



Title	The Origin of Spodumene Pegmatites Associated with the Leinster Granite in Southeast Ireland
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39 Rare-element pegmatites display very diverse chemical signatures and are enriched in elements
40 that are usually present in low concentrations in other rocks. These features make them important
41 sources of a wide range of commodities, including industrial minerals, gemstones and strategic
42 metals. Two alternative petrogenetic processes have been proposed to explain the formation of rare-
43 element pegmatites: 1) extensive fractionation of fertile parental granitic magma, with rare-element
44 pegmatites representing the residual phase in this process; or 2) partial melting of rare-element-rich
45 source rocks. Pegmatites geographically associated with granitic batholiths have often led to support
46 for the first hypothesis that granitic rocks and pegmatite magmas have a comagmatic relationship.
47 Several studies have successfully demonstrated the link between parent granite and pegmatites,
48 including examples in Canada (Černý *et al.* 2012) and Spain (Roda-Robles *et al.* 2012). However age
49 and/or geochemical incompatibilities, such as pegmatites being significantly older than granite, or an
50 apparent absence of parental granite, are often documented (*e.g.* Walker *et al.* 1989, Muller *et al.*
51 2015).

52 One way to approach and resolve the petrogenetic problem is to use quantitative trace element
53 modeling and mathematically test different models. We use this approach for a pegmatite belt in
54 southeast Ireland, which is associated with the peraluminous Leinster Granite. A genetic link between
55 them has been suggested based on age and geochemical similarities (O'Connor *et al.* 1991, Whitworth
56 & Rankin 1989), but the exact relationship between them remains uncertain. As a result of new
57 lithium exploration drilling being carried by Blackstairs Lithium Ltd., an extensive lithochemical
58 database is available that provides drill core of the entire thickness of several pegmatite dykes.
59 Coupled with textural and mineralogical observations on the drill core, these are used for geochemical
60 modeling to better constrain the origin of these rare metal pegmatites.

61

62 GEOLOGICAL SETTING AND PREVIOUS STUDIES

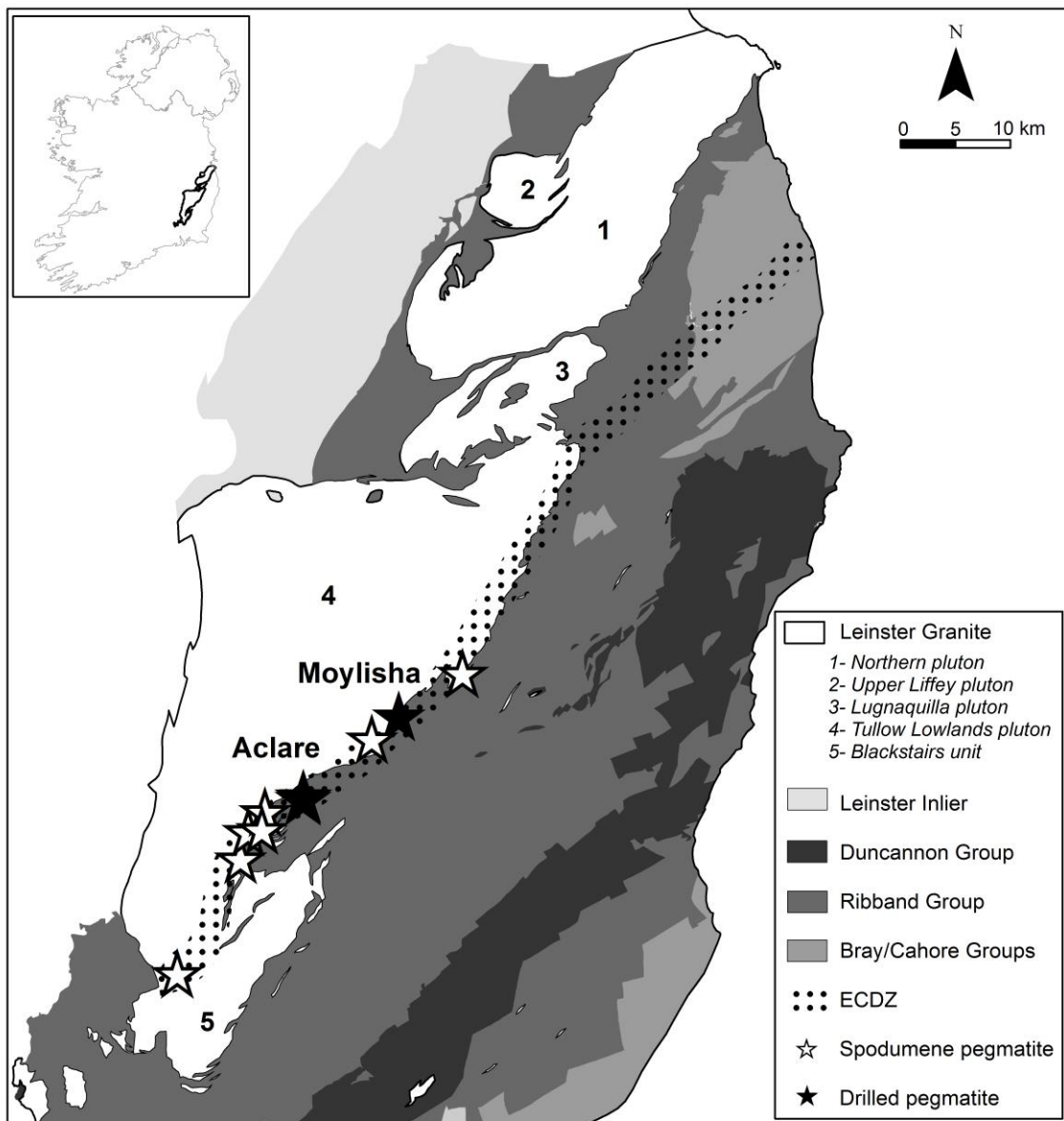
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64 The geology of the southeast of Ireland is dominated by Paleozoic basement and igneous
65 intrusions (Fig. 1). The basement is composed of sedimentary and associated volcanic rocks
66 regionally metamorphosed to lower greenschist facies and is subdivided into the following units and
67 lithologies: greywacke and shale of the Cambrian Bray and Cahore Groups; mudstone, siltstone and
68 sandstone of the Ordovician Ribband Group; deep marine shale, now commonly slate, with abundant
69 intermediate to acidic volcanic rocks of the Ordovician Duncannon Group; and the deep marine
70 turbidite sequence of the Silurian Leinster Inlier (Graham & Stillman 2009, Holland 2009).

71 The largest of the younger igneous intrusions is the Leinster Granite, a Caledonian batholith
72 composed at least in part of sheeted intrusions (Grogan & Reavy 2002) of S-type two-mica granitic
73 rocks (predominantly granodiorite) intruded into the metasedimentary basement. The batholith is part
74 of the Trans-Suture Suite (Brown *et al.* 2008), which spans the trace of the Iapetus suture in Britain

75 and Ireland. According to the same authors, the origin of these granitic magmas is related to orogen-
76 wide sinistral transtension during the early Devonian, triggering lamprophyre magma generated by
77 melting of Avalonian lithospheric mantle to rise and transfer heat to the lower part of the Avalonian
78 crust, forming partial melts with S-type granitic composition. The Leinster Granite comprises five
79 plutons aligned with the NE-SW regional strike of the Lower Paleozoic supracrustal country rocks
80 (McConnell & Philcox 1994). Despite its size, the batholith is poorly exposed. Most available data are
81 from the better exposed Northern pluton, including a U-Pb monazite age of 405 ± 2 Ma (O'Connor *et*
82 *al.* 1989). It has been assumed that all plutons intruded and crystallized simultaneously (Brindley
83 1973) and therefore that this age applies to the whole batholith. However a wide variability in initial
84 Sr and Nd isotope ratios, both between and within plutons, has been interpreted as resulting from the
85 generation of different magma batches by partial melting of an isotopically heterogeneous
86 sedimentary source (Mohr 1991). In addition, disequilibrium textures in plagioclase phenocrysts
87 indicate a multi-stage and multi-scale acid-acid mixing of magma batches during ascent and
88 emplacement (Grogan & Reavy 2002).

89 A network of lithium pegmatite dykes, metres to tens of metres thick, is known from at least nine
90 localities along the eastern margin of the Tullow Lowlands pluton and within the schist septum that
91 separates it from the Blackstairs Unit to the south (Fig. 1). There are almost no outcrops of these
92 dykes and the localities have been identified by concentrations of glacial boulders, in several cases
93 confirmed by mineral exploration drilling (Steiger & von Knorring 1974, Steiger 1977). Most of the
94 dykes so far detected roughly follow the NE-SW regional structure of the Caledonian East Carlow
95 Deformation Zone (McArdle & Kennedy 1985). They intrude the Tullow pluton, minor granitic
96 bodies related to the Leinster Granite and quartz-mica schist formed by contact metamorphism of the
97 Leinster Granite with the Ribband Group. Rb-Sr isotope studies of the pegmatites (O'Connor *et al.*
98 1991) yielded a whole-rock errorchron age of 396 ± 7 Ma (MSWD = 5.54), constructed using
99 spodumene pegmatites, associated lepidolite greisens and barren pegmatites. This corresponds to an
100 age of 402 ± 7 Ma when recalculated using the recent IUGS recommendation on ^{87}Rb half life (Villa
101 *et al.* 2015). The pegmatite errorchron initial Sr ratio of 0.705 ± 0.005 (O'Connor *et al.* 1991) lies
102 within the Leinster Granite range obtained by Mohr (1991). Moreover, results from fluid inclusion
103 analysis (Whitworth & Rankin 1989) and mineral chemistry (Whitworth 1992) suggest a genetic link
104 between the Leinster Granite and lithium pegmatites. Despite the indistinguishable Caledonian ages of
105 granitic rocks and pegmatites and their similar initial Sr isotope ratios, it is debatable whether lithium
106 pegmatites represent the residual magma after crystallization of the Leinster Granite, or were formed
107 from separate partial melts of similar or different metasedimentary sources. Here we aim to better
108 constrain the petrogenesis of the pegmatite belt and its relationship with the Leinster Granite.



