

Beryllium in Silicic Magmas and the Origin of Beryl-Bearing Pegmatites

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INTRODUCTION

This chapter addresses the portion of the geochemical cycle of beryllium that entails its transfer through the continental crust by the generation and migration of silicic magmas, which will be referred to loosely as granitic but will include all of the textural variants of rocks derived from the granitic bulk compositions. Within the granite system, we will examine the distribution of Be between anatectic melts and residual crystals, and we will track the accumulation of Be in melt via crystal fractionation, culminating in the saturation of pegmatite-forming melts in the mineral beryl. Beryl is unquestionably the most common manifestation of Be enrichment, and the most abundant of the phases that contain essential beryllium (see Černý this volume). To explain the presence of beryl in pegmatites, we pose four questions:

1. How much Be does it take to saturate a granitic (pegmatite-forming) melt in beryl?
2. How much Be is likely to be incorporated in silicic partial melts at their source?
3. Consequently, how much fractionation of a typical granitic magma must occur to achieve beryl saturation in pegmatitic derivatives (or is beryl saturation possible without extended magmatic fractionation)?
4. And should all granitic magmas be expected to achieve beryl saturation eventually, or are there special attributes of the source material or crystalline phases in the magma that predetermine the likelihood of forming beryl in pegmatitic differentiates?

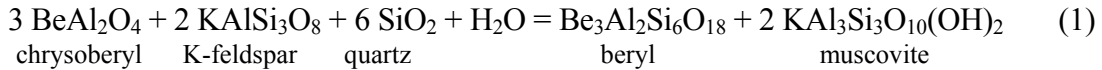
To address these questions, we start with a review of beryl saturation in pegmatites. We then examine the likely abundance of Be derived by anatexis at the sources of granitic magmas. With the two ends of the system bracketed, we can then subject Be fractionation to conventional modeling methods to ascertain how much fractionation must take place to bring granitic melts to beryl saturation. As a review, this chapter relies as much as possible on readily available publications (monographs and journal articles, as opposed to abstracts, unpublished dissertations, and other more obscure works) that bear on these questions. We will, however, introduce new results from our laboratory that provide much of the partitioning data needed to model the fractionation of Be from anatectic source to pegmatite. We will not address liquidus effects, viscosity, and the volumetric properties of Be in granitic melts (Hess et al. 1996; Evensen et al. 1999), nor will we assess the stability or occurrence of beryllium phosphates or borates in this chapter (see Burt 1975; Charoy 1999; Černý this volume).

BERYLLIUM CONTENTS OF PEGMATITIC MELTS AT BERYL SATURATION

Experimental evidence

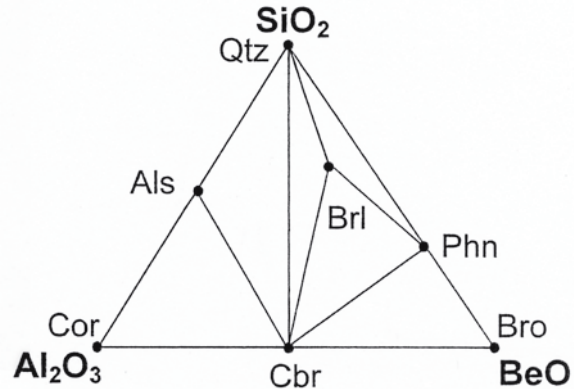
Beryl (Brl) occurs principally in granitic pegmatites, but it is also found in topaz rhyolite, in hydrothermal veins associated with high-level silicic porphyries, in metamorphic rocks (gneiss), and less commonly in leucogranites (see Beus 1966 and

Černý, this volume). The widespread occurrence of beryl rather than other Be-silicates in granitic pegmatites stems from the stability relations of Be phases in quartz-saturated portions of the system BeO-Al₂O₃-SiO₂-H₂O (Burt 1978; Franz and Morteani 1981; Barton 1986; Cemič et al. 1986). Divariant assemblages containing beryl + quartz occur over the range of moderate pressures (~100-400 MPa) and temperatures (~400°-700°C) pertinent to the consolidation of pegmatites (Barton 1986; Černý 1991b). Figure 1 predicts three fields containing 3-phase assemblages among beryl, phenakite (Be₂SiO₄), chrysoberyl (BeAl₂O₄), and quartz. Beryl-bearing pegmatites are commonly peraluminous in composition, and therefore the mineral assemblage beryl + chrysoberyl (Cbr) + quartz is a possible (though uncommon) high-temperature assemblage (Burt 1978). In natural systems, however, chrysoberyl reacts with alkali feldspar plus quartz (or their components of melt) upon cooling and an increase in *a*_{H₂O},

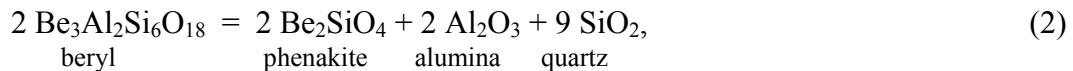


to generate the common association of beryl and muscovite. The assemblage Cbr + Brl + Phn (phenakite) is unlikely in quartz-saturated systems such as granitic pegmatites, but it is of interest because it contains most of the important Be phases in the quaternary system cited above.

Figure 1. Proposed phase relations in the system BeO-Al₂O₃-SiO₂-H₂O at the moderate *P-T* conditions investigated in this study (685°-850°C, 200 MPa). Abbreviations are Cor (corundum), Qtz (quartz), Bro (bromelite), Als (aluminosilicate), Cbr (chrysoberyl), Brl (beryl), Phn (phenakite). The tie line Cbr-Qtz is stable at the experimental conditions investigated by Evensen et al. (1999). Bertrandite [Be₄Si₂O₇(OH)₂] and euclase [BeAlSiO₄(OH)], not shown here, become stable at lower temperatures. Mineral assemblages are from Burt (1978) and Barton (1986).



Evensen et al. (1999) determined the solubility of beryl-, chrysoberyl-, and phenakite-bearing assemblages in metaluminous to peraluminous granitic melts, and hence addressed the first question posed above: what is the Be content needed to saturate granitic melts in these phases at *P-T* conditions (200 MPa H₂O) that pertain to hydrous granites and pegmatites? They employed two approaches to beryl saturation, one in which forward (thermally prograde) and reverse (thermally retrograde) experiments utilized beryl as the source of Be, and replicate experiments in which a chemically equivalent assemblage containing phenakite was substituted for beryl:



The granitic compositions included a metaluminous mineral mixture made to the 200 MPa H₂O granite minimum (Tuttle and Bowen 1958), a similar mixture made strongly peraluminous (ASI ~ 1.3) by addition of synthetic boehmite, and the chemically complex peraluminous rhyolite obsidian from Macusani, Peru (Barnes et al. 1970; London et al. 1988).

The resultant Be mineral assemblages were the same as those predicted from the topology of Figure 1. Quartz-undersaturated experiments contained the assemblage Phn + Brl + Cbr. As predicted (by Burt 1978; Barton 1986), the assemblage Cbr-Qtz is stable,

and the tie line between these two phases precludes assemblages of Brl + Als at moderate P and T. Beryl occurred with and without Cbr in quartz-saturated experiments.

The experimental study by Evensen et al. (1999) assessed Be saturation of melt not only as a function of T (at constant $P_{\text{H}_2\text{O}} = 200 \text{ MPa}$), but also as functions of the activities of other melt components. Evensen et al. (1999) recognized that beryl saturation should be determined as an activity product, i.e.,

$$[a_{\text{Be}_3\text{Al}_2\text{Si}_6\text{O}_{18}}]_{\text{beryl}} = [a_{\text{BeO}}]_{\text{melt}}^3 [a_{\text{Al}_2\text{O}_3}] [a_{\text{SiO}_2}]^6, \quad (3)$$

in which the activity of a beryl component in melt is a function of the activities of its constituent oxides. This representation emphasizes the importance of the activities of silica and of alumina (monitored through the parameter of ASI) on the BeO content of melt at beryl saturation. It allowed Evensen et al. (1999) to consider the effects of these and other components (e.g., B, P, and F in the Macusani obsidian) on the activities of the individual beryl-forming components.

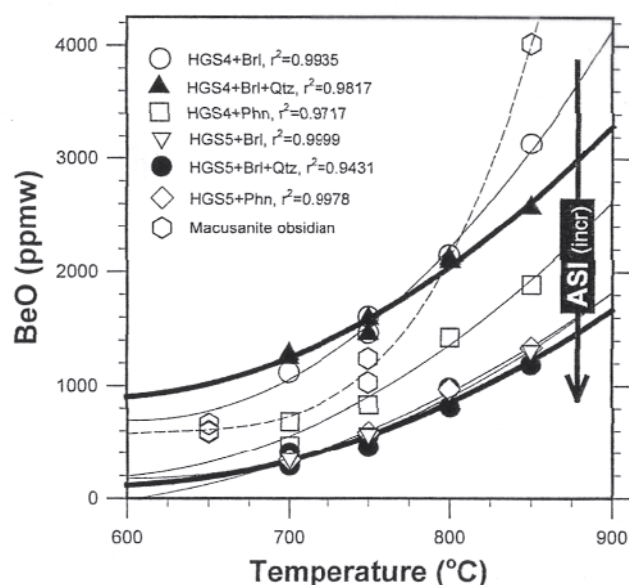


Figure 2. BeO contents of granitic melts at beryl saturation from 650–850°C and 200 MPa H_2O , from Evensen et al. (1999). The thick curves are for quartz-saturated experiments. The arrow shows the direction of increasing melt ASI from 1.00 in the simple metaluminous (HGS4) to 1.28 in the peraluminous (HGS5) melts. The experimental trend using the Macusani obsidian, with an ASI = 1.27, lies in the opposite direction of the arrow, at higher BeO contents of beryl-saturated melt, for reasons explained in the text. From Evensen et al. (1999).

Effect of silica activity. The activity of the beryl component in melt varies to the sixth power of silica activity. Consequently, variations in the activity of SiO_2 can effect major changes in the BeO content of melt at beryl saturation. This relationship is evident in the data for metaluminous granite + Brl with and without quartz saturation (Fig. 2). Without added quartz, the metaluminous haplogranite composition HGS4 + Brl, which is quartz-saturated at the solidus, becomes increasingly silica-undersaturated with increasing temperature above the solidus. When sufficient quartz was added to the mineral mixture HGS4 + Brl + Qtz to maintain quartz saturation at all the temperatures above the solidus, the resultant Be content of melt decreased, and divergence between the two saturation curves increased with increasing temperature (Fig. 2). Though the experiments illustrate an effect of silica activity on beryl saturation, the effect is negligible only because the activity of silica does not change appreciably over the range of temperature investigated. In addition, beryl almost invariably occurs with quartz in natural systems, so the activity of silica in melt or vapor is normally buffered.

Effect of alumina activity. The solubility of beryl varies to the first power of the activity of alumina in melt. In contrast to silica, however, the activity of alumina in melt, as measured by the Aluminum Saturation Index ASI ($= \text{mol Al}_2\text{O}_3 / [\text{Na}_2\text{O} + \text{K}_2\text{O} + \text{CaO}]$) varies substantially from ASI values of ~ 1.0 to ~ 1.3 . At the high ASI of the chrysoberyl-saturated runs, the BeO contents of peraluminous melts were displaced systematically

(parallel to metaluminous results) to lower values. The lowest values were obtained in experiments with haplogranite compositions (HGS5 + Brl + Qtz) that were saturated in both chrysoberyl and quartz. Increasing the ASI of melt from 1.0 to 1.3 served to reduce the BeO content of melt at beryl saturation by over 65% at 700°C.

Effects of other components. The Macusani glass is high in silica and strongly peraluminous with an ASI = 1.27 (including Li, Rb, and Cs in the total of alkalis). In contrast to the experiments with similarly peraluminous haplogranite ± Qtz, however, the Macusani obsidian yielded the highest BeO contents of any experiments (with Cbr ± Brl). The disparity between Macusani obsidian and the peraluminous haplogranite (HGS5, with a similarly high ASI value) indicates that speciation reactions among the more numerous components of the Macusani glass (Li, B, P, F, Rb, and Cs—all at 10⁴ to 10⁵ ppmw levels) and any or all of the beryl-forming components lead to a net reduction of activity for beryl. Among these components, F and P are believed to react with Al to form melt species (e.g., Manning et al. 1980; Mysen and Virgo 1985; Wolf and London 1994, 1997). Hence, in reducing the activity of alumina, these components raise the BeO content of melts needed to achieve saturation in beryl or chrysoberyl. Speciation reactions involving Be, F and P may also play a role (Toth et al. 1973; Wood 1991; Charoy 1999).

Summary effects of blk composition. Evensen et al. (1999) found that at a given temperature (Fig. 2), the solubility of beryl is lowest in simple quartz-saturated peraluminous melt compositions (~200 ppmw BeO, or ~70 ppmw Be at 700°C). The added components of the Macusani glass, whose composition is similar to many evolved Li-rich pegmatites, reduce the activities of beryl-forming components, requiring higher BeO concentrations (~900 ppmw BeO, or ~325 ppmw Be) to achieve beryl saturation in complex lithium pegmatites.

Effects of temperature. Although the activities of silica, alumina, and other added components influence the solubility of beryl, the effect of temperature is significantly greater over the range of the conditions studied by Evensen et al. (1999). For example, the Be content of melt saturated in Cbr ± Brl increases nearly an order of magnitude between 750 and 850°C. The low-temperature range of Figure 2, however, is most relevant to pegmatites. The slopes of the beryl saturation curves decrease substantially and tend to converge toward very low temperature.

COMPARISON TO BULK COMPOSITIONS OF PEGMATITES

The coarse grain size and heterogeneous zonation within common and rare-element granitic pegmatites renders estimates of bulk composition imprecise. Consequently, there are not many published attempts to derive pegmatite bulk compositions. Table 1 lists those pegmatite whole-rock estimates that were found to include Be in the totals. Methodologies for the assessments vary in detail but in most cases they utilized cored or quarried samples from which different minerals or whole-rock samples of recognizable zones were analyzed for Be, which in most cases was represented as the oxide BeO. Representative bulk compositions were obtained by assessing the Be content of a zonal assemblage and weighting its proportion to other zones. Among those sources cited in Table 1, the information presented by Stilling (1998) for the Tanco pegmatite, Manitoba, is undoubtedly the most accurate, as it is derived from data logged from 1,355 surface and underground drill cores. Interested readers should consult each source for additional details. For comparison, Table 1 also contains analyses of Be in melt inclusions found in pegmatitic quartz from a pegmatite mineralized in Sn, W, P, and F at Ehrenfriedersdorf, central Erzgebirge, Germany (Webster et al. 1997).

Table 1. Average BeO content of granitic pegmatites.

Location	Pegmatite type (where known)	Average BeO, ppm	Source
Tanco Manitoba, Canada	complex Li petalite	452	Stilling (1998)
Sparrow Pluton Yellowknife, NWT, Canada	beryl	722	Kretz et al. (1989)
Helen Beryl Black Hills, SD USA	beryl	254	Staatz et al. (1963)
Tin Mountain Black Hills, SD USA	complex Li spodumene	619	Staatz et al. (1963)
Souchon Kamativi mine, Zimbabwe	complex Li lepidolite	560	Gallagher (1975) Rijks & van der Veen (1972)
Augustus near Salisbury, Zimbabwe	complex Li lepidolite	560	Gallagher (1975) Ackermann et al. (1968)
Mistress near Salisbury, Zimbabwe	complex Li lepidolite	1200	Gallagher (1975) Ackermann et al. (1968)
Benson 1 Mtoko district, Zimbabwe	complex Li lepidolite	1600	Gallagher (1975) Ackermann et al. (1968)
Benson 4 Mtoko district, Zimbabwe	complex Li lepidolite	210	Gallagher (1975) Ackermann et al. (1968)
Benson 2 Mtoko district, Zimbabwe	complex Li lepidolite	100	Gallagher (1975) Ackermann et al. (1968)
Al Hayat Bikita district, Zimbabwe	complex Li spodumene	350	Gallagher (1975) Ackermann et al. (1968)
Bikita Main Bikita district, Zimbabwe	complex Li petalite	500	Gallagher (1975) Ackermann et al. (1968)
Beryl Rose Darwin district, Zimbabwe		420	Gallagher (1975)
unnamed pegmatite Gobi Field, E. Gobi, Mongolia	complex Li lepidolite	430	Rossovskiy & Matrosov (1974)
Ehrenfriedersdorf Erzgebirge, Germany	complex Li Li-mica & topaz	472	Webster et al. (1997)

The compositionally simple beryl-rich pegmatites (Helen Beryl, Table 1) have BeO contents that match the experimental values expected of muscovite-bearing pegmatites at ~700°C. Similarly low BeO values are evident in some of the less-evolved Li pegmatites (lepidolite-bearing but low in Li minerals overall) of the Benson claims (Table 1). These are primarily noted for their cassiterite contents, and are not manifestly beryl-rich; thus, the low BeO values could mean that the pegmatite magmas became saturated in beryl only after extended crystal fractionation. The highly fractionated, complex Li pegmatites (Tanco, Tin Mountain, and Bikita, Table 1) possess about half of the BeO expected for such evolved melt compositions, especially considering that the pegmatites are beryl-rich. These values may be indicative of melts that crystallized at low temperatures, perhaps substantially lower than the likely liquidus temperatures near 650°C (London

1992, 1996). A few beryl pegmatites, such as the most Be-rich body associated with the Sparrow pluton in the Yellowknife field, Canada, are higher than anticipated for such chemically simple pegmatites. Overall, however, the pegmatite compositions fall within the range expected from the experimental results of Evensen et al. (1999).

