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TOURMALINE IN PETALITE-SUBTYPE GRANITIC PEGMATITES: EVIDENCE OF FRACTIONATION AND CONTAMINATION FROM THE PAKEAGAMA LAKE AND SEPARATION LAKE AREAS OF NORTHWESTERN ONTARIO, CANADA

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ABSTRACT

Tourmaline in petalite-subtype granitic pegmatites from the Pakeagama Lake and Separation Lake areas of northwestern Ontario provides evidence for the compositional evolution of pegmatite-forming melts through magmatic crystallization and pegmatite melt – host-rock interactions. At the former locality, tourmaline from the pegmatite units is essentially magmatic, whereas at the latter locality, tourmaline in many of the Fe-suite pegmatites displays characteristics (such as elevated Mg, Ti and Ca contents) that indicate significant contamination through the incorporation and digestion of components of amphibolite (mafic metavolcanic units) and banded iron-formation (BIF) host-rocks. The presence of tourmaline in mafic host-rocks indicates that cation migration also involved the introduction of B, Li and Al from pegmatite-generated fluids to form metasomatic haloes. The Li + Mn *versus* Mg + Ti + Ca diagram is introduced to discriminate tourmaline with a dominantly magmatic signature from tourmaline with a signature influenced by pegmatite – host-rock interaction. The behavior of Ca in tourmaline is also shown to be potentially useful in recognizing late-stage Ca-enrichment in pegmatite-forming melts as a result of Ca–F complexing, but can give similar trends to tourmaline that has suffered Ca-metasomatism as a result of interaction with granitic host-rocks. The tourmaline data suggest that at Separation Rapids, pegmatites mainly crystallized in an open system (*i.e.*, pegmatite – host-rock interaction was common), whereas at Pakeagama Lake, the pegmatite units were predominantly unaffected by interaction with host rocks (*i.e.*, they crystallized in a closed system). This has implications for concentrating rare-elements such as Rb and Cs, which otherwise would be dispersed throughout pegmatite and host rocks (as at Separation Lake). The potential for a buried pollucite deposit associated with the SE pegmatite at Pakeagama Lake has increased as a result of these observations, particularly as Cs is elevated in associated K-feldspar and mica, and is also present in a late pollucite-bearing vein.

Keywords: tourmaline, granitic pegmatite, petalite subtype, host-rock interaction, Pakeagama Lake, Separation Lake, Ontario.

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SOMMAIRE

La tourmaline dans les massifs de pegmatite granitique, sous-type à pétalite, dans les régions des lacs Pakeagama et Separation, dans le nord-ouest de l'Ontario, fournit de bons indices de l'évolution des venues magmatiques qui ont cristallisé sous forme de pegmatite et, dans certains cas, été affectées par une réaction entre le magma et les roches encaissantes. Au premier endroit, la tourmaline des unités pegmatitiques est essentiellement magmatique, tandis qu'au deuxième endroit, la tourmaline de plusieurs pegmatites enrichies en fer montrent, par leurs teneurs élevées en Mg, Ti et Ca, des indications d'une contamination par l'incorporation et la digestion d'amphibolite (unités mafiques métavolcaniques) et de formation de fer rubannée. La présence de tourmaline dans les roches encaissantes mafiques démontre l'importance de la migration de cations par introduction de B, Li et Al par l'intermédiaire d'une phase fluide issue de la pegmatite pour former une auréole métasomatique. Le diagramme Li + Mn *versus* Mg + Ti + Ca est utile pour effectuer une discrimination entre compositions de tourmaline ayant une dérivation magmatique dominante et celles à dérivation plus complexe impliquant une interaction entre magma évolué et roches encaissantes. Le comportement du calcium pourrait aussi servir d'indice d'enrichissement tardif du magma à cause d'une complexation impliquant Ca–F, mais peut aussi signaler une métasomatose impliquant le Ca par implication avec les roches-hôtes granitiques. Les données recueillies montrent qu'au lac Separation, les pegmatites auraient surtout cristallisé dans un système ouvert, c'est-à-dire, que l'interaction avec les roches-hôtes était courante. En revanche, au lac Pakeagama, les unités pegmatitiques sont restées généralement non contaminées par de telles interactions, et la cristallisation a eu lieu dans un système plutôt fermé. Cette conclusion a des implications importantes pour la concentration des éléments rares comme le Rb et le Cs qui, au lac Separation, seraient dispersés dans les pegmatites et les roches hôtes. Le potentiel pour un gisement enfoui de pollucite associé à la suite de pegmatites dite SE au lac Pakeagama a augmenté à la suite de nos observations, surtout que la teneur en Cs est élevée dans le feldspath potassique et le mica, remarquée aussi dans une veine tardive à pollucite.

(Traduit par la Rédaction)

Mots-clés: tourmaline, pegmatite granitique, sous-type de la pétalite, interaction avec les roches hôtes, lac Pakeagama, lac Separation, Ontario.

INTRODUCTION

Because tourmaline can incorporate a large variety of cations in terms of size and charge (Dietrich 1985), it obtains a geochemical signature of the medium from which it crystallizes. For this reason, tourmaline has been shown to provide useful petrogenetic information about both metamorphic and igneous environments (Henry & Guidotti 1985, Henry & Dutrow 1996). In the pegmatitic environment, tourmaline has been used to map fractionation trends, and where strong internal zonation occurs, to provide evidence for internal processes such as disequilibrium crystallization, wallrock contamination and reaction with a compositionally evolving fluid (Jolliff *et al.* 1986, 1987, Selway *et al.* 2000a, b).

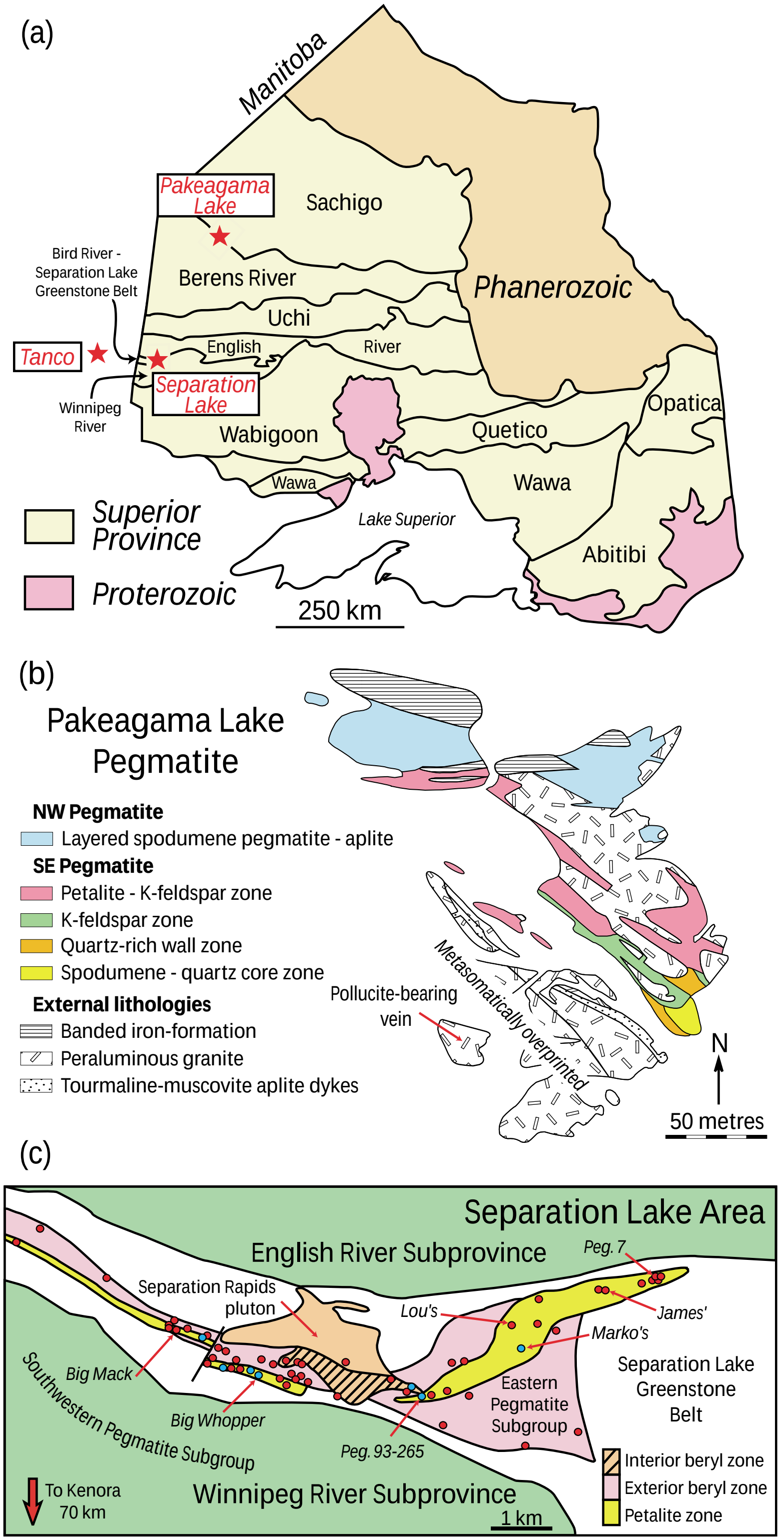
The lithium-bearing tourmaline elbaite is a typical major to accessory mineral in many rare-element pegmatites. Its composition, along with that of associated tourmaline species, has been studied in some detail from elbaite-subtype (Novák & Povondra 1995, Novák *et al.* 1999) and lepidolite-subtype pegmatites (Selway *et al.* 1999). More recently, Selway *et al.* (2000b) have examined tourmaline in the Tanco (Manitoba) pegmatite, a petalite-subtype pegmatite, and shown it to vary from foitite–schorl through a sequence of elbaite subspecies to a Ca-bearing "fluor-elbaite"–rossmanite composition (hypothetical species in quotation marks).

The composition of tourmaline species has also proved useful in the examination of metasomatic alteration of exocontact rocks surrounding pegmatites. Because pegmatite-forming melts and coexisting aqueous fluids are commonly enriched in relatively incompatible elements such as Li, Rb, Cs, B, Ta and Nb, many of these elements modify or replace original compositions of the primary mineral phases of surrounding host-rocks (Morgan & London 1987) where there is outward transport of fluid. Such transport can lead to extensive modification of the original minerals and results in the formation of exomorphic aureoles (Shearer *et al.* 1986, Selway *et al.* 2000a).

Apart from the Tanco pegmatite (Selway *et al.* 2000b), previous work on tourmaline compositions in petalite-subtype pegmatites is restricted to brief descriptions of the Urubu pegmatite, Minas Gerais, Brazil (Quéméneur *et al.* 1993) and to Canadian and Swedish examples (Selway *et al.* 1998a).

Tourmaline is a common accessory mineral in the Pakeagama Lake granitic pegmatite and its surrounding exocontact rocks (Fig. 1). In contrast, the Separation Lake pegmatites and their exocontact rocks contain relatively little tourmaline, although both are petalite-subtype pegmatites. In the two major economic pegmatites

FIG. 1. (a) Map of the Superior Province in Ontario annotated with locations of rare-element pegmatites (red stars) discussed in the text (derived from Breaks *et al.* 1998). (b) Geological map of the Pakeagama Lake pegmatite (derived from Breaks *et al.* 1999a). (c) Distribution of pegmatites in the Separation Lake area. Individual pegmatites marked by dots (red: Fe-suite, blue: Mn-suite).


(a)
Manitoba
Pakeagama Lake
Sachigo
Bird River - Separation Lake Greenstone Belt
Berens River
Phanerozoic
Uchi
Tanco
English
River
Separation Lake
Winnipeg River
Opatica
Wabigoon
Quetico
Wawa
Wawa
Lake Superior
Abitibi
Superior Province
Proterozoic
250 km
(b)
Pakeagama Lake Pegmatite
NW Pegmatite
Layered spodumene pegmatite - aplite
SE Pegmatite
Petalite - K-feldspar zone
K-feldspar zone
Quartz-rich wall zone
Spodumene - quartz core zone
External lithologies
Banded iron-formation
Peraluminous granite
Tourmaline-muscovite aplite dykes
Pollucite-bearing vein
Metasomatically overprinted
N
50 metres
(c)
Separation Lake Area
English River Subprovince
Peg. 7
Separation Rapids pluton
Lou's
James'
Marko's
Separation Lake Greenstone Belt
Big Mack
Southwestern Pegmatite Subgroup
Eastern Pegmatite Subgroup
Big Whopper
Peg. 93-265
Interior beryl zone
Exterior beryl zone
Petalite zone
To Kenora 70 km
Winnipeg River Subprovince
1 km

of the area (the ore mineral is petalite), it is essentially absent.

We make use of a large electron-microprobe dataset of tourmaline compositions (approaching 2500) and investigate the chemical evolution of tourmaline in these granitic pegmatites and adjacent host-rocks. We provide evidence for mobility of a range of elements both into and out of the Pakeagama Lake and Separation Lake pegmatite systems. We summarize the substitution schemes operating in tourmaline from these pegmatites and relate the compositional trends to processes operating under magmatic, post-magmatic and hydrothermal conditions. Table 1 lists tourmaline species encountered in this work, both those actually present and those belonging to an end-member of a substitution scheme for which some data from Pakeagama Lake or Separation Lake are available.

EXPERIMENTAL METHODS

Mineral compositions were performed in the Department of Earth Sciences at the Open University with a Cameca SX100 microprobe operating in wavelength-dispersion mode. We used the following standards and X-ray lines: synthetic LiF (F$K\alpha$), jadeite (Na$K\alpha$ and Al$K\alpha$), forsterite (Mg$K\alpha$), feldspar (Al$K\alpha$, Si$K\alpha$ and K$K\alpha$), synthetic KCl (Cl$K\alpha$), rutile (Ti$K\alpha$), bustamite (Mn$K\alpha$ and Ca$K\alpha$), hematite (Fe$K\alpha$), and willemite (Zn$K\alpha$). An operating voltage of 20 kV and probe current of 20 nA (measured on a Faraday cage) were used. The diameter of the beam was 10 μm. Count times varied from 20 to 80 seconds per element, and data were corrected using a PAP correction procedure (Pouchou & Pichoir 1985). The Separation Lake dataset was supplemented with over 200 compositions collected in the Department of Geological Sciences, University of Manitoba, on a Cameca SX50 microprobe using closely similar operating conditions (Selway *et al.* 1999).

Structural formulae were calculated on the basis of 31 anions, assuming stoichiometric amounts of H_2O as (OH), *i.e.*, OH + F = 4 *apfu* (atoms per formula unit), B_2O_3 (3 *apfu* B) and Li_2O (as Li^+) (Burns *et al.* 1994, MacDonald *et al.* 1993). The amount of Li assigned to the *Y* site corresponds to the ideal sum of the cations occupying the $T + Z + Y$ sites (15 *apfu*) minus the sum of the cations actually occupying these sites [Li = 15 – ($T + Z + Y$) or Li = 15 – (Si + Al + Mg + Fe + Mn + Zn + Ti)]; the calculation was iterated to self-consistency. All Fe and Mn were assumed to be divalent. We were unable to estimate Fe^{3+} or O^{2-} (substitution for OH), but we believe both to be insignificant. However, we note that from a dataset of nearly 2500 compositions, 5% of the data has more than 6.05 *apfu* Si, and 0.6% has more than 6.09 *apfu* Si, suggesting that our method of calculation is not perfect. The discrepancies could be due to underestimated Li in data for elbaite and to minor variation in Fe^{2+}/Fe^{3+} in a sample of the selvedge (entrained sliver of xenolithic material) with a schorl–feruvite composition. Only four compositions with more than 6.05 *apfu* Si are from the Separation Rapids pegmatites.

The tourmaline nomenclature used follows that of Hawthorne & Henry (1999) and Selway & Novák (1997). For an individual composition of tourmaline, if the ratio of two end-members is between 4:6 and 6:4 (*i.e.*, close to the 50:50 dividing line), both end-member names are given, *e.g.*, schorl–elbaite, with the first name designating the dominant component.

A Microsoft Excel™ worksheet that performs structural formula calculations for tourmaline is available from the following web address: http://tabitha.open.ac.uk/tindle/AGTHome.html

GEOLOGICAL ENVIRONMENT

Pakeagama Lake pegmatite

The Pakeagama Lake granitic pegmatite, located 160 km north of Red Lake, is one of the largest and most evolved rare-element pegmatites in northwestern Ontario (Fig. 1a). It is mostly contained within the northwestern part of an elongate, 2 to 3 by 15 km stock of peraluminous, muscovite–biotite granite named the Pakeagama Lake pluton (Breaks *et al.* 1998). The stock, delineated by Stone *et al.* (1993) and Stone (1998, Map P.3382), represents one of nine peraluminous granite plutons regionally distributed within the Berens River – Sachigo Subprovince boundary zone. Such plutons have been documented by Stone (1998) to occur over a strike length of 140 km in this zone. The pluton has a granoblastic texture, resulting from shearing and recrystallization induced by deformation along the Bearhead Lake fault system, delineated by Stone *et al.* (1993). In

TABLE 1. TOURMALINE END-MEMBER COMPOSITIONS DISCUSSED IN THE TEXT

Species*	(X)	(Y_3)	(Z_6)	T_6O_{18}	$(BO_3)_3$	V_3	W
			Alkali tourmaline				
Dravite	Na	Mg_3	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	(OH)
Schorl	Na	$Fe^{2+}{}_3$	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	(OH)
Elbaite	Na	$Li_{1.5}Al_{1.5}$	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	(OH)
"Fluor-elbaite"	Na	$Li_{1.5}Al_{1.5}$	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	F
			Calcic tourmaline				
Uvite	Ca	Mg_3	$MgAl_5$	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	F
Feruvite	Ca	$Fe^{2+}{}_3$	$MgAl_5$	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	F
			***X*-site-vacant tourmaline**				
Rossmanite	□	$LiAl_2$	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	(OH)
Foitite	□	$Fe^{2+}{}_2Al$	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	(OH)
			Mn^{2+}-bearing tourmaline				
"Mn-foitite"	□	$Mn^{2+}{}_2Al$	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	(OH)
"Oxy-Mn-foitite"	□	$Mn^{2+}Al_2$	Al_6	Si_6O_{18}	$(BO_3)_3$	$(OH)_3$	O

The general formula of a tourmaline can be expressed as $XY_3Z_6(T_6O_{18})(BO_3)_3V_3W$ (Hawthorne & Henry 1999). In such a formula, X = Na, Ca, (K) and vacancies (□), Y = Li, Al, Fe, Mg, (Ti), (Mn), (Zn); Z = Al, Mg, T = Si, Al; V = O, OH and W = O, OH, F. Species listed above are either actually present or feature as end-members of substitution schemes. * Species in quotation marks are not IMA-approved, but have been suggested as being appropriate (Hawthorne & Henry 1999).

close proximity to the pegmatite, the pluton hosts a series of randomly oriented tourmaline-rich minor intrusions and metasomatic veins (Fig. 2). At its northwestern end, the pegmatite is in contact with banded iron-formation (BIF) and metapelite host-rocks that form part of the Makataiamik assemblage, North Spirit Lake greenstone belt (Corfu *et al*. 1998). The pegmatite is also within 1 km of a mafic metavolcanic suite belonging to the Setting Net assemblage, Favourable Lake greenstone belt (Thurston *et al*. 1991). The Makataiamik assemblage is characterized by polymict conglomerates overlain by cross-bedded sandstone, mudstone and marble representing alluvial, fluvial and lacustrine deposits. The Setting Net assemblage includes a basal sequence of komatiites and basalts overlain by siltstone, sandstone, marble and ferruginous chert and by 2925 Ma intermediate pyroclastic rocks, lava flows and submarine intrusions representing a caldera-filling sequence (Corfu *et al*. 1998). An exocontact zone consisting of muscovite–tourmaline-bearing granitic pegmatite dykes emplaced into the mafic metavolcanic and metasedimentary rocks occurs adjacent to the northwestern end of the pluton.