109

110 Fig 1: Spodumene pegmatite occurrences in southeast Ireland. Dykes are restricted to the eastern
 111 margin of the Tullow Lowlands pluton, spatially associated with the East Carlow Deformation Zone
 112 (ECDZ). Pegmatites intrude both the Tullow pluton and metasedimentary rocks of the Ribband
 113 Group.

114

115 CHARACTERIZING THE LEINSTER RARE-ELEMENT PEGMATITE BELT AND COUNTRY ROCKS

116

117 A recent mineral exploration drilling programme has provided an opportunity to study the Leinster
 118 spodumene pegmatites, barren (spodumene-free) pegmatites and their immediate country rocks. The
 119 rock types identified in the drill cores in the study areas of Aclare and Moylisha (Fig. 1) are:

120

121

- (1) Spodumene pegmatite: coarse-grained (crystals up to 10 cm), composed of spodumene (10-40%), albite (25-35%), quartz (15-20%), Li-muscovite (10-15%), spessartine (5%) and minor K-

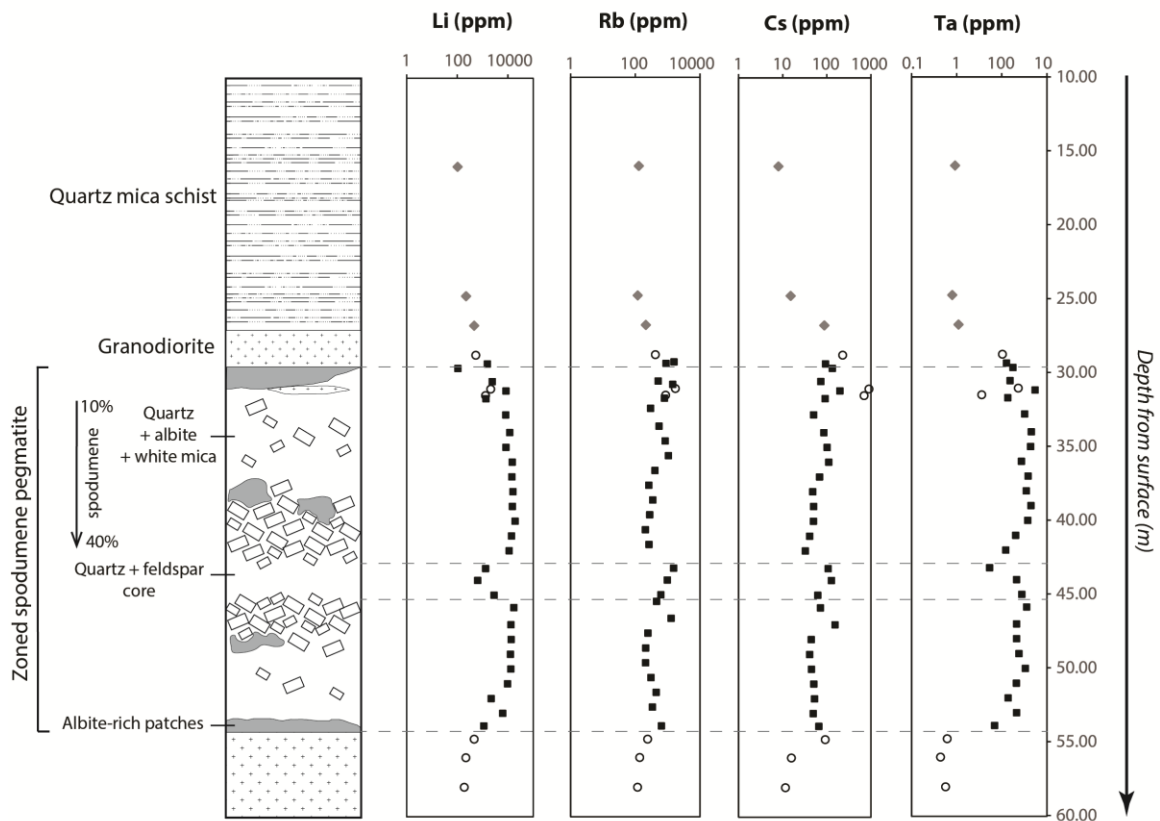
122 feldspar, apatite, cassiterite and sphalerite (all less than 5%). Spodumene, albite and Li-muscovite are
123 present as well-formed subhedral crystals, while quartz is interstitial. The dykes can be unzoned, or
124 present zoning when thicker than 10 meters, with an increase in modal spodumene towards the quartz
125 and feldspar core (Fig. 2). In some dykes, crystals of spodumene are aligned perpendicular, and
126 sometimes parallel, to the dyke margins. These observations suggest that each pegmatite dyke has
127 formed by the progressive *in situ* crystallization of a single body of magma. Fine grained albite-rich
128 (85-90%) aplitic patches are common within all zones and include around 10% combined quartz and
129 muscovite, with accessory apatite, cassiterite, spessartine, beryl and Mn,Fe-phosphate.

130 (2) Barren (spodumene-free) pegmatite: coarse-grained (typically 4 cm, maximum 10 cm),
131 consisting of K-feldspar (10-20%), albite (20-30%), quartz (20-35%), muscovite (15-20%) and
132 spessartine (2-5%). K-feldspar and albite occur as subhedral prismatic crystals, with later quartz and
133 muscovite. Quartz-feldspar intergrowth (graphic texture) is common.

134 (3) Granodiorite (Leinster Granite): medium-grained granodiorite consisting of quartz (30-45%),
135 oligoclase (30-40%), microcline (10-20%), biotite (5-10%) and muscovite (2-5%), locally porphyritic
136 with occasional 2 cm plagioclase phenocrysts. Sparse tourmaline is observed in some of the contact
137 zones with spodumene pegmatite and its veinlets.

138 (4) Quartz-mica schist (Ribband Group): fine- to medium-grained schist consisting of biotite (40-
139 50%), muscovite (20-30%), quartz (20-25%) and minor arsenopyrite, titanite and apatite, with a
140 foliation defined by mica; late tourmaline porphyroblasts with poikiloblastic texture are present up to
141 20 cm from the contact with spodumene pegmatite.

142 Spodumene pegmatite dykes crosscut both granodiorite and quartz-mica schist in Aclare, and
143 granodiorite only in Moylisha. The contacts between spodumene pegmatite and Leinster Granite are
144 defined by irregular planes and narrow (up to 10 cm) zones of interaction between the two rock types.
145 Contacts between spodumene pegmatite and mica schist are sharp and parallel to the foliation in the
146 mica schist. Spodumene-free pegmatites occur only within granodiorite, against which their contacts
147 are diffuse.



148

149 Fig. 2: Mineral and chemical profiles of zoned spodumene pegmatite (drill core ACL13-04). Modal
 150 spodumene (open rectangles) increases towards the quartz-feldspar core. Albite-rich aplite occurs as
 151 patches throughout the dyke. Whole-rock chemical compositions measured for homogeneous intervals
 152 of rock correlate with the mineralogical variation.

153

154 METHODS

155

156 *Sampling and geochemical analysis*

157

158 Eight mineral exploration drill cores with close to 100% recovery from two localities, Aclare (6
 159 cm diameter) and Moylisha (4 cm diameter), were split in half and divided into homogeneous rock
 160 parts, between 7 cm and 3.05 m long, resulting in 281 samples. These parts were then crushed,
 161 decomposed using a four acid digestion and analysed for 48 elements by ICP-MS by ALS Minerals
 162 (Loughrea, Co. Galway, Ireland). Routine practices were used to ensure data quality control: sample
 163 duplicates (1 in every 20 samples), homogeneous quartz pebbles (1/40) and certified standards (1/20).
 164 Results showed reproducibility between duplicates within 15% for most elements, except Ta, Zr and
 165 Ce, and no contamination problems. Detection limits for the elements analyzed range between 0.002
 166 and 100 ppm. Whole-rock geochemical analysis of pegmatites can be problematic because of their
 167 large crystal size, demanding large volume samples for representative results. However, the
 168 pegmatites sampled from drill core in the present study have a typical grain size of around 2 cm, with

169 the largest, rare, crystals having grain sizes less than 10 cm, which facilitates representative whole-
170 rock sampling.