The data recorded from melt inclusions, such as in the beryl-bearing Ehrenfriedersdorf pegmatite (Webster et al. 1997), ostensibly provide the most reliable measures of trace-element abundances in magma, devoid of subsolidus hydrothermal remobilization. Melt inclusions in the Ehrenfriedersdorf pegmatite contain an average of 472 ppmw BeO (170 ppmw Be), which is comparable to other pegmatites shown in Table 3 (below). What is peculiar about these data, however, is that Be does not correlate well with other lithophile trace elements such as F, Sn, Rb, and Li (see R^2 values in Fig. 3, and further discussion below). Moreover, other element pairs that might be expected to correlate well (e.g., Li and Sn, Li and Rb) do not (Fig. 3e,f).

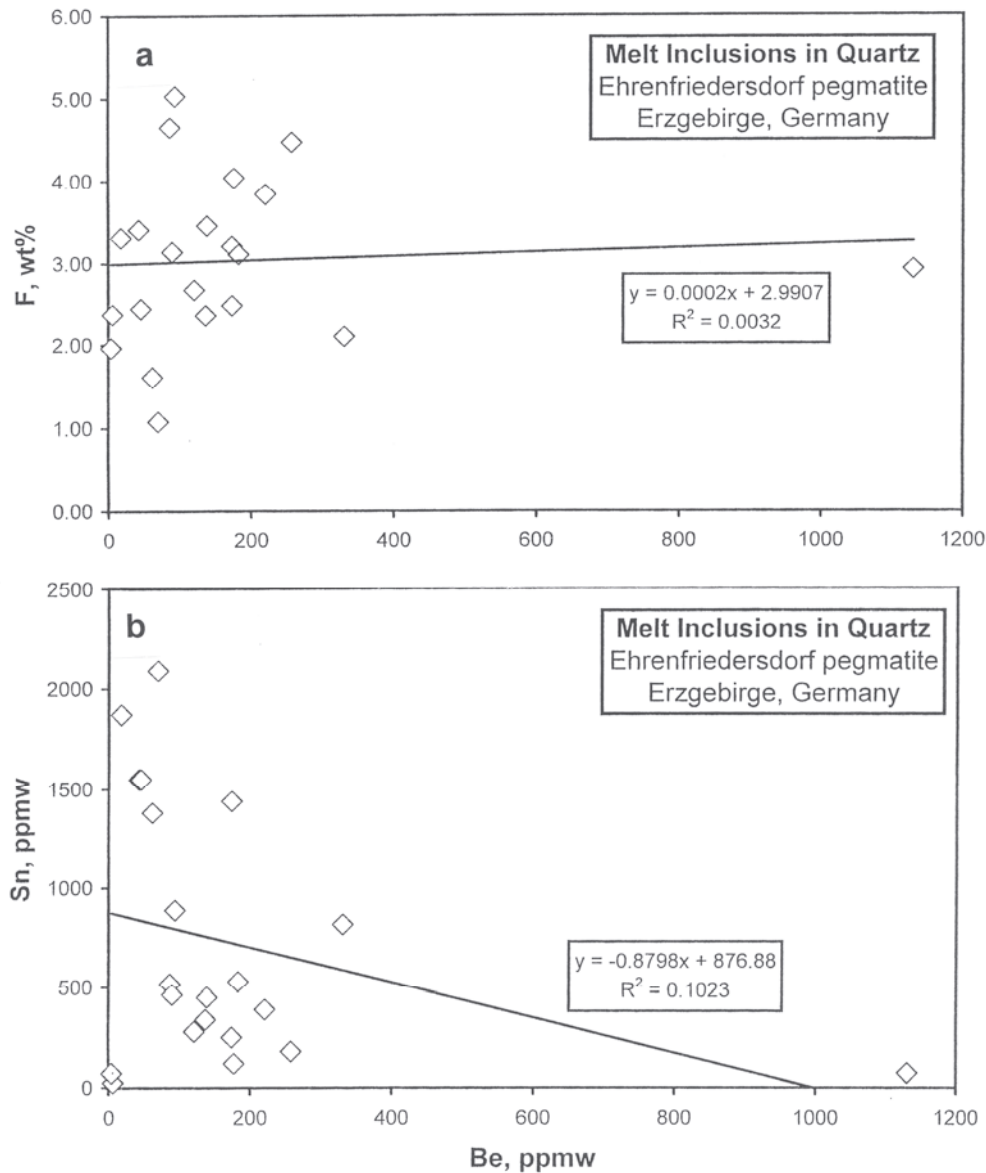
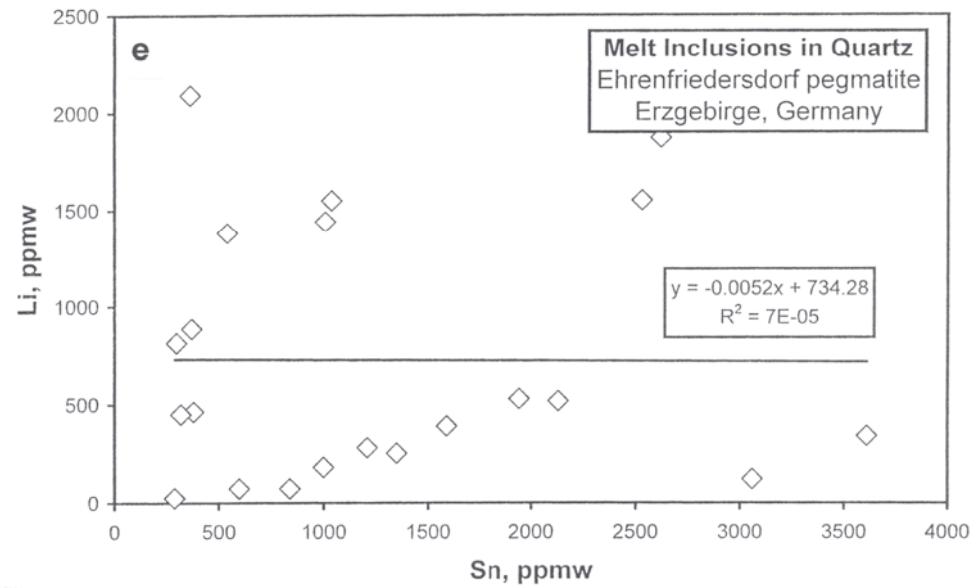
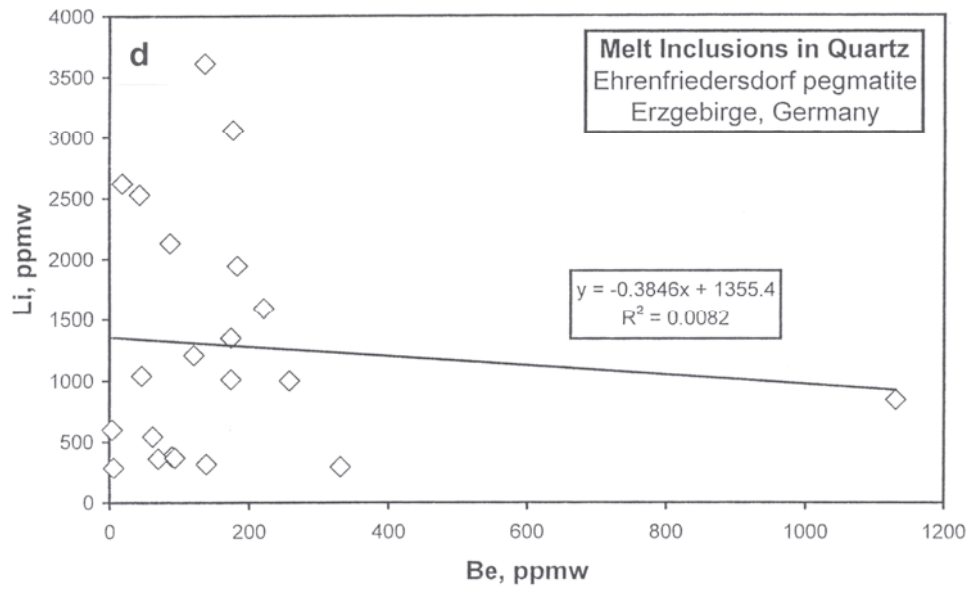
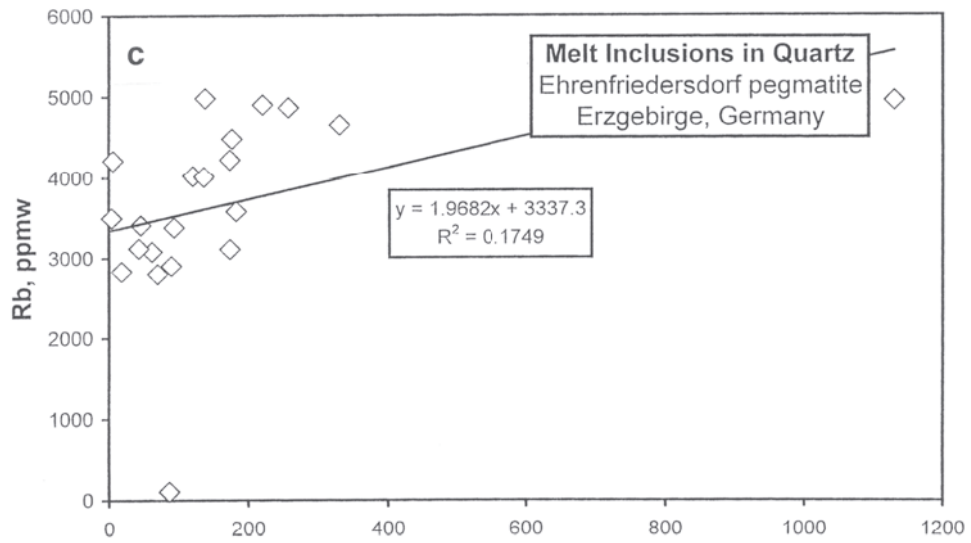
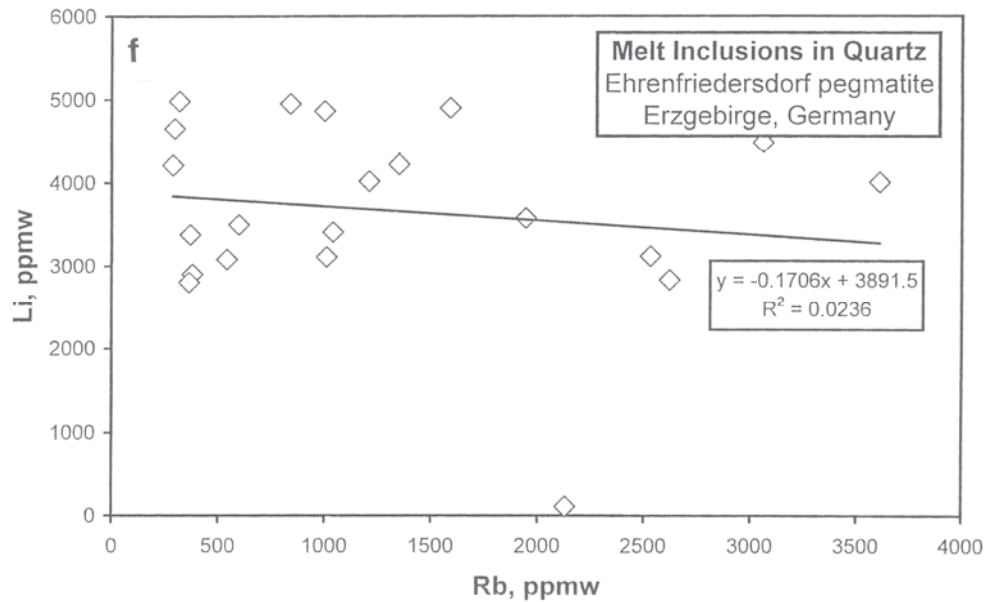


Figure 3. Plots of Be versus other elements and chemical indices of 21 melt inclusions hosted by quartz from the beryl-bearing, Sn-W Ehrenfriedersdorf pegmatite, Erzgebirge, Germany (from Webster et al. 1997). **See following pages for 3c-3f.**





BERYLLIUM BUDGETS FOR MAGMAS AT THEIR ANATECTIC SOURCES

Beryllium contents of protoliths

Having established how much Be is required to achieve beryl saturation in granitic magmas over a suitable range of temperatures, we now examine the likely sources of magma to assess the starting Be budget. Granitic magmas can arise by extended fractionation of mafic or even ultramafic precursors (e.g., Bowen 1928), but more felsic crustal materials of igneous (I-type) or sedimentary (S-type) origin are the most productive sources because their bulk compositions are closer to those of granites. Among these potential protoliths, mantle rocks are conspicuously poor sources of Be (<1 ppmw Be), and oceanic crust, whether fresh or altered by seawater, is negligibly higher (Ryan, this volume). Psammitic sedimentary rocks (arkose, greywacke) represent fertile sources of granitic magmas, but taken as a whole, psammitic and even pelitic sediments do not manifest substantial enrichment in Be (~2-5 ppmw; Grew, this volume). From the limited sources of data (see Grew, this volume), Be does not appear to be conspicuously mobile during prograde metamorphism: the Be content of metasedimentary rocks remains essentially unchanged up to conditions of the granulite facies of metamorphism. Consequently, mafic, felsic, and micaceous metamorphic rocks arrive at the amphibolite to granulite facies of metamorphism—where anatectic reactions can commence—with a bulk Be content of ~0-5 ppmw Be.

Mineral/melt partition coefficients

The abundance of Be in partial melts will vary with the proportions of phases that participate in the melting reactions and the distribution of Be between them and the melt phase. Mineral/melt partition coefficients are applicable if the minerals and melts closely approach chemical equilibrium. Though melting intervals may be too short for mineral-melt equilibrium to be achieved by intracrystalline diffusion (Bea 1996), some minerals, such as the micas, tend to re-equilibrate rapidly by simultaneous dissolution and reprecipitation of new phases in equilibrium with the melt (Johannes et al. 1994; Icenhower and London 1995). Mineral/mineral partition coefficients are more useful for

the purposes of estimating trace element abundances in systems that fail to achieve equilibrium between crystals and melt (Bea 1996). In either case, the likely Be content of an anatectic melt can be ascertained with knowledge of where Be resides in the phases of the protolith, and the proportions of phases involved in the melting reactions.

Relevant data for the distribution of Be among coexisting minerals or mineral/melt pairs are sparse. Table 2 lists reported average values, which are derived from only a few and very disparate geochemical systems and sources (i.e., experimental versus natural, mafic systems to ultrafractionated leucogranites). For phases that may be major contributors to granitic melts (e.g., quartz, feldspars, micas, and aluminous ferromagnesian phases) or restitic phases in equilibrium with those melts, the Be content of the coexisting phases decreases in the order:

Table 2. Partition coefficients for beryllium

Source	D[Be]	Reference
Phases	Avg (s.d.)	
phenocryst/groundmass of ongonite, Mongolia		Kovalenko et al. (1977)
K-feldspar/groundmass	0.53(0.19)	
albite/groundmass	1.56(1.46)	
Li-"mica"/groundmass	6.11(2.76)	
K-feldspar/albite	0.34	
Li-"mica"/K-feldspar	11.53	
Li-"mica"/albite	3.92	
restite/leucosome Peña Negra, Spain		Bea et al. (1994b)
K-feldspar/leucosome	3.05(0.3)	
plagioclase/leucosome	3.13(0.4)	
biotite/leucosome	15.5(0.9)	
garnet/leucosome	3.23(0.25)	
cordierite/leucosome	29.1(2.1)	
experimental synthetic metapelite		Evensen and London (1999)
quartz/melt	0.24	London and Evensen (2001)
K-feldspar/melt	0.14(0.11)	Evensen (2001)
albite/melt	0.19(0.13)	
oligoclase-andesine (An ₃₁)/melt	1.84(0.21)	
biotite/melt	0.46(0.07)	
muscovite/melt	1.35	
cordierite/melt	201 to 6.7	
experimental basalt-andesite		Brenan et al. (1998a)
olivine/melt	0.002	
orthopyroxene/melt	0.02	
clinopyroxene/melt	0.02	mafic melt
clinopyroxene/melt	0.01	intermediate melt
amphibole/melt	0.23	

quartz monzonite		Beus (1966)
K-feldspar/plagioclase	0.25	
K-feldspar/quartz	2.86	
K-feldspar/biotite	2.00	
biotite/quartz	1.43	
biotite/plagioclase	0.13	
coarse-grained biotite granite		Beus (1966)
K-feldspar/plagioclase	2.5	
K-feldspar/quartz	25	
T 2 cont'd K-feldspar/biotite	0.5	
biotite/quartz	50	
biotite/plagioclase	5	
muscovite granite		Beus (1966)
muscovite/plagioclase	5	
muscovite/quartz	250	
plagioclase/quartz	50	
coarse-grained "albitized" granite		Beus (1966)
muscovite/feldspar	2.45	
muscovite/quartz	49	
feldspar/quartz	20	
coarse-grained "greisenized" muscovite granite		Beus (1966)
muscovite/feldspar	2	
muscovite/quartz	31	
feldspar/quartz	16	

cordierite » calcic oligoclase \geq muscovite > biotite > quartz \cong albite \cong K-feldspar (4)

The position of plagioclase in this order depends on its anorthite content. The Be content varies with the An content and reaches a maximum around the oligoclase-andesine boundary (Beus 1966; Kosals et al. 1973). In our recent experimental work, we have established a partition coefficient of 1.84 for plagioclase of An₃₁ (Evensen 2001), which compares with a value of 0.19 for albite (Table 2). The partitioning sequence of (4) above is essentially the same for natural versus experimentally determined values except in the relative position of quartz. Our recent experimental results (London and Evensen 2001) show Be to be more compatible in quartz than in the alkali feldspars, but this is not the case in natural minerals (Grew, this volume). We do not know if this is a vagary of our experiments, or if natural quartz tends to lose Be during retrograde recrystallization, which is prevalent in igneous and especially metamorphic rocks and manifested by ductile deformation, healed cracks, etc. The distribution of Be between quartz phenocrysts and their associated melt inclusions in volcanic ignimbrites could resolve the question if the quartz-inclusion pairs truly represent a quenched magmatic system near equilibrium, but those data do not appear to exist at the present time.