The geology of the Pakeagama Lake pegmatite (Fig. 1b) has been described previously by Breaks *et al*. (1998, 1999a). In summary, the pegmatite has a minimum strike-length of 260 m, with northwestern and southeastern parts open along strike. The width of the northwestern arm of the pegmatite (layered spodumene pegmatite–aplite) varies from 30 to 70 m. It is likely that the pegmatite system extends an additional 300 m to the southeast, where a 3-m-wide ferrotapiolite–tourmaline–apatite-bearing aplite dyke outcrops. Using a laser ^{40}Ar/^{39}Ar technique on muscovite, Smith *et al*. (1999) recorded a primary cooling age of 2672 ± 6 Ma in a tourmaline-bearing vein cross-cutting the pluton. This corresponds well with a columbite–tantalite U–Pb LA–MC–ICP–MS age of 2673 ± 8 Ma (S.R. Smith, pers. commun.) from the petalite – K-feldspar zone of the southeastern part of the pegmatite.

The five internal zones of the Pakeagama Lake pegmatite are: 1) layered spodumene pegmatite–aplite (NW pegmatite): a coarse petalite (retrograded to spodumene + quartz), K-feldspar pegmatite zone interlayered with a creamy white aplite, 2) a K-feldspar-rich zone, with generally blocky K-feldspar exceeding 80 vol.%, occurs with petalite (up to 0.7 by 1.1 m and retrograded to spodumene + quartz), 3) a petalite – K-feldspar zone, perhaps representing a variant of the K-feldspar-rich pegmatite zone, but with 30–50% modal spodumene–quartz intergrowths after petalite, 4) a quartz-rich wall zone, containing 70–90% grey to white quartz, blocky K-feldspar, albite, muscovite, lepidolite, and spodumene, and 5) a spodumene–quartz-rich core zone, consisting of 60% spodumene and 40% quartz in aggregates indicative of replacement after petalite.

On the basis of columbite–tantalite compositions (Breaks *et al*. 1999a), the NW part of the pegmatite is considered to represent the earliest intrusive phase. Collectively, the other four zones are referred to as the SE pegmatite as they have similar columbite–tantalite compositions. A summary of the mineralogy of the Pakeagama Lake pluton and pegmatite is presented in Table 2.

The Pakeagama Lake pluton contains equant black tourmaline (mainly schorl) that in thin section is zoned from light brown to greyish blue. Partial alteration of the tourmaline has produced a bright blue tourmaline (dark grey in the back-scattered electron image) of foitite composition (Figs. 3a, b).

In the NW pegmatite, tourmaline is black to deep blue, representing a shift in composition to elbaite. In thin section, colors are bright blue to almost colorless (Figs. 3c, d). Tourmaline is scarce in the K-feldspar-rich and petalite – K-feldspar zones, except for the later cross-cutting veins. Tourmaline (elbaite) is more abundant in the more evolved zones (quartz-rich wall zone and spodumene–quartz-rich core zone), where it forms equant or stubby prisms of bluish black or deep blue color; in thin section, it seems very similar to the blue tourmaline from the NW pegmatite.

Tourmaline from late veins cutting pegmatite and pluton is much more variable in color, shape and localized abundance, likely because of different histories of crystallization in the various veins. Black to deep blue tourmaline is most common, and in thin section some of the tourmaline has a dark blue (inclusion-rich) core of schorl – "fluor-elbaite" composition, mantled by a later generation of pale blue "fluor-elbaite" (Figs. 3e, f). One small (30 cm thick) vein of K-feldspar – quartz – albite aplite, situated 120 m south of the main pegmatite mass within the Pakeagama Lake pluton (Fig. 1b), contains pollucite and a distinctly green tourmaline. A similar green tourmaline was found in albitized veinlets cross-cutting the petalite – K-feldspar zone of the pegmatite. Both have "fluor-elbaite" compositions.

The banded iron-formation host-rocks close to the contact and also in entrained selvedge material contain high concentrations of black tourmaline (modally up to 40% and of foitite–schorl composition) which, in thin section, is brown or greenish brown. A zoned foitite – schorl – elbaite grain from the NW pegmatite but only 2 mm away from selvedge material also shows this coloration (Fig. 3g).

Separation Rapids pegmatites

The Separation Rapids group of rare-element granitic pegmatites (Fig. 1c) is located 70 km north of Kenora in northwestern Ontario and lies almost entirely within the Separation Lake metavolcanic belt, which forms part of the boundary zone between the high-grade metasedimentary-rock-dominant English River Subprovince to the north (Breaks 1991, Breaks & Bond 1993) and the granite–tonalite-dominant Winnipeg River Subprovince to the south (Beakhouse 1991). On the basis of a striking similarity in geological setting, age of emplacement and mineralogy, the Separation Rapids pegmatites are regarded as the easterly extension of the Winnipeg River – Cat Lake pegmatite field of Manitoba and can be linked along strike with probably the greatest number of complex-type, petalite-subtype granitic pegmatite occurrences in Canada. These include the Greer Lake and Bernic Lake (Tanco) pegmatite groups (Černý *et al*. 1981, Breaks & Tindle 1997).

All the petalite-subgroup pegmatites are exterior to the Separation Rapids pluton and predominantly intrude gabbroic and mafic metavolcanic rocks, although locally some cut banded iron-formation. Two major clusters of pegmatites comprise the Separation Rapids pegmatite group. Both are spatially related to the Separation Rapids pluton (Breaks 1993, Breaks & Tindle 1994, 1996a, b, 1997, Breaks & Pan 1995, Breaks *et al*. 1999b). Complex-type, petalite-subtype and beryl pegmatites are

FIG. 2. Outcrop photograph of the Pakeagama Lake granite showing a variety of randomly oriented and, in some cases, cross-cutting series of tourmaline-bearing minor intrusions and metasomatic veins. Camera case is 18 cm across.

TABLE 2. MINERAL SPECIES FOUND IN GRANITIC PEGMATITE AT SEPARATION LAKE AND PAKEAGAMA LAKE

	Pakeagama Lake					Separation Lake				
	1	2	3	4	5	6	7	8	9	10
Ferrocolumbite						●	●			●
Ferrotantalite						(●)	●			●
Manganocolumbite		●	●	●	●	(●)		●	●	●
Manganotantalite		●	●	●	●	●		●	●	●
Ferrotapiolite						(●)			(●)	
Ferrowodginite						(●)	●		(●)	
Wodginite				(●)			(●)	●	●	(●)
Titanowodginite									●	(●)
"Ferrotitanowodginite"								●		
Tungsteniferous wodginite								●	●	
Microlite			(●)	(●)	(●)		●	●	●	●
Stibiomicrolite				(●)			●		●	
Uranmicrolite					●		●			
Cassiterite	(●)	●	●	●	●	●	●	●	●	●
Strüverite									●	
Gahnite						●				●
Nigerite							●			●
Pyrite						●	●		●	
Löllingite	(●)						●		●	
Arsenopyrite	(●)						●	●		●
Chalcopyrite						●	●			
Monazite					●	●	●		●	●
Xenotime							●			●
Amblygonite–montebrasite			●	●						
Epidote						●	●		●	
Fluorite				(●)	(●)	●		●		●
Topaz								●		●
Beryl		●	●	●	●	●	●	●	●	●
Tourmaline										
Dravite							●			
Foitite	●						●			
Schorl	●	●			●		●			
Elbaite		●	●	●	●			●	●	
"Fluor-elbaite"		●	●	●	●				●	
Cordierite–Sekaninaite						●				
Garnet										
Almandine (>50%)	●					●	●	(●)		(●)
Spessartine (> 50%)	(●)						●	●	●	●
Siderophyllite–Zinnwaldite	●					●				●
Muscovite	●	●	●	●	●	●	●	●	●	●
Lepidolite		●	●	●	●		●	●	●	●
Nanpingite				●					●	
Spodumene		●	(●)	●	●				●	●
Petalite							●		●	●
Eucryptite										●
Pollucite					●				●	

Column headings: 1 Pakeagama Lake pluton, 2 NW pegmatite, 3 SE pegmatite, quartz-rich wall zone, 4 SE pegmatite, quartz–spodumene core zone, 5 minor intrusions and veins. All rock types also contain quartz, albite, K-feldspar, fluorapatite (especially in Pakeagama Lake pegmatites), and zircon. Additional minor species include: lithiophilite in 3, stibiotantalite and native antimony in 4; sphalerite, ilmenite, alluaudite and allanite in 6, scheelite, uraninite, purpurite, chrysoberyl and bityite in 7, bismutomicrolite in 8, stibiobetafite in 9; yttropyrochlore and cookeite in 10. In 2, native bismuth, löllingite and arsenopyrite also are found, but only along the pegmatite – banded iron-formation host-rock contact. In the Big Mack pegmatite (not listed above), holmquistite and bikitaite are further species. (●): found in one sample only.

found in both clusters, termed the Eastern subgroup (7.5 km^2) and the Southwestern subgroup (6 km^2). Beryl and petalite pegmatites having essentially identical U/Pb columbite–tantalite ages of 2649 ± 4 and 2642 ± 7.6 Ma, respectively, were reported by Smith *et al.* (2000). In total, 70 rare-element mineral occurrences (pegmatites or aplites) have been documented in the Separation Lake area.

In the Eastern subgroup, 11 complex-type, petalite-subtype pegmatites, ranging in size from 1 by 14 m to 8 by 130 m, are almost entirely enveloped by a zone of beryl pegmatites (Fig. 1c). The largest of the petalite-subtype pegmatites is Marko's pegmatite, which exhibits striking internal zonation, and is host to a unique assemblage of tantalum-bearing species (summarized in Table 2). The tantalum-bearing species found in this and other Separation Rapids pegmatites have been described by Tindle & Breaks (1998), Tindle *et al.* (1998) and Tindle & Breaks (2000a, b). Marko's pegmatite, pegmatite 93–265 and one other beryl pegmatite define a Mn-suite on the basis of their manganocolumbite–manganotantalite assemblage [*i.e.*, $Mn/(Mn + Fe) > 0.5$], and are considered to be derived from evolved parental compositions relatively enriched in fluorine, as suggested by the presence of the F-rich species topaz and microlite (Tindle & Breaks 1998). Other petalite-subtype pegmatites in the eastern subgroup have a more primitive signature; on the basis of their ferrocolumbite and ferrotantalite content, they have been termed Fe-suite pegmatites (Tindle & Breaks 1998). The Fe-suite pegmatites have been further divided on the basis of their columbite-group mineralogy into Group 1 with $0 < Mn/(Mn + Fe) < 0.25$ and Group 2 with $0.25 < Mn/(Mn + Fe) < 0.5$ (Tindle & Breaks 2000a).

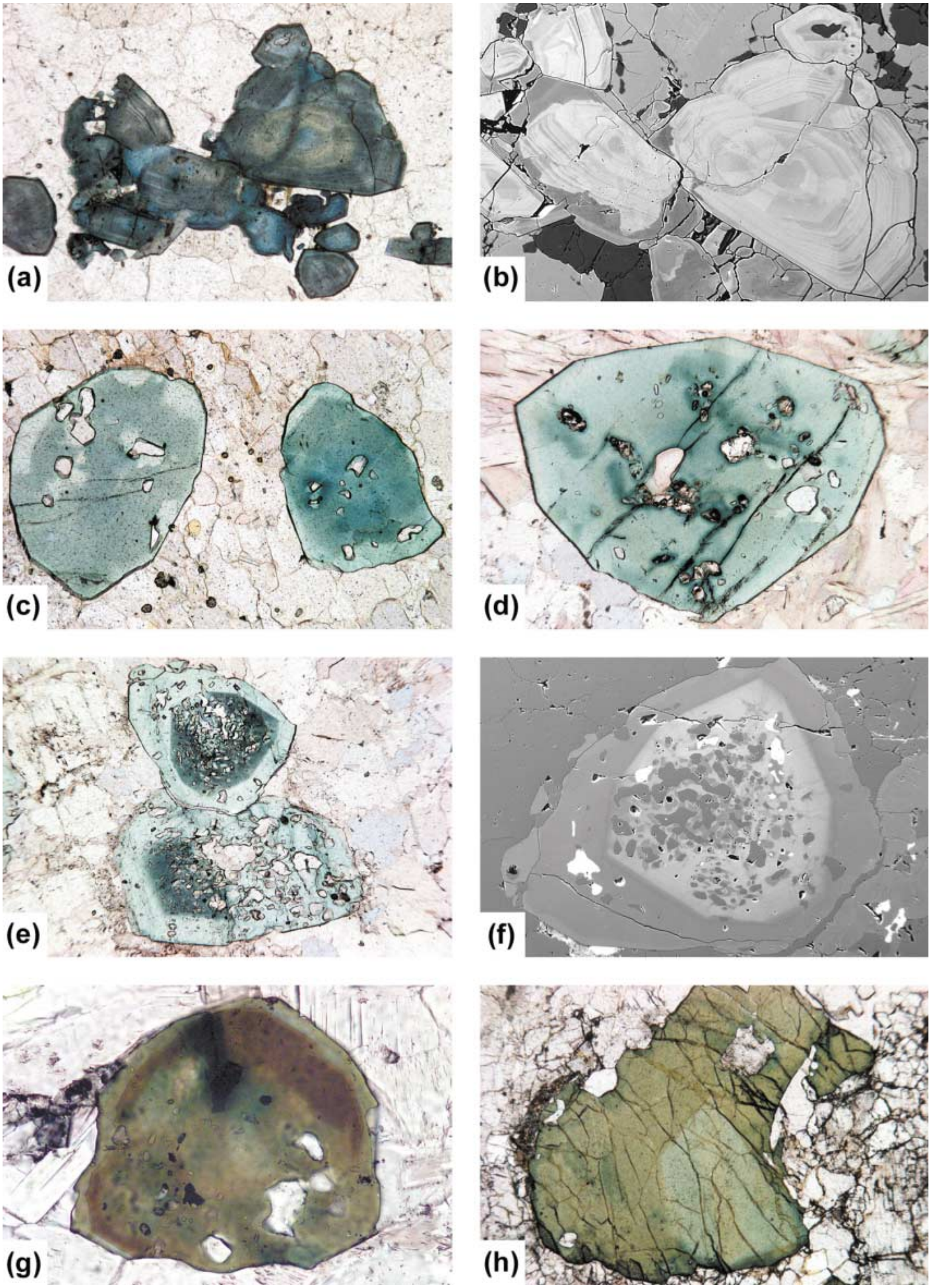
The Southwestern subgroup contains a swarm of small petalite-bearing pegmatites and nine relatively large deformed pegmatites ranging in size from 6 by 56 m to 60 by 650 m. The largest of these is the Big Whopper pegmatite, in which petalite occurs in a K-feldspar assemblage (minor quartz and muscovite) and, less commonly, in monomineralic lenses in a garnet–muscovite aplite (Breaks & Tindle 1997). The other major petalite-bearing pegmatite belonging to the subgroup is the Big Mack pegmatite (Breaks *et al.* 1999b), notable for being the only pegmatite described here to host all of the following lithium-rich species: petalite (major), eucryptite, spodumene, bikitaite and holmquistite. The presence of bikitaite indicates that crystallization processes were active down to 275°C between load pressures of 1 to 2 kbar (Vidal & Goffé 1991). Apart from a single isolated crystal, tourmaline does not occur in any of the larger pegmatites. Columbite–tantalite data indicate that the southwestern subgroup, like the eastern subgroup, contains both Fe-suite and Mn-suite pegmatites.

Tourmaline (elbaite) from Marko's pegmatite and pegmatite 93–265 is relatively uncommon, forming dark blue to dark green prisms in hand specimen (dark to light blue in thin section). Tourmaline from other Separation Rapids pegmatites form black stubby prisms that in thin section vary from green to brownish green. Some exhibit simple zonation (Fig. 3h), and the majority have the composition of schorl. There is little evidence of any exocontact tourmaline in the entire Separation Lake area. In one thin petalite-bearing dyke (97–71), however, black Ca-rich dravite–schorl associated with Rb- and Cs-enriched mica has been found in and around entrained selvedge material.

COMPOSITIONAL TRENDS IN TOURMALINE

Lithium clearly plays a role in the development of petalite-subgroup granitic pegmatites. Figures 4–11 indicate a large variation in the content of Li in tourmaline from both the Pakeagama Lake and Separation Rapids pegmatites. In addition, because Li is absent (or at least very low in abundance) in the host rocks and enriched in the pegmatites, its presence in tourmaline is potentially useful in comparing characteristics of magmatic processes, such as crystal fractionation, with other processes, such as pegmatite – host-rock interaction. The only alternative to Li when trying to illustrate these processes is Al, an element that also resides at the *Y* site of tourmaline (as well as occupying the *Z* site). Whereas we have confirmed in all cases that there is little difference between trends in Li and Al, it is Li that illustrates the trends best. The only reservation in using Li data

FIG. 3. Tourmaline in thin section. (a) – (g) Pakeagama Lake, (h) Separation Lake. (a) Foitite–schorl core to schorl–foitite to schorl rim, from parent granite; width 1.0 mm (99–FWB38). (b) Back-scattered electron image of selected grains in (a) showing sector-zoned compositional variation in foitite–schorl to schorl and with foitite replacement (dark grey), particularly around grain on left; width 750 μm. (c) Elbaite–schorl (blue) to "fluor-elbaite" – schorl (almost colorless) from the layered spodumene pegmatite – aplite (NW pegmatite); width 1.0 mm (99–FWB36). (d) Elbaite–schorl core to "fluor-elbaite" rim containing quartz, lepidolite, fluorapatite and mixed albite – K-feldspar (crystallized melt?) inclusions, from vein cutting wall zone of SE pegmatite; width 2.8 mm (98–AT5). (e) Inclusion-rich Ca-bearing "fluor-elbaite" – schorl core to Ca-rich "fluor-elbaite" (both deep blue) to relatively inclusion-free "fluor-elbaite" rim (light blue) containing many inclusions of quartz and a few of K-feldspar throughout the grain, from vein cutting wall zone of SE pegmatite; width 2.8 mm (98–AT6). (f) Back-scattered electron image of one grain from (e) showing abundant quartz inclusions in the core region (dark grey) and a few scattered inclusions of K-feldspar (white). Reaction of the core of the grain with the inclusions has locally modified the composition of the tourmaline (medium grey); width 1600 μm. (g) Foitite–schorl core to schorl–foitite to schorl–elbaite to thin elbaite–schorl rim from NW pegmatite, 2 mm from selvedge; width 0.5 mm (99–FWB20). (h) Mg-rich schorl from Fe-suite petalite pegmatite, in which the core is more Fe-rich than the rim; width 2.8 mm (95–117D).


(a)
(b)
(c)
(d)
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(g)
(h)

from tourmaline analyzed by electron microprobe is that concentrations have to be calculated on stoichiometric criteria rather than actually being measured, as the electron microprobe cannot detect Li. The calculation procedure used here is, however, well established and tested (Morgan & London 1987, Novák & Povondra 1995, Novák *et al*. 1999, Selway *et al*. 1999, Teertstra *et al*. 1999). Aurisicchio *et al*. (1999) reported that Li values quantified with the ion microprobe "show excellent agreement with the stoichiometric ones, testifying both to the accuracy of the SIMS calibration procedure and to the good match between ion and electron microprobe sampling".