171 Pegmatite drill cores were subdivided for whole-rock analysis by lithology into aplite, spodumene-
172 rich pegmatite, pegmatite without spodumene, granodiorite and quartz-mica schist. Weighted mean
173 compositions of each rock type were then calculated for each drill core. For example, in a drill core in
174 which granodiorite constituted a total length of 4.8 m, three samples of lengths 1.6 m, 1.2 m and 2.0 m
175 were analysed. Mean concentrations were then calculated by weighting analyses proportional to the
176 rock volumes in these three core lengths. In the case of pegmatite dykes, the calculation comprised all
177 the intervals from the upper to the lower contact with the country rocks. Contact zones with visible
178 and/or chemical haloes in country rocks and granitic lenses within pegmatites were excluded from the
179 estimated bulk compositions. The method assumes that the drill core samples are representative of the
180 pegmatite bodies as a whole and therefore does not allow for possible chemical variation along strike
181 or down dip within the dykes. It is also assumed that each pegmatite dyke crystallized from a single
182 batch of magma and that bulk pegmatite concentrations of the key elements remained constant during
183 and after crystallization. To highlight possible heterogeneities within rock types between localities,
184 whole-rock data is presented as the weighted mean of each rock type per drill core, for Aclare and
185 Moylisha separately.

186

187 *Modeling of pegmatite origin by in situ crystallization of granitic magma*

188

189 Calculated bulk concentrations of key elements were used to test whether the Leinster pegmatites
190 could have been derived as a residual phase of continuous crystallization of Leinster Granite
191 granodiorite magma. The elements Ba, Sr, Li, Rb and Cs were chosen as petrogenetic tracers, the first
192 two as compatible elements in granitic minerals and the others as incompatible in granodioritic
193 magma but enriched in pegmatites. To estimate the mean concentrations of these key elements for the
194 Tullow Lowlands pluton, the overall mean of all granitic intervals in all drill cores was considered,
195 weighted by volume as core diameters are different for drill core from Aclare and Moylisha. The same
196 was done for pegmatites and schist, to obtain representative concentrations assuming intrusion of one
197 batch of Li-pegmatite magma and one batch of barren pegmatite magma per dyke.

198 The equation for Rayleigh fractionation has frequently been used for trace element modeling of the
199 formation of pegmatites from granitic magmas (e.g. Shearer *et al.* 1992, Roda-Robles *et al.* 2012).
200 However, the idea of crystals separating from melt through gravity is most likely only applicable to
201 ultramafic melts (Rollinson 1993). In granites and pegmatites solidification most likely starts from the
202 intrusion margins inwards and in pegmatites experimental data suggest that this is a rapid process (e.g.
203 Webber *et al.* 1997, 1999, London 2008, Nabelek *et al.* 2010). In this case, a more adequate approach
204 is the quantitative *in situ* crystallization model proposed by Langmuir (1989), in which a solidification
205 zone occurs between the crystallized margins and the central convecting magma batch in a chamber.

206 Langmuir (1989) considers the case where once solidification starts, all liquid remaining in the
207 solidification front is ultimately separated from the crystal mush and returns to the interior of the
208 magma chamber, which is a reasonable approximation of the current understanding of granite and
209 pegmatite crystallization, and from this assumption the derived equation is $C_L/C_0 = (M_L/M_0)^{(f(D-1)/[D(1-
210 f)+f])}$, where C_L = concentration of the element in the differentiated magma; C_0 = initial concentration
211 of the element in the magma; M_L = mass of the differentiated magma; M_0 = initial mass of the magma
212 chamber; f = fraction of magma allocated to the solidification zone which returns to the unfractionated
213 magma chamber; and D = bulk partition coefficient. The bulk partition coefficient was calculated
214 through the equation $D = \Sigma (X_A K_{dA} + X_B K_{dB} + X_C K_{dC} + \dots)$, where X_A = weight fraction of mineral
215 A in the rock and K_{dA} = mineral A / liquid partition coefficient. This *in situ* crystallization equation
216 also yields the result for simple Rayleigh fractionation, for the condition $f = 1$.

217

218 *Modeling of pegmatite origin by partial melting*

219

220 Direct partial melting of a sedimentary source has previously been proposed as an alternative
221 mechanism to form rare element pegmatite magma (*e.g.* Jolliff *et al.* 1992, Shearer *et al.* 1992). The
222 most suitable process to generate the felsic melts discussed is batch melting (Shaw 1970), that can be
223 modelled by the equation $C_L/C_0 = 1/[D_{RS} + F(1 - D_{RS})]$, where C_L = concentration of the element in
224 the generated melt; C_0 = concentration of the element in the unmelted source; D_{RS} = bulk partition
225 coefficient of the residual solid; and F = weight fraction of melt produced. The bulk partition
226 coefficient is calculated in the same way as for the *in situ* crystallization calculations. Minimum PT
227 conditions of partial melting can be estimated based on the minimum temperature of 700 °C to
228 produce granitic composition melts (MacRae & Nesbitt 1980) and the spodumene stability field,
229 yielding a minimum pressure around 350 MPa. Conditions considered are in agreement with the
230 thermal model for generation of S-type granitic magmas within a transtensional pull-apart zone
231 proposed by Brown *et al.* (2008).

232

233 RESULTS

234

235 *Whole-rock geochemistry*

236

237 Ranges and means of whole-rock trace and major element concentrations for the four rock types
238 and the relevant study areas are given in Table 1. The granodiorite is classified as peraluminous using
239 the index of Shand (1943) and carries the signature of syn-collisional granites according to the trace
240 element discrimination diagram of Pearce *et al.* (1984) (Fig. 3). Granodiorite in Aclare has lower
241 alkali contents thus higher A/CNK and slightly lower Y and Nb contents when compared to the same
242 rock type in Moylisha. Both types of pegmatites are also peraluminous and syn-collisional, but

243 spodumene-free pegmatites exhibit the greater scatter in Rb, Y and Nb values. Spodumene pegmatites
244 in Aclare present a high scatter of A/CNK values and higher than Moylisha.

245 Multi-element diagrams per locality are presented on Fig. 4. Average trace element concentrations
246 for granodiorite show relative abundances decreasing towards the more incompatible elements to the
247 right of the plot and a strong negative Nb anomaly. Spodumene pegmatites and barren pegmatites
248 present a multi-peak signature, with low Ba, Th, La, Ce, Sr and Zr and Y, strong negative Ti anomaly
249 and positive Rb, Ta and Hf anomalies. In comparison with both types of pegmatites, the Leinster
250 Granite granodiorite is enriched in the LIL elements Ba and Sr, which are compatible in feldspars, and
251 in the light rare-earth elements La and Ce.

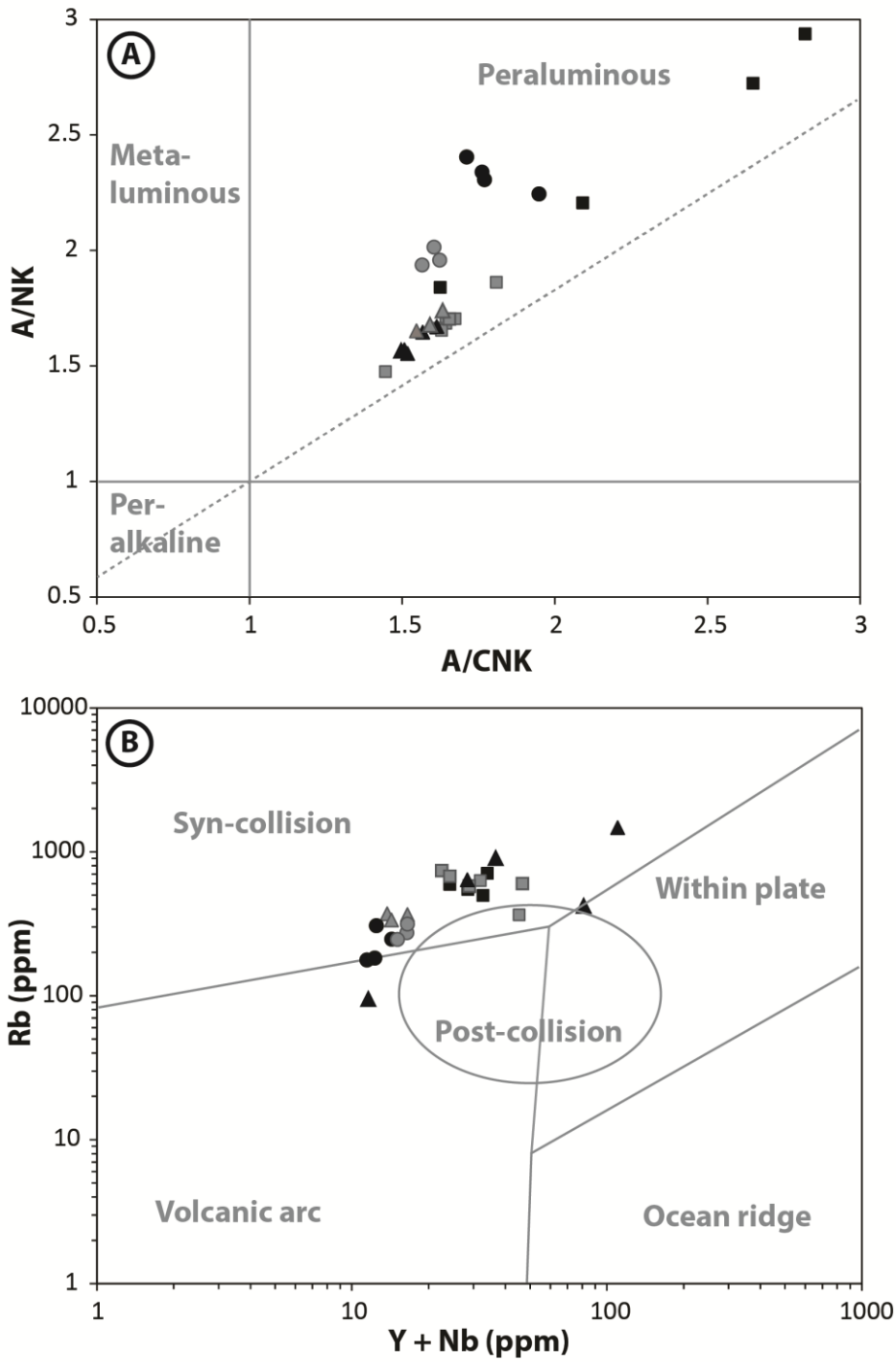
252 As Li is a suitable element to differentiate the three granitic rock types (granodiorite, spodumene
253 free pegmatite and spodumene pegmatite), incompatible and compatible elements were plotted against
254 this element (Fig. 5). The three rock types cluster separately without well-defined trends between
255 them. Spodumene pegmatites in Aclare have higher mean Li when compared with the same rock type
256 in Moylisha, while spodumene-free pegmatites in Moylisha present a higher mean Li concentrations
257 than those from Aclare. Granodiorite has the lowest mean Rb concentration and highest mean Sr and
258 Ba, while concentrations of Li and Cs are intermediate between spodumene-free and spodumene
259 pegmatites.