Values of $D_{\text{Be}}^{\text{mica/fds}}$ are on the order of 1 to 10, so that mica-rich protoliths (metapelites) generally represent the most fertile sources of Be-enriched magmas. In natural micas, $D_{\text{Be}}^{\text{musc/biot}}$ appears to be greater than 1 (Grew, this volume), which is consistent with the experimental results reported by London and Evensen (2001) in Table 2.

Partition coefficients for garnet are similar to those of the feldspars (Bea et al. 1994b).

The role of cordierite during anatexis. Cordierite stands out as an important reservoir of Be. Among the common rock-forming minerals, Be is highly compatible in cordierite, and partition coefficients for Be between cordierite and all other coexisting minerals are large. The experimentally derived values of $D_{\text{Be}}^{\text{Crd/melt}}$ cited in Table 2 (Evensen and London 1999) range from 201 to 7 between 700° and 850°C at 200 MPa H₂O. These data fit the linear regression,

$$D_{\text{Be}}^{\text{Crd/melt}} = -1.37 T + 1145, \quad (5)$$

with $R^2 = 0.88$ over this temperature interval. In cordierite, Be goes into a T^[4] site for Al, with two different substitution mechanisms: Na_{channel}BeAl₋₁ or BeSiAl₂ (Hölscher and Schreyer 1989; Hawthorne and Huminicki, this volume; Grew, this volume). Experiments reveal that these two substitution mechanisms vary inversely with temperature: Na_{channel}BeAl₋₁ predominates at low temperature whereas BeSiAl₂ predominates at high temperature (Evensen and London 1999). At the temperatures of anatexis, the exchange component BeSiAl₂ controls the compatibility of Be in cordierite and hence the distribution of Be between cordierite and melt. There is extensive miscibility between beryl and cordierite or its hexagonal high-temperature polymorph indialite, such that complete miscibility exists between these phases above 900°C at 200 MPa H₂O (Fig. 4).

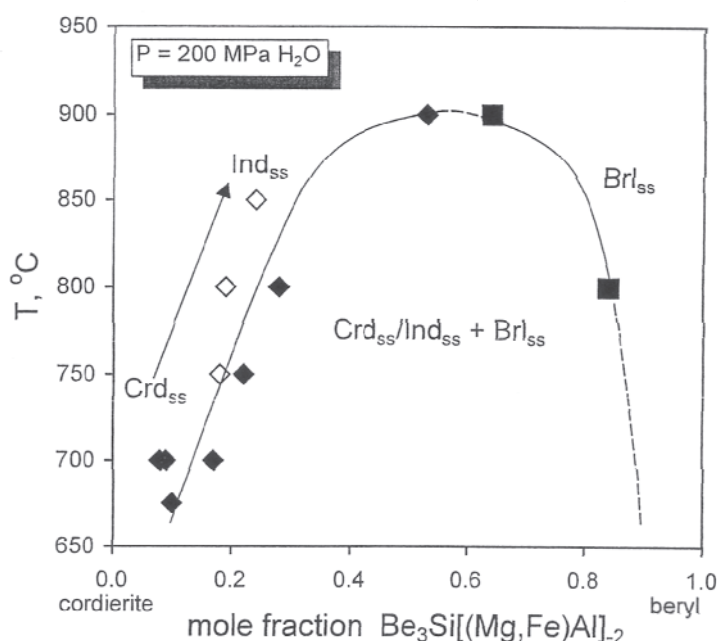


Figure 4. Experimental miscibility relations between cordierite (Crd) or indialite (Ind) [diamonds] and beryl [■] in granitic melt as a function of temperature at 200 MPa H₂O. Coexisting Crd/Ind and beryl are solid symbols; experiments resulting in only Crd/Ind are open diamonds. Uncertainties in temperature and analytical errors are smaller than the symbols used. The exchange operator which converts Crd (at X = 0.0) to Brl (at X = 1.0) accounts for ~65% to 85% of the composition of the synthetic minerals. The solvus is hand-drawn. Modified from Evenson (2001) based on the SIMS analyses of experimental run products.

Summary of partitioning data . The values of $D_{\text{Be}}^{\text{min}^1/\text{min}^1}$ summarized in Table 2 are mostly similar among the various studies. There is generally good agreement between the natural and experimental values of $D_{\text{Be}}^{\text{min}^1/\text{min}^1}$, which lends some support for the experimental $D_{\text{Be}}^{\text{min}^1/\text{melt}}$ values as well (Evensen and London 1999). The distribution of Be between minerals and groundmass or leucosome (Table 2) stand out as anomalous because they show moderately to strongly compatible behavior for Be in all rock-forming minerals except K-feldspar in the ongonites (Kovalenko et al. 1977; Bea et al. 1994b). Were these values correct, then partial melts starting with a few ppmw Be could never attain beryl saturation. We suggest that the experimental values (Evensen and London 1999; London and Evensen 2001; Evensen 2001) constitute a more reliable data set.

The Be content of aluminous anatectic melts will be controlled strongly by the

participation of micas and cordierite in melting reactions. White micas are expected to melt or decompose completely at most conditions of anatexis, and the process of incomplete melting of micas followed at higher temperature by reactions such as



can effectively transfer a large fraction of the trace element content of white micas (dark micas, too) to a small fraction of partial melt (London 1995). In cordierite-bearing rocks, virtually all Be will reside in cordierite, and the Be budget of partial melts will be almost entirely controlled by the degree to which this phase participates in melting reactions. For the cordierite-bearing migmatites of Peña Negra, central Spain, Bea et al. (1994b) cite an average of 2.3 ppmw Be in the paleosome of the migmatites, and 6.0 and 1.2 ppmw Be respectively in the melanosome (restite-rich) and leucosome (melt-rich) portions of the migmatitic rocks. This yields an apparent restite(melanosome)-melt(leucosome) distribution coefficient of 5, which is close to the partition coefficient for Be between cordierite and melt at high temperature (Evensen and London 1999). Del Moro et al. (1999) also document the distribution of Be between various lithologic facies of migmatites in Italy, but the Be contents are so low throughout the different rock types (reported as 0.00-2.87 ppmw Be) that no trends are evident.

Using partition coefficients to estimate the Be contents of melts. The partition coefficients presented in Table 2 could be used to assess the Be content of melts by the combination of mineral/mineral coefficients with mineral/melt values. For example, values of $D_{\text{Be}}^{\text{min}/\text{K-fds}}$ are known, as are the value of $D_{\text{Be}}^{\text{K-fds}/\text{melt}}$ from the experiments by Evensen and London (1999). Thus, in relation to one of the best and most widely used indicators of chemical fractionation in granite-pegmatite systems (K-feldspar; e.g., Černý 1994), the concentration of Be in melt could be ascertained and used, for example, as a prospecting tool for Be-rich pegmatites. Taking the range of 200 to 900 ppmw BeO for beryl-saturated melts, K-feldspar in equilibrium with those melts should contain ~5 to 27 ppmw BeO. These values closely match the range of values for K-feldspar samples that have been analyzed for Be (see citations in Parsons (1994) and Roda Robles et al. (1999) for pegmatites that are not distinctly beryl-rich). This method does offer promise for exploration and evaluation of pegmatitic Be deposits specifically, but it cannot be tested with the existing database.

THE BERYLLIUM CONTENT OF SILICIC IGNEOUS ROCKS

The data base for Be in rhyolites and granites is quite robust in comparison to that of pegmatites. However, Be is not routinely analyzed in whole-rock studies (because x-ray fluorescence methods dominate in modern analytical laboratories), and Be does not appear as a keyword in citations of relevant petrologic articles. Finding these data, therefore, is hit-or-miss. The data sources gleaned for this review encompass about 400 analyses in all. The purposes of this literature survey are to establish mean Be contents of obsidians, rhyolites, and granites at various degrees of chemical fractionation, and to explore correlations among Be, other elements, and the protolith (source type) or tectonic setting of the magma source.

Obsidians and rhyolites

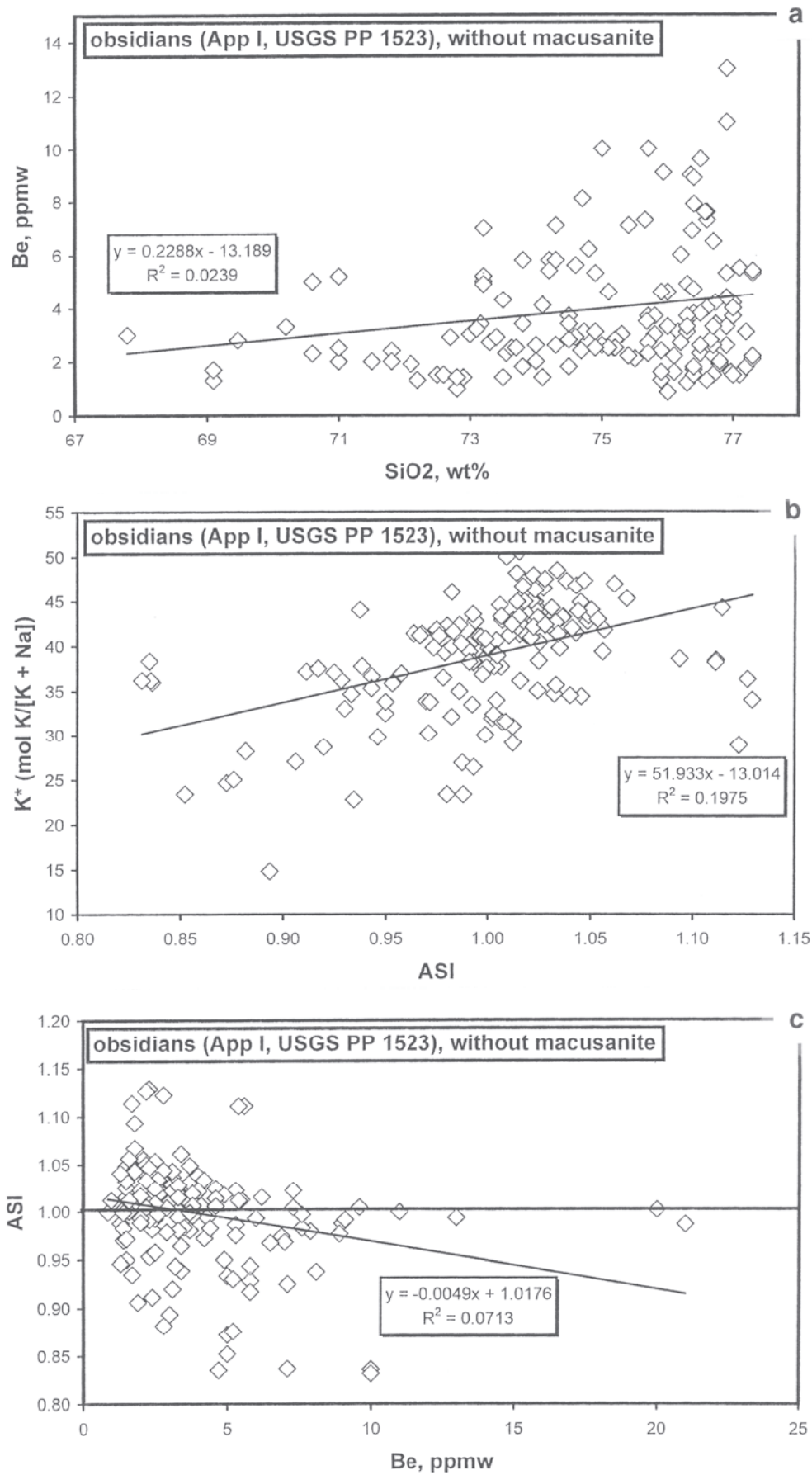
As a whole, obsidians represent the closest approximation to silicic melt compositions that are readily available in the geological literature. Though trapped melt inclusions may more accurately preserve the volatile components of melts, analyses of these are comparatively few.

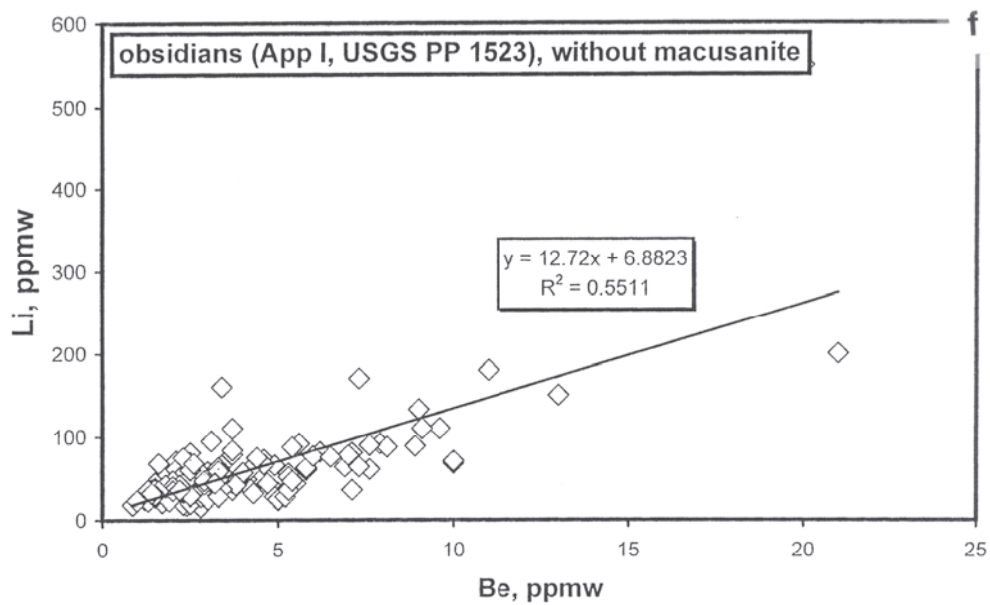
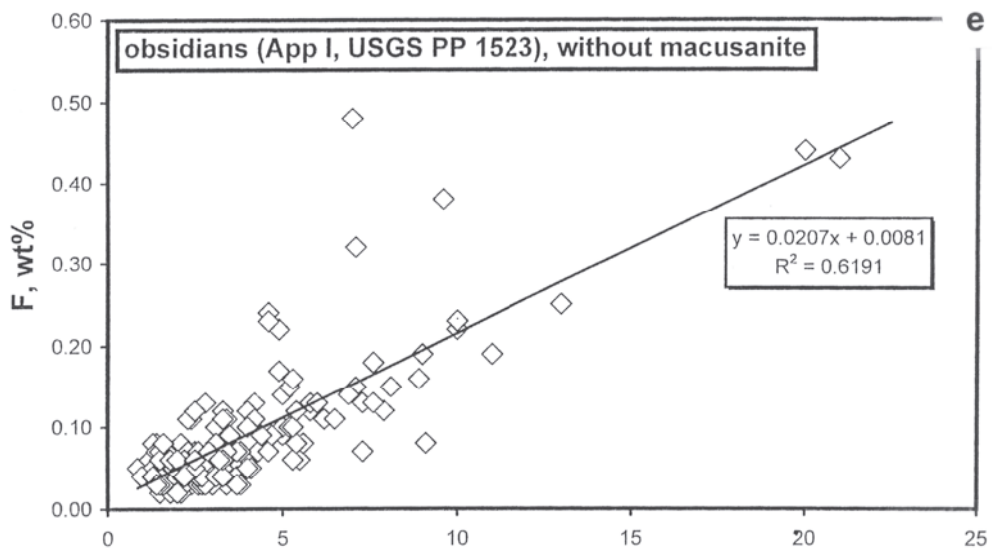
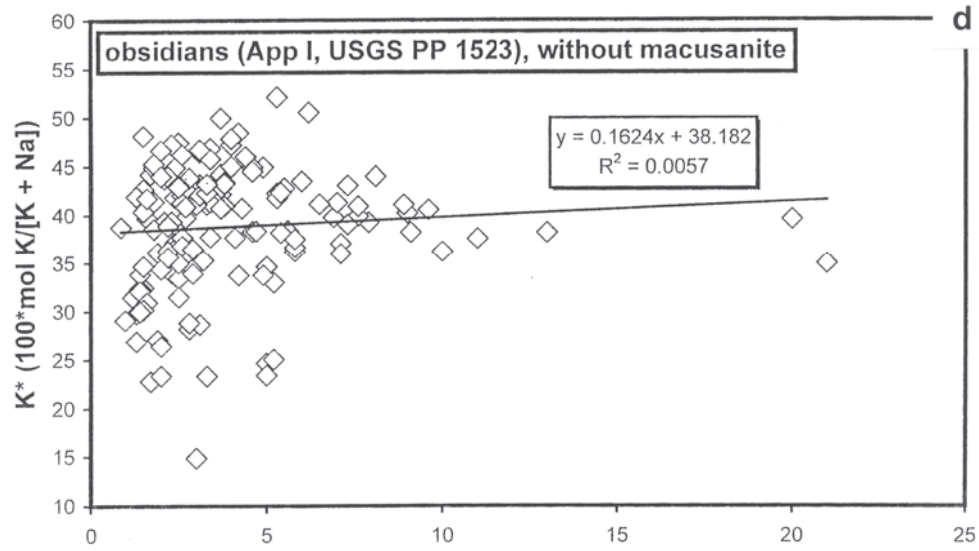
Shalkaline silicic rhyolites . Macdonald et al. (1992) compiled a large chemical database for subalkalic silicic obsidians, which will be referred to here as the USGS (for