We now examine Li variation in tourmaline from a wide range of rock types from Pakeagama and Separation Lakes and correlate the behavior of Li with that of other elements. These variations will be tied into the main schemes of substitution operating in tourmaline once the overall composition is established. This approach also has the advantage that the complexity of tourmaline nomenclature can, for the time being, be minimized. As a reference, some tourmaline end-member names have been included on Figures 1–8, but because tourmaline nomenclature demands that a number of elements be considered, no single two-element plot can be used to classify tourmaline directly. For this reason, it is important to note that no data from Pakeagama or Separation Lakes correspond to feruvite, uvite or rossmanite, even though the data fall on or close to end-member positions on Figures 4–11.

Li–Fe covariation

The negative correlation between Li and Fe^{2+} is striking for the Pakeagama pegmatite (Fig. 4a). Samples from the SE quartz–spodumene core zone fall slightly off the correlation line (fortuitously between the foitite and elbaite end-members on Fig. 4a, despite the fact that the data are later shown to pertain to schorl–elbaite compositions!). Tourmaline data from the Pakeagama pluton do not define a continuum with the pegmatite, but show relatively elevated Fe^{2+} contents (Fe^{2+} at Y site: 2.0–2.5 *apfu*). Minor intrusions and metasomatic veins (too narrow in many cases to be described as aplitic or pegmatitic dykes) from the Pakeagama Lake area, with few exceptions, fall between schorl–elbaite and foitite–elbaite compositions (Fig. 4b), but the exocontact and selvedge data define a trend toward the dravite end-member, indicating a major substitution of Mg for Fe^{2+}. Some pegmatitic veins (*e.g.*, 98–SS21B and 99–PKL12) have compositions of tourmaline close to those of tourmaline in the Pakeagama Lake pluton that they intrude, and they are therefore considered to be primitive. Others, such as the pollucite-bearing vein and an albitized veinlet (hosted in the petalite – K-feldspar zone, 98–FWB37) are more Li-enriched.

Figure 4c shows the data on tourmaline from the Separation Rapids pegmatites defining a weak trend along the foitite–elbaite line, with significant deviation toward dravite compositions (up to 1.1 *apfu* Fe^{2+}) in the Group-1 Fe-suite pegmatites (seven petalite-subtype pegmatites from the eastern subgroup comprise this group). Tourmaline with a high Li content from Marko's pegmatite also deviates from the main correlation line (dropping to 0.3 *apfu* Fe^{2+}), toward the rossmanite end-member. Tourmaline from the Tanco and Urubu pegmatites closely mimic those from the Pakeagama Lake pegmatite and the more evolved (Li-enriched) pegmatites from Separation Lake (Fig. 4c), but both continue to higher Li concentrations with elbaite–rossmanite substitution. Compositions from replacement veinlets of tourmaline (replacing an earlier generation of tourmaline) in Mn-suite pegmatites (actually foitite and schorl–foitite) and from selvedge-rich pegmatite 97–71 (actually Ca-rich dravite–schorl), both from the Separation Lake area, cluster around the foitite end-member, unlike most of the tourmaline from Tanco exocontact rocks, which are enriched in Li and trend toward the elbaite end-member (Fig. 4d).

Li–Mg covariation

Magnesium is uniformly low in tourmaline from the Pakeagama pegmatite (Fig. 5a). No samples have more than 0.04 *apfu*. In the Pakeagama Lake pluton, tourmaline is only slightly more enriched (Mg < 0.14 *apfu*). This is not the case for tourmaline from mafic exocontact and selvedge samples, where Mg attains 1.7 *apfu* (Fig. 5b). Tourmaline in veins from the Pakeagama Lake area shows little enrichment in Mg, although those samples with relatively high Fe^{2+} also have an elevated Mg content (to 0.14 *apfu*), with the highest values concentrated in the two primitive pegmatitic veins cutting the Pakeagama Lake pluton (98–SS21B and 99–PKL12).

Compared to tourmaline in the Pakeagama pegmatite, with <0.04 *apfu* Mg, the pattern is very different in the Separation Lake pegmatites, where the concentration of Mg rises to 1.4 *apfu* in Group-1 Fe-suite pegmatites and to 0.5 *apfu* in one sample from Marko's pegmatite collected 2–5 cm away from the BIF host-rocks (Fig. 5c). These pegmatites have tourmaline compositions much more comparable with those from the mafic exocontact samples at Pakeagama Lake. Tourmaline in the only Group-2 Fe-suite pegmatite (a single pegmatite from the eastern subgroup, pegmatite 7), however, has a low Mg content (up to 0.1 *apfu*), similar to that in the Pakeagama pegmatite. The bulk of the tourmaline data from the Mn-suite pegmatites (pegmatite 93–265 and Marko's pegmatite) also have low levels of Mg (<0.1 *apfu*), which is similar to tourmaline in the Pakeagama pegmatite. Tourmaline data from the Tanco and Urubu pegmatites have many similarities with the Separation Rapids pegmatite data, with most data plotting along the Li axis in Figure 5c because of extremely low concentrations of Mg. In addition, tourmaline with

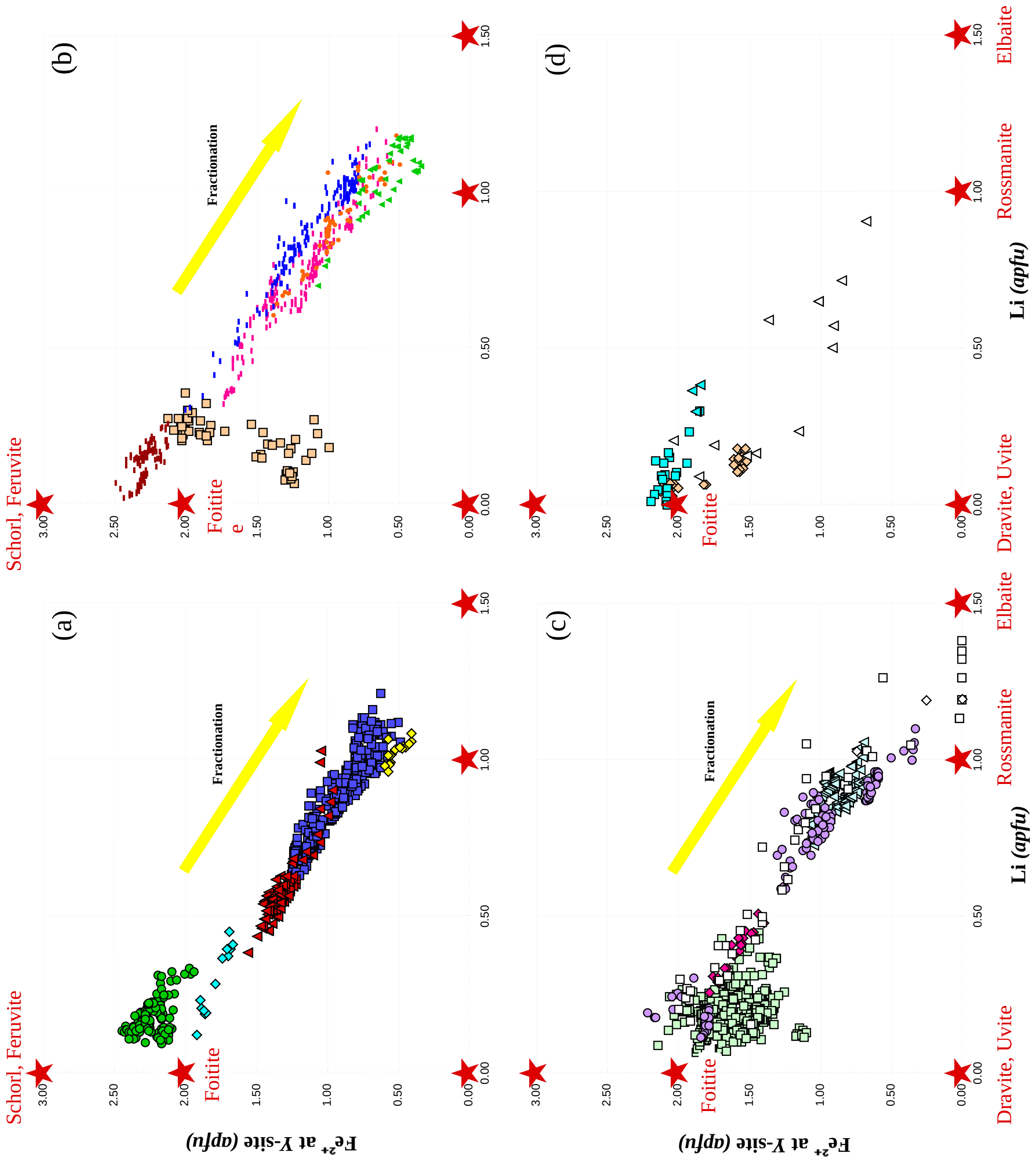

FIG. 4. Variation in Li *versus* Fe^{2+} (atoms per formula units, *apfu*) variation in tourmaline. (a) Pakeagama Lake data; pluton (dark green circles), pluton, replacement-related compositions (bright blue diamonds) (see also Figs. 3b, c), NW pegmatite (red triangles), SE quartz-rich wall zone (dark blue squares), SE quartz–spodumene core zone (yellow diamonds). (b) Pakeagama Lake data; mafic host-rocks and selvedge caught up in the pegmatite (fawn squares), primitive pegmatitic veins cutting pluton, 98–SS21B and 99–PKL12 (brown dashes), pollucite-bearing vein (orange dots), albitized veinlet in petalite – K-feldspar zone, 98–FWB37 (green triangles), other minor intrusions in exocontact rocks (pink dashes), metasomatic veins (blue dashes). (c) Separation Rapids data; Group-1 Fe-suite pegmatites from the eastern subgroup (pale green squares), Group-2 Fe-suite pegmatite (red diamonds), Mn-suite pegmatites: Pegmatite 93–265 (pale blue triangles), Marko's pegmatite (lilac circles), Tanco pegmatite, Manitoba (Černý *et al.* 1996, 1998, Selway *et al.* 2000b) (white squares) and Urubu pegmatite, Minas Gerais, Brazil (Quéméneur *et al.* 1993) (white diamonds). (d) Separation Rapids data; replacement veins cutting tourmaline in pegmatite 93–265 (bright blue triangles) and Marko's pegmatite (bright blue squares), selvedge associated with pegmatite 97–71 (fawn diamonds), Tanco exocontact samples (white triangles). Tourmaline end-member positions are marked with red stars, where appropriate. The yellow arrow points in the direction of increasing fractionation, as determined from the Tanco (Selway *et al*. 2000b) and Bob Ingersoll (Jolliff *et al*. 1986) pegmatites.

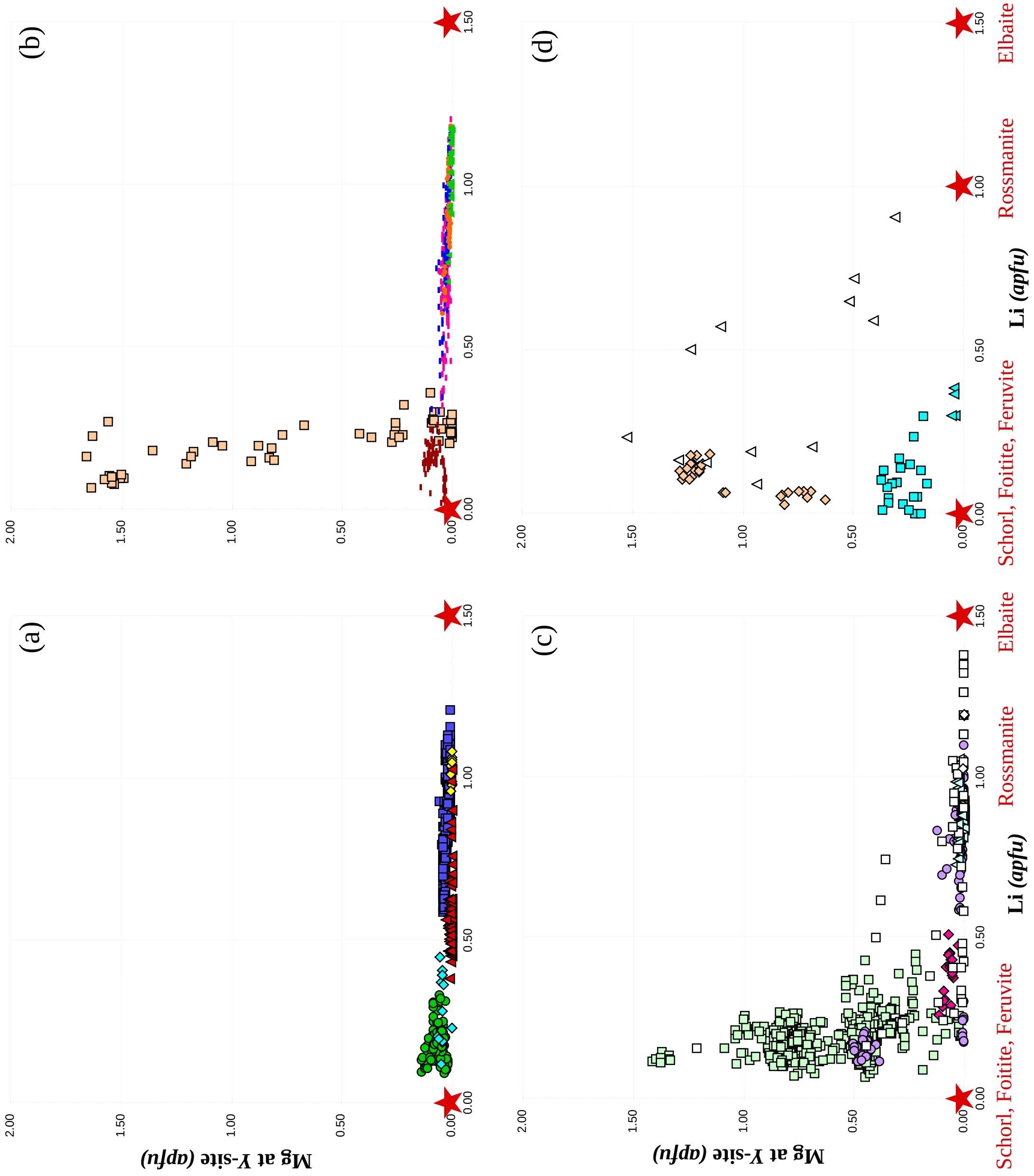

FIG. 5. Variation in Li *versus* Mg (at the *Y* site) (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4. In addition to Mg at the *Y* site, selvedge samples 99–FWB24 and 97–71 have Mg at the *Z* site.

low Li (0.1–0.3 *apfu*) and relatively high Mg (to 1.2 *apfu*) is reported from the Tanco pegmatite (Selway *et al*. 2000b). The level of magnesium in tourmaline from selvedge-entrained pegmatite 97–71 (Fig. 5d) closely matches that in the selvedge and the mafic exocontact at Pakeagama Lake (including being further enriched in Mg to 0.2 *apfu* at the *Z* site) and from Tanco exocontact rocks, but late replacement veins of tourmaline contain little Mg (Marko's pegmatite; <0.1 *apfu*) or are only moderately enriched (pegmatite 93–265; 0.2–0.4 *apfu*).

Li–Ti covariation

The behavior of Ti is similar to that of Mg in tourmaline from both areas (Fig. 6). There is a slight enrichment of Ti in tourmaline from the Pakeagama Lake pluton (<0.04 *apfu*) compared to the Pakeagama pegmatite, which varies from ~0.02 *apfu* in the NW pegmatite to <0.01 *apfu* in the SE quartz–spodumene core zone (Fig. 6a). This pattern is repeated in the minor intrusions, although there is some scattering of data, up to 0.05 *apfu* Ti in some cases, such as in the primitive pegmatitic veins 98–SS21B and 99–PKL12 (Fig. 6b), all of which intrude the Pakeagama Lake pluton. As with Mg, it is in tourmaline from mafic exocontact and selvedge samples that the greatest enrichment of Ti occurs (0.0 to 0.14 *apfu*). In the Pakeagama Lake pluton, replacement tourmaline is significantly lower in Ti (<0.01 *apfu*) than the original tourmaline.

At Separation Lake, tourmaline from the Group-1 Fe-suite pegmatites are enriched in Ti, and a continuum from 0.0 to 0.11 *apfu* rising to 0.14 *apfu* is observed (Fig. 6c). One sample from Marko's (Mn-suite) pegmatite, collected 2–5 cm away from BIF host-rocks is, perhaps not unexpectedly, also Ti-enriched (to 0.11 *apfu*). This is the same sample that also had high Mg relative to the rest of Marko's pegmatite. Other tourmaline from the wall zone of Marko's pegmatite have higher Li and Ti contents (<0.05 *apfu* Ti) despite the same samples having low Mg (<0.1 *apfu*). Only pegmatite 7 (the Group-2 Fe-suite pegmatite) and pegmatite 93–265 (Mn-suite) have low Ti contents (<0.02 *apfu*) comparable with the Pakeagama Lake pegmatite (Fig. 6c). Surprisingly, the distribution of Ti in tourmaline from the Tanco pegmatite is somewhat erratic, although a broad decrease with increasing Li is noted. Figure 6d shows Ti enrichment, to 0.11 *apfu*, in tourmaline from exocontact and selvedge samples from Separation Lake and from Tanco, but in replacement veinlets from Separation Lake, there is little increase in Ti.

Li–Mn covariation

So far, all the elements discussed appear to indicate that at Pakeagama Lake, there is a broad continuum of compositions, perhaps beginning with tourmaline from the pluton (low Mn), followed by a succession through the main pegmatite units, NW pegmatite, SE quartz-rich wall zone and SE spodumene – quartz core zone (increasing Mn). Manganese is the first element to deviate from this simple pattern, as Figure 7a shows the NW pegmatite data to trend toward low levels of Mn (0.02 *apfu*), whereas the two SE pegmatite zones trend toward higher levels (up to 0.22 *apfu* Mn). The sinusoidal shape of Mn variation in the Pakeagama pegmatite is also found in the minor intrusions (Fig. 7b), together with a major split into low- and high-Mn groups. As the high-Mn group closely matches the composition and pattern of the Pakeagama pegmatite, we consider it to be essentially a magmatic grouping, whereas data in the low-Mn group are predominantly from tourmaline-rich metasomatic veins that cross-cut the Pakeagama Lake pluton close to the contact with the SE pegmatite zones. We contend that these samples have an origin that involved interaction with host rocks. Implicit in this model is the assumption that late- to post-magmatic albitization produces tourmaline with an evolved signature. Falling between these two main groups are data from the pollucite-bearing vein.

A sinusoidal trend in tourmaline data from the Separation Rapids pegmatites is also observed (Fig. 7c), but this time to much higher concentrations (Mn to 0.39 *apfu* in a Li mica – bladed albite pod from the eastern end of Marko's pegmatite). The lowest Mn contents are recorded in the Group-1 Fe-suite pegmatites (0.0–0.07 *apfu* Mn), followed by the Group-2 Fe-suite pegmatite (0.07–0.14 *apfu* Mn) and pegmatite 93–265 (0.07–0.23 *apfu* Mn). Marko's pegmatite again spans almost the entire range of tourmaline compositions, with samples from close to the host rocks having the lowest Mn contents (*e.g.*, 96–AT19A: 0.27–0.32 *apfu*). Apart from high-Li tourmaline (Li > 1.3 *apfu*), there is good correlation of Tanco compositions with those from the Separation Rapids pegmatites. Replacement veinlets, exocontact and selvedge tourmaline from Pakeagama Lake, Separation Lake and Tanco are uniformly low in Mn (<0.05 *apfu*; Figs. 7b, d).