260 The ratio K/Rb versus Cs is controlled by K-feldspar and micas and is a useful petrogenetic index
261 in granite-pegmatite systems (Černý *et al.* 1981) and values for the studied rock types are shown on
262 Fig. 6. Granodiorites present the highest K/Rb ratios (50-120) and can be locally enriched in Cs as
263 represented by one sample from Aclare. Spodumene pegmatites and barren pegmatites show lower
264 K/Rb ratios of 20-60 and 20-90, respectively. The data do not fit on the expected evolution trend for
265 the case of a parental granite magma fractionating to residual pegmatitic magmas.

266 Table 1: Geochemical data for the studied rock types in the localities Aclare and Moylisha. Values in ppm. LOD = limit of detection. Duplicate analysis of 15
 267 of the samples shows maximum percentage differences between the higher and lower concentration presented in the last column of the data table.

	LOD (ppm)	Aclare												Moylisha						<i>Estimation of reproducibility (%)</i>			
		Quartz mica-schist			Granodiorite			Spodumene pegmatite			Spodumene-free pegmatite			Granodiorite			Spodumene pegmatite				Spodumene-free pegmatite		
		mean	min	max	mean	min	max	mean	min	max	mean	min	max	mean	min	max	mean	min	max		mean	min	max
Li	0.2	236.8	97.2	445.1	395.7	188.3	569.3	7020.4	1678	10794.9	430.8	26.8	1076.5	528.4	400.3	587.7	2670.0	601.8	4144.9	204.1	151.4	221.1	<i>8.1</i>
Na	100	11845.9	10201.8	13752.6	29734.6	26516.7	32012.6	27310.9	18738.9	40469.7	37577.1	33686.3	57400	26201.9	25650.1	28160.2	34556.4	27540	37405.1	29523.9	28480	31939.3	<i>4.8</i>
K	100	24651.7	24158.2	25449.1	17492.6	15403.6	18966.7	16236.6	15481.4	17165.2	24691.2	4100	35500	29341.7	27455.5	30158.6	21950.7	16133.4	32401.4	29939.7	28562.2	30968.3	<i>4.9</i>
Rb	0.1	150.2	110.3	205.4	248.2	177.4	306.2	593.1	496.6	707.8	725.7	94.6	1470	281.1	245.4	314.2	559.6	364	738.9	361.0	335.4	371	<i>8.7</i>
Cs	0.05	26.2	6.6	61.2	125.1	30.4	277.4	77.9	63.8	99.1	77.1	15.6	418	50.6	39.4	64.7	65.5	45.6	100.8	36.4	35.8	36.8	<i>8.2</i>
Mg	100	11176.7	10687.1	12609.1	6669.4	5861.0	7666.7	262.0	112.3	933.9	358.4	200	400	4566.0	3962.2	4840.1	181.8	80	381.3	696.3	624.7	724.6	<i>5.4</i>
Ca	100	3786.0	2664.3	5370.6	14090.5	6366.7	17842.0	1787.3	969.7	6408.9	1760.6	1366.5	3200	12189.2	10457.1	12784.3	1130.6	799.9	1791.4	3436.8	3086.7	3553.9	<i>10.2</i>
Sr	0.2	119.8	95.1	153.7	227.0	133.9	281.1	15.3	12	29.1	16.3	9.5	40.4	158.6	137.1	171.6	16.3	11	29.9	35.3	27.6	38.5	<i>10.0</i>
Ba	10	499.9	482.1	520	335.0	279.2	357.3	16.7	9.7	35.9	36.4	20	50	368.2	321.0	378.3	15.3	8.4	32.9	60.6	59.3	62.3	<i>6.9</i>
Ti	50	4585.7	4323.3	5016.9	2094.2	2066.7	2178.6	11.1	2.9	32.6	6.0	0	100	2211.5	1914.2	2308.9	33.5	13.3	116.3	290.1	221.6	335.6	<i>2.9</i>
Mn	5	696.8	495.2	1190.3	500.2	416.4	590.9	935.6	741.4	1009.6	302.6	108	649	565.6	499.0	584.7	783.5	656.2	1041.1	761.7	636	902.9	<i>8.0</i>
Fe	100	48192.0	46395.4	50366.7	20568.5	19728.1	21000.9	4316.3	3513.3	4600	4152.4	2100	4863.1	17271.1	15905.3	17838.6	3292.2	2863.8	4007.1	5952.1	5775	6145.5	<i>10.2</i>
Y	0.1	15.9	14.8	18.1	6.4	5.7	7.1	0.3	0.1	1.5	0.6	0.5	0.8	8.2	7.8	8.4	0.4	0.1	2.5	4.8	3.2	6.3	<i>13.3</i>
Zr	0.5	105.0	97.6	115.2	37.1	34.7	40.6	12.0	9.2	15	12.2	7.6	23.2	97.5	77.2	105.3	13.4	7.8	20	21.0	19	22	<i>15.9</i>
Nb	0.1	13.9	13.3	14.2	6.7	5.3	7.2	28.0	24.1	33.5	33.8	11.1	109.5	8.0	7.3	8.3	34.3	22.1	45.2	10.3	9.8	11.1	<i>7.0</i>
Hf	0.1	3.0	2.7	3.2	1.2	1.2	1.3	1.4	1.2	1.8	1.4	1	3	2.8	2.4	3.2	2.1	1.4	2.7	1.2	1.1	1.2	<i>8.7</i>
Ta	0.05	1.0	0.9	1.1	2.5	0.5	3.4	27.6	17.9	50.1	36.2	16.3	180	1.7	1.3	1.9	33.9	20.9	44.2	4.4	4.1	5.1	<i>26.4</i>
Al	100	86710.3	84007.1	89132.5	75597.8	69600.0	78509.0	68487.7	65249.5	70403	68118.4	50800	78300	74074.3	73271.9	74682.2	63341.4	53760.2	71578.5	67780.4	64332.3	71249.3	<i>7.2</i>
P	10	427.5	384.3	494.9	557.6	375.8	655.3	669.0	489.1	1038.9	677.4	430	1530	864.6	751.3	922.6	503.4	386.4	745.1	537.3	461.9	659.6	<i>5.5</i>
La	0.5	33.9	32.7	35.9	13.9	12.1	14.9	0.2	0	1	0.3	0.1	0.7	25.7	21.6	27.9	0.2	0	0.9	3.8	2.6	4.6	<i>8.8</i>
Ce	0.01	73.0	71.6	74.6	28.4	23.9	29.8	0.7	0.4	2.2	1.0	0.8	1.4	57.3	48.2	60.9	0.6	0.2	2.2	8.5	6	10.4	<i>40.1</i>
Th	0.2	10.5	9.2	11.4	4.9	4.7	5.6	0.6	0.5	0.8	0.9	0.4	3.1	11.5	9.4	12.2	2.6	2.3	3	2.5	1.8	3	<i>13.3</i>

268
 269 Values indicate minimum and maximum concentrations calculated for rock types in each drill core and the mean value is calculated as the mean of all
 270 analysed samples from each locality weighted by volume. The volumes of core that contributed to these calculated mean concentrations are: quartz-mica
 271 schist – Aclare and total 0.075 m³; granodiorite – Aclare 0.11 m³, Moylisha 0.13 m³, total 0.24 m³; spodumene pegmatite – Aclare 0.15 m³, Moylisha 0.05 m³,
 272 total 0.20 m³; spodumene-free pegmatite – Aclare 0.02 m³, Moylisha 0.04 m³, total 0.06 m³.