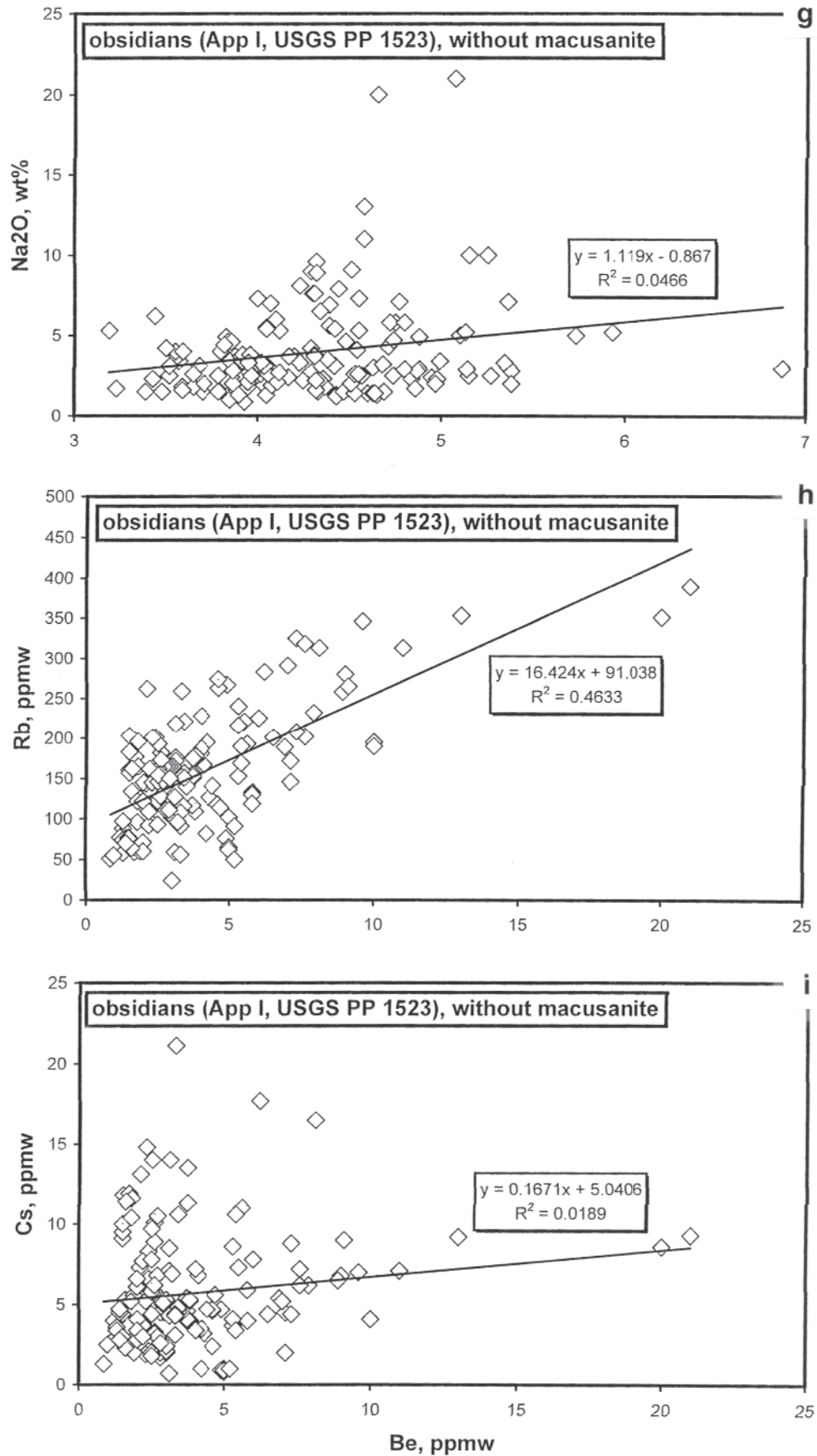
U S Geological Survey) data set. Their Appendix I, though heavily skewed toward North American localities, contains 152 analyses that include Be along with other trace and major elements. For the total data set, the average Be content is 4 ppmw with a standard deviation (s.d.) of 3 ppmw. In terms of tectonomagmatic groups recognized by Macdonald et al. (1992), rhyolite obsidians from continental interiors contain the highest average Be content with 6.9 ppmw (range: 1.8-32 ppmw). Note that the Be contents of melt inclusions in quartz from undifferentiated Paleozoic tonsteins (kaolinized K-rich metaluminous to mildly peraluminous rhyolites of continental origin) are similar with a mean 7.6 ppmw Be (s.d. = 4.6, N = 27; Webster et al. 1995). Next highest are obsidians from islands over “oceanic extensional zones”, with an average 4.7 ppmw Be and a narrower range of 1.9-5.8 ppmw. Obsidians derived from subduction-related volcanism at continental margins and mature island arcs mostly fall well below the average for the entire data set, with means of 2.6 (1.3-5.5) and 1.6 (0.86-3.2) ppmw Be, respectively. As a whole, these trends broadly reflect the more fractionated compositions of continental rocks, and perhaps more specifically that continental crustal rocks contain a larger fraction of minerals that can more readily accommodate Be, such as the micas.

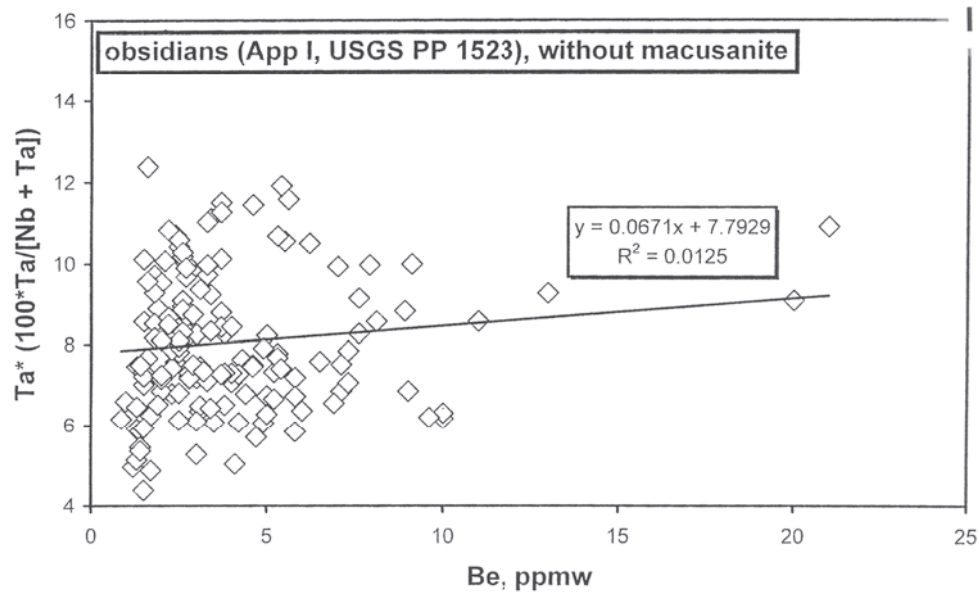
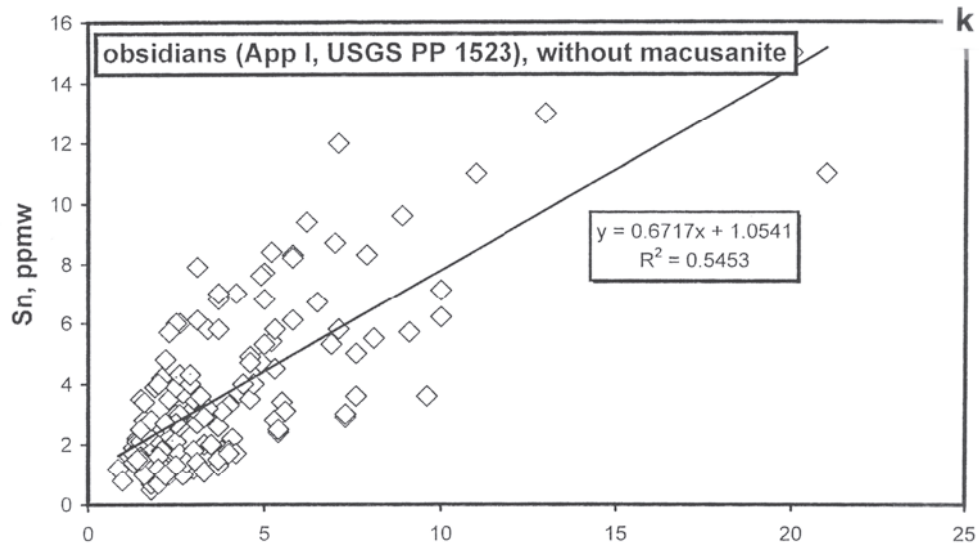
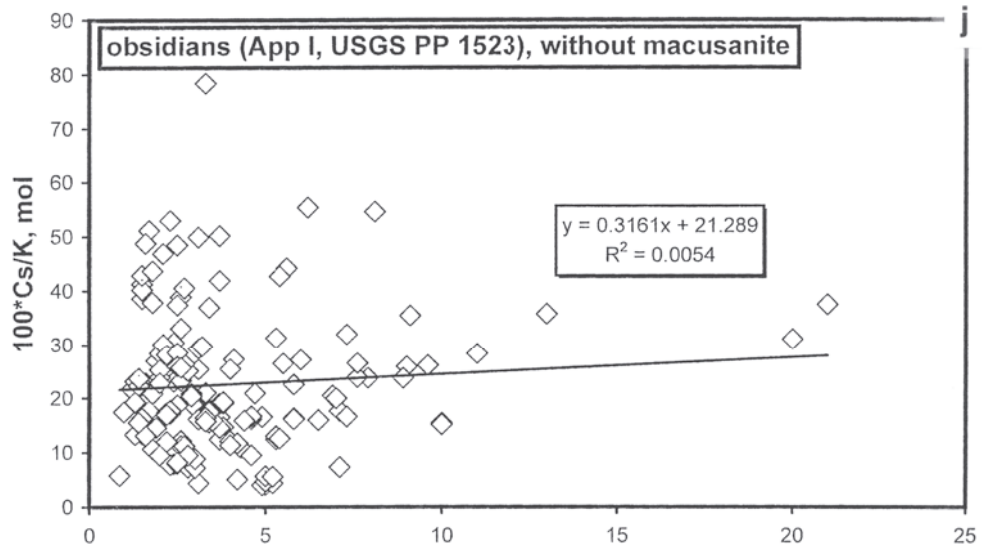
A correlation matrix for the USGS data set (Macdonald et al. 1992) reveals the most positive associations between Be and F (0.82), Sn (0.82), Rb (0.75), Pb (0.74) U (0.70), Li (0.68), and the REE (see Figs. 5e-f, 5h, and 5k). Positive correlations of Be with Sn, Rb, U, and Li are well documented in these and other geochemical discrimination plots, but these do not uniquely distinguish the sources of the magmas. Enrichment in this suite of elements is as characteristic of intracontinental rift-related rhyolites (A-types) as of collision-related metasedimentary S-types (e.g., Christiansen et al. 1986; Černý et al. 1985; Černý and Meintzer 1988; Černý 1991a; London 1995). The USGS data set can be scanned as a whole, including the data from “continental interiors” and “continental margins”, to identify magmas with a large S-type component, as these are distinctly peraluminous and tend to have high K^* (atomic $K/[Na+K]$) (White and Chappell 1983). The data set contains very few peraluminous obsidians (ASI or $A/CNK \geq 1.1$) with the exception of the green glass pebbles from Macusani, Peru (Noble et al. 1984), which have been omitted from the plots in Figure 5. The poorly defined trend of decreasing ASI with increasing Be (Fig. 5c) may be best ascribed to Be enrichment in the mildly alkaline A-type rhyolites, which are included in the USGS data set but are discussed separately below. Although there is a weak positive correlation between K^* and ASI (Fig. 5b), which might be indicative of mica-rich sources, we note that the peraluminous rhyolites with $ASI > 1.1$ actually lie below the trend line for the entire USGS data set. To further identify the S-type protoliths in the USGS data, the values for Be were plotted against Cs^* (atomic $100 \cdot Cs/K$) and Ta^* (atomic $Ta/[Ta+Nb]$). In addition to Li (Fig. 5f), these ratios are diagnostic of the suite of mostly S-type, granite-associated ore deposits with the LCT (Li-Cs-Ta) chemical signature (Černý 1991a). Figures 5j and 5l show no correlation of Be with Cs^* , or Ta^* for the USGS data. In addition, the mean values of Cs^* or Ta^* are low, 45 and 8 respectively, in comparison, for example, to the strongly peraluminous and distinctly S-type Macusani rhyolite obsidian (e.g., Pichavant et al. 1988), for which $Cs^* = 4670$ and $Ta^* = 30$. We conclude that the only S-type obsidian in the USGS data set is from the Macusani rhyolite, Peru.

Figure 5. Plots of Be versus other elements and chemical indices of 151 subalkalic silicic obsidians presented in Appendix 1 of Macdonald et al. (1992). Though reported by Macdonald et al. (1992), these plots do not contain the green obsidian glass from the Macusani volcanic province of southeastern Peru (Barnes et al. 1970) because of the extreme skewness that results from including this obsidian in the plots. **Figures 5a-l are on the following four pages.**









A-type rhyolites. The topaz (A-type, or intracontinental rift-related) rhyolites of the western U.S. and Mexico are notably Be-rich. Some of these volcanic units in Utah have produced gem red beryl from hydrothermal veins (Keith et al. 1994), and others enriched in bertrandite comprise the largest Be deposits in North America (Lindsey 1977). Rhyolites from Utah exclusive of Honeycomb Hills contain an average 14.1 ppmw Be (s.d. = 7.5, N = 7; Christiansen et al. 1986). Honeycomb Hills is anomalous with an average 87.7 ppmw Be (s.d. = 91.3, N = 6; Congdon and Nash 1991), and Congdon and Nash (1991) proposed that the high Be contents of some samples, up to 267 ppmw, result from subsolidus hydrothermal concentration. Note that the high Be content of Honeycomb Hills is not matched by similarly high F, Li, or Sn, and hence supports the hypothesis that anomalously high values in the Honeycomb Hills are not the result of igneous fractionation. In the topaz rhyolite data set, Be shows weakly positive correlations with Rb, Cs, and Sn (Figs. 6d and 6g,h); there is no correlation of Be with Li or F (Figs. 6b,f). Melt inclusions from quartz phenocrysts in tin-rich topaz rhyolites from the Sierra Madre Occidental, central Mexico, contain an average of 18.0 ppmw Be (s.d. = 4.4, N = 35) (Webster et al. 1996), which is close to the whole-rock values for similar

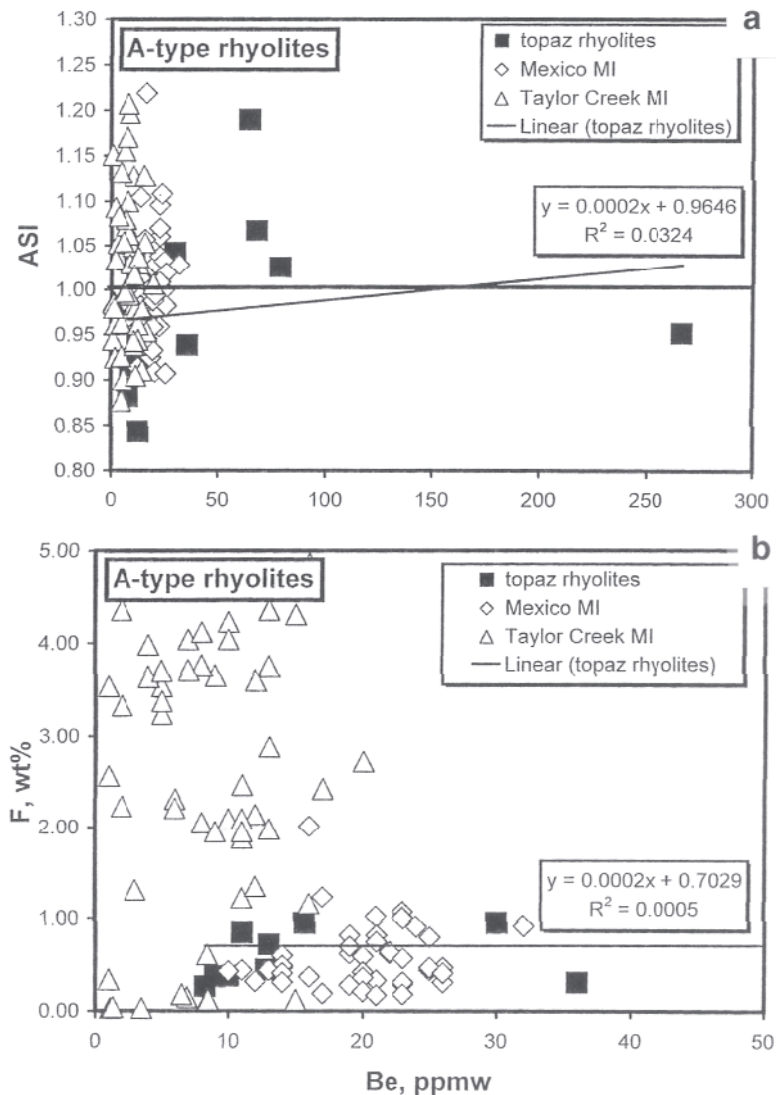


Figure 6. Plots of Be versus other elements and chemical indices of topaz rhyolites from the western U.S. (from Christiansen et al. 1986), and vitreous melt inclusions (MI) from F-rich rhyolites of New Mexico (Webster et al. 1991a) and Mexico (Webster et al. 1996).