Li–Zn covariation

Broad, subparallel trends of decreasing Zn with increasing Li are found in tourmaline from the Pakeagama pegmatite (Fig. 8a), with the NW pegmatite offset to much higher concentrations (up to 0.2 *apfu* Zn) compared to the SE pegmatite zones (<0.12 *apfu* Zn). Data from the Pakeagama Lake pluton (0.02–0.05 *apfu* Zn) or replacement compositions show no obvious relationship with either pegmatite unit, but tourmaline from a primitive pegmatitic vein cutting the pluton (99–PKL12; Fig. 8b) has comparable Zn, as well as Fe^{2+} and Mg contents, to those in the pluton, suggesting a common origin. However, a second primitive vein (98–SS21B) cutting the Pakeagama Lake pluton shows little affinity with either the pegmatite or the pluton. Jolliff *et al.* (1986) predicted that under ideal conditions, the maximum enrichment of Zn in tourmaline will occur at an intermediate stage of crystallization, and so the pattern from low Zn (in the pluton) to high Zn (in the NW pegmatite), to low Zn (in the SE pegmatite zones) could represent a normal pattern due to fractionation. Our data support this conclusion to some extent (particularly if data from Fig. 8b are included), but because data from the NW pegmatite are offset from data from the SE pegmatite zones, there is likely to be an additional factor involved. In our opinion, the NW pegmatite crystallized from a slightly different parental composition to that which gave rise to the SE pegmatite zones.

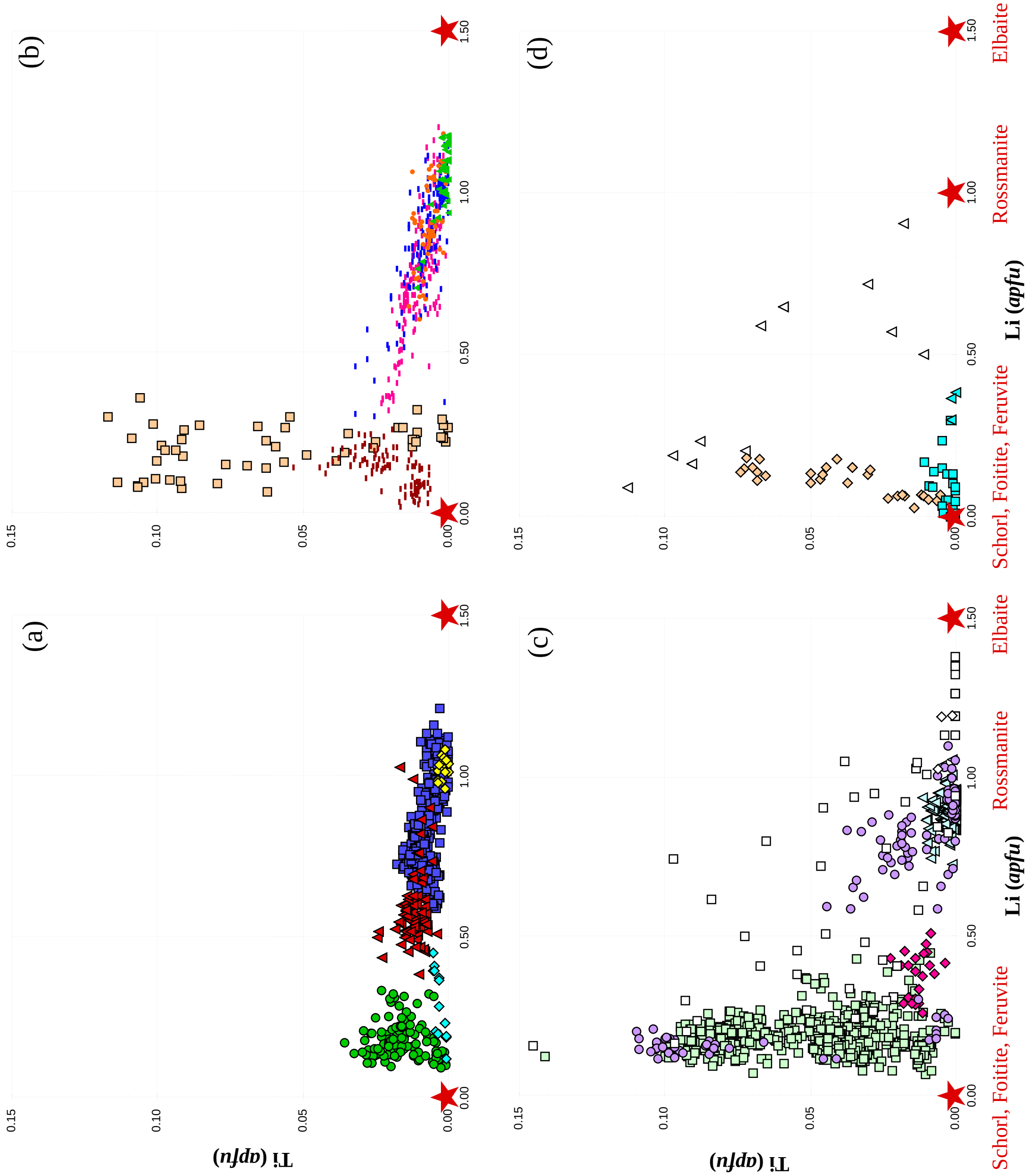

FIG. 6. Variation of Li *versus* Ti (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4.

The behavior of Zn in tourmaline from the minor intrusions at Pakeagama Lake (Fig. 8b) is much more complex, perhaps because some were derived from the NW pegmatite (>0.12 *apfu*), whereas others were derived from the SE part of the pegmatite (0.03–0.10 *apfu* Zn). As with Figure 8a, the maximum contents of Zn are found at intermediate Li contents.

Clearly defined trends in Zn are only apparent in metasomatic veins cross-cutting the Pakeagama Lake pluton (Zn decreasing systematically from 0.03 to 0.005 *apfu*). Data from these veins and the pollucite-bearing vein all have much lower Zn contents than the Pakeagama pegmatite (Figs. 8a, b).

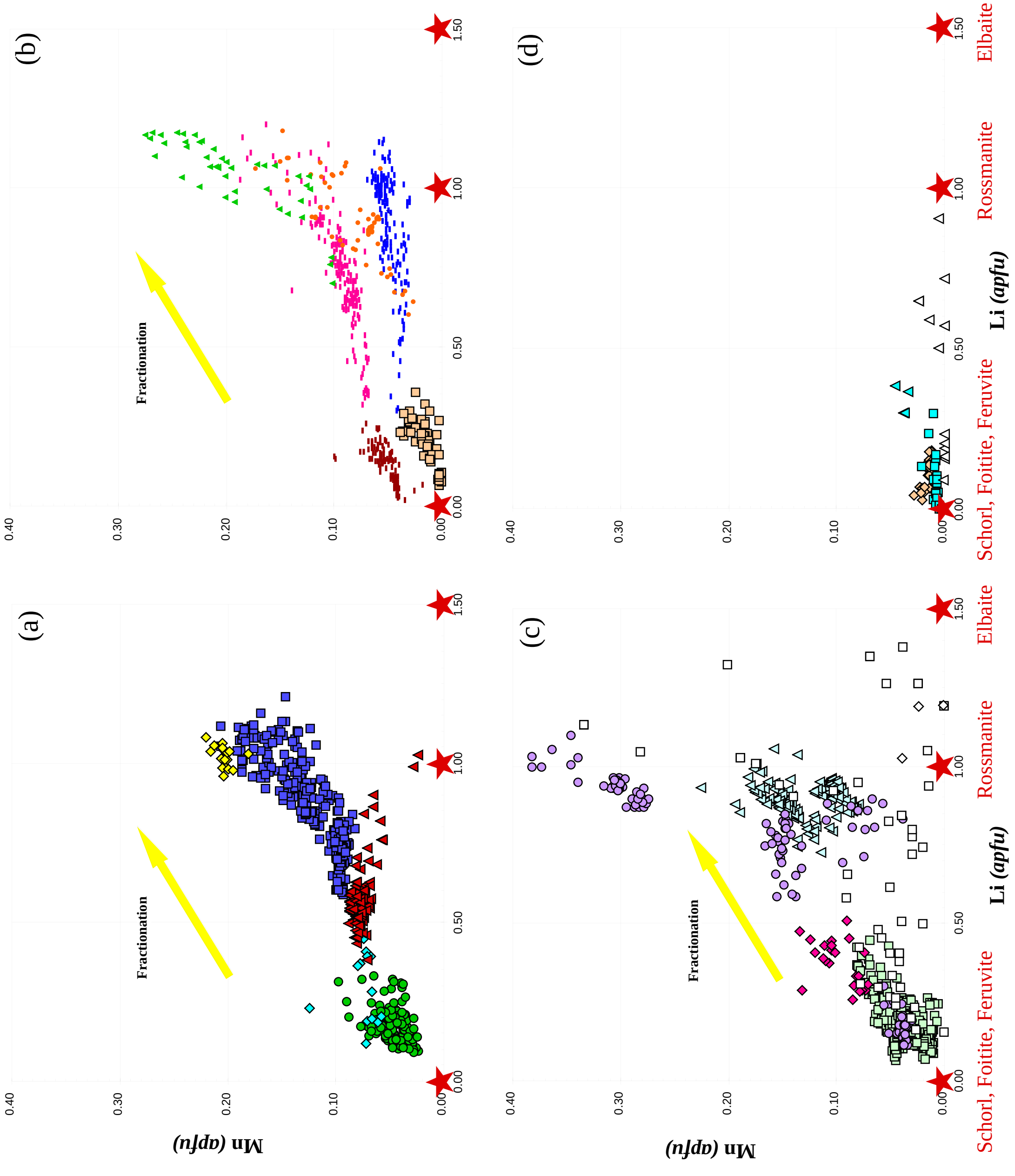

FIG. 7. Variation of Li *versus* Mn (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4.

At Separation Lake (and Tanco), the distribution of Zn in tourmaline shows very little systematic behavior (Fig. 8c), although the Group-1 Fe-suite pegmatites show some enrichment comparable (but at lower concentrations) to that observed for Mg and Ti. Tourmaline from pegmatite 93–265 is more enriched overall in Zn than Marko's pegmatite. Tourmaline from exocontact and selvedge samples and replacement veins (Figs. 8b, d) from both areas (and Tanco) are uniformly low in Zn (<0.04 *apfu*).

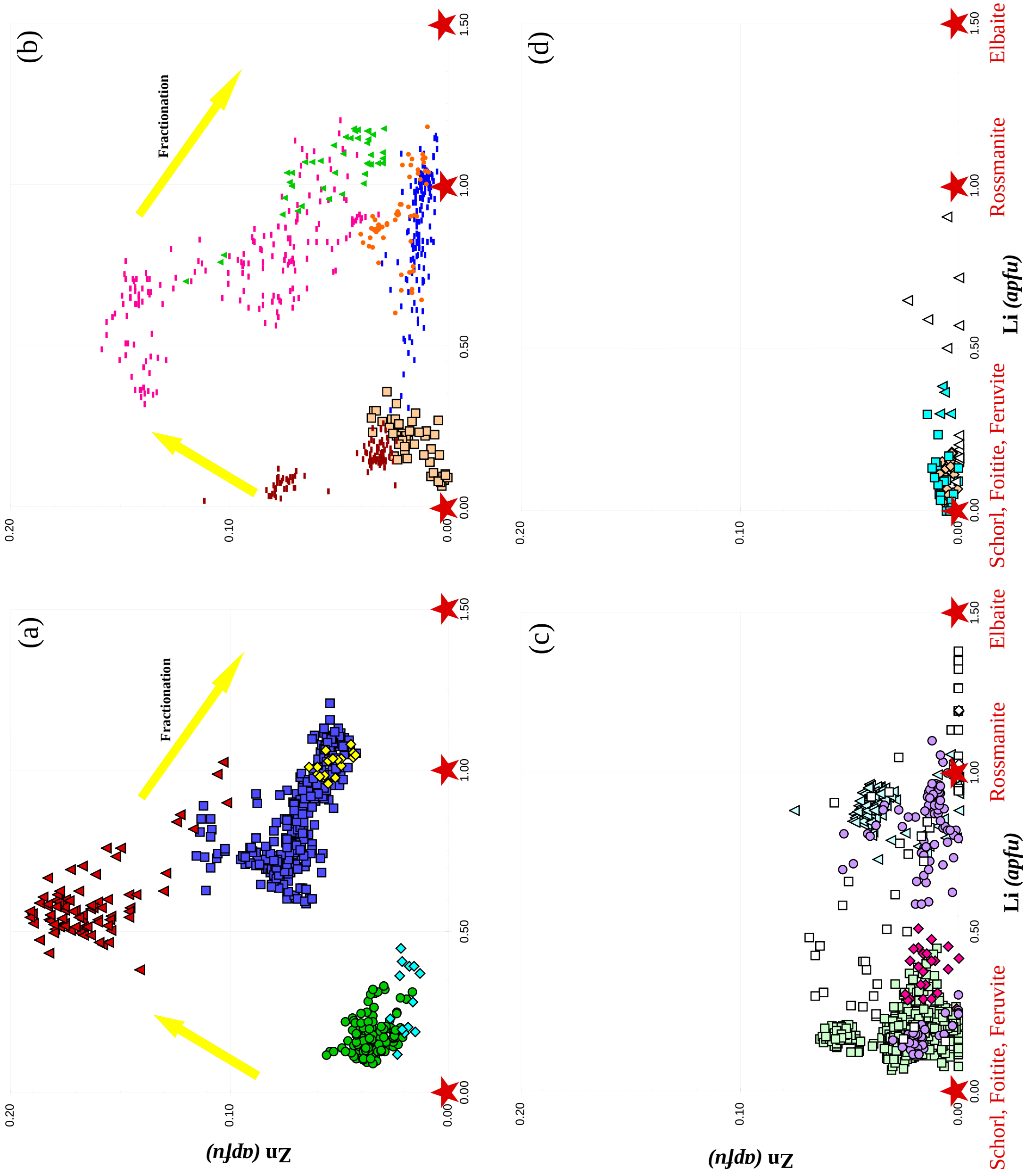

FIG. 8. Variation of Li *versus* Zn (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4.

Li–Ca covariation

Calcium in tourmaline from the Pakeagama pegmatite varies systematically with increasing Li (Fig. 9a). There is a small but perceptible increase in Ca from 0.01 *apfu* in the NW pegmatite to 0.05 *apfu* in the SE spodumene–quartz core zone. This trend is the opposite of what might be expected from normal processes of crystal fractionation, as Ca is usually consumed by the early crystallization of plagioclase. Most samples from the Pakeagama Lake pluton contain higher levels of Ca (up to 0.18 *apfu*) than the pegmatite. Similar, or slightly greater Ca-enrichment (up to 0.29 *apfu*) is found in tourmaline from mafic exocontact and selvedge samples from Pakeagama Lake, Separation Lake and, to a lesser extent, Tanco (Figs. 9b, d).

Tourmaline compositions from most of the minor intrusions at Pakeagama Lake closely match those in the Pakeagama pegmatite, with Ca contents increasing from 0.01 to 0.07 *apfu* in the pollucite-bearing vein and albitized veinlets in the petalite – K-feldspar zone of the SE pegmatite (Fig. 9b). In the two primitive veins cutting the Pakeagama Lake pluton (98–SS21B and 99–PKL12), Ca departs from this simple trend and rises to a maximum of 0.1 *apfu* with minimal Li variation in each sample.

The envelope of data defined by tourmaline from Ca-enriched metasomatic veins at Pakeagama Lake and replacement tourmaline in the Pakeagama Lake pluton is unusual in having a bounding line between Li and Ca = 0 and Li = 2, Ca = 0.5 *apfu* (Figs. 9a, b). This is not a line describing a known substitution (such as to the liddicoatite end-member), but occurs again in tourmaline data from Mn-suite pegmatites from the Separation Lake area, where Ca attains 0.22 *apfu* (Fig. 9c). In all the Group-1 Fe-suite pegmatites and the sample of Marko's pegmatite collected 2–5 cm away from BIF host-rocks, there is a broad spread of Ca-enrichment, up to 0.12 *apfu*. This pattern is distinct from that found in Pakeagama Lake, Tanco and Urubu pegmatites, where Ca is uniformly low in most cases (although some tourmaline from Tanco with high Li contents, is Ca-enriched (to 0.16 *apfu*).

Considering the data as a whole, six groupings emerge: (i) slight enrichment in Ca with increasing Li, best developed in the Pakeagama pegmatite and minor intrusions, (ii) major enrichment in Ca (up to 0.3 *apfu*) with low Li, best developed in mafic host-rocks and selvedge from both areas, (iii) moderate enrichment in Ca (up to 0.12 *apfu*) with low Li, best developed in the Pakeagama Lake pluton and the primitive veins cutting it (98–SS21B and 99–PKL12), Separation Lake Fe-suite pegmatites, and a contact-zone sample from Marko's pegmatite, (iv) major enrichment in Ca (up to 0.3 *apfu*) and Li (up to 1.2 *apfu*) developed in metasomatic veins at Pakeagama Lake, (v) major enrichment of Ca (up to 0.22 *apfu*) and Li (0.5 to 1.0 *apfu*) in Mn-suite pegmatites at Separation Lake (where Ti is also elevated), and (vi) moderate enrichment of Ca (up to 0.16 *apfu*) and Li (>1.0 *apfu*) in evolved pegmatite at Tanco.

Li–Na covariation

In common with Mn, Na increases steadily (but erratically) in abundance in tourmaline from the Pakeagama Lake pluton (0.6 *apfu*) through compositions found in the NW pegmatite (typically 0.7 *apfu*), to compositions in the SE quartz-rich wall zone (0.8 *apfu*). In the SE spodumene–quartz core zone, Na is slightly depleted, to 0.7 *apfu* (Fig. 10a). There is also a drop in Na to 0.4 *apfu* in some samples from the pluton (particularly the replacement compositions) that mimic the pattern observed in mafic exocontact and selvedge samples (Fig. 10b) and is consistent with substitution toward foitite (*X*-site vacancy). Although some minor intrusions, and in particular the pollucite-bearing vein, follow the Pakeagama pegmatite trend, there are many examples where Na is relatively depleted; these include tourmaline in albitized veinlets cutting the petalite – K-feldspar zone of the SE pegmatite (98–FWB37) and, most noticeably, in metasomatic veins cutting the pluton close to the SE pegmatite (Fig. 10b).

Tourmaline data from pegmatites at Separation Lake show essentially the same pattern as those at Pakeagama Lake (Fig. 10c), but there is a much greater spread of data, comparable to that at Tanco (Selway *et al.* 2000b). Replacement veinlets of tourmaline in pegmatite 93–265 and Marko's pegmatite exhibit the most extreme depletion of Na; coupled with their low Ca content, the depletion places them toward the foitite end-member (Fig. 10d).

Li–F covariation

So far, the Li data from tourmaline in the Pakeagama pegmatite and, to a lesser extent, some data from the Pakeagama Lake pluton, have defined clear linear trends with other elements at the *Y* site (Fe^{2+}, Mg, Ti, Mn, Zn) and *X* site (Ca, Na), corresponding to increasing evolution of the pegmatite-forming melt. Figure 11a shows that this relationship also applies to the *W* site, with Li correlating well with F. Tourmaline data from the Pakeagama Lake pluton fall at the low-F end (with some data slightly off the trend, perhaps because of detection-limit problems), and there is a steady progression to F = 0.65 *apfu* in the SE pegmatite zones. Those samples with F > 0.5 *apfu* are "fluor-elbaite", according to Hawthorne & Henry (1999).

All data from minor intrusions, metasomatic veins and mafic exocontact and selvedge samples from Pakeagama Lake and Separation Lake (Figs. 11b, d) follow the same pattern of increasing F with Li. Only in a few cases from the pollucite-bearing vein, mafic exocontact and selvedge samples and replacement compositions of tourmaline from the Pakeagama Lake pluton is there any divergence at all.

A systematic increase of F with Li is also observed in tourmaline data from the Separation Rapids pegmatites (Fig. 11c), with the Group-1 Fe-suite pegmatites falling at the low-F end of the trend and the Mn-suite pegmatites, at the high end (0.7 *apfu* F). Most samples from the Tanco and Urubu pegmatites follow the same pattern, although above 1.3 *apfu* Li, the relationship discontinues because rossmanite at these localities tends to be F-poor.

Summary statements

The variation in tourmaline composition from the pegmatite bodies examined is consistent with the following conclusions:

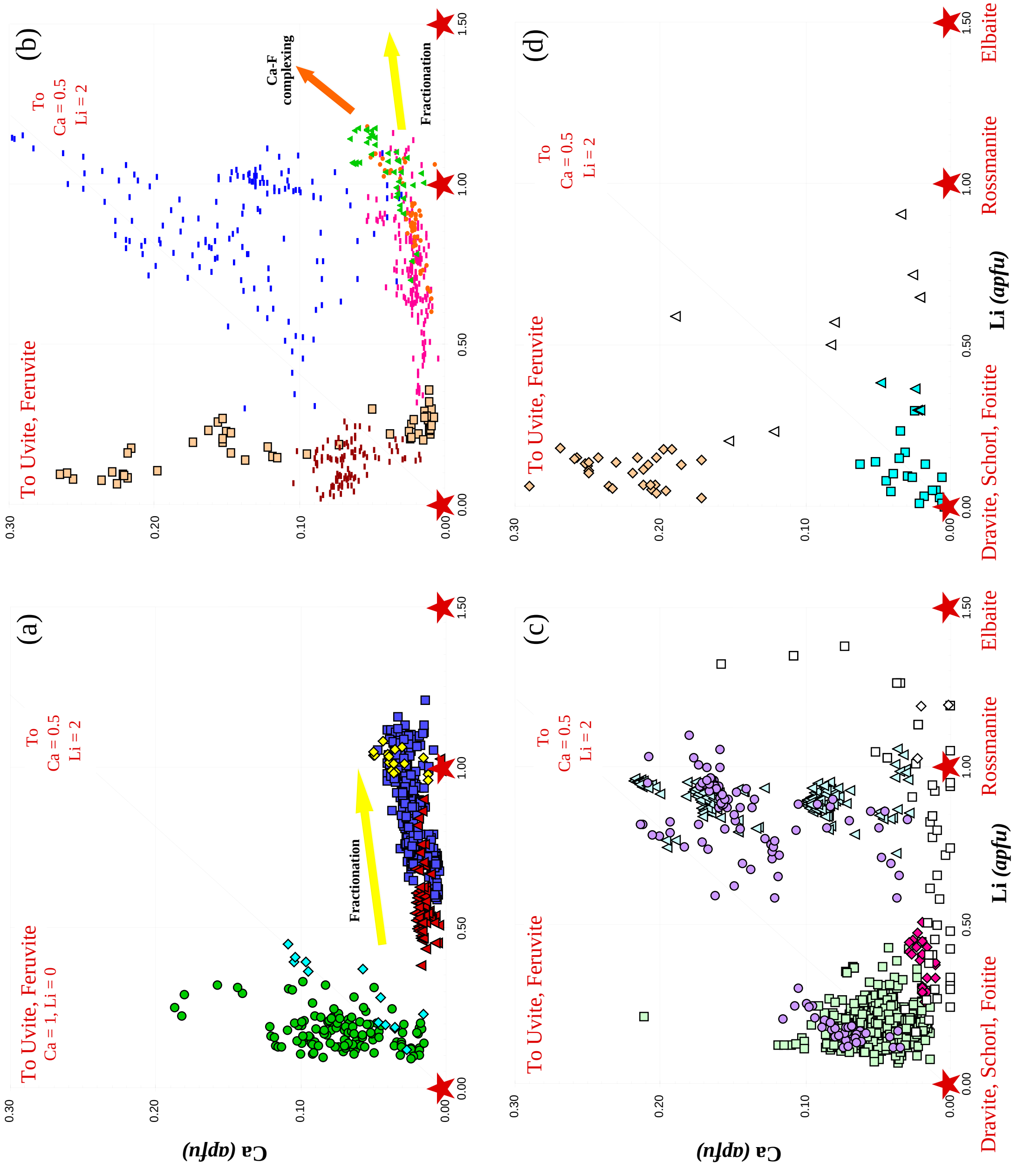

FIG. 9. Variation of Li *versus* Ca (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4.

1. The Pakeagama Lake pegmatite represents an evolving magmatic sequence in which a progressive increase in Li content is correlated with increasing levels of Mn, Ca, Na, and F, and decreasing levels of Fe^{2+}, Mg, and Ti. Crystal fractionation is the likely process controlling most of this variation.

2. At Pakeagama Lake, the SE spodumene–quartz core zone is expected to be the last of the main pegmatite units to crystallize, and hence, the tourmaline should have the most evolved compositions. However, it does not have the highest Li content, and falls off otherwise linear trends in plots against Fe^{2+}, Mn and Na. At least

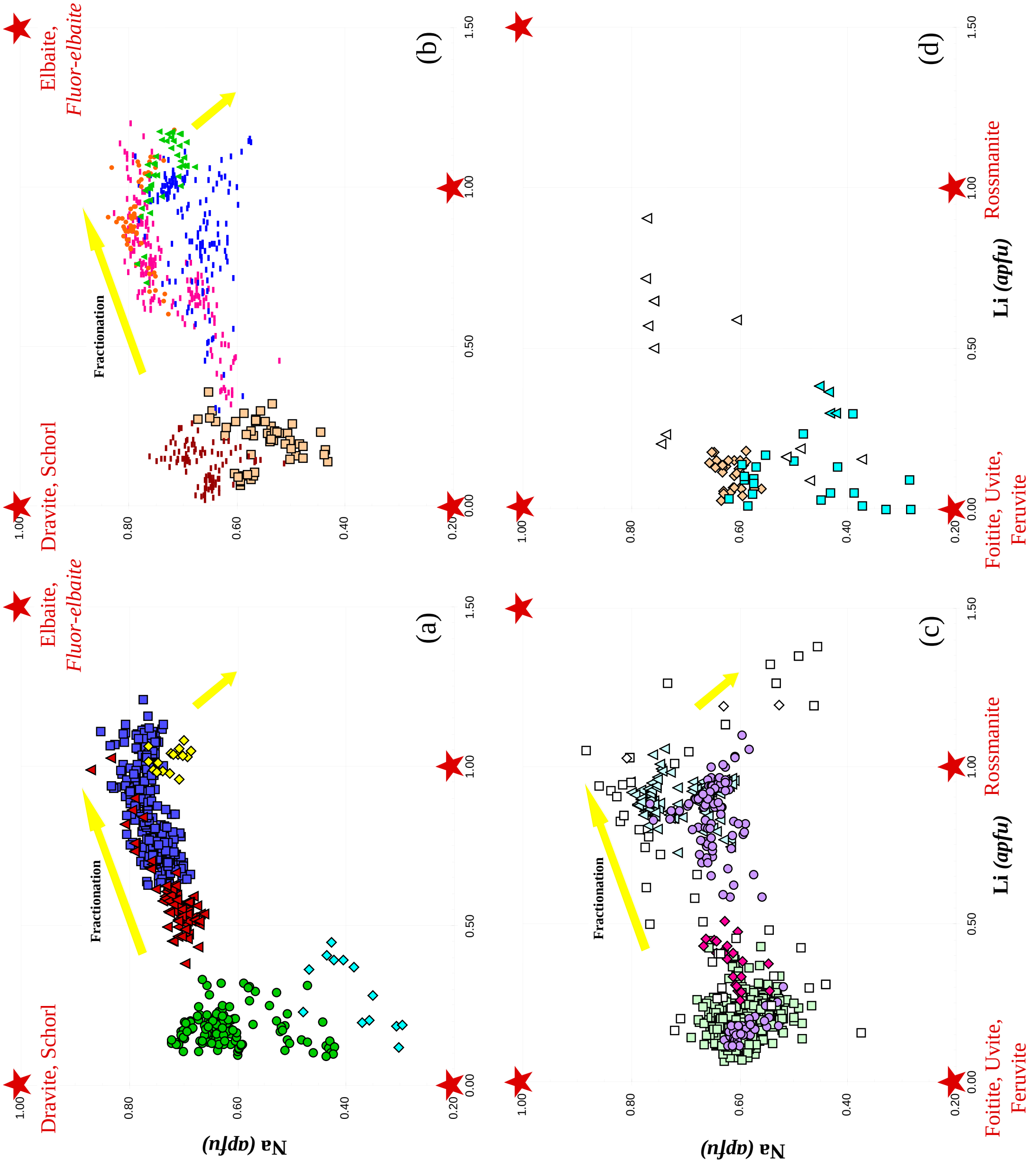

FIG. 10. Variation of Li *versus* Na (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4. Hypothetical composition in italics.

two schemes of substitution are thus operating on tourmaline from the Pakeagama pegmatite.

3. The NW pegmatite probably had a slightly different parental composition to the SE pegmatite zones as there are discontinuities in the Zn (and, to a lesser extent, Mn) data. This conclusion is supported by columbite–tantalite data, where the NW and SE pegmatite zones have Mn/(Mn + Fe) = 0.7 and in the range 0.8–0.95, respectively (Tindle & Breaks 2000a). The interpretation is complicated because Zn is expected to

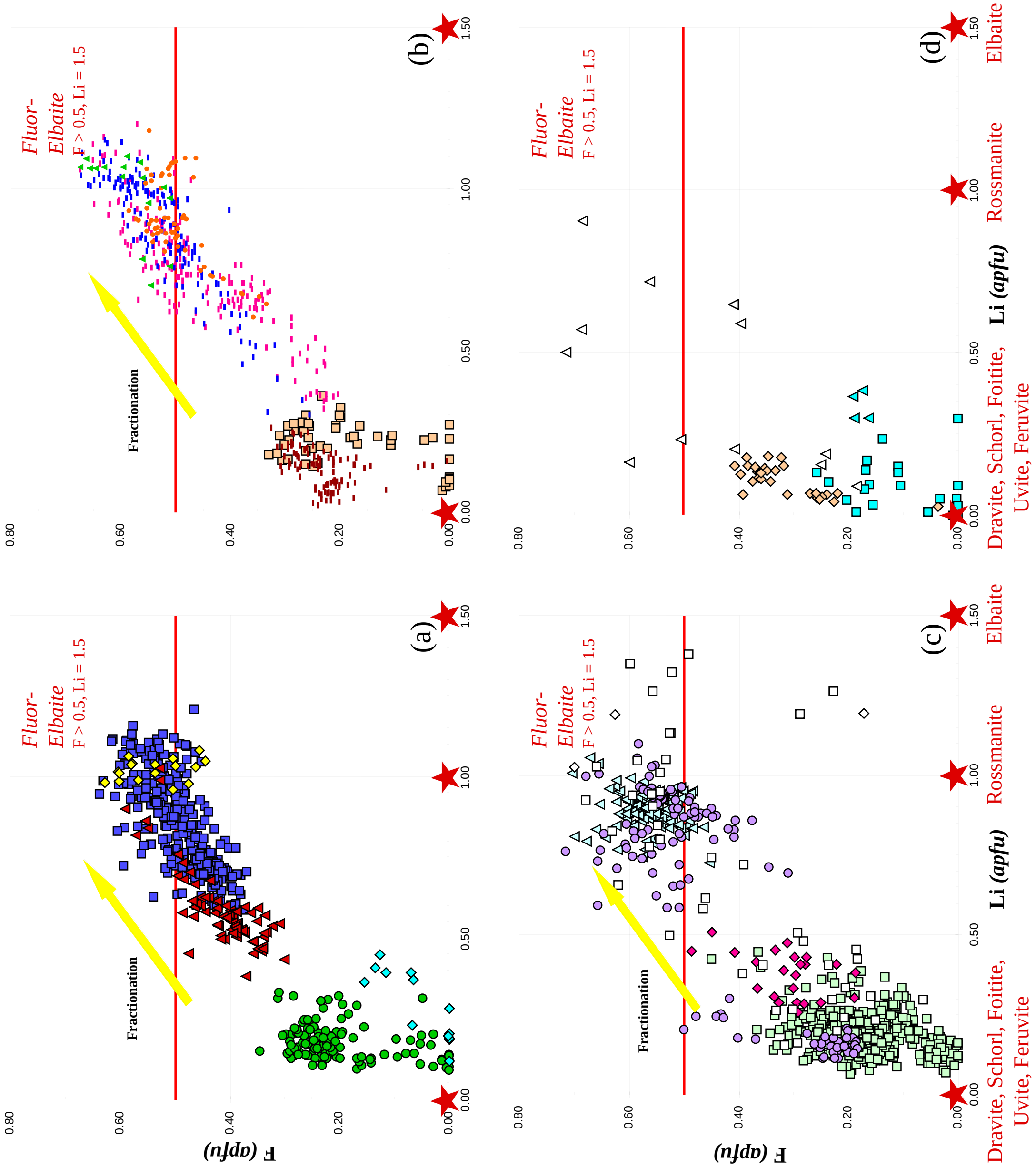

FIG. 11. Variation of Li *versus* F (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4. Hypothetical composition in italics.

reach its maximum abundance at an intermediate stage of fractionation and hence produce nonlinear trends (Jolliff *et al*. 1986).

4. Some tourmaline data from the Pakeagama Lake pluton (such as for Mn, Na and F) form continuous trends with data from the Pakeagama Lake pegmatite and could therefore represent precursor (or parental) compositions. However, discontinuities with other elements (*e.g.*, Fe^{2+} and Ca) preclude a simple link *via* fractionation. We propose some pluton – host-rock interaction.

5. Mafic exocontact and selvedge samples from in and around the Pakeagama Lake and Separation Rapids pegmatites have tourmaline compositions with elevated levels of Mg, Ti, and Ca, and depleted levels of Fe^{2+}, Mn, Zn, and Na, relative to the main pegmatite units at Pakeagama Lake. Their formation is due to the expulsion of boron-rich "fluids" from the pegmatite. This fluid phase reacted with (Mg,Ti,Ca)-rich host-rocks before tourmaline began to crystallize.

6. Tourmaline data from primitive vein samples (98–SS21B and 99–PKL12), which cut the Pakeagama Lake pluton, do not match any other vein or pegmatite compositions encountered in this study. On the basis of relatively high Fe^{2+} and low Li, we tentatively suggest that these samples may be derived from an earlier concentration of boron-rich "fluids" in the pluton prior to the generation of the Pakeagama pegmatite. Elevated Mg, Ti and Ca contents in tourmaline in these veins indicate they may also have exchanged fluids with mafic host-rocks.

7. Many other minor exocontact pegmatite and aplite intrusions (from thick dykes to thin veins) cutting the Pakeagama Lake pluton have essentially the same tourmaline compositions as the Pakeagama pegmatite and are considered to have a dominantly magmatic signature. Comparable Zn data suggest some were derived from the NW pegmatite, whereas others have compositions more like the SE pegmatite zones.

8. Metasomatic veins cutting the Pakeagama Lake pluton contain tourmaline in which there is an increase in Fe^{2+}, Ca, (+Ti, Mg), and a decrease in Mn, Zn and Na, relative to the Pakeagama pegmatite. We believe that interaction of pegmatite-forming melt (or, more likely, "fluids") with predominantly granitic host-rocks affected these samples. Marble from the Makataiamik assemblage is also a possible source for the Ca.

9. The pollucite-bearing vein that cuts the Pakeagama Lake pluton contains tourmaline whose composition closely matches that from the SE quartz-rich wall zone of the pegmatite, the zone from which it was most likely derived. Further enrichment in Cs is found (A.G. Tindle, unpubl. data) in associated rubidian K-feldspar (to 2700 ppm Cs), lepidolite (to 1.1 wt.% Cs_2O) and rare nanpingite (20 wt.% Cs_2O).

10. Albitized veinlets (98–FWB37) cutting the petalite – K-feldspar zone of the Pakeagama pegmatite contain tourmaline with high Li, Mn, Na, and F, and very low Fe^{2+}, Mg, and Ti contents. These compositions fall along the same trends defined by the quartz-rich wall zone and probably have an origin involving advanced *in situ* fractional crystallization of a pegmatite-forming melt followed by albitization.

11. Tourmaline in Group-1 Fe-suite pegmatites from the Separation Lake area share many characteristics with mafic exocontact and selvedge tourmaline. These include relatively low levels of Fe^{2+} and Mn, and high levels of Mg, Ti, and Ca. One sample from Marko's (Mn-suite) pegmatite, collected 2–5 cm away from BIF host-rocks, also has relatively high Mg, Ti and Ca. We interpret this occurrence as evidence that these samples have exchanged "fluids" with, or digested, host rocks such as amphibolite or banded iron-formation, and that elements released from these rocks became available for incorporation in tourmaline.

12. The Mn-suite pegmatites at Separation Lake (apart from the sample described in 11 above) contain tourmaline that closely mimics the magmatic pattern displayed by the Pakeagama Lake pegmatite. There is some indication (relatively high Ti and Ca), however, that even in these samples there has been some exchange with the host rocks.

13. Pegmatite 7, the Group-2 Fe-suite pegmatite, seems to be the only petalite-subtype pegmatite at Separation Lake to have a truly magmatic signature preserved in its tourmaline. For all elements (apart from Zn), it has an intermediate composition between uncontaminated Pakeagama Lake pluton and NW pegmatite compositions, although it is spatially unrelated to either.

14. The Tanco and Urubu granitic pegmatites are the only other petalite-subtype pegmatites for which tourmaline data are available (Selway *et al*. 2000b, Quéméneur *et al*. 1993). Both have tourmaline compositions that follow more closely the pattern of the Pakeagama Lake pegmatite than those from the Separation Rapids pegmatites. In a few samples from both Tanco and Urubu, tourmaline has been found with relatively high Mg and Ti and low Fe^{2+}, suggestive of interaction with the host rocks. Because of this, the Pakeagama Lake pegmatite is considered to be the most uncontaminated example of a petalite-subgroup pegmatite studied to date.

15. Micrometric veinlets of tourmaline cross-cutting earlier tourmaline from the two Mn-suite pegmatites in the Separation Rapids suite probably indicate a low-temperature hydrothermal event in which boron was either locally remobilized or came from fluids that evolved after melt saturation. Such tourmaline has low concentrations of Li, Mg, Ti, Mn, Zn, Ca, Na and F. Extensive *X*-site vacancies and Fe^{2+} close to 2 *apfu* characterize them as being of foitite composition.

All these observations have been used to construct Figure 12, which defines empirically the boundaries of magmatic and host-rock-dominated tourmaline compositions in petalite-subgroup pegmatites and separates them from those samples with complex origins. The area near the origin of Figure 12 is where either primitive magmatic tourmaline, or primitive tourmaline only slightly affected by pegmatite – host-rock interaction (or a combination) would plot.

Figures 12a, b and d place tourmaline from the Pakeagama Lake pegmatite, many of the minor intrusions, the pollucite-bearing vein and the albitized veinlets cutting the petalite – K-feldspar zone clearly in the magmatic field, and the mafic exocontact and selvedge material in the host-rock-dominated field. The Pakeagama Lake pluton data are considered to have a primi-

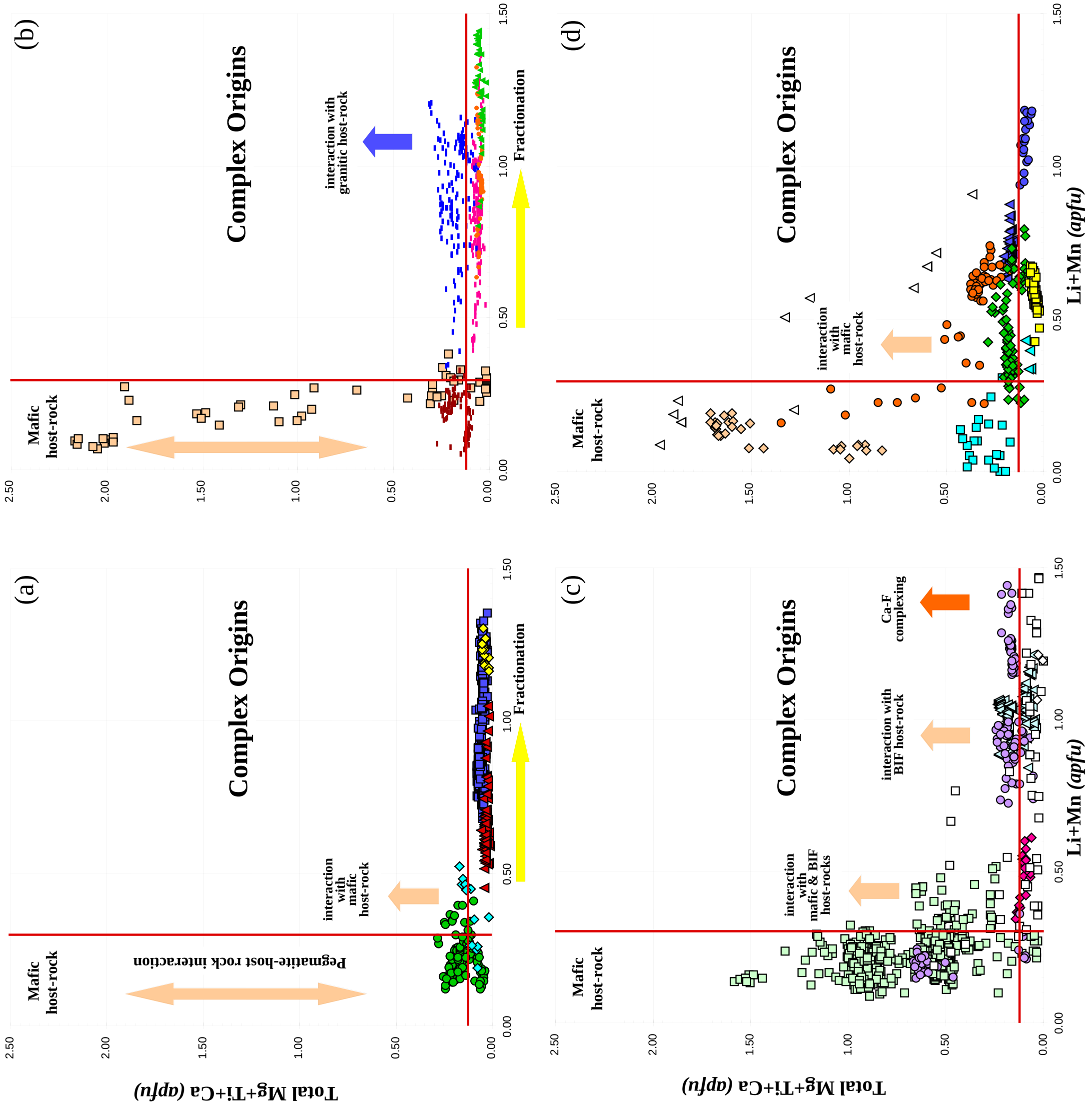

FIG. 12. Variation of (Li + Mn) *versus* (Mg + Ti + Ca) (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4. This diagram has been constructed to discriminate tourmaline data with a magmatic signature from data dominated by pegmatite – host-rock interaction. Mg = ^{Y}Mg + ^{Z}Mg. Boundaries have been set at Li + Mn = 0.3, total Mg + Ti + Ca = 0.14. Additional symbols on (d): Pakeagama Lake data: sample 99–FWB20 from the NW pegmatite, blue magmatic tourmaline (yellow squares), greenish brown tourmaline affected by pegmatite – host-rock interaction (dark green diamonds), brownish green tourmaline in selvedge (orange circles); pegmatitic vein 98–AT5, tourmaline rims are magmatic (dark blue circles) and cores are affected by pegmatite – host-rock interaction (dark blue triangles). Host rock is BIF for 99–FWB20 and granite for 98–AT5.

tive magmatic signature, but with a minor host-rock signature, whereas the metasomatic veins cutting the pluton are derived from a more evolved parental source that had also interacted to a limited extent with the host rocks (presumed to be granite).

Tourmaline from the Group-1 Fe-suite pegmatites at Separation Rapids is clearly affected by pegmatite – host-rock interaction, but the Group-2 Fe-suite pegmatite is not (Fig. 12c). Some data from the Group-1 Fe-suite pegmatites (predominantly rim compositions) cross into the complex origin field. These data are considered to have been strongly influenced by pegmatite – host-rock interaction, but once available Mg + Ti + Ca from the host rock was used up, tourmaline with a more typical magmatic signature began crystallizing.

The Separation Lake Mn-suite pegmatites are perhaps the most difficult group to classify as most of the data plot close to the magmatic fractionation – complex origin line on Figure 12c. Tourmaline data from Marko's pegmatite with Li + Mn < 0.5 *apfu* have elevated Mg, Ti and Ca contents clearly affected by pegmatite – host-rock interaction. Between Li + Mn > 0.5 and < 1.0 *apfu*, only Ti and Ca contents are elevated, and where Li + Mn exceed 1.0 *apfu*, it is only Ca and F that are elevated. As Marko's is one of the most strongly zoned pegmatites, this could mean that as fractionation progressed, migration of Mg, Ti and Ca and its availability for tourmaline formation were arrested at different times, perhaps because of falling temperature or because these elements were preferentially incorporated into other phases. A further possibility for those tourmaline compositions with elevated Ca and F contents alone is that they crystallized from a relatively Ca-rich melt. Late-stage Ca-enrichment in granitic melts is possible if Ca is stably incorporated in the melt structure as fluoride-bearing complexes. This relationship with F (as observed in tourmaline from Marko's pegmatite) means it is not available for plagioclase formation (Weidner & Martin 1987) and can be conserved until late in the crystallization sequence. Such late-stage Ca-enrichment has been proposed for the High Grade Dike, Manitoba (Teertstra *et al.* 1999) and the Tanco pegmatite (Selway *et al.* 2000b), but the process appears to be exceptionally rare, as it is not reported from other pegmatites in the same region (such as the Odd West, Buck, Pegli and Central Claim pegmatites: references in Teertstra *et al.* 1999). For evolved samples from Marko's pegmatite, the best explanation for data above the magmatic line in Figure 12c (Mg + Ti + Ca > 0.14 *apfu*) appears to be limited interaction with the host rock (where Li + Mn < 1.0 *apfu*) followed by Ca–F complexing (where Li + Mn > 1.0 *apfu*). Whatever the mechanisms involved, they also produce almandine–spessartine compositions that are Ca-enriched (to 0.11 *apfu*) in those evolved samples that also contain Ca-enriched tourmaline (A.G. Tindle, unpubl. data).

We cannot say that we fully understand the Ca-enrichment patterns in tourmaline from the Mn-suite pegmatites from the Separation Rapids pegmatites or from the metasomatic veins in the Pakeagama Lake pluton, but like Selway *et al.* (2000b) did for the Tanco pegmatite, we do favor a magmatic origin for the late-stage Ca-enrichment observed in the Pakeagama Lake SE pegmatite zones, the pollucite-bearing dyke and the albitized veinlets cutting the petalite – K-feldspar zone (Fig. 9).

It remains to be tested whether the boundaries on Figure 12 apply to tourmaline from other types of rare-element pegmatites, particularly those containing the Ca-rich tourmaline liddicoatite, but two complex examples from Pakeagama Lake are now used to test its applicability. Sample 99–FWB20 is one of a number of samples from the NW pegmatite differing only in the presence of a small fragment of selvedge. A single thin section was prepared that contained approximately a 2-cm strip of pegmatite and a 1-cm strip of selvedge, and all data pertain to this small volume of material. In the pegmatite portion of the sample, the tourmaline is deep blue in thin section and forms equant or stubby zoned crystals parallel to the selvedge contact. Approximately 2 mm from the selvedge contact, the grain size and abundance of the tourmaline increase, and its color becomes greenish brown (Fig. 3g). In the selvedge itself, tourmaline is abundant and approaches 50 modal %. It is fine-grained, brownish green in color and accompanied by holmquistite (restricted to a few tens of μm along the actual contact only), muscovite and quartz.

On Figure 12d, the blue tourmaline plots well within the magmatic field, whereas the greenish brown tourmaline from the pegmatite, adjacent to the selvedge, falls above the magmatic – complex origin boundary. Data from the selvedge itself fall in both the pegmatite – host-rock interaction field (typically core compositions) and the complex origin field (typically rim compositions). The sample therefore provides evidence of migration of boron-rich fluids from the pegmatite into the selvedge, and of localized migration of Mg + Ti + Ca from the selvedge down their concentration gradients into the margin of the pegmatite.

The second complex example (98–AT5; Figs. 3e, f) is from a vein not dissimilar to the metasomatic veins cutting the Pakeagama Lake pluton. Figure 13 shows a back-scattered-electron image of tourmaline from this sample. A lighter grey core (relatively high mean atomic number) in the grain on the right seems to be in the process of interacting with the darker rim composition. The crystal on the left has a more straightforward core–rim relationship, across which data from a 50-point traverse (10-μm intervals) have been collected. Results from the traverse, presented in Figure 14, show the core of the grain to be Fe-, Mg-, and Ca-rich, whereas the rim is Li- and Mn-rich. In terms of Figure 12d, core compositions plot in the complex origin field, and rim compositions plot in the magmatic field. Our interpretation is that the tourmaline initially crystallized from a pegmatite-forming melt (or fluid) locally contaminated with Mg + Ti + Ca from the host rocks, or contaminated by a small selvedge of host rock incorporated and digested by the melt or fluid. Once these elements were depleted, crystallization continued under essentially uncontaminated magmatic conditions.

SUBSTITUTION SCHEMES

Nomenclature, classification and an insight into the substitution mechanisms operating on the tourmaline

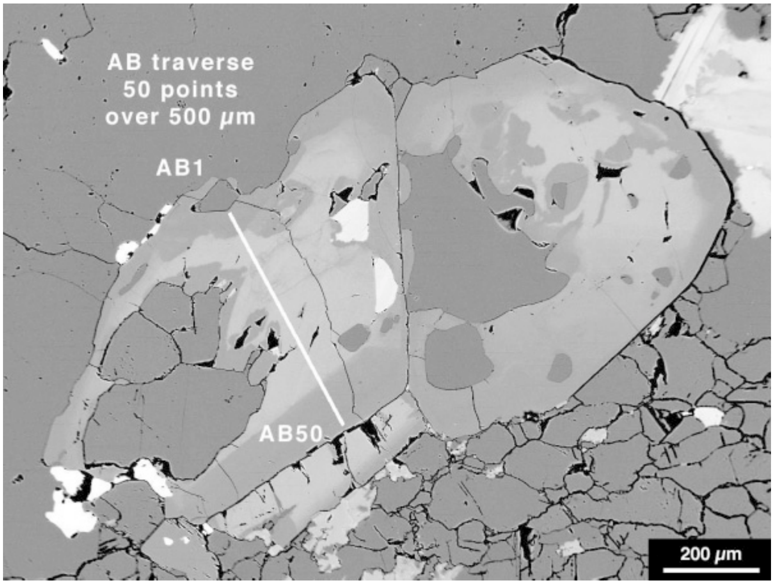

FIG. 13. Back-scattered electron image of two grains of tourmaline (schorl–elbaite) from pegmatitic vein 98–AT5. The right-hand grain shows evidence of interaction between an earlier (lighter grey: higher mean atomic number) core with a darker rim composition. The left-hand grain has more symmetrically zoned core and rim compositions. The line marks a 500-μm-long traverse along which the data of Figure 14 were collected. White regions are fluorapatite, other inclusions are quartz.

described here have benefitted greatly from the thorough systematic review of the tourmaline group by Hawthorne & Henry (1999). Although we were unable to follow through their suggestion of analyzing for H, Li, B and Fe^{3+}, we have attempted to follow this scheme as closely as possible. A further difficulty with the electron microprobe is that it is not possible to measure some components at the *W* site of tourmaline. Hawthorne & Henry (1999) recognized that tourmaline has hydroxyl-, fluor- or oxy-end members, and classification requires these to be known (or at least estimated). The scheme, although not IMA-approved, is most appropriate as it is the only one that comprehensively covers all the more recently discovered species of tourmaline, such as foitite (MacDonald *et al*. 1993), feruvite (Grice & Robinson 1989), and rossmanite (Selway *et al*. 1998b).

An initial division of tourmaline species is based on *X*-site occupancy (Hawthorne & Henry 1999). The majority of compositions considered here consist of alkali tourmaline because of dominant Na at this site. A few compositions of tourmaline have their *X* site mostly vacant; they occur in the Pakeagama Lake pluton, replacement compositions in the pluton, selvedge material entrained in the Pakeagama Lake NW pegmatite, Separation Rapids Group-1 Fe-suite pegmatites, and replacement veinlets in pegmatite 93–265 and Marko's pegmatite. No calcic tourmaline has been encountered in this study.

Figure 15 shows two of the dominant substitutions operating on our tourmaline dataset: Na ⇔ □ at the *X* site and Al ⇔ Fe at the *Y* site. All the *X*-site-vacant tourmaline data plot in the foitite field, whereas alkali tourmaline falls in the schorl or elbaite fields. Selway *et al*. (1999) have shown that tourmaline with >0.1 *apfu* Mg should not be plotted on this diagram, as those data normally belong to the dravite–schorl series (substitution of Fe^{2+} ⇔ Mg at the *Y* site) and cannot be represented correctly. From the Pakeagama and Separation Rapids pegmatites, over 800 tourmaline compositions have >0.1 *apfu* Mg, including samples from the Pakeagama Lake pluton, exocontact and selvedge material from the Pakeagama Lake and Separation Rapids pegmatites, veinlets of replacement tourmaline, Marko's pegmatite and, most importantly, the Separation Rapids Group-1 Fe-suite pegmatites. The data have been included on Figure 15 because virtually all of these data have compositions closer to the schorl end-member than to the dravite end-member, but Mg-rich tourmaline should not generally be plotted on this diagram.

Also on Figure 15 are three vectors (1 to 3) that closely follow those described for lepidolite-subgroup pegmatites (Selway *et al*. 1999) and for the Tanco

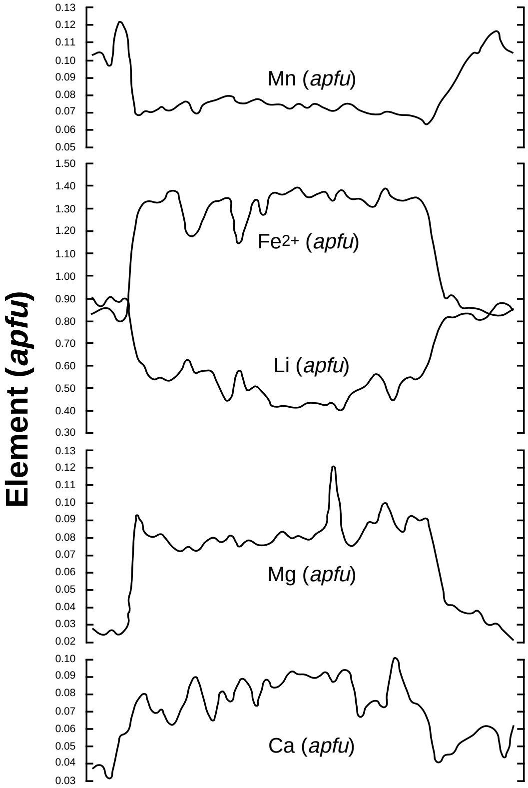

FIG. 14. Compositional variation in tourmaline along the 500-μm-long traverse line shown on Figure 13. Data were collected at 10-μm intervals and have been smoothed to highlight the main trends.

petalite-subgroup pegmatite (Selway *et al.* 2000b). Tourmaline from the only other petalite-subgroup pegmatite for which data are available (Urubu pegmatite, Minas Gerais, Brazil; Quéméneur *et al.* 1993), mainly falls along the elbaite–rossmanite trend (3). These vectors describe an evolutionary series from foitite to schorl to elbaite to rossmanite, corresponding to the sequence of earliest to latest zones, and represent increasing fractionation of the pegmatite-forming melt. They correspond to three solid-solution series: foitite–schorl (trend 1): ${}^{X}\square + {}^{Y}\mathrm{Al} \Leftrightarrow {}^{X}\mathrm{Na} + {}^{Y}\mathrm{Fe}^{2+}$, schorl–elbaite (trend 2): ${}^{Y}\mathrm{Fe}^{2+}_{3} \Leftrightarrow {}^{Y}\mathrm{Al}_{1.5} + {}^{Y}\mathrm{Li}_{1.5}$, and elbaite–rossmanite (trend 3): ${}^{X}\mathrm{Na} + {}^{Y}\mathrm{Li}_{0.5} \Leftrightarrow {}^{X}\square + {}^{Y}\mathrm{Al}_{0.5}$.

Tourmaline data from the Pakeagama Lake pegmatite and its postulated parent, the Pakeagama Lake pluton (Breaks *et al.* 1998), are presented on Figure 15a and in Table 3. As might be expected, the Pakeagama Lake pluton and compositions attributed to replacement have the most "primitive" foitite–schorl compositions, some of which approximate to trend 1. A larger grouping of data from the pluton lies within the schorl field and defines a subvertical trend close to and slightly crossing to the left of the schorl field. Tourmaline data from the NW pegmatite, which on the evidence of columbite–tantalite data is the earliest pegmatite zone to crystallize at Pakeagama Lake (Breaks *et al.* 1999a), plot at the beginning of trend 2 and varies from schorl to elbaite in composition. More evolved than this, but also falling along trend 2, are data from the quartz-rich wall zone of the SE pegmatite. The latter unit and the SE spodumene–quartz core zone are part of the second major magmatic pulse at Pakeagama Lake, and on Figure 15a they complete the crystallization sequence along trend 3. Tourmaline from the SE pegmatite is exclusively elbaite in composition, but with >0.5 *apfu* F at the *W* site (Fig. 11), many compositions should perhaps be referred to as "fluor-elbaite" (Hawthorne & Henry 1999). Unlike the case of Tanco (Selway *et al.* 2000b), compositions do not continue into the rossmanite field. Perhaps most significant from this diagram is the lack of continuity of tourmaline compositions from the pluton to the pegmatite.

Other tourmaline occurrences from the Pakeagama Lake area are illustrated on Figure 15b. Together they mimic the pattern described in Figure 15a, but differ in forming a "bent-trumpet-shaped" volume with a narrow "mouthpiece" end in the elbaite field, which progressively open up into the "horn" of the trumpet within and to the left of the schorl field. Significantly, the bulk of the selvedge and mafic host-rock samples fall at the "horn" end of the volume (schorl). We consider this "horn" to be evidence for interaction of pegmatite-derived "fluids" with mafic host-rocks. The double-ended arrow on Figure 15 therefore represents an exchange vector between a relatively Na-rich, Al-poor (and Mg-rich) host-rock component and a Na-poor, Al-rich magmatic component, with schorl–dravite substitution more dominant than foitite–schorl. The vector does not suggest higher Na contents in the host rocks, as Na in tourmaline is strongly chemically linked with F, and low F will depress the Na content of early-formed magmatic tourmaline. Crystal fractionation away from the variable host-rock-contaminated schorl composition leads to a narrowing of the trend towards elbaite ("fluor-elbaite") compositions as the magmatic component becomes dominant (most extreme in the albitized veins).

Tourmaline data from the Separation Rapids pegmatites display a pattern similar to that of tourmaline from the Pakeagama Lake pegmatite, with a few minor differences (Fig. 15c, Table 4). Most of the data are of schorl composition and were collected from seven (out of a total of 11) petalite-bearing pegmatites of the eastern subgroup (the Group-1 Fe-suite pegmatites). Together, data from these samples define a broad spread subparallel to the foitite–schorl "tram-lines" and with a similar composition to many of the Pakeagama Lake selvedge and host-rock samples. Only a few compositions fall in the foitite field, and there is little evidence

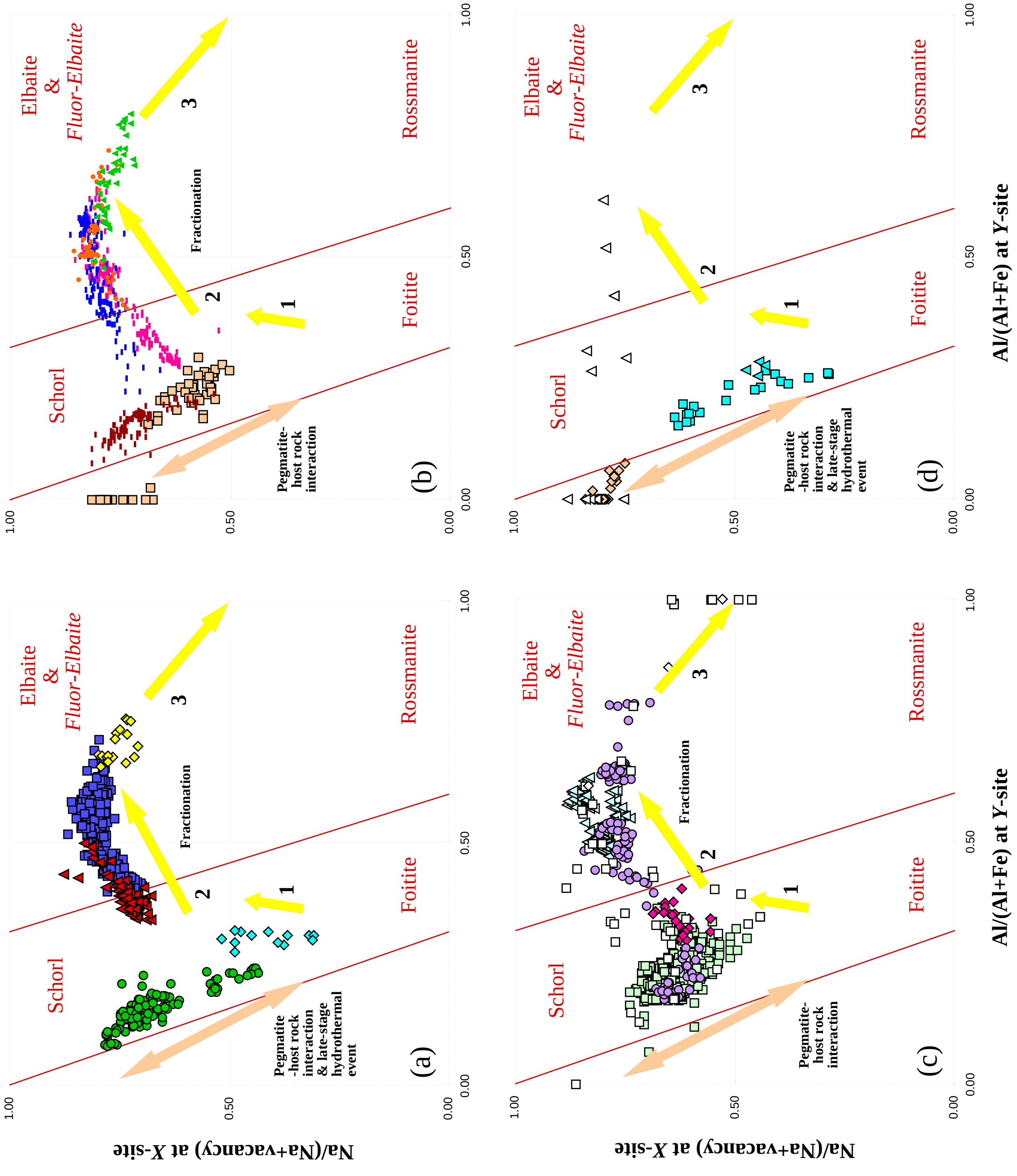

FIG. 15. Plot of Na/(Na + □) at the *X* site *versus* Al/(Al + Fe) at the *Y* site in tourmaline. For description of symbols, see caption to Figure 4. Yellow arrows closely mimic the direction of increasing fractionation, as determined from lepidolite-subtype pegmatites (Selway *et al.* 1999) and the petalite-subtype Tanco pegmatite (Selway *et al.* 2000b). The fawn, double-ended arrow broadly represents variation caused by pegmatite – host-rock interaction. Figure 15b, in particular, contains appreciable data with elevated Mg contents (dravite–schorl compositions) that cannot be represented correctly and that, under normal circumstances (*i.e.*, for classification purposes), would not be plotted here. Hypothetical composition in italics.

TABLE 3. COMPOSITION OF TOURMALINE FROM THE PAKEAGAMA LAKE PEGMATITE, VEINS AND EXOCONTACT ROCKS

	1	2	3	4	5	6	7	8	9	10	11	12	13
SiO_2	35.33	34.62	35.59	35.29	35.92	37.88	33.37	35.98	37.11	37.47	37.12	35.08	36.46
TiO_2	0.03	0.08	0.01	0.08	0.11	0.02	0.09	0.10	0.03	0.00	0.04	0.73	0.74
Al_2O_3	33.30	33.22	34.42	35.55	37.24	38.98	32.89	34.02	37.37	37.98	36.30	32.75	29.55
FeO	15.09	15.56	13.19	10.48	7.68	3.11	16.63	10.99	6.37	2.95	7.48	9.04	9.17
MgO	0.12	0.49	0.16	0.06	0.11	0.00	0.13	0.09	0.05	0.02	0.04	4.73	7.30
CaO	0.13	0.39	0.20	0.10	0.14	0.21	0.43	0.08	0.15	0.19	1.22	1.21	1.47
MnO	0.19	0.20	0.50	0.53	0.76	1.58	0.28	0.58	0.88	1.96	0.26	0.09	0.02
ZnO	0.27	0.35	0.12	1.27	0.60	0.37	0.64	1.28	0.19	0.25	0.12	0.16	0.01
BaO	0.00	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00
Na_2O	1.28	1.82	0.91	2.20	2.45	2.31	2.02	2.00	2.55	2.28	2.03	1.35	1.81
K_2O	0.01	0.04	0.01	0.02	0.03	0.01	0.03	0.03	0.02	0.02	0.02	0.02	0.05
F	0.03	0.27	0.00	0.67	1.15	1.00	0.41	0.52	1.02	1.16	1.14	0.63	0.00
Cl	0.00	0.00	0.01	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00
H_2O*	3.52	3.42	3.56	3.29	3.13	3.31	3.29	3.34	3.24	3.18	3.17	3.30	3.64
B_2O_3*	10.25	10.27	10.32	10.46	10.66	10.98	10.10	10.39	10.79	10.80	10.75	10.44	10.54
Li_2O*	0.18	0.17	0.28	0.70	1.11	1.67	0.05	0.72	1.41	1.70	1.55	0.27	0.15
Sum	99.74	100.90	99.26	100.73	101.10	101.44	100.36	100.12	101.18	99.97	101.23	99.81	100.90
O=F	0.01	0.11	0.00	0.28	0.48	0.42	0.17	0.22	0.43	0.49	0.48	0.27	0.00
Total	99.73	100.79	99.26	100.44	100.62	101.02	100.18	99.90	100.76	99.48	100.75	99.54	100.90
Structural formula based on 31 anions (O, OH, F)													
T Si	5.990	5.857	5.993	5.863	5.855	5.997	5.743	6.017	5.977	6.029	6.003	5.842	6.015
Al	0.010	0.143	0.007	0.137	0.145	0.003	0.257	0.000	0.023	0.000	0.000	0.158	0.000
B 3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
Z Al	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	5.745
Mg	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.255
Y Al	0.645	0.481	0.826	0.824	1.010	1.271	0.414	0.705	1.071	1.202	0.919	0.269	0.000
Ti	0.004	0.010	0.001	0.010	0.013	0.002	0.012	0.013	0.004	0.000	0.005	0.091	0.092
Mg	0.030	0.124	0.041	0.015	0.027	0.000	0.033	0.022	0.012	0.005	0.010	1.174	1.540
Mn	0.027	0.029	0.071	0.075	0.105	0.212	0.041	0.082	0.120	0.266	0.036	0.013	0.003
Fe^{2-}	2.140	2.201	1.858	1.456	1.047	0.412	2.393	1.537	0.858	0.397	1.012	1.259	1.265
Zn	0.034	0.044	0.015	0.156	0.072	0.043	0.081	0.158	0.023	0.030	0.014	0.020	0.001
Li*	0.122	0.114	0.188	0.466	0.727	1.061	0.028	0.483	0.914	1.099	1.006	0.176	0.099
ΣY	3.002	3.002	3.000	3.002	3.001	3.001	3.002	3.000	3.001	3.000	3.000	3.002	3.001
X Ca	0.024	0.071	0.035	0.018	0.024	0.036	0.079	0.014	0.026	0.033	0.211	0.216	0.260
Ba	0.000	0.000	0.000	0.001	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Na	0.421	0.597	0.296	0.709	0.774	0.709	0.674	0.648	0.796	0.711	0.636	0.436	0.579
K	0.002	0.009	0.002	0.004	0.006	0.002	0.007	0.006	0.004	0.004	0.004	0.004	0.011
□	0.553	0.324	0.667	0.268	0.195	0.253	0.240	0.331	0.174	0.252	0.148	0.343	0.151
OH	3.984	3.856	3.997	3.648	3.407	3.497	3.777	3.725	3.480	3.412	3.417	3.668	4.000
F	0.016	0.144	0.000	0.352	0.593	0.501	0.223	0.275	0.520	0.588	0.583	0.332	0.000
Cl	0.000	0.000	0.003	0.000	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Name	Foitite	Schorl	Foitite	Schorl	“Fluor-Elbaite”	“Fluor-Elbaite”	Schorl	Schorl	“Fluor-Elbaite”	“Fluor-Elbaite”	“Fluor-Elbaite”	Schorl	Dravite

Compositions are quoted in weight %, and structural formulae, in atoms per formula unit. *: Amount inferred from considerations of stoichiometry. Column headings: 1 Pluton, sample 99-FWB38A; 2 Pluton, sample Pak91-10; 3 Pluton (replacement), sample 99-FWB38A; 4 NW pegmatite, sample 99-FWB35; 5 SE quartz-rich wall zone, sample 99-FWB25; 6 SE quartz–spodumene core zone, sample 99-FWB46C; 7 Magmatic vein, sample 98-SS21B; 8 Magmatic vein, sample 99-FWB14B; 9 Pollucite-bearing vein, sample 99-SS128; 10 Albitized veinlet, sample 98-FWB37; 11 Metasomatic vein, sample 98-FWB28; 12 Exocontact with banded iron-formation, sample 99FWB15S; 12 Selvedge, sample 99-FWB24.

for trend 1. The start of trend 2 is best developed in pegmatite 7, the Group-2 Fe-suite pegmatite. A few compositions from the Group-1 Fe-suite also fall along trend 2, even though the bulk of their data mostly fall in the Group-1 Fe-suite field. Trend 2 is mainly defined by evolved samples from pegmatite 93–265 and Marko’s pegmatite, the Mn-suite pegmatites, whereas trend 3 is defined by data from Marko’s pegmatite alone. Like at Pakeagama Lake, the more evolved tourmaline from Separation Lake is “fluor-elbaite” rather than

TABLE 4. COMPOSITION OF TOURMALINE FROM SEPARATION RAPIDS PEGMATITES, ASSOCIATED INTRUSIONS AND EXOCONTACT ROCKS, AND PAKEAGAMA LAKE TEST SAMPLES

	1	2	3	4	5	6	7	8	9	10	11	12	13	14
SiO_2	35.70	35.75	35.57	36.75	34.40	35.40	35.80	35.59	35.48	35.69	34.86	35.99	36.62	37.24
TiO_2	0.68	0.38	0.08	0.02	0.81	0.15	0.03	0.03	0.39	0.10	1.13	0.59	0.13	0.12
Al_2O_3	31.90	33.83	36.02	36.77	32.88	38.20	39.20	34.00	29.42	35.83	33.95	32.89	35.61	36.70
FeO	8.41	11.36	10.64	7.12	12.57	7.22	2.56	14.76	11.18	9.65	12.26	8.85	8.54	6.17
MgO	5.70	1.93	0.26	0.05	1.77	0.04	0.00	0.82	5.43	0.08	0.50	4.87	0.30	0.15
CaO	0.48	0.21	0.11	1.02	0.51	1.11	1.20	0.05	1.38	0.04	0.11	0.42	0.45	0.33
MnO	0.15	0.26	0.89	0.72	0.27	1.17	2.79	0.04	0.10	0.32	0.23	0.01	0.53	0.75
ZnO	0.03	0.22	0.15	0.30	0.14	0.00	0.06	0.02	0.05	0.76	0.35	0.05	0.39	0.42
BaO	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00
Na_2O	1.99	1.63	2.02	2.09	1.74	1.88	1.94	1.19	1.81	2.39	2.04	1.78	2.26	2.37
K_2O	0.03	0.02	0.00	0.03	0.03	0.01	0.00	0.00	0.02	0.03	0.03	0.02	0.03	0.02
F	0.39	0.21	0.93	0.99	0.38	1.01	1.08	0.00	0.70	0.83	0.49	0.49	1.04	1.16
Cl	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00
H_2O*	3.42	3.50	3.20	3.24	3.35	3.22	3.21	3.58	3.21	3.23	3.35	3.40	3.17	3.16
B_2O_3*	10.45	10.44	10.54	10.74	10.25	10.72	10.78	10.38	10.26	10.51	10.37	10.53	10.63	10.75
Li_2O*	0.19	0.37	0.68	1.43	0.31	1.23	1.60	0.08	0.16	0.92	0.59	0.25	1.17	1.49
Sum	99.52	100.12	101.08	101.27	99.43	101.36	100.24	100.54	99.59	100.38	100.25	100.14	100.89	100.83
O=F	0.16	0.09	0.39	0.42	0.16	0.43	0.45	0.00	0.29	0.35	0.21	0.21	0.44	0.49
Total	99.36	100.03	100.69	100.86	99.27	100.93	99.79	100.54	99.30	100.03	100.05	99.93	100.45	100.34
Structural formula based on 31 anions (O, OH, F)														
T Si	5.936	5.950	5.867	5.946	5.832	5.739	5.775	5.959	6.008	5.903	5.842	5.942	5.991	6.021
Al	0.064	0.050	0.133	0.054	0.168	0.261	0.225	0.041	0.000	0.097	0.158	0.058	0.009	0.000
B	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000	3.000
Z Al	6.000	6.000	6.000	6.000	6.000	6.000	6.000	6.000	5.871	6.000	6.000	6.000	6.000	6.000
Mg	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.129	0.000	0.000	0.000	0.000	0.000
Y Al	0.188	0.586	0.869	0.958	0.401	1.037	1.227	0.669	0.000	0.888	0.548	0.341	0.856	0.993
Ti	0.085	0.048	0.010	0.002	0.103	0.018	0.004	0.004	0.050	0.012	0.142	0.073	0.016	0.015
Mg	1.413	0.479	0.064	0.012	0.447	0.010	0.000	0.205	1.242	0.020	0.125	1.199	0.073	0.036
Mn	0.021	0.037	0.124	0.099	0.039	0.161	0.381	0.006	0.014	0.045	0.033	0.001	0.073	0.103
Fe^{2-}	1.169	1.581	1.468	0.963	1.782	0.979	0.345	2.067	1.583	1.335	1.718	1.222	1.168	0.834
Zn	0.004	0.027	0.018	0.036	0.018	0.000	0.007	0.002	0.006	0.093	0.043	0.006	0.047	0.050
Li*	0.122	0.244	0.448	0.930	0.212	0.797	1.036	0.049	0.105	0.609	0.392	0.160	0.767	0.969
ΣY	3.002	3.002	3.002	3.001	3.002	3.001	3.001	3.002	3.001	3.001	3.002	3.002	3.001	3.000
X Ca	0.086	0.037	0.019	0.177	0.093	0.193	0.207	0.009	0.250	0.007	0.020	0.074	0.079	0.057
Ba	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.000
Na	0.642	0.526	0.646	0.656	0.572	0.591	0.607	0.386	0.594	0.766	0.663	0.570	0.717	0.743
K	0.006	0.004	0.000	0.006	0.006	0.002	0.000	0.000	0.004	0.006	0.006	0.004	0.006	0.004
□	0.267	0.432	0.335	0.161	0.329	0.214	0.186	0.605	0.151	0.220	0.311	0.352	0.197	0.196
OH	3.795	3.889	3.515	3.493	3.793	3.482	3.449	4.000	3.625	3.566	3.740	3.744	3.459	3.407
F	0.205	0.111	0.485	0.507	0.204	0.518	0.551	0.000	0.375	0.434	0.260	0.256	0.538	0.593
Cl	0.000	0.000	0.000	0.000	0.003	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.003	0.000
Name	Dravite	Schorl	Schorl	“Fluor-Elbaite”	Schorl	“Fluor-Elbaite”	“Fluor-Elbaite”	Foitite	Schorl	Elbaite	Schorl	Schorl	“Fluor-Elbaite'	“Fluor-Elbaite”

Compositions are quoted in weight %, and structural formulae, in atoms per formula unit. *: Amount inferred from considerations of stoichiometry. Column headings: 1 Separation Rapids pegmatites, Group 1, Fe-suite, Lou's pegmatite; 2 Group 1, Fe-suite, sample 94-80i; 3 Group 2, Fe-suite, pegmatite 7; 4 Pegmatite 93-265, Mn-suite, sample 93-265Nii; 5 Marko's pegmatite, Mn-suite, sample 96-AT20; 6 Marko's pegmatite, Mn-suite; 7 Marko's pegmatite, Mn-suite; 8 Replacement veinlet, sample 93-265-97-1B; 9 Selvedge, sample 97-71; 10 Pakeagama Lake test sample, 2 cm into pegmatite, sample 99-FWB20; 11 Pakeagama Lake test sample, 2 mm from selvedge, sample 99-FWB20; 12 Pakeagama Lake test sample, selvedge, sample 99-FWB20S; 13 and 14 Pakeagama Lake test samples, pegmatite vein, core and rim compositions, respectively, sample 98-AT5.

elbaite. The overall pattern is essentially identical to that of tourmaline at the Tanco pegmatite.

Finally, the two most evolved Separation Rapids pegmatites (93–265 and Marko's) contain elbaite and “fluor-elbaite” cross-cut by replacement veinlets of foitite–schorl 10–30 μm wide (Fig. 15d). Similar foitite–schorl compositions are reported from late-stage pockets in pegmatites at Dobrá Voda, Czech Republic (Novák & Taylor 1996) and interpreted as evidence of crystallization from late Fe-rich hydrothermal fluids. It is likely that compositions of replacement tourmaline from both Pakeagama Lake and Separation Lake areas (and illustrated in Figs. 4–12, 15, 16 with bright blue symbols) all have similar histories. However, the compositions are not identical, a reflection perhaps of the differences in primary compositions.

Tourmaline from petalite-subtype pegmatites is therefore capable of completing a geochemical loop of compositions starting with foitite, through schorl, elbaite and "fluor-elbaite" compositions, to rossmanite (only present at Tanco) and finally back to foitite and schorl. This circle of compositions is produced by a combination of pegmatite "fluids" entering the host rocks, host-rock contamination of pegmatite-forming melt, crystal fractionation and later infiltration of Fe-rich hydrothermal fluid into solidified pegmatite. The intensity of these individual processes varies among individual pegmatites.

Figure 16 illustrates the importance of the Fe^{2+}-for-Al substitution in our tourmaline compositions, with the majority of the data falling along a line corresponding to ${}^{Y}Fe^{2+}_{3} \Leftrightarrow {}^{Y}Al_{1.5} + {}^{Y}Li_{1.5}$, *i.e.*, the schorl–elbaite trend of Figure 15. The limited foitite–schorl and elbaite–rossmanite substitutions do not show up well on Figure 16, but the fourth major scheme of substitution, that of schorl–dravite (substitution of ${}^{Y}Fe^{2+}_{3} \Leftrightarrow {}^{Y}Mg^{2+}_{3}$), is evident off the main trend toward the dravite end-member.

Tourmaline from the Pakeagama Lake pluton and pegmatite are well correlated along the schorl–elbaite line (Fig. 16a), although a minor disturbance is observed toward the schorl end-member, where foitite–schorl substitution elevates some data from the pluton (and replacement compositions) to above the schorl–elbaite line. Data for tourmaline from minor intrusions from Pakeagama Lake, with few exceptions, also fall along the schorl–elbaite line (Fig. 16b), but most of the mafic exocontact and selvedge samples define a trend between the dravite end-member and an intermediate schorl composition with $Fe^{2+} = 2$ and ${}^{Y}Al = 0.5$ *apfu*. This trend is considered to be a mixing line between a Mg-rich source (mafic metavolcanic rocks, amphibolite or banded iron-formation) and unfractionated pegmatite-forming melt. A few data points from selvedge material entrained in the NW pegmatite at Pakeagama Lake and from selvedge at Separation Lake are not plotted on Figures 16b, d as they have excess Mg that place them in substitution schemes (schorl–uvite and schorl–feruvite) that cannot be represented satisfactorily on this diagram. Their composition will be discussed later.

Tourmaline data from the Separation Rapids Group-1 Fe-suite pegmatites are well correlated with data from mafic exocontact and selvedge samples from the Pakeagama Lake area (Figs. 15b, c, 16b, c). Again, we interpret this correlation as evidence for major interaction of pegmatite-derived "fluids" with host rocks to these pegmatites. Pegmatite 7 and the Mn-suite pegmatites at Separation Lake follow the main schorl–elbaite trend, which is consistent with little or no pegmatite – host-rock interaction. Data from Tanco and Urubu pegmatites show all the features of the Separation Rapids pegmatites, including poorly defined foitite–schorl substitution, well-developed schorl–elbaite and schorl–dravite substitutions; in addition, an elbaite–rossmanite substitution is well developed in the most evolved samples (Fig. 16c). Replacement veinlets in two Separation Rapids Mn-suite pegmatites fall close to or above the schorl–elbaite line despite being of foitite–schorl composition because both compositions contain elevated Al contents, and the diagram does not account for *X*-site contents (Fig. 16d).

Figure 17 shows a second pair of important substitutions operating on tourmaline in petalite-subtype pegmatites: Na ⇔ Ca at the *X* site and $Fe^{2+} \Leftrightarrow Mg$ at the *Y* site. Those samples with major foitite–schorl, schorl–elbaite and elbaite–rossmanite substitutions are not plotted on this diagram. What data remain are from three samples of exocontact and selvedge material from Pakeagama Lake, the sample of Marko's pegmatite collected 2–5 cm away from BIF host-rocks and, most importantly, virtually all of the Separation Rapids Group-1 Fe-suite pegmatites. The last two groups of data belong to the schorl–dravite series, although of all the pegmatites examined, only data from Lou's pegmatite fall in the dravite field. The remainder of the data have schorl–dravite or schorl compositions. Two samples of selvedge with Ca-rich schorl–dravite compositions have Mg at the *Z* site ($Al \Leftrightarrow Mg^{2+}$ substitution), indicating additional minor dravite–uvite (99–FWB24) and schorl–feruvite (97–71) substitutions, respectively. Tourmaline from exocontact sample 99–FWB15S (originally BIF) does not contain Mg at the *Z* site and, therefore, consists of schorl.

A further scheme of substitution operating on tourmaline in both Pakeagama Lake and Separation Rapids pegmatites involves ${}^{X}Na + {}^{Y}Li_{1.5} + {}^{W}OH \Leftrightarrow {}^{X}\square + {}^{Y}Mn^{2+} + {}^{Y}Al_{0.5} + {}^{W}O$ substitution and affects evolved samples from both areas (Fig. 18a). The most extreme Mn-rich composition is a Ca-rich (Mn, Fe)-bearing "fluor-elbaite" from the eastern end of Marko's pegmatite $(Na_{0.61}Ca_{0.20}o_{0.19})(Mn_{0.38}Fe_{0.35}Al_{1.23}Li_{1.02})Al_6(Si_{5.78}Al_{0.22}O_{18})(BO_3)_3OH_{3.45}F_{0.55}$, which potentially could give rise to the hypothetical end-member "oxy-Mn-foitite" (Hawthorne & Henry 1999) if more extreme compositions were found. This composition is quite distinct from "tsilaisite", a non-valid manganese tourmaline first described by Schmetzer & Bank (1984).

To understand Mn incorporation further, published data (Rossman & Mattson 1986, Burns *et al.* 1994, Aurisicchio *et al.* 1999, Morgan & London 1999, Novák *et al.* 1999, Teertstra *et al.* 1999, Akizuki *et al.* 2001) from Mn- and Ca-rich tourmaline have been plotted alongside subsets of our data in Figures 18a and 18b. It is apparent that no tourmaline continues towards "oxy-Mn-foitite" much beyond 0.5 *apfu* Mn; instead, Mn-enrichment continues toward Mn and Na = 1 *apfu* to

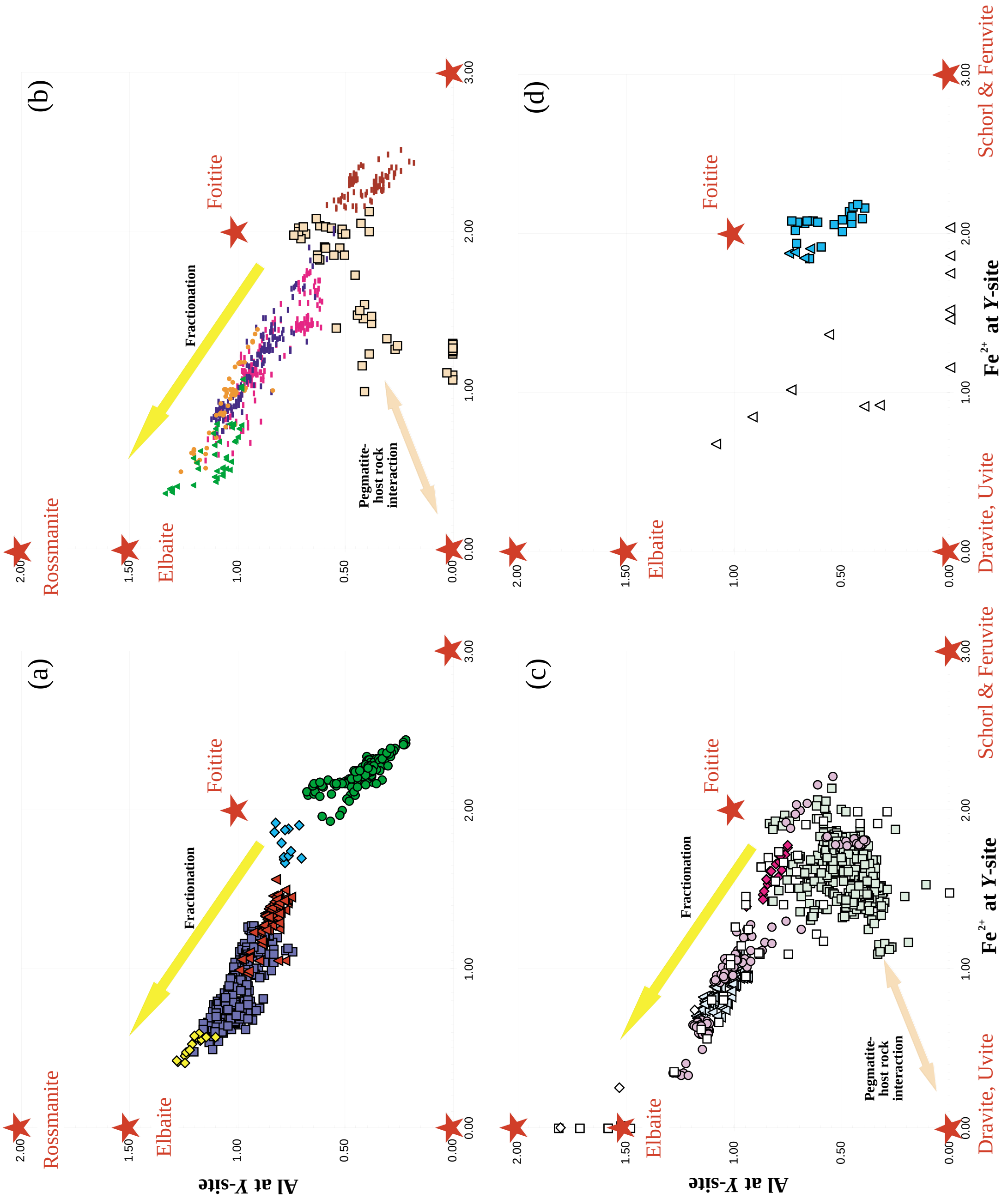

FIG. 16. Variation in Fe^{2+} *versus* Al at the *Y* site (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4. Data from selvedge samples 99–FWB24 and 97–71, which have Mg at the *Z* site corresponding to schorl–uvite and schorl–feruvite substitutions, are not plotted.

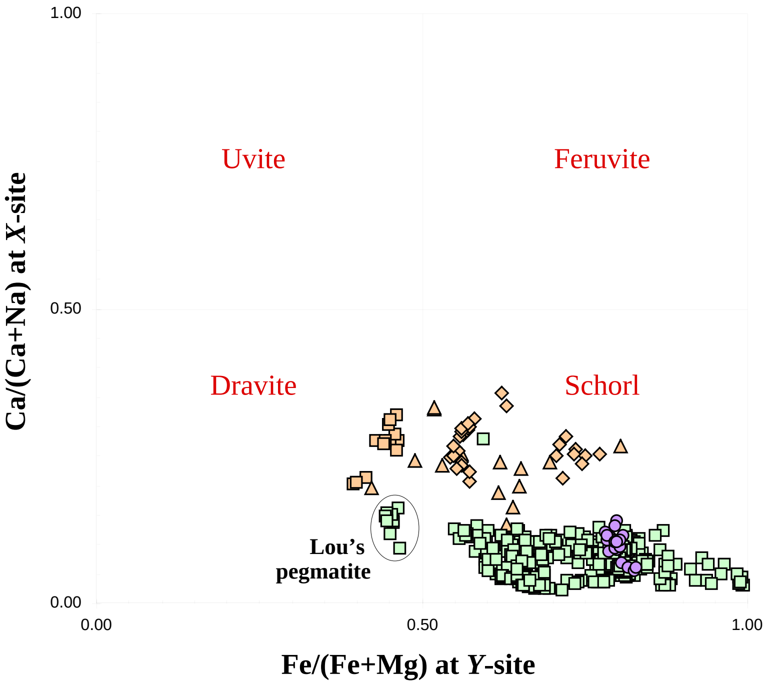

FIG. 17. Variation in $Fe^{2+}/(Fe^{2+} + Mg)$ at the *Y* site *versus* Ca/(Ca + Na) at the *X* site in tourmaline. Separation Rapids data; Group-1 Fe-suite pegmatites from the Eastern subgroup (pale green squares); Marko's pegmatite, sample collected 2–5 cm away from banded iron-formation (BIF) host-rocks (lilac circles); Ca-rich schorl–dravite in selvedge caught up in the pegmatite (97–71: fawn diamonds). From all pegmatites examined, only one (Lou's pegmatite, a Separation Rapids Group-1 Fe-suite pegmatite) has a dravite composition: Pakeagama Lake data; Ca-rich schorl in BIF (99–FWB15S: fawn triangles), Ca-rich dravite–schorl in selvedge (also originally BIF) caught up in the pegmatite (99–FWB24: fawn squares).

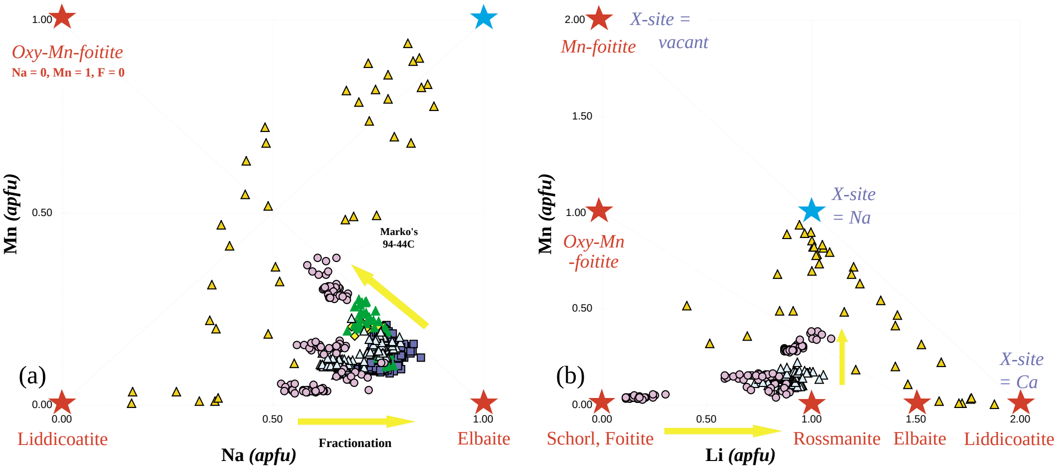

FIG. 18. (a) Variation in Na *versus* Mn (*apfu*) in tourmaline. (b) Variation in Li *versus* Mn (*apfu*) in tourmaline. For description of symbols, see caption to Figure 4. Additional symbol: orange triangles: Mn- and Ca-rich tourmaline compositions from the literature (Rossman & Mattson 1986, Burns *et al.* 1994, Aurisicchio *et al.* 1999, Morgan & London 1999, Novák *et al.* 1999, Teertstra *et al.* 1999, Akizuki *et al.* 2001). Yellow arrows represent fractionation vectors. Hypothetical compositions in italics. Blue star corresponds to the composition $Na(Mn,Li,Al)Al_6(Si_6O_{18})(BO_3)_3(OH)_3F$.

give a composition $Na(Mn,Li,Al)Al_6(Si_6O_{18})(BO_3)_3(OH)_3F$. Further evolution can only occur by replacing Na in the *X* site with Ca or by leaving it vacant. If the former, then Mn content is reduced and liddicoatite results; if the latter, then Mn content increases and "Mn-foitite" results (Fig. 18b). Currently there is no evidence for the hypothetical tourmaline end-member "Mn-foitite", but the liddicoatite – "Mn-foitite" substitution line gives support to its validity.

At Pakeagama and Separation Lake, tourmaline does not come very close to this substitution, mainly because appreciable Fe is still present at the *Y* site, which suppresses Mn-enrichment. If all the Fe were replaced with Mn, then much of the evolved tourmaline from these pegmatites would fall very tightly along the liddicoatite – "Mn-foitite" substitution line.

Table 5 summarizes the interpretations made for magmatic crystallization and pegmatite – host-rock interaction and matches these with the resulting schemes of substitution operating on tourmaline compositions. Data from the Pakeagama Lake and Separation Lake areas are then compared with data from the Tanco and Urubu pegmatites to give as full a picture as possible of the generality of these processes in petalite-subtype pegmatites.

CONCLUSIONS

Tourmaline data provide evidence that petalite-subgroup pegmatites, such as those at Pakeagama Lake and Separation Lake, do react with their host rocks, during which time Mg, Ti and Ca are introduced into the pegmatite-forming melt primarily from adjacent mafic metavolcanic, amphibolite or banded iron-formation host-rocks. Such reaction can occur by the digestion of fragments of host rock (which if arrested leaves a metasomatic selvedge behind). There is also fluid exchange in the opposite direction, with Li, B and Al all being expelled from the crystallizing pegmatite into the host rocks, which produces metasomatic exocontact rocks enriched in dark brown or black schorl, holmquistite, muscovite and quartz. "Biotite" enriched in Rb and Cs accompanies this assemblage in the Separation Rapids pegmatites.

The influence of pegmatite – host-rock interaction is quite variable, but has been observed in certain units from both Pakeagama Lake and Separation Lake areas, as well as from Tanco (Manitoba), and Urubu (Brazil). Of these, the Pakeagama Lake pegmatite is the most closed to pegmatite – host-rock interactions. Instead, the magma fractionated *in situ* and gave up very little of its rare-element content to its host rocks. This factor has economic implications, since the association of the SE pegmatite with a pollucite-bearing vein could imply a buried Cs deposit nearby. Tourmaline data from veinlets at Pakeagama Lake suggest that late-stage albitization is a continuation of the *in situ* fractionation process. It is only in the metasomatic veins cutting the Pakeagama pluton, that limited pegmatite – host-rock interaction occurred, leaving behind Ca-enriched tourmaline. The source of the Ca was either the pluton itself or, less likely, marble from the Makataiamik assemblage of the North Spirit Lake greenstone belt.

At Separation Lake, the story is very different, and the majority of petalite-subgroup pegmatites containing tourmaline (*i.e.*, most of the Fe-suite) appear to have been open to pegmatite – host-rock interactions. In a few cases, Ca-enrichment in tourmaline from mafic exocontact and selvedge samples gave rise to minor schorl–uvite and schorl–feruvite substitutions in addition to schorl–dravite substitution. Even in the more evolved Mn-suite pegmatites, there is some indication that pegmatite – host-rock interaction occurred, although late-stage Ca-enrichment in the melt due to fluoride complexing complicates the interpretation of these rocks.

The evolution of tourmaline compositions in the Pakeagama Lake pegmatite follows a pattern similar to those observed in more evolved lