), upper and lower quartiles (25%; gray boxes), maximum and minimum values (upper and lower whiskers) and outliers (black circles). Note that n reflects the total number of deposits considered for this summary plot.