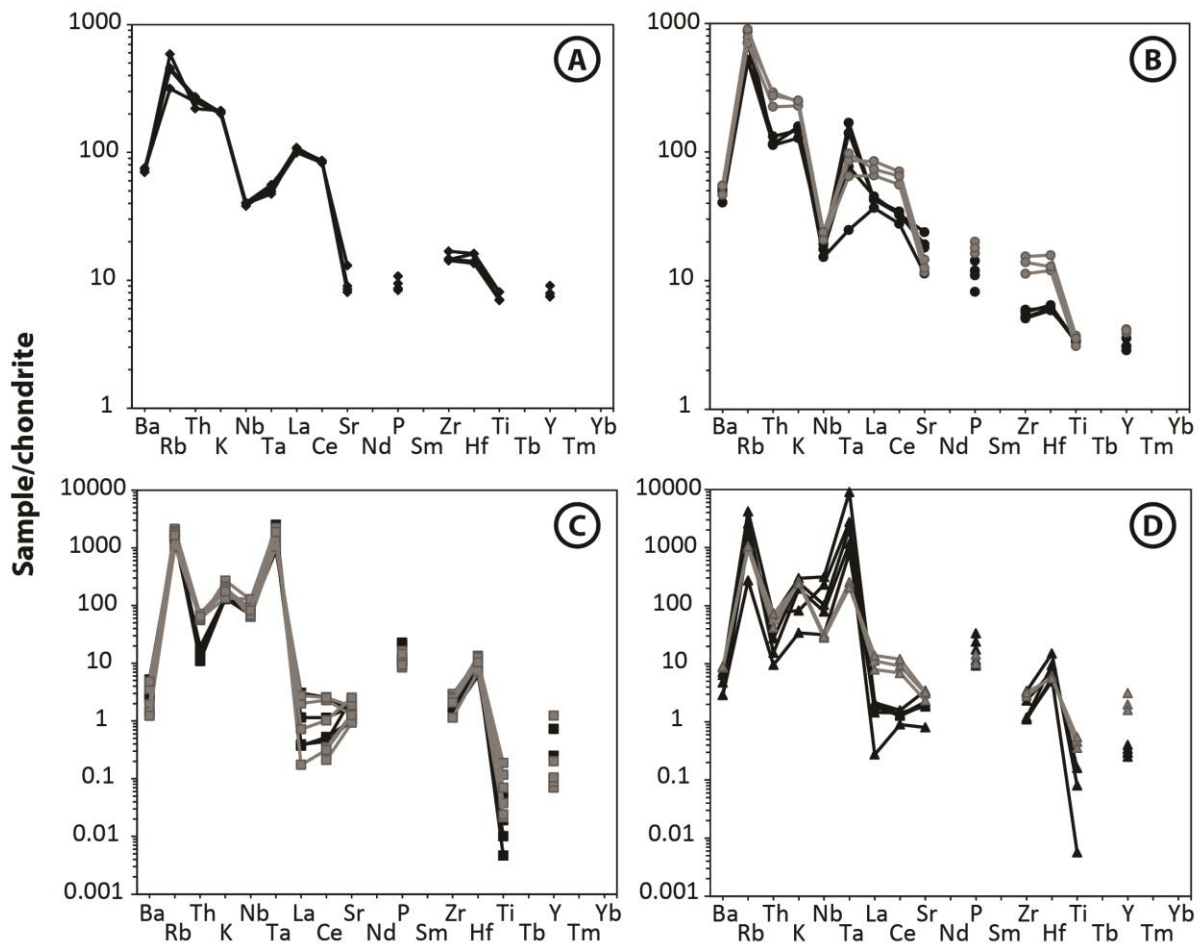
- Granodiorite (Aclare) ▲ Spodumene-free pegmatite (Aclare) ■ Spodumene pegmatite (Aclare)
- Granodiorite (Moylisha) ▲ Spodumene-free pegmatite (Moylisha) ■ Spodumene pegmatite (Moylisha)

274

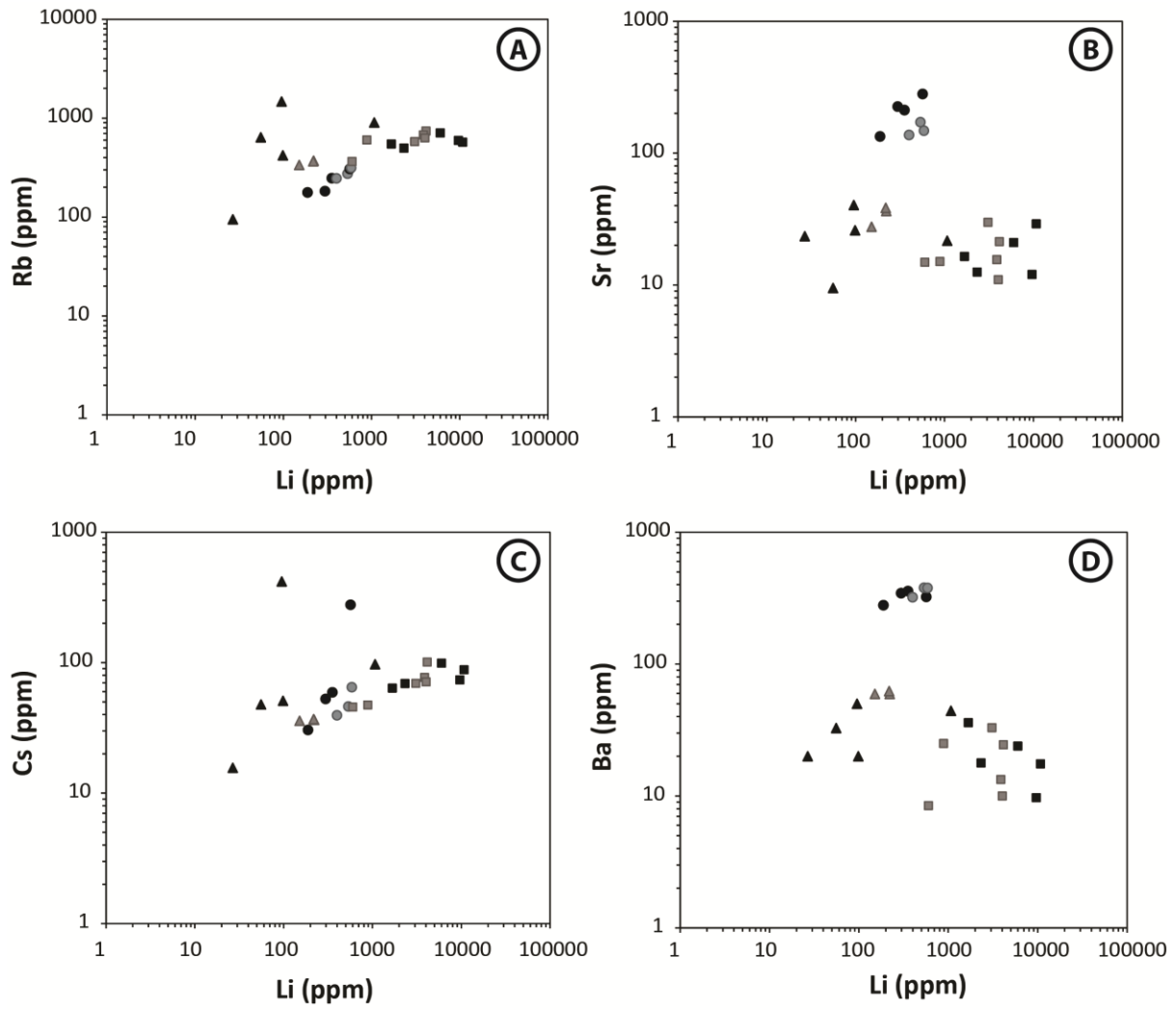
275 Fig. 3: A) A/CNK versus A/NK plot (Shand 1943) showing the peraluminous character of

276 granodiorite and pegmatites. B) Trace element discrimination diagram of Pearce *et al.* (1984) showing

277 syn-collisional signature of the rock types. Data points represent mean concentrations weighted by
 278 drill core volume of each rock type in individual drill cores.



279
 280 Fig. 4: Trace element concentrations are normalized to the composition of chondritic meteorites given
 281 by Thompson (1982). Samples in black are from Aclare and in grey from Moylisha. The diagrams are
 282 for A) quartz-mica schist; B) granodiorite; C) spodumene pegmatites and D) spodumene-free
 283 pegmatites. Gaps indicate no available data.

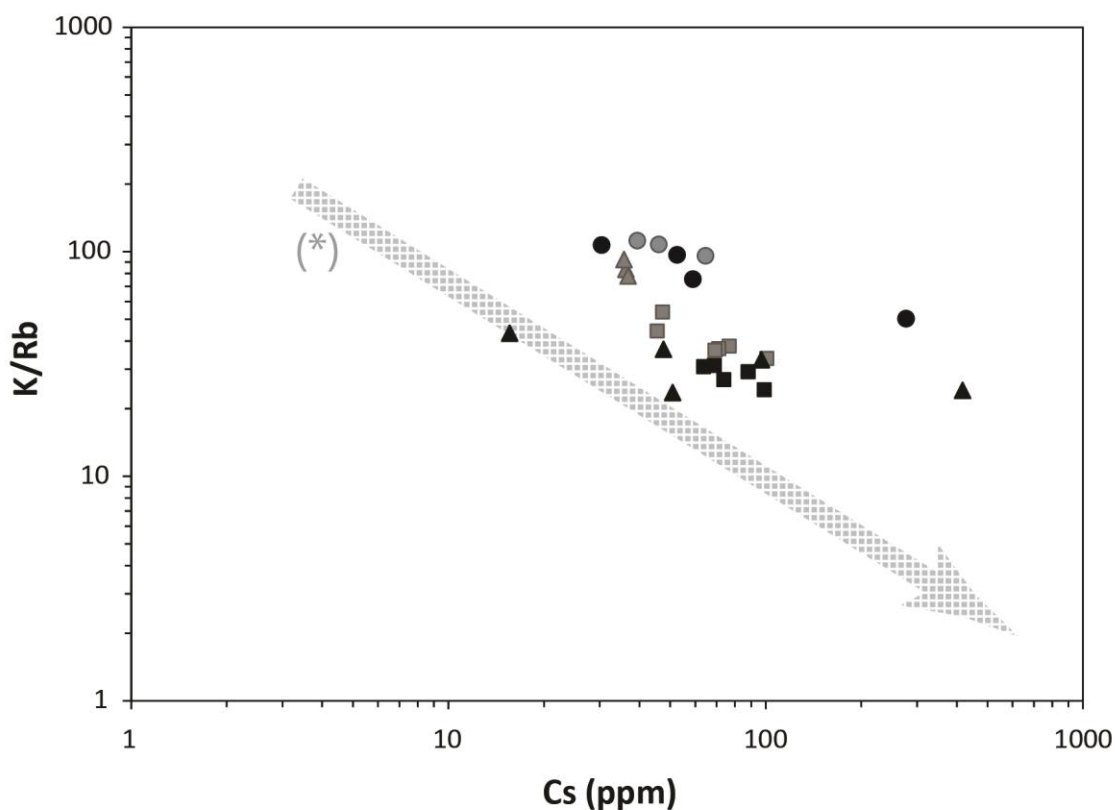


- Granodiorite (Aclare) ▲ Spodumene-free pegmatite (Aclare) ■ Spodumene pegmatite (Aclare)
- Granodiorite (Moylisha) ▲ Spodumene-free pegmatite (Moylisha) ■ Spodumene pegmatite (Moylisha)

284

285 Fig. 5: Bivariate plots of Li versus A) Rb, B) Sr, C) Cs and D) Ba. Samples in black are from Aclare

286 and in grey from Moylisha.



- Granodiorite (Aclare) ▲ Spodumene-free pegmatite (Aclare) ■ Spodumene pegmatite (Aclare)
- Granodiorite (Moylisha) ▲ Spodumene-free pegmatite (Moylisha) ■ Spodumene pegmatite (Moylisha)

287

288 Fig. 6: Cs versus K/Rb as an index of chemical evolution of pegmatites. (*) For comparison, the
 289 arrow indicates data presented in London (2008) for variation within a granite-pegmatite group (Red
 290 Cross Lake, Manitoba) with a proven petrogenetic link.

291

292 *Partition coefficients used in modeling*

293

294 Modeling of both *in situ* crystallization and batch melting requires calculation of bulk partition
 295 coefficients. As already debated in other pegmatite modeling studies (e.g. Jolliff *et al.* 1992, Shearer
 296 *et al.* 1992), uncertainty of the appropriate values of bulk partition coefficients can arise for several
 297 reasons. Several variables can affect mineral/melt partition coefficients, including temperature,
 298 confining pressure and melt composition (Rollinson 1993), but none of these are considered to be
 299 major issues in the studied granite-pegmatite systems. The partition coefficients and their published
 300 sources chosen for the key elements in granitic and pegmatitic minerals are presented in Table 2.
 301 Values were chosen to be representative of high-Si crystallizing magmas, consistent with the
 302 predominant granodiorite in Leinster Granite, and maximum values available were used for

303 calculations in cases where the reference presented a range. Bulk partition coefficients were
 304 calculated considering the mineral assemblages discussed below.

305

306 Table 2: Crystal / melt partition coefficients used for quantitative modeling.

Mineral/melt	Albite	Muscovite	Quartz & aluminosilicate	K-feldspar	Biotite
Li	0.1	1.67	0.05	0.05	1.65
<i>reference</i>	9	3	8	9	3
Rb	0.06	1.75	0.016	0.74	5.3
<i>reference</i>	6	3	7	1	1
Cs	0.44	0.24	0.044	0.13	3.1
<i>reference</i>	6	3	7	2	1
Sr	3.31	0.5	0.01	5	0.06
<i>reference</i>	4	3	<i>assumed</i>	5	3
Ba	0.19	5.5	0.015	6.7	7
<i>reference</i>	6	3	7	1	1

References: 1 Mahood & Hildreth (1983); 2 Icenhower & London (1996); 3 Icenhower & London (1995); 4 Drake & Weill (1975); 5 Long (1978); 6 Bea *et al.* (1994); 7 Nash & Crecraft (1985); 8 Jollif *et al.* (1992); 9 Walker *et al.* (1989).

307

308

309 *In situ* crystallization modeling

310

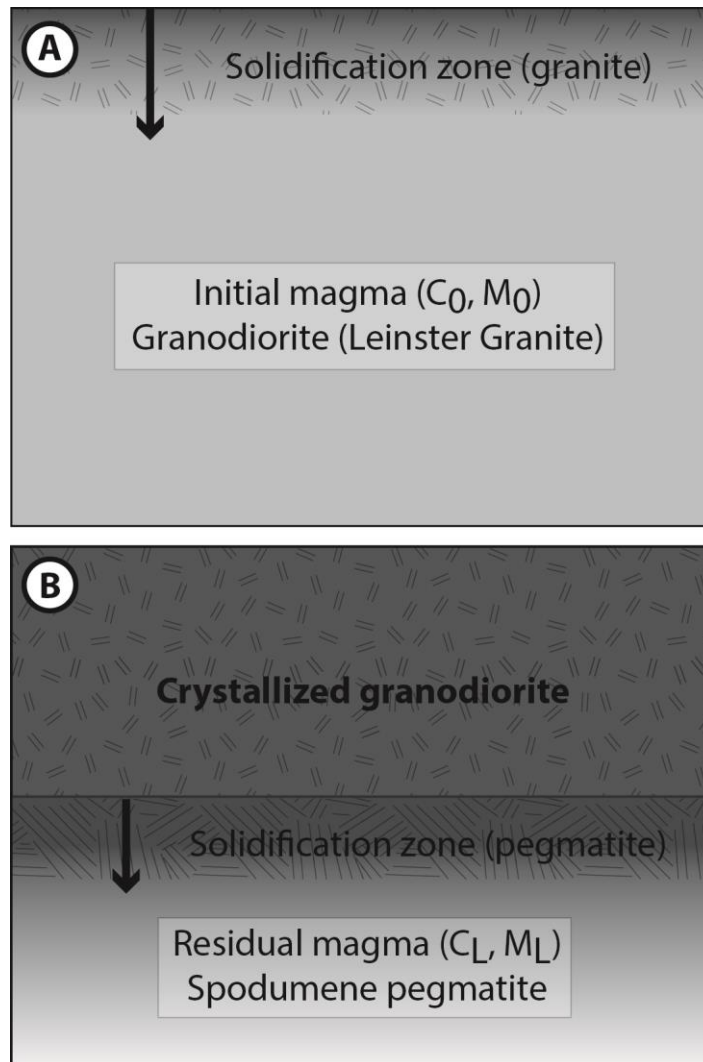
311 *In situ* crystallization calculations test the hypothesis that pegmatites and granodiorite are
 312 genetically linked through a continuous evolution model in a closed system (Fig. 7). Whole-rock
 313 concentrations of Li, Rb, Cs, Sr and Ba were estimated for granodiorite, spodumene pegmatite, barren
 314 pegmatite and schist as means weighted by total drill core volume of all analyses from all drill cores,
 315 for each rock type. These means for granodiorite are then considered to represent the initial magma
 316 from which residual pegmatitic fluids might have evolved in the late stages of crystallization, and
 317 subsequently escaped to be intruded as dykes; concentrations for schist are presented for comparison
 318 only in this scenario. Fractionation paths with various combinations of parameters were calculated, to
 319 show whether it is possible to reach bulk concentrations for the residual magma that are similar to
 320 mean spodumene pegmatite and barren pegmatite chemical compositions. The crystallizing
 321 assemblage considered is plagioclase (40%), K-feldspar (25%), quartz (25%), biotite (5%) and white
 322 mica (5%).

323 In *in situ* crystallization, the enrichment of a trace element in the residual melt relative to the
 324 parent melt (C_L/C_0) varies according to the mass of residual melt relative to the total mass of the
 325 magma chamber (M_L/M_0), the fraction of liquid returned to the main magma body from the

326 solidification zone (f) and the bulk partition coefficient (D). From lithogeochemical data, the values of
327 C_0 were considered to be the bulk trace element concentration calculated for granodiorite as the initial
328 magma. The fraction M_1/M_0 is equivalent to the fraction of residual magma and values between 0.01
329 (99% residual magma, start of crystallization) and 0.99 (1% residual magma, end of crystallization)
330 were considered. Values of f range between 0.01 (1% liquid returned from solidification zone,
331 representing very rapid crystallization) and 1 (all liquid returns from the solidification zone,
332 representing Rayleigh fractionation). The bulk partition coefficient, and therefore different values of
333 partition coefficients for the elements studied, can contribute substantially to variations in the final
334 result and this will be discussed later.

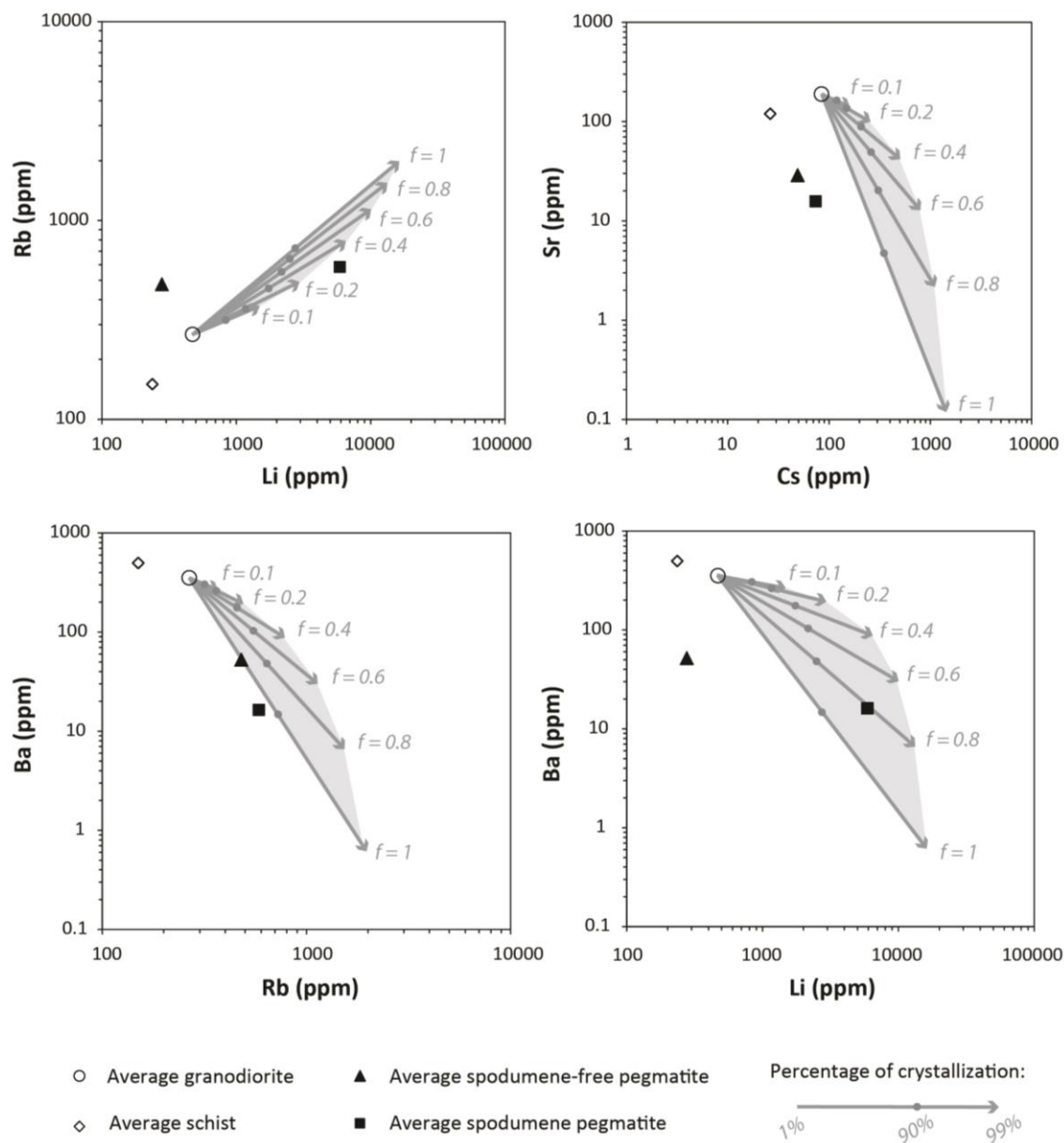
335 The ranges of possible enrichment and depletion of the five key elements, with granodiorite as
336 model initial magma, are plotted on Fig. 8. Average concentrations for key elements in pegmatites are
337 not always within the ranges of possible concentrations reached by *in situ* crystallization and the
338 maximum limiting Rayleigh fractionation. The magnitude of depletion in Ba and Sr and enrichment in
339 Rb and Li by *in situ* crystallization is sufficient to reach the mean composition of spodumene
340 pegmatite. However, very different crystallization conditions are required to individually explain the
341 elements analysed; for example ~95% crystallization at $f \sim 0.8$ can produce mean spodumene
342 pegmatite composition on the Ba-Li plot, whereas at least 99% crystallization is required at $f \sim 0.3$ to
343 produce mean spodumene pegmatite on the Rb-Li plot; there are no conditions which can account for
344 the mean Cs concentration of the spodumene pegmatites. Spodumene-free pegmatite compositions are
345 even less well reproduced by *in situ* crystallization (Fig. 8).

346



347

348 Fig. 7: *In situ* crystallization model (Langmuir 1989) applied to the Leinster Granite granodiorite -
 349 pegmatites system, depicting continuous granodiorite-to-pegmatites evolution within the same magma
 350 batch. A) Solidification starts from the margins of the magma chamber inwards, with a fraction of
 351 magma returning to the main magma body. The initial magma's composition is equivalent to the bulk
 352 composition of the crystallized granodiorite. B) After extensive crystallization, the residual magma
 353 would have become concentrated in incompatible elements and have a composition equivalent to the
 354 most evolved bulk spodumene pegmatite dykes. The same process could also generate the (less
 355 evolved) spodumene-free pegmatites as an intermediate member or as another end-member.



356

357 Fig. 8: Bivariate plots with the results of *in situ* crystallization modeling with the average Leinster
 358 Granite granodiorite as the initial magma. The arrows show the range from minimum (10%) to
 359 maximum (100%) fractions of liquid returned (f) to the main magma body from the solidification
 360 zone considered in this system – 100% returned is equivalent to Rayleigh fractionation. The grey area
 361 shows the range of values with other values of f between 0.1 and 1.

362

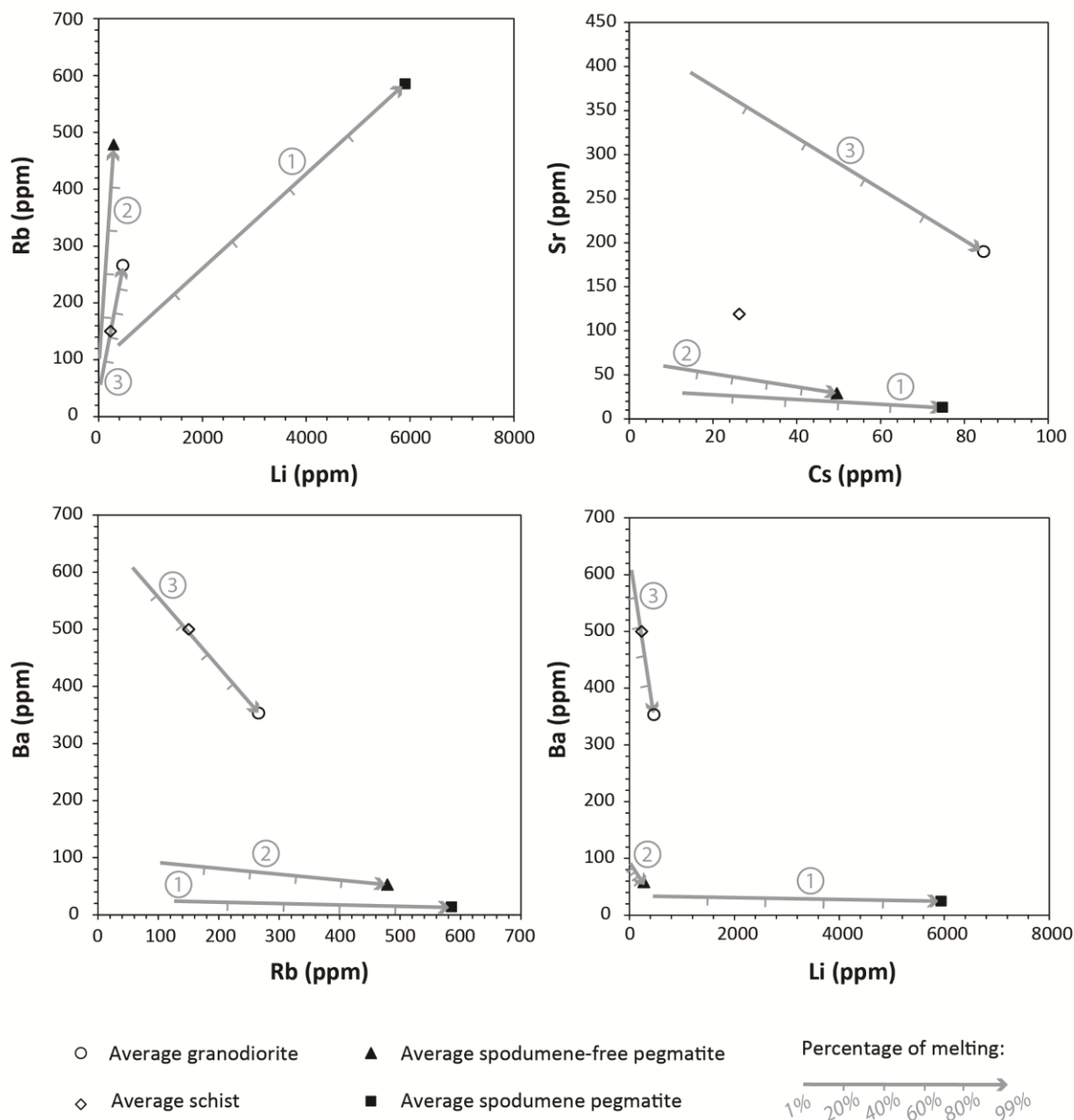
363 *Batch melting modeling*

364

365 The range of suitable initial compositions of sedimentary sources that underwent batch partial
 366 melting forming the three rock types were also calculated to assess whether a common origin for them
 367 is possible. In batch melting, the concentration of an element in the generated melt compared with the
 368 concentration of the element in the unmelted source (C_L/C_0) varies with the weight fraction of melt

369 produced (F) and the bulk partition coefficient of the residual solid in the source (D_{RS}). The weighted
370 mean concentrations measured in the three rock types were considered as different values for C_L ,
371 allowing calculation of the range of necessary concentrations in the source C_0 to generate melts with
372 the measured compositions. The range of initial concentrations was calculated from 1% melting ($F =$
373 0.01) to 99% melting ($F = 0.99$). In order to calculate D_{RS} , the source was estimated to be quartz
374 feldspar-rich sediment, since partial melting of pelitic sediments would result in Li, Rb and Cs being
375 retained in the mica-rich residual solid as partition coefficients are high for these elements (Jolliff *et*
376 *al.* 1992 and references therein). The residual solid considered would be mainly composed of quartz
377 (25%), feldspars (50%) and aluminosilicate (25%), thus allowing for all mica to be melted. Variations
378 in the percentages of these minerals in the residual solid and from the partition coefficients chosen do
379 not have major impact on the final results.

380 Possible initial concentrations in sources that can form melts with the compositions of average
381 granodiorite and pegmatites are featured on Fig. 9. Considering a range of 1 to 40% of partial melting
382 to form the three rock types (1 to 10% for pegmatites and 10 to 40% for granodiorite), the lines of
383 initial concentrations converge to a restricted area representing little variation in the source for Rb (50
384 to 130 ppm), Li (30 to 430 ppm) and Cs (around 10 ppm). For Ba and Sr, initial concentrations
385 converge to restricted areas for pegmatites (30 to 90 ppm Ba, 30 to 60 ppm Sr), but indicate a source
386 more enriched in those elements for granodiorite (around 600 ppm Ba and 400 ppm Sr).



387

388 Fig. 9: Bivariate plots with the results of partial melting modeling. The arrows show the possible
 389 initial concentrations of the magma source rocks for spodumene pegmatite (line 1), spodumene-free
 390 pegmatite (line 2) and granodiorite (line 3) if they underwent 1 to 99% melting. Average composition
 391 of the schist is plotted for comparison, representing metasedimentary rocks of the Ribband Group.

392

393 **IMPLICATIONS FOR SPODUMENE PEGMATITE FORMATION**

394

395 The strong negative Nb anomaly in Leinster Granite granodiorite indicates that a crustal
 396 component is involved in its formation, which can also be inferred for pegmatites, but with a less
 397 pronounced negative anomaly. Pegmatites do not always carry higher concentrations of incompatible
 398 elements than granodiorite (e.g. concentrations of Zr, Ti and Y) and no clear fractionation trends are
 399 observed among incompatible or compatible elements, or ratios that indicate chemical evolution.

400 Plotted geochemical data is scattered rather than representing a chemically closed or semi-closed
401 continuous evolution system. Although the key elements Li and Cs have not been analysed in samples
402 from elsewhere in the Leinster Granite, the chemical signature of granodiorites within the batholith
403 varies between localities, probably reflecting the multi-intrusion character of the batholith and some
404 variation in source rock composition (Mohr 1991, Sweetman 1987, Grogan & Reavy 2002).

405 With the chosen partition coefficients it is not possible to reach spodumene pegmatite mean
406 concentration considering all elements, therefore a single crystallizing system from granodiorite to
407 pegmatite. If, however, the bulk partition coefficient for Ba (D_{Ba}) is double that listed in Table 2, by
408 consequence of higher $Kd_{Ba}^{biotite/liquid}$ and $Kd_{Ba}^{K-feldspar/liquid}$, the required depletion of this element from
409 granodiorite to pegmatites can be attained. Such high values were obtained experimentally by
410 Icenhower & London (1996), for example. For Rb, a slightly lower bulk partition coefficient makes
411 possible the enrichment observed in spodumene pegmatites. This would be mainly controlled by a
412 lower $Kd_{Rb}^{biotite/liquid}$ and such lower values have been obtained experimentally (*e.g.* Icenhower &
413 London 1995). However, the spodumene pegmatite Sr and Cs concentrations cannot be reached using
414 reasonable partition coefficients for these elements. They could only be reached with a much higher
415 Cs bulk partition coefficient. Extreme Li concentrations found in spodumene pegmatite can be
416 reached towards high fraction of magma return from the solidification front, or Rayleigh fractionation,
417 and with less than 5% magma left, but this scenario would require the combination of cumulate-type
418 crystallization followed by effective extraction of the residual magma. Therefore granodiorite-to-
419 pegmatite magmatic evolution seems highly unlikely taking account of all five elements.

420 One possibility to explain the discrepancy observed when trying to correlate concentrations of key
421 elements through the Langmuir *in situ* crystallization equation is a significant loss of the mobile
422 elements Rb, Cs and Li at some stage between generation, ascent and emplacement of pegmatite
423 magma. However, very narrow exomorphic haloes (up to 20 cm) into the granodiorite and the
424 permeable quartz-mica schist do not support this hypothesis. Cs mobility does however seem possible
425 as high-Cs granodiorite is observed (Figs. 5 and 6). The mean Cs concentration for the initial magma
426 could be lower if this locally high-Cs granodiorite is considered to be part of an exomorphic halo (and
427 therefore excluded from bulk composition calculations), but still considerably higher D_{Cs} would be
428 needed to reach pegmatite concentrations. Another possibility is contamination from country rocks,
429 especially contamination of spodumene pegmatites by the schist, but its mean composition with low
430 Li (<250 ppm), Rb (<150 ppm) and Cs (<30 ppm) cannot explain the deviations of *in situ*
431 crystallization trends away from pegmatite compositions.

432 On the other hand, with a narrow range of high source rock incompatible element concentrations
433 and reasonable assumptions for degrees of partial melting, it is possible to form melts with the
434 average composition of granodiorite, spodumene-free and spodumene pegmatites. However, source
435 compatible element concentrations have to be highly variable if the three lithologies share a common
436 source. As bulk concentrations of Ba and Sr would strongly depend on the amount of feldspars in

437 metasedimentary rocks, this variation can be explained by the presence of feldspar-rich and feldspar-
438 poor source rocks, generating granodiorite and pegmatites, respectively. If pegmatite magmas were
439 generated at the same time or later than the granodioritic magmas that formed the Leinster Granite, as
440 implied by cross-cutting relationships, the required source rock heterogeneity likely existed on at least
441 a kilometre scale. Otherwise, it is difficult to envisage the pegmatite source rocks remaining unmelted
442 during Leinster Granite granodiorite magma formation.

443

444 CONCLUSIONS AND FUTURE WORK

445

446 Observing patterns on multi-element diagrams and bivariate plots, the behaviour of incompatible
447 elements and the absence of fractionation trends among elements, the hypothesis that pegmatite
448 magmas formed by the continuous evolution of the same magmatic system as the Leinster Granite
449 granodiorite is not a satisfactory explanation for the origin of these rocks. Specifically, *in situ*
450 crystallization modeling shows that it is highly unlikely that pegmatites represent the residual magma
451 from crystallization of Leinster Granite granodiorite. This requires that granodiorite and pegmatite
452 magmas were derived in separate partial melting events, the pegmatites presumably representing a
453 later event because they are observed to intrude granodiorite. However the source rocks required to
454 account for the geochemistry of pegmatites and the Leinster Granite granodiorite must be different,
455 with considerably more feldspar required in the granodiorite source rocks than in the pegmatites'
456 source rocks. An unresolved question is why magmas that formed igneous rocks in close spatial
457 association (granodiorite plutons and pegmatite dykes) were not apparently formed by partial melting
458 of the same source rocks. In an alternative scenario the voluminous Leinster Granite could represent
459 mixing between mantle-derived mafic magma and crustal melts of similar rocks that alone generated
460 pegmatite magmas. Precise dating and radiogenic isotope studies to define the sources are planned
461 towards a better understanding of the story behind the formation of pegmatites in southeast Ireland.

462

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464

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