Figure 6

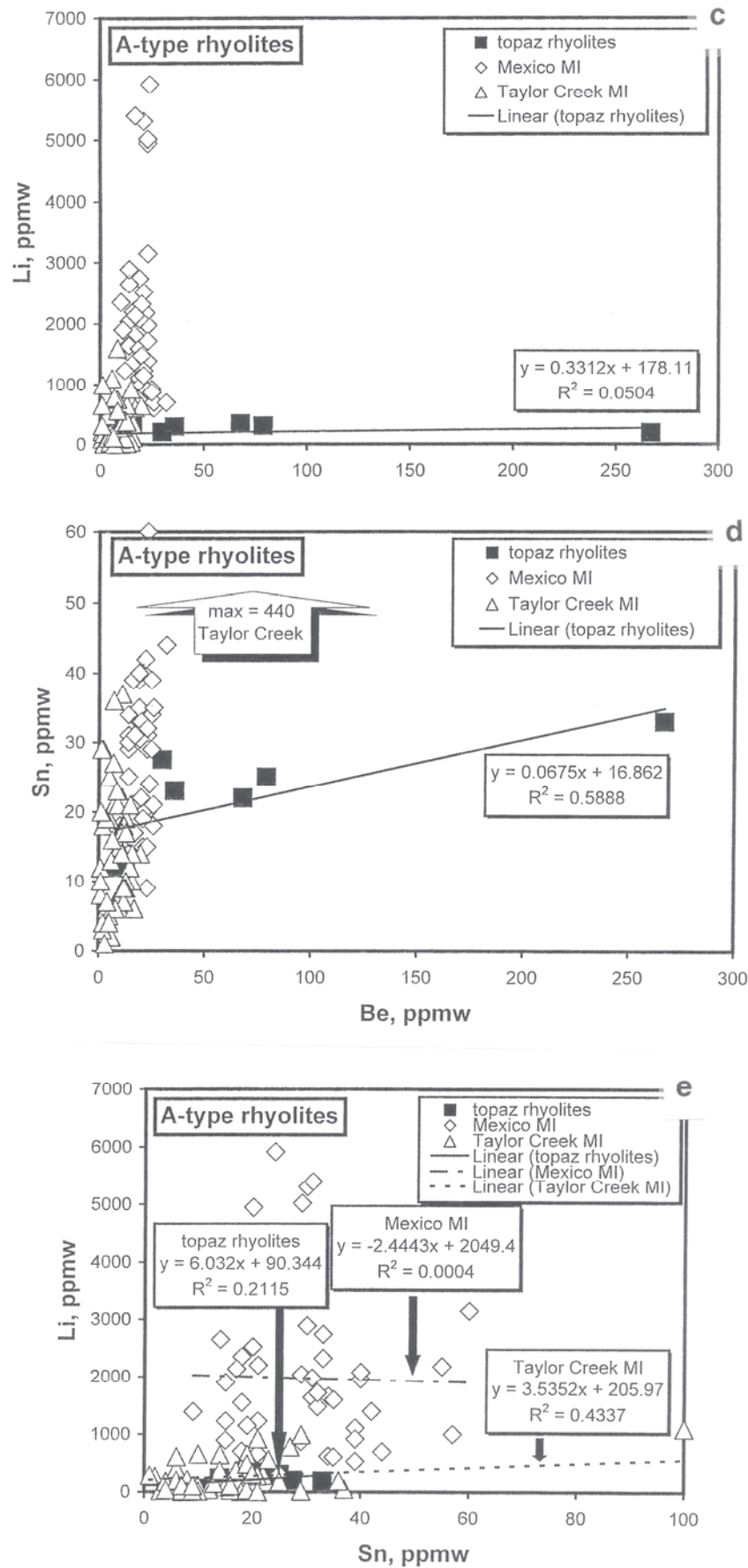
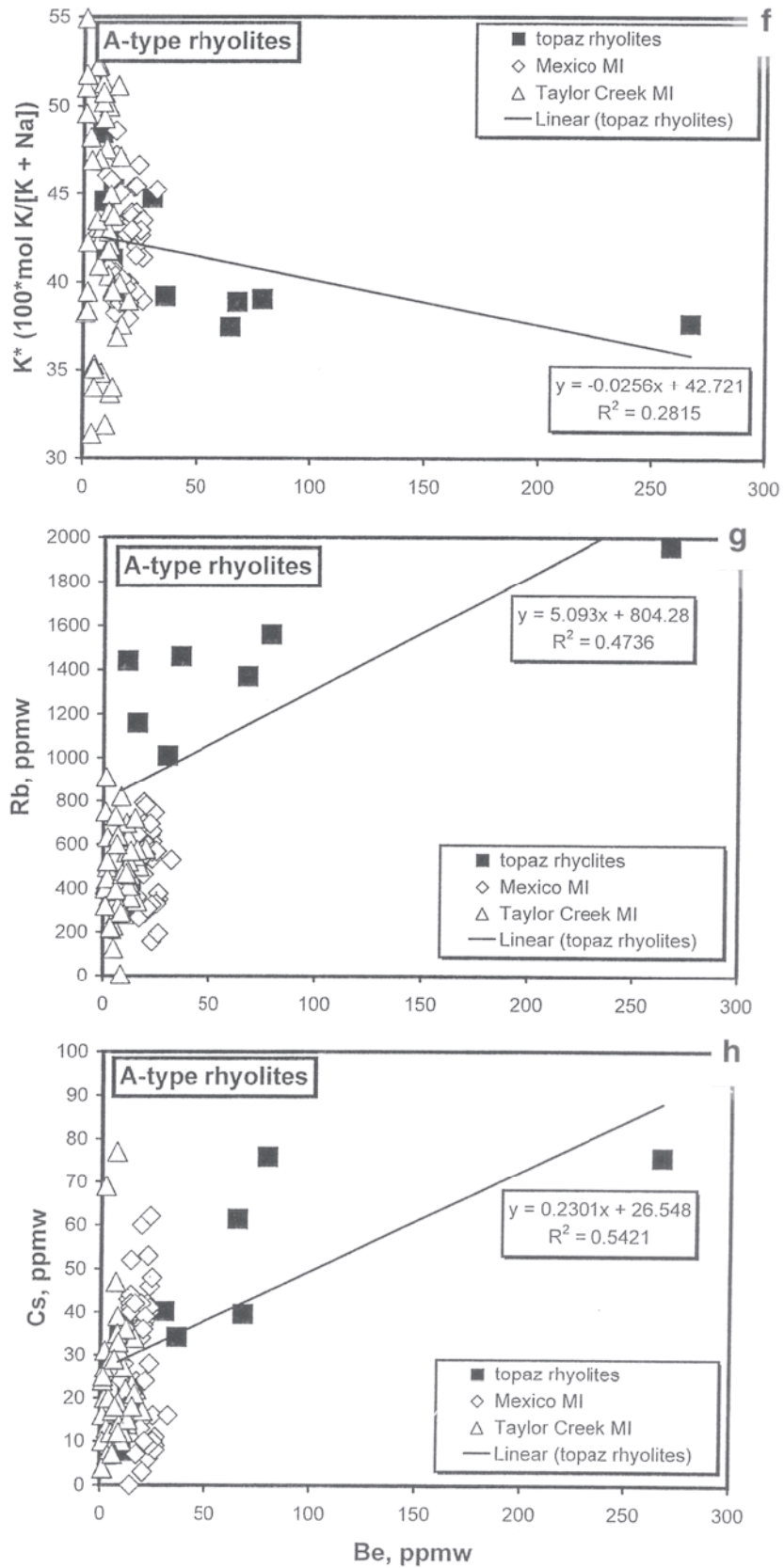


Figure 6



rhyolites in Utah. Analyses of melt inclusions from the F- and Sn-rich Taylor Creek rhyolite, New Mexico, are grossly similar to those from the Mexican localities except for lower average Be contents of 8 ppmw (s.d. = 4.7, N = 53; Webster and Duffield 1991a,b). Figure 6 illustrates, however, that there is no correlation of Be with any of the typical fractionation indices in the melt inclusions of either data set because there is relatively little change in Be abundance while there are large variations in the other fractionation indices. The lack of correlation of Be with the other incompatible lithophile trace elements is surprising, and distinct from the trends of the USGS data set (cf. Figs. 5 and 6). While plots such as Figures 6c and 6d might give the impression that elements such as Li and Sn should be highly correlated in the melt inclusions, Figure 6e demonstrates that this is not necessarily so. Though there is a positive correlation of Li with Sn in the Taylor Creek inclusions, there is no correlation in the melt inclusions from the topaz rhyolites of Mexico. In total, the melt inclusion data appear to reflect a bulk crystal/melt distribution coefficient of 1 for Be, which is possible when, for example, calcic oligoclase represents a large component of the fractionating crystalline assemblage (discussed further below). It is not our purpose, however, to interpret the melt inclusion data in relation to the other geochemical aspects of these rhyolites. Overall, the F- and Sn-rich A-type rhyolites and their trapped melt inclusions establish a mean Be content of about 14-18 ppmw in these evolved melts, and that is the important point for this assessment.

S-type rhyolites. The highest single value in the USGS obsidian data set (32 ppmw Be) comes from the green glass nodules from the Macusani volcanic center in southeastern Peru. The less-altered tuffs and pumices from the Macusani province contain 28 (s.d. = 9, N = 4) ppmw, with a high of 37 ppmw Be (Noble et al. 1984). These rhyolites are S-type (metasedimentary protoliths) in overall chemical character with high $^{87}\text{Sr}/^{86}\text{Sr}_o$ of 0.7309, strongly peraluminous compositions, and trace-element patterns that follow from an abundance of argillic or micaceous material at the source (Pichavant et al. 1988). To the southeast in the Bolivian tin belt, the voluminous Los Frailes and Morococala volcanic fields consists of similarly peraluminous S-type ignimbrites (Ericksen et al. 1990; Morgan et al. 1998). The Los Frailes volcanics vary from dacites to rhyolites; they are, as a group, less fractionated than the Morococala volcanics, which range in composition from quartz latites to cordierite-andalusite rhyolites to muscovite-andalusite rhyolites (Morgan et al. 1998). This is evident in the trace-element systematics of the two fields. The mean Be content of the Los Frailes volcanics is 4.3 ppmw (s.d. = 1.0, N = 26; Luedke et al. 1997), whereas that of the Morococala field is 10.0 ppmw Be (s.d. = 4.6, N = 12; Luedke et al. unpublished data). Other fractionation indices, e.g., Li and Rb, similarly reveal the greater chemical fractionation of the Morococala field over that of Los Frailes (Fig. 7a). Note that although Li and Rb are well correlated in the Morococala field, this is not true of the Los Frailes volcanics. Figure 7b illustrates that Be increases with ASI, which corresponds to the general trend of increasing fractionation in both volcanic fields. The correlation of Be with ASI is more pronounced for the Los Frailes rocks than for the Morococala suite, because all of the Morococala units possess high ASI values near the magmatic saturation limit (discussed below under granites). Increasing Be is weakly to negatively correlated with K^* (Fig. 7b), as is the case for the A-type rhyolites (Fig. 6f). A decrease in K^* with increasing fractionation, as is seen in both S- and A-type volcanics, mirrors a similar trend observed in granitic rocks. The decrease in K^* is thought to stem from homogeneous reactions in melt of anions such as F, P, and B with Na, which drives residual liquids to Na-richer compositions as more potassic phases (K-feldspar, joined by mica) increasingly dominate the crystalline mode (London 1997). The correlation of Be with Li, Rb, and Sn in the two Bolivian volcanic fields (Fig. 7c) ranges from good (with Rb in the Los Frailes volcanics) to poor to negative (with Rb in the Morococala field).

Both Bolivian volcanic fields are chemically less evolved than those of the Macusani

province to the northwest (cf. Pichavant et al. 1988 with Luedke et al. 1997; Morgan et al. 1998). Given that peraluminous S-type rhyolites are, globally, comparatively few in number, the much-studied Macusani volcanics clearly should not be construed as typical of these magma types. Using the available data, the Be contents for these S-type volcanics in Bolivia and Peru span the full range seen for all obsidians in the USGS data set.

Summary of data on rhyolites . Without further assessment of the individual sources, the averages for the four groupings used by Macdonald et al. (1992) probably represent suitable starting points for modeling the fractionation of Be in most large-volume silicic magma bodies. All of the obsidians in the USGS data set possess negative Eu anomalies, which become more pronounced toward the magmas from “continental interiors”. Hence, various degrees of restite separation and crystal-liquid fractionation have occurred in all of these magmas. The most fractionated rhyolites include the topaz rhyolites of the western U.S.A. and Mexico, and the Macusani rhyolite and obsidian from Peru (e.g., Christiansen et al. 1986; Pichavant et al. 1988). Their Be contents are the highest among the rhyolite data sets. We now compare these values with those from a diverse suite of plutonic rocks.

Granites

Unlike the data sets for rhyolites, which collectively give good coverage of Be in obsidians, rhyolites, and associated melt inclusions from only a few published sources, the sources of data on Be in granites are more numerous but less comprehensive—meaning that the data sets are essentially limited to individual plutons. Also, in contrast with the USGS obsidian suite, peraluminous and probably S-type granites comprise a large proportion of the data cited in Table 3. The sources of the peraluminous granites are skewed to European localities, most of which contain accessory cordierite or are likely to have originated in cordierite-bearing sources. In Figure 8, Be is plotted against the same geochemical parameters as in Figures 5-7 for most of the granite sources.

Because of the high compatibility of Be in cordierite, we can recognize in Table 3 granites that have achieved a high degree of chemical fractionation but are poor in Be. These include the granites of Spain and the Czech Republic, where at least some facies in each data set can be regarded as highly fractionated (Bea et al. 1994a; Breiter et al. 1991; Breiter and Scharbert 1995; Ramirez and Grundvig 2000). When grouped as representative of a region, the Jalama and Pedrobernardo plutons in Spain (Bea et al. 1994a; Ramirez and Grundvig 2000) and the Argemela microgranite dikes in Portugal (Charoy and Noronha 1996) display positive correlations of Be with Li, Rb, and Sn (Fig. 8a). This is actually an artifact of the two different populations, the Spanish granites on one end and the Portuguese dikes on the other. As typical representatives of the central Iberian granite system, the Jalama and Nisa-Albuquerque plutons are similar: they are peraluminous with cordierite-bearing interior facies and evolved tourmaline-bearing marginal facies (Ramirez and Menendez 1999; Ramirez and Grundvig 2000). Cordierite (whether magmatic or restitic) likely constitutes the principal mineralogical reservoir of Be, and hence variations of Be with fractionation are relatively small (i.e., note the small standard deviations for Be in Table 3). There is no correlation of Be with the typical trace elements cited above for the rhyolite data set. In the Nisa-Albuquerque batholith, for example, the correlation (R^2) of Be with Rb is 0.10 and with Sn it is 0.03. In the Pedrobernardo pluton of central Spain, Bea et al. (1994a) proposed that the uppermost of three internal facies was derived from the lower portions by a melt extraction process

Figure 7 (on the next page). Plots of Be versus other elements and chemical indices in S-type peraluminous volcanic rocks from the Los Frailes and Morococala volcanic fields of Bolivia (Luedke et al. 1997; Morgan et al. 1999).

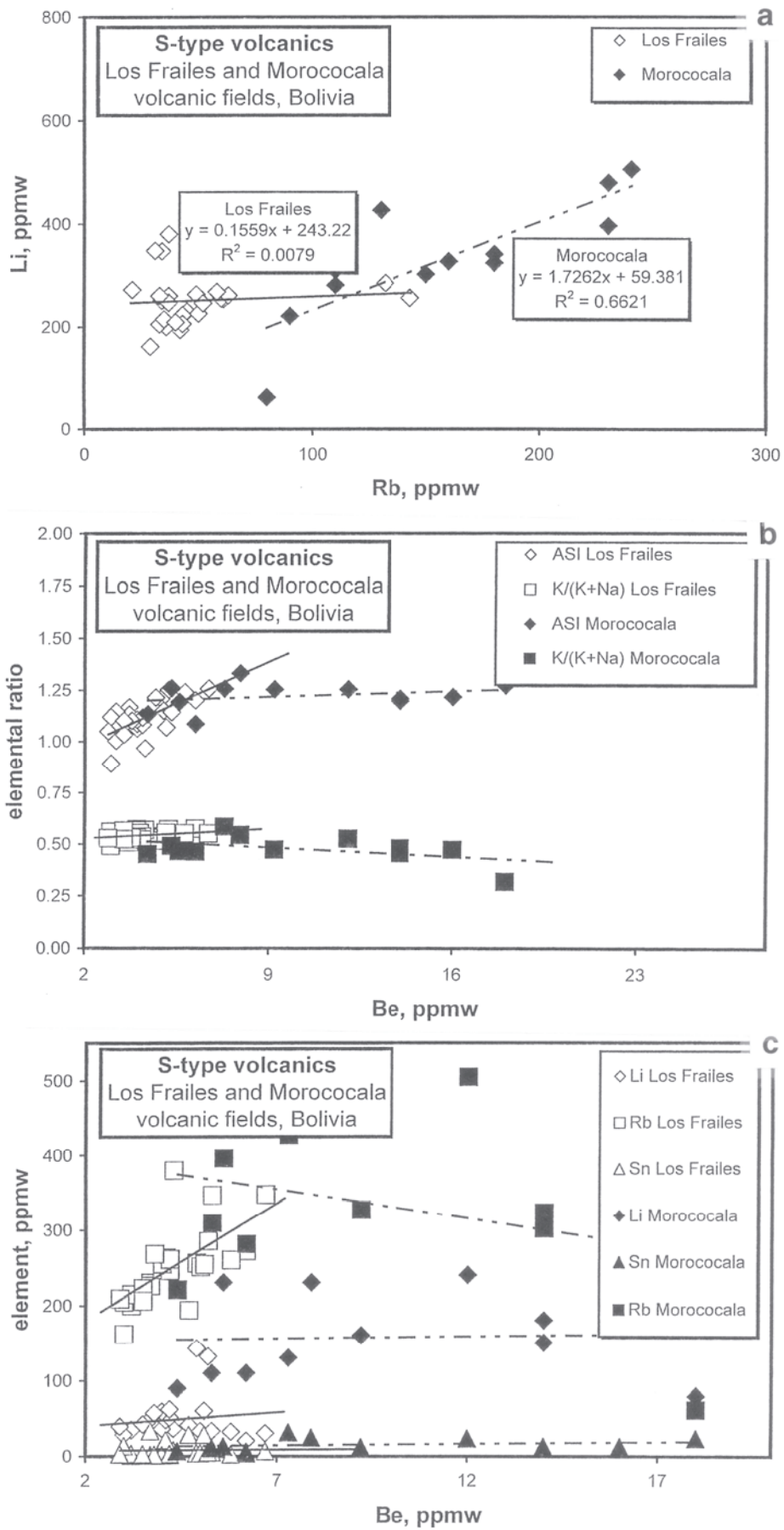


Table 3. Average beryllium content of granites.

Location	Granite type Element enrichment Other comments	Average Be ppmw (S.D.) ^N	Source
Pedrobernardo Spain	S-Type P Crd source	4.1(0.6)11	Bea et al. (1994a)
Jalama Spain	S-type P	7.2(3.1)13	Ramirez & Grundvig (2000)
Nisa-Alburquerque Portugal and Spair	S-type Crd-bearing	5.0(3.0)9	Ramirez & Menendez (1999)
Argemela Portugal	S-type P beryl-bearing	130(99)10	Charoy & Noronha (1996)
Beauvoir Massif Cent'l, Fr.	S-type Li, Sn, Ta, P	101.6(116.0)45	Raimbault et al. (1995)
Bohemia Czech Republic	S-type? Sn, P	12(10)10	Breiter et al. (1991)
Homolka Czech Republic	S-type P Crd source	2.3(0.7)10	Breiter & Scharbert (1995)
Milevsko Czech Republic	peraluminous felsite dikes	23(19)7	Vrána (1999)
Pelhrimov Czech Republic	felsite dikes	5.5(ND)4	Vrána (1999)
Lasenice Czech Republic	felsite dikes	1.6(ND)11	Vrána (1999)
East Transbaikalia Russia	S-type (?) Li, Ta, F	28(41)11	Zaraisky et al. (1997)
Fawwarah Saudi Arabia	peraluminous Sn	6.3(2.0)18	Kamilli & Criss (1996)

¹ Crd = cordierite
ND = only average value and number of analyses cited

such as filter pressing. Hence, most fractionation indices jumped up discontinuously between the upper facies and the rocks below. This jump is evident for Li, Rb, and Sn, but there is no change in Be among any of the layers (Fig. 8b). Constant Be values speak to a buffering reaction, as a small quantity of cordierite or a large fraction of calcic oligoclase or white mica might accomplish for a large volume of magma. In contrast, the microgranitic dike swarm at Argemela, Portugal (Charoy and Noronha 1996) is strongly enriched in Be (Table 3, Fig. 8c). Argemela is notable because beryl is reported as an accessory magmatic phase in some dikes; the Be contents of individual dikes reach 385 ppmw, and we presume that these values correspond to the beryl-saturated rocks.

Table 3, continued

Ghost Lake	S-type	3.0(1.3)14	Breaks & Moore (1992)
Ontario, Canada	Li, Sn		
	Crd and garnet		
Jarna, Dala group	I-type	1.6(0.8)22	Ahl et al. (1999)
Sweden			
Siljan, Dala group	I-type	4.15(1.62)13	Ahl et al. (1999)
Sweden	high-level fractionated		
Garberg, Dala group	I-type	4.9(2.32)10	Ahl et al. (1999)
Sweden	fractionated		
Eurajoki	A-type	6(4)10	Haapala (1997)
Finland	Sn		
Silvermine	A-type	5.5(0.71)2	Lowell & Young (1999)
Missouri, USA	mafic enclaves		
Red Bluff	A-type	5.73(0.46)2	Shannon et al. (1997)
Texas, USA	mafic and felsic		
Oka	NYF	3.1(3.3)20	Gold et al. (1986)
Quebec, Canada	peralkaline		
Strange Lake	NYF	160(115)14	Salvi
Quebec-Labrador	peralkaline		& Williams-Jones (1996)
Khaldzen-Buregtey	NYF	33-92	Kovalenko et al. (1995)
western Mongolia	peralkaline		
Tuva	alkali granites	3.4(ND)30	Popolitov et al. (1967)
Siberia-Mongolia	"Phase II"		
Tuva	alkali granites	5.3(ND)15	Popolitov et al. (1967)
Siberia-Mongolia	"Phase III"		
Tuva	plagiosyenites	1.7(ND)29	Popolitov et al. (1967)
Siberia-Mongolia			
Tuva	nepheline syenites	2.9(ND)29	Popolitov et al. (1967)
Siberia-Mongolia			
Fantale	pantellerite	10.0(1.2)19	Webster et al. (1993)
Ethiopia	melt inclusions		

¹ Crd = cordierite

ND = only average value and number of analyses cited

Although the dikes at Argemela contain tourmaline as the only mafic silicate, a few studies of likely basement sources and peraluminous Hercynian plutons of Portugal report that garnet, rather than cordierite, is the prevalent aluminous mafic silicate (Godinho 1974; Reavy 1987). The dominance of garnet rather than cordierite at the source could explain the sharply higher Be contents of the Portuguese rocks compared to those in Spain, and for the occurrence of beryl-bearing pegmatites in Galicia (Fuertes-Fuente and Martin-Izard 1998), to the north of the Portuguese locality.

From the Bohemian Massif of the Czech Republic, analyses of mostly S-type granites show a large variation of Be, from anomalously low Be in the Homolka granite

(average 2.3 ppmw Be, Table 3) to values as high as 60 ppmw Be in one of a suite of evolved felsite porphyry dikes (Vrána 1999) not far from Homolka. The low values for such highly fractionated plutons as the Homolka granite correlate with their cordierite-bearing source rocks (Breiter and Scharbert 1995). By implication, those granites and dikes with substantially higher Be contents had fundamentally different, or at least cordierite-absent, source materials. With the highest Be values reported in the Bohemian Massif, the Milevsko felsite porphyries (devitrified subvolcanic dikes) described by Vrána (1999), range from 8 to 60 ppmw. Beryllium, however, does not correlate with F ($R^2 = 0.15$) or with Sn ($R^2 = 0.14$).

Among the highly fractionated granites cited in Table 3, the Beauvoir granites of the Massif Central, France, stand out for their Be enrichment. Values cited for the various facies of the Beauvoir granite in the GPF drill core range from 5.7 to 494 ppmw Be (Raimbault et al. 1995). In the Beauvoir series, fractionation increases from a basal B3 unit through B2 to an upper B1 facies. These three units are interpreted as a stacked sequence of successively more fractionated magmas, and in support of this argument, the large increase of Be correlates positively, though variably, with Li and Rb from B3 to B1 (Fig. 8d). Beryl is reported as a very rare phase in pegmatitic segregations within the B1 facies (Cuney et al. 1992). A large fraction of the Be resides in micas (up to 150 ppmw Be: Monier et al. 1987; Cuney et al. 1992, Ru et al. 1992), but at least locally, the phosphates beryllonite and hydroxylherderite contribute to the high whole-rock Be values (Charoy 1999). Thus, the paucity of beryl in this Be-rich magmatic suite stems from the increased compatibility of Be in Li-micas (Grew, this volume), and from the high activity of phosphorus in the system. A large portion of the ASI values reported by Raimbault et al. (1995) lies above the likely magmatic limit, 1.4 (Fig. 8j,k). This limit is based on experimentation involving silicate liquids in equilibrium with peraluminous crystalline phases including white micas and tourmaline, where ASI values of melt reach 1.3-1.4 (e.g., Icenhower and London 1995; Wolf and London 1997; London et al. 2000, 2001).

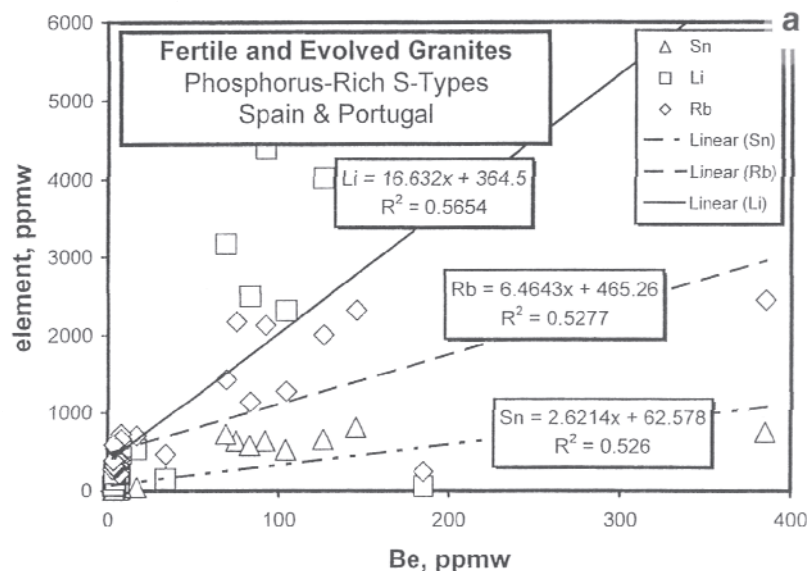
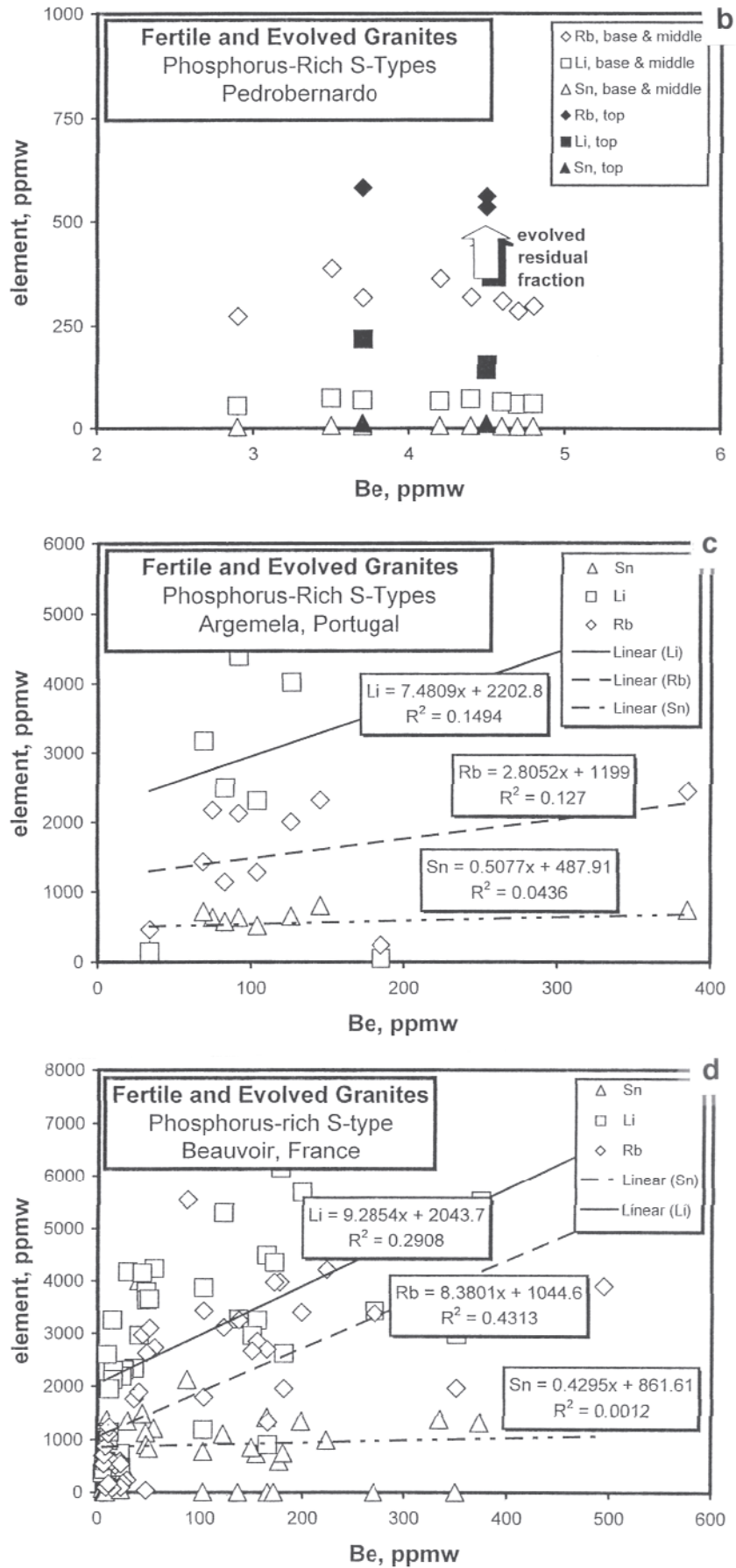
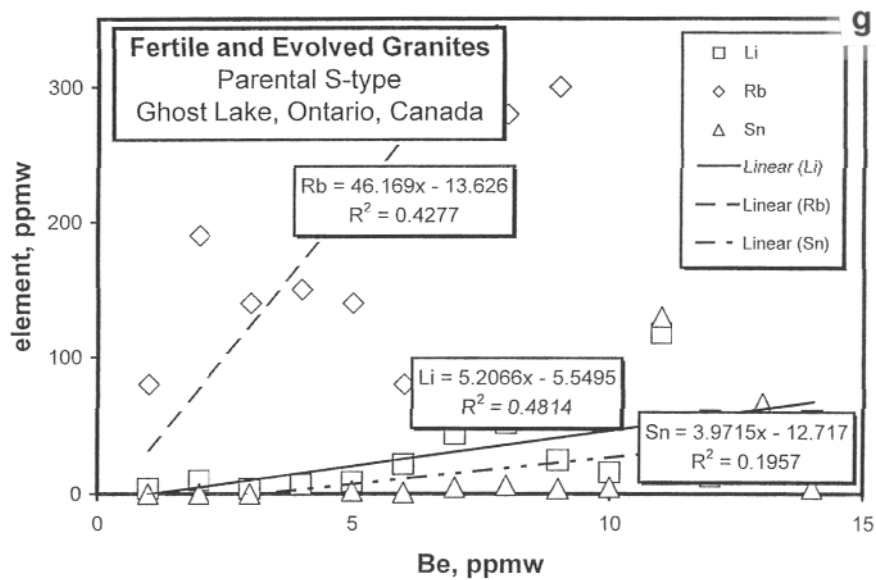
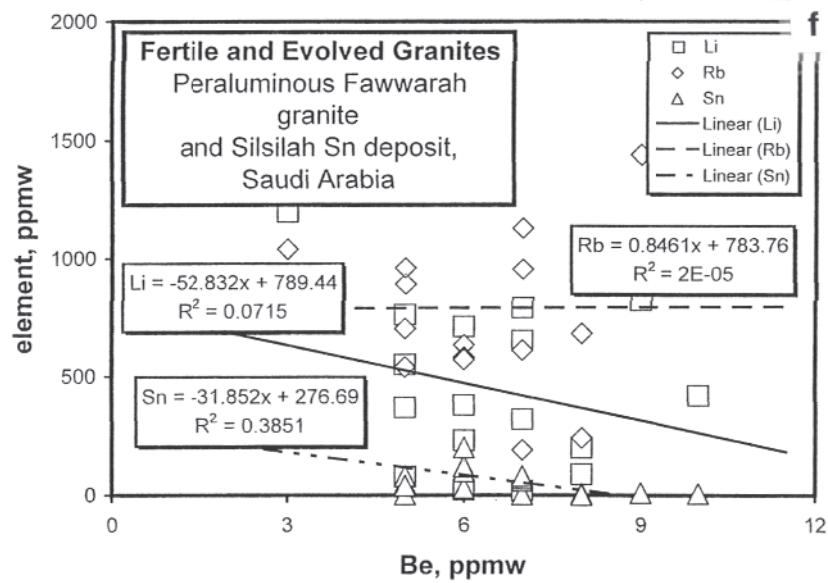
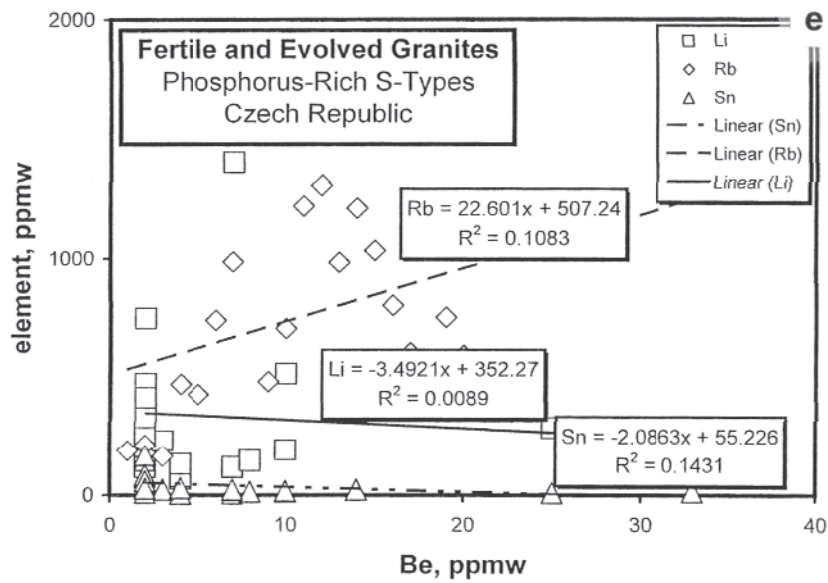
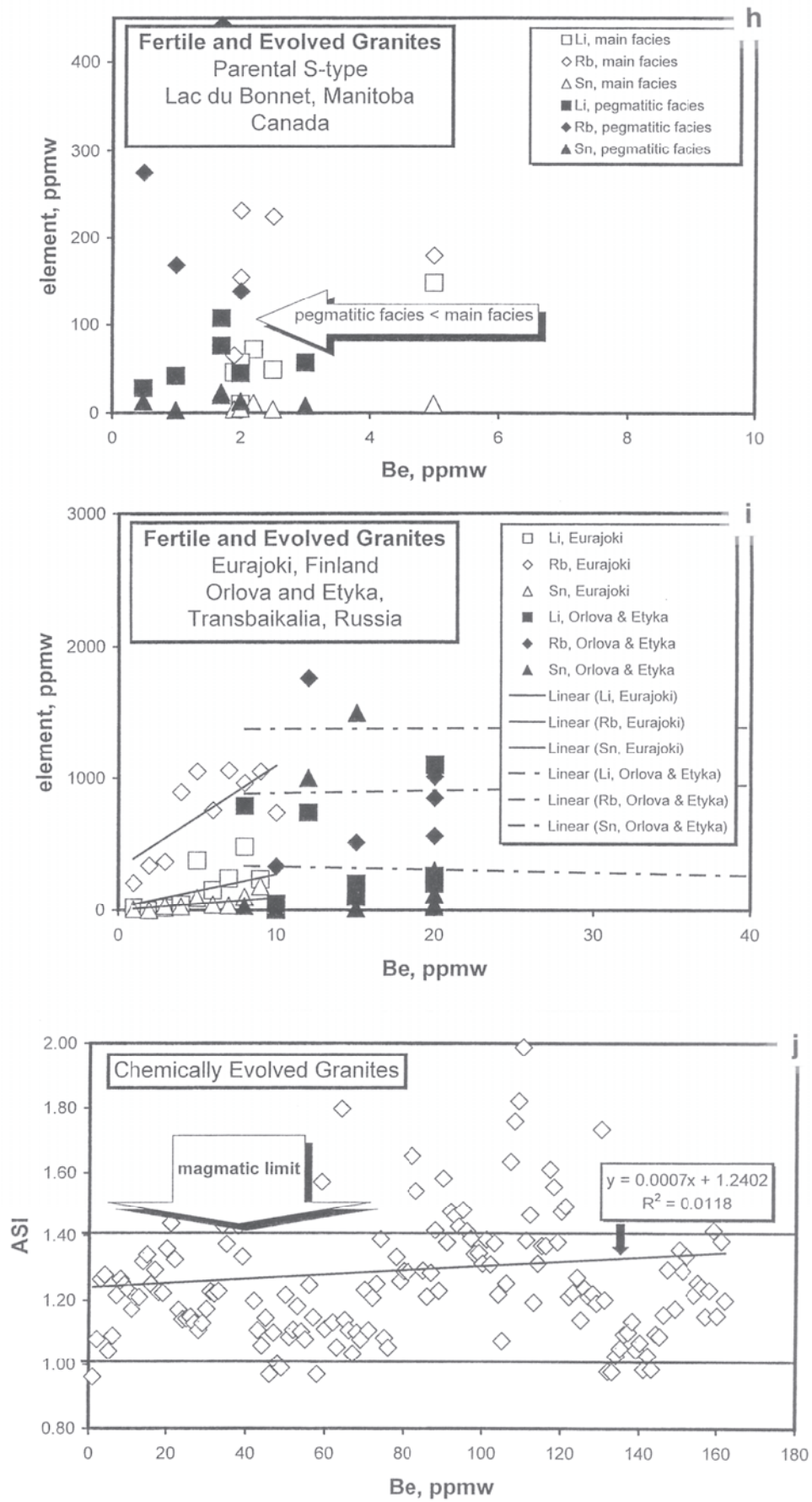
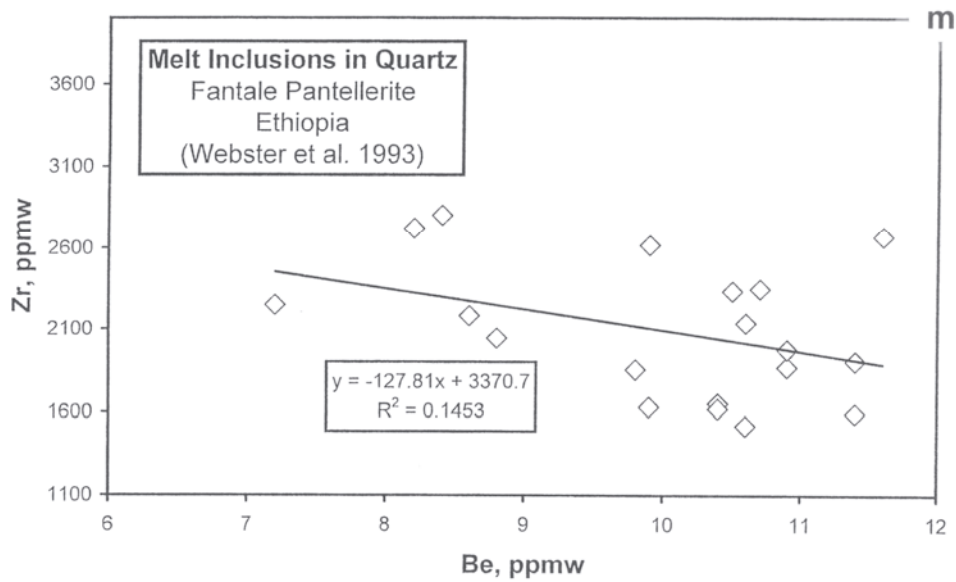
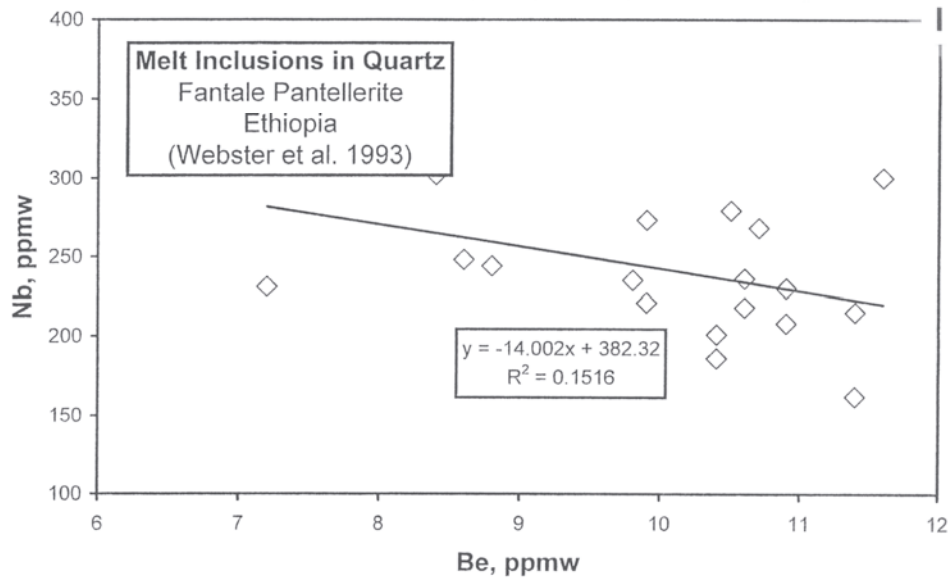
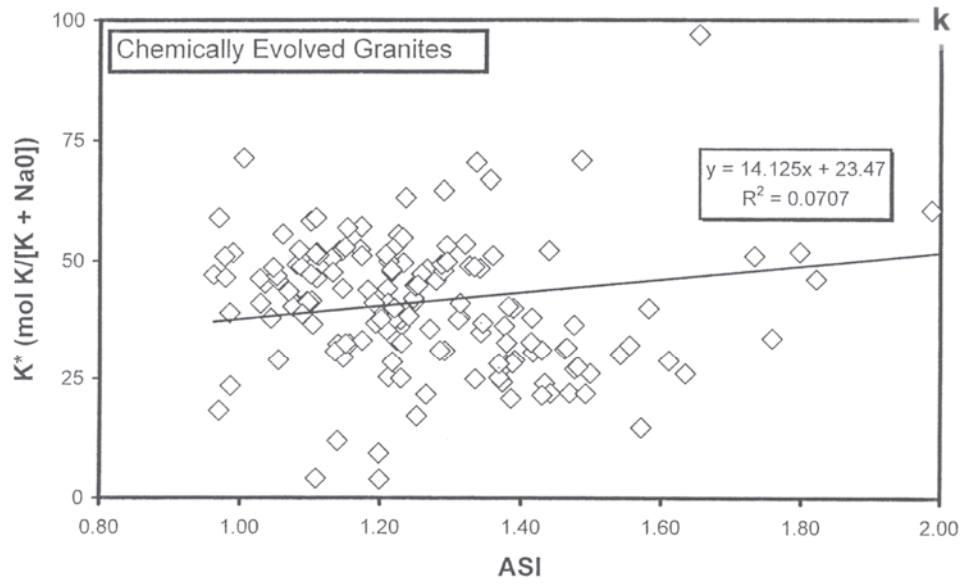


Figure 8. Plots of Be versus other elements and chemical indices in granitic rocks. Sources include the Jalama (Ramirez and Grundvig 2000) and Pedrobernardo (Bea et al. 1994a) plutons in Spain; microgranite dikes at Argemela, Portugal (Charoy and Noronha 1996); the Homolka granite (Breiter and Scharbert 1995) and various other localities in Bohemia, Czech Republic (Breiter et al. 1991); the Fawwarah granite and Silsilah Sn deposits of Saudi Arabia (Kamilli and Criss 1996); parental granites at Ghost Lake, Ontario (Kretz et al. 1989) and Lac du Bonnet, Manitoba, Canada (Černý and Meintzer 1988); rapikivi granites from Eurajoki, Finland (Haapala 1997); and peraluminous Ta-rich granites from Orlova and Etyka, Transbaikalia, Russia (Zaraisky et al. 1997). **Cont'd next 4 pages.**









It is likely that the most aluminous and Be-rich rocks at Beauvoir contain a significant hydrothermal overprint. Raimbault and Burnol (1998), however, identified a highly fractionated rhyolitic dike at Richemont, Haute-Vienne, France, which they interpret as a quenched magmatic phase of the Beauvoir granites. Like the Beauvoir granites and the Argemela dikes, this dike possesses high ASI (1.45), Be (91.6 ppmw), Li (3194 ppmw), Sn (487 ppmw), etc. Also like the Beauvoir granites, the Richemont dike is enriched in P (1.18 wt % P_2O_5) and F (1.30 wt %), and hence contains abundant Li-micas and various phosphates, but not beryl.

Beryllium contents are uniformly low in the Lac du Bonnet, Greer Lake, Osis Lake, and Ghost Lake granites in Canada (Goad and Černý 1981; Černý and Meintzer 1988; Breaks and Moore 1992), all of which are linked to beryl-bearing pegmatite fields. The beryl-rich Shatford Lake pegmatite group (Buck et al. 1999) is derived from the biotite facies of the Lac du Bonnet granite (1.9 ppm Be; Goad and Černý 1981, Černý and Meintzer 1988), and the nearby but genetically independent Greer Lake (3.8 ppm) and Osis Lake (0.8 ppm) leucogranites (Goad and Černý 1981; Baadsgaard and Černý 1993) also yielded Be-rich pegmatite aureoles. It may seem logical that, as likely sources of the beryl-rich pegmatitic melts, these granites are themselves depleted in Be and other incompatible components.

I- and A-type granites. The Järna granitoids (Sweden), which are comprised of deep-level, slightly fractionated I-type monzodiorites to monzogranites, contain exceptionally low Be contents (Ahl et al. 1999). The Siljan and Garberg granite centers, which are more fractionated I-types emplaced at higher levels (Ahl et al. 1999), have relatively higher Be contents but are still near the granite and rhyolite averages of ~4-6 ppmw (Table 3), and similar to the A-type Eurajoki rapakivi granite (Fig. 8I; Haapala 1997). In the USA, two A-type granites, the Silvermine Granite (Lowell and Young 1999) in the St. Francois Mountains, Missouri, and the Red Bluff Granite suite in west Texas (Shannon et al. 1997) contain the typical and normal Be levels at 5-6 ppmw (Table 3).

Peralkaline granites and syenites. Peralkaline granites and related rocks (including syenites, pegmatites, and carbonatites) evolve toward a suite of enriched lithophile elements that includes Be along with the distinctive trilogy of Nb, Y, and F (NYF; Černý 1991a). Enrichment in Be is not diagnostic of the NYF type of ore deposits (Černý 1991a), as the following two examples illustrate (Table 3). The Oka alkaline igneous complex in southeastern Quebec, Canada (Gold et al. 1966; Treiman and Essene 1985) carries the typical NYF enrichment, but all of the various peralkaline igneous rocks (mafic to felsic) and carbonatites are low in Be (Gold et al. 1986). In contrast, the Strange Lake NYF deposit (Miller 1996; Salvi and Williams-Jones 1996) is exceeding rich in Be (Table 3), though Be does not correlate with Li ($R^2 = 0.00$), and it shows a weakly negative correlation with F ($R^2 = 0.31$). Alkali granites, both mineralized (Nb, Zr, and REE) and nonmineralized, in Siberia and Mongolia (Popolitov et al. 1967; Kovalenko et al. 1995) manifest similar contrasts in Be abundance (Table 3).

A data set of melt inclusions sampled from quartz in pantellerite from Fantale, Ethiopia, provides the same sort of controls on magmatic processes as opposed to subsolidus hydrothermal effects in a high-silica (≥ 70 wt % SiO_2) peralkaline magma (Webster et al. 1993). These data are included with granites (Table 3) because they reflect the compositions of melts that coexisted with phenocrysts at plutonic conditions. Abundances of Zr, Nb, Y, and Ce show strong positive correlations (Webster et al. 1993), but Be correlates negatively with these elements (Fig. 8I,m).

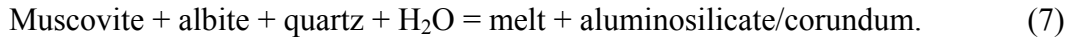
Summary of data on granites. One obvious difference between the volcanic and plutonic sources is that the granites, as a group, are far more aluminous than the rhyolites (see Fig. 8j) This is mostly due to the prevalence of S-type sources in the granite database

(Table 3), but also to alkali loss via sericitic or argillic alteration that is more pervasive in plutonic rocks. The correlations of Be with the fractionation indices of Li, Rb, and Sn are poor in the granite data base as a whole, especially in comparison the USGS data set on obsidians. Regression of the data to exponential or polynomial functions does not yield much better fits. The poorer correlations follow from the more variable mineralogy of the granites, especially in the proportions and compositions of the micas, and from the likelihood of more pervasive subsolidus alteration in the granites, which may be selective by mineralogy. Nevertheless, the data for Be in granites from different ages, sources, and degrees of fractionation are grossly comparable to those of the obsidian and rhyolite data sets in these ways:

1. The correlations of Be with other incompatible elements, though poor, are generally positive. Thus, Be behaves incompatibly and increases with fractionation. One notable exception is the melt inclusion data from Fantale, Ethiopia (Fig. 81,m), in which Be varies inversely with the other fractionation indices.
2. The Be contents of the granite data set are, on average, similar to the obsidians. Single-digit values of Be in ppmw are the norm in large plutons. Double-digit values (a few tens of ppmw Be) are typical of small, highly evolved plutons and dikes.

THE BERYLLIUM BUDGET FROM MIGMATITE TO PEGMATITE

Consider two starting melt compositions, one that corresponds to the minimum in the H₂O-saturated metaluminous haplogranite system Ab-Or-Qtz at 200 MPa, and another that reflects the 200 MPa H₂O minimum for melting of a muscovite-bearing protolith via the reaction:



We choose the pressure of 200 MPa because the compositions of minimum-melting liquids are well constrained for Reaction (7) at that pressure (Icenhower and London 1995). For the H₂O-saturated haplogranite minimum composition at this pressure, Ab₃₈Or₂₈Qz₃₄, the bulk distribution coefficient for Be as derived from our experimental data in Table 2 is 0.19 (Table 4). At 200 MPa, the melt produced by Reaction (7) above has a normative composition close to Ab₃₇Ms₂₀Or₁₀Qz₃₃ (Icenhower and London 1995), for which the bulk distribution coefficient of Be is 0.43 (Table 4). Figure 9 illustrates that,

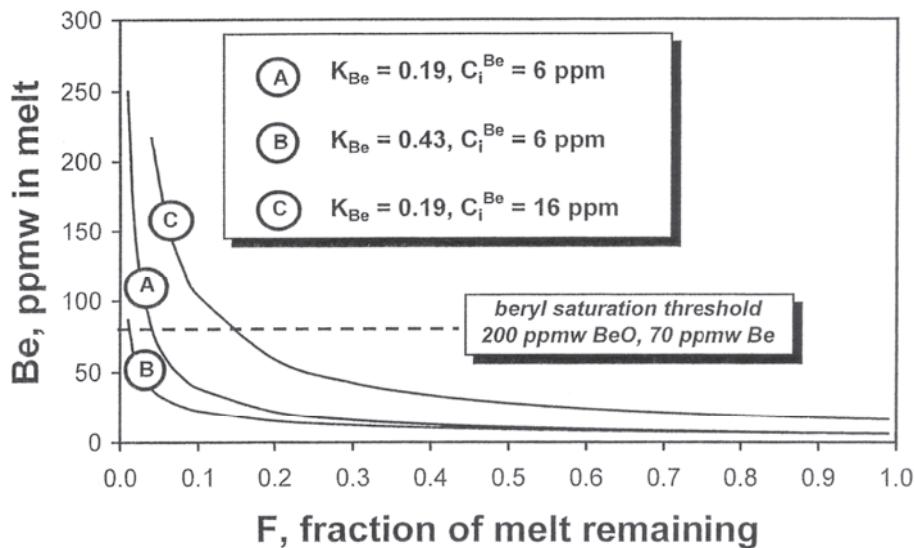


Figure 9. Rayleigh fractionation model for melts containing initially 4 or 16 ppmw BeO and bulk distribution coefficients of 0.19 (haplogranite minimum composition) and 0.43 (peraluminous haplogranite minimum). See the text for explanation.

Table 4. Bulk distribution coefficients for beryllium.

Phase	$D_{\text{Be}}^{\text{mineral/melt}}$	Normative wt fraction	Distribution coefficient $K_{\text{Be}}^{\text{mineral/melt}}$
haplogranite minimum at 200 MPa H₂O			
Qtz	0.24	0.34	0.08
Pl _{An1}	0.19	0.38	0.07
Kf	0.14	0.28	0.04
K_{Be}			0.19
muscovite-saturated granite minimum at 200 MPa H₂O			
Qtz	0.24	0.33	0.08
Pl _{An1}	0.19	0.37	0.07
Kf	0.14	0.10	0.01
Ms	1.35	0.20	0.27
K_{Be}			0.43
muscovite-saturated Ca-bearing granite at 200 MPa H₂O			
Qtz	0.24	0.33	0.08
Pl _{An31}	1.84	0.37	0.68
Kf	0.14	0.10	0.01
Ms	1.35	0.20	0.27
K_{Be}			1.04

taking 6 ppmw Be as a starting concentration, neither magma reaches what we consider the minimum beryl saturation threshold, ~70 ppmw Be, until they are >95% crystallized. At ~80% crystallization, either magma would contain ~20 to 30 ppmw Be, a range of values that corresponds to some of the more evolved rhyolites and obsidians cited above. If that liquid were efficiently extracted, for example by filter pressing (Bea et al. 1994a), then continued fractionation of that small volume of residual melt could approach beryl saturation with ~20% melt remaining. That last fraction of residual melt, corresponding to the final few percent of the original melt volume, would correspond to the stage where pegmatite dikes are derived from an apical pluton or the cupola of a large body. There is nothing unique about the value of melt extraction at 20% of melt remaining at each stage, except that the liquids at these steps in the fractionation model correspond well with natural rocks. The extraction of ~20% or less of residual granitic melt appears to be quite feasible, especially if some shear stress is applied to the crystal-rich magma (Brown et al. 1995; Vigneresse et al. 1996).

This scenario conforms in large measure to the observed field relations: pegmatite fields commonly emanate from small, texturally and chemical evolved pegmatitic plutons (Goad and Černý 1981; Černý and Meintzer 1988; Černý 1991c), and these in turn appear to have batholithic-scale sources (Černý and Brisbin 1982; Mulja et al. 1995).

Because pegmatite fields extend above and beyond their sources, the connections between pegmatites and larger plutons or batholiths are often difficult to establish unless exposures constitute crustal cross sections. However, Kosals and Mazurov (1968) report Be values in the vertical evolution of a stacked and zoned plutonic sequence from southwest Baikalia. The Be contents, from 7 ppmw Be in the batholithic source to 21 ppmw Be in the evolved apophyses higher in the plutonic system, correspond well with the modeled values described above.

We can now answer the third and fourth questions posed at the beginning of this chapter. The plausible protoliths for granitic plutons—amphibolites, gneisses, and metapelitic schists—contain similar (order of magnitude) and uniformly low Be abundances (Grew, this volume). Despite generally low mineral-melt partition coefficients, these source rocks appear to be capable of imparting only a few ppmw Be to anatectic melts (based on the data for rhyolites and relatively undifferentiated granites presented here). This observation corroborates an important concept expressed recently by Bea (1996): that the time frames of anatexis are generally not sufficient to achieve a redistribution of trace elements between residual crystals and melt via diffusion through the residual crystalline phases. Consequently, partition coefficients are $\cong 1$ and are determined by the proportions in which crystalline phases participate in melting. With the conclusion that most protoliths can and do impart about the same Be content to partial melts, it would appear that no one granite source rock is uniquely or specially predetermined to produce beryl-bearing pegmatites. There are, however, attributes of mineralogy at the source and during fractional crystallization of magma that do affect the accumulation of Be in derivative melts.

The composition of plagioclase. Kosals et al. (1973) noted that the Be content of plagioclase reaches a peak (in natural rocks) at $\sim\text{An}_{30}$, and decreases in abundance in both directions away from this composition. That observation has been corroborated by the recent experimental work of Evensen and London (2001a) and Evensen (2001): as reported in Table 2, the partition coefficient for Be drops from 1.84 at An_{31} to 0.19 at An_{01} . Because of the compatibility of Be in calcic oligoclase, the Be concentration in melt cannot increase appreciably until plagioclase fractionation has removed virtually all Ca from the granitic melt.

Micas. Micas, especially white micas near muscovite in composition, represent important reservoirs of Be in crustal rocks (Grew, this volume). When white micas contribute to melting reactions such as (7) above, they can transfer a large mass fraction of Be available from the source rock into the melt. The melting of white mica also imparts a high ASI value to the anatectic melt (e.g., Icenhower and London 1995), which ultimately lowers the Be content needed to saturate derivative melt fractions in beryl. However, the crystallization of abundant white mica from granitic melts consumes Be and forestalls beryl saturation (e.g., the Li-rich granites of the Massif Central, France). Indeed, a model granitic magma that crystallizes with the assemblage of calcic oligoclase and white mica in the proportions observed in experiments would possess a distribution coefficient for Be near 1 (Table 4), and such granites would not experience any increase of Be with crystallization. The compatibility of Be in calcic oligoclase and white mica probably explains the lack of variation of Be with fractionation in the three granite facies at Pedrobernardo, Spain (Fig. 8b). The An content of plagioclase remains moderately high, with cores of An_{20-30} and rims of An_{5-10} , throughout the crystallization of the (predominant) lower and middle zones (Bea et al. 1994a). These granites also contain appreciable muscovite (6 to 14 modal %).

The special role of cordierite. Miscibility between cordierite or indialite and beryl increases with temperature, and solid solution between indialite and beryl appears to be

complete at high temperatures (Fig. 4). At the same time, the partition coefficient for Be between cordierite and granitic partial melt is extremely high (Table 2), though it decreases with increasing temperature (Evensen and London 1999). It is evident that if melting occurs in the medium- to low-pressure regime of cordierite (Mukhopadhyay and Holdaway 1994), then cordierite controls the fate of Be.

When S-type granite magmas originate from previously unmelted metapelites, then reactions similar to (7) above (but including biotite) largely determine both the volume and the Be contents of the melt, even if cordierite is present in the melting assemblage. This is because normative calculations based on experimental liquids and their crystalline products indicate that only a small fraction (<4 wt %) of cordierite is imparted to the melt up to 800°C (Evensen 2001), and hence most Be remains in residual cordierite. Melting reactions that produce cordierite (from the prograde reaction of biotite + sillimanite + quartz: Pereira and Bea 1994; Clark 1995) similarly generate granitic liquids that are low in Be and in normative Crd component.

We have already noted that restitic cordierite can be highly enriched in Be (Schreyer et al. 1979), and becomes more so as reactions (not necessarily melt-producing) consume cordierite with increasing metamorphic grade. The situation with sapphirine, another phase that concentrates Be (Grew, this volume), and the beryllosilicate surinamite (e.g., Baba et al. 2000), is less obvious. Grew et al. (2000) proposed that Be-enriched pegmatites in Enderby Land, Antarctica, formed at temperatures near 1000°C by partial melting of metapelites containing Be-bearing sapphirine, but work in progress on these rocks suggests an alternative scenario whereby the Be pegmatites formed as sapphirine was breaking down to Be-poor phases.

Cordierite versus garnet sources. The parental magmas of most beryl-bearing pegmatites have sources in the stability field of garnet ± aluminosilicate, rather than cordierite. We have noted already a possible connection between Be-poor granites in Spain and Be-rich granites in Portugal with the distinction of cordierite- or garnet-bearing source rocks. Continental terranes where cordierite-bearing S-type granites are prevalent (western Europe and eastern New South Wales, Australia) are conspicuously devoid of beryl-rich pegmatites. Conversely, in regions where peraluminous granites are marked by accessory garnet but not cordierite (North America), beryl-rich pegmatites are commonplace.

Vapor transport. So far, we have addressed only the distribution of Be between minerals and melt. It is pertinent, however, to address the partitioning and transport of beryllium by hydrothermal fluids, at least cursorily (see Ryan, this volume; Morris et al. this volume). Wood (1991) has calculated that the solubility of Be chloride complexes in equilibrium with bertrandite or phenakite is <1 ppmw in hydrothermal fluids to 300°C. Though Wood (1991) proposed that Be is far more soluble as a fluoride complex, the experimental data on melt-vapor partitioning in halide-rich systems does not reveal a strong tendency for Be to partition into the vapor phase. In the relevant experiments, London et al. (1988) examined the behavior of Be and other trace elements using the F-rich Macusani obsidian as starting material, and Webster et al. (1989), employed the F-rich Spor Mountain vitrophyre, with and without added Cl. In both experimental programs, the vapor/melt partition coefficient for Be was generally ≤ 1 over a range of fluid compositions, salinities, temperatures, and pressures. From the recent work of Evensen et al. (1999), we know that silicate melts are capable of dissolving hundreds to thousands of ppmw of Be. Thus, we conclude that silicate melts are more efficient agents for the transfer of Be than are aqueous fluids. This does not mean that hydrothermal fluids are unimportant in the movement of Be through the Earth's crust (Tatsumi and Isoyama 1988; Morris et al. 1990; Bebout et al. 1993; You et al. 1994; Leeman 1996;

Brenan et al. 1998a,b; Ryan, this volume), only that they are not as effective as silicate melts on an equal-volume basis.

Beryl saturation in pegmatites

In reviewing the likely conditions for beryl saturation by magmatic processes, Evensen et al. (1999) stressed the relation of beryl solubility to temperature. The Be content of melt necessary to achieve saturation in beryl decreases sharply with temperature. Recent studies of pegmatite cooling histories indicate very low temperatures of crystallization (e.g., ~400°-450°C: London 1992, 1996; Morgan and London 1999; Webber et al. 1999) for small dikes that are likely to be farthest from source (Baker 1998).

TEMPERATURE:	700°C	600°C	500°C	400°C
<u>Be content of melt, ppmw</u>				
Initially in melt:	140	140	140	140
In equilibrium with beryl at T:	140	60	50	40
Be content of minerals, ppmw, & mineral modal percent				
quartz (30 wt%)	34	14	12	10
K-feldspar (20 wt%)	27	8	7	5
plagioclase (40 wt%)	252 (An ₃₁)	6 (An ₁)	5 (An ₁)	4 (An ₁)
muscovite (10 wt%)	189	81	68	54
Total Be in RFM, ppmw:	135	16	14	11
Be in beryl, % of total Be:	4%	89%	90%	92%
Pegmatite Class:	muscovite	beryl	rare-element	rare-element
	NO BERYL	BERYL SATURATED		

Figure 10. Model for the distribution of Be between rock-forming minerals and melts in a zoned pegmatite dike system, and the formation of beryl-bearing pegmatites. See the text for explanation.

Figure 10 illustrates how the distribution of Be among rock-forming minerals and the abundance of beryl might vary in a dike system that experiences the thermal gradient shown, from 700°C to 400°C. The model considers a peraluminous granitic melt of uniform composition with 140 ppmw Be (≈ 400 ppmw BeO, a representative value for the beryl class of rare-element pegmatites; Table 1) that is injected along its entire length and cools to the temperatures shown before crystallization commences. The melt is then allowed to crystallize, and partition coefficients determine the distribution of Be between rock-forming minerals. The model utilizes our experimentally measured partition coefficients presented in Table 2. The beryl saturation curve for peraluminous granitic liquid compositions (Fig. 1) is extrapolated linearly at 10 ppmw Be per 100°C from 600° to 400°C. The mineral mode (wt %) is set at 40% plagioclase, 30% quartz, 20% K-feldspar, and 10% muscovite, which is more feldspathic than the minimum melt composition in the muscovite-bearing granite system at 200 MPa H₂O (Icenhower and London 1995) but also more typical of “two-mica” granite-pegmatite systems.

Because Be is compatible in moderately calcic plagioclase (we used An₃₁, for which we have partitioning data, in this model) and in muscovite, the rock that crystallizes at 700°C is essentially beryl-undersaturated. Using the partition coefficients and the modes cited above, all but 4% of the initial Be content of the melt is accounted for in the rock-forming mineral assemblage. Upon falling to 600°C, however, the melt becomes saturated in beryl (at ~60 ppmw Be in melt), partly because of the drop in temperature and because we have adjusted the plagioclase composition to near the albite end member (An₁, again corresponding to our experimental compositions). Such an abrupt decrease in the An-content of plagioclase does have natural precedent, for example in the Pedrobernardo and Albuquerque plutons of Spain (Bea et al. 1994a; London et al. 1999). In both cases, plagioclase of the main plutonic body is calcic oligoclase (An₁₀ at Albuquerque, An₂₀₋₃₀ at Pedrobernardo), which falls sharply to An₁ in the marginal or upper facies of each pluton.

At 600°C, the rock-forming mineral assemblage accounts for only 16% of the total Be content, and the remainder is in beryl. The fraction of Be in beryl is largely controlled by, and varies inversely with, the abundance of white mica at this stage of the modeling. Note also in Figure 10 that the Be concentrations in mica and alkali feldspars fall sharply from the beryl-free proximal pegmatites (700°C) to the beryl-bearing distal ones (≤600°C). This pattern is in fact observed in natural occurrences (Smeds 1992). Because the mineral modes and partition coefficients are held constant, the changes in Be concentrations in the rock-forming minerals and the fraction of Be in beryl change only slightly with falling temperature according to the projection of the beryl saturation surface to 500°C and then at 400°C.

The absence of beryl in the proximal mica-rich pegmatites arises from the higher temperatures required to saturate the melt in beryl, and from the relative compatibility of Be in muscovite and in plagioclase that is typically in the range of oligoclase in composition (Kosals and Mazurov 1968; Kosals et al. 1973). The compatibility of Be in these two phases effectively suppresses Be accumulation and beryl saturation in the melt. These proximal muscovite-rich pegmatites are considered ‘barren’ in some exploration models (Trueman and Černý 1982). With a drop of only 100°C to 600°C, a peraluminous melt containing 140 ppmw Be and depleted in Ca would crystallize beryl in profusion. Thus, it is not surprising that the pegmatite type normally recognized just beyond the ‘barren zone’ of muscovite pegmatites is the beryl type (Trueman and Černý 1982). If partition coefficients do not change with temperature or composition, then the abundance and distribution of beryl in the distal pegmatites would resemble those of the beryl class. The most distal pegmatites, however, are more chemically evolved, and from the work of Evensen et al. (1999) we would expect higher concentrations of Be to saturate these melts in beryl. The net effect might be that beryl saturation occurred later in the consolidation of the pegmatites, and more concentrated and segregated into the latest internal units.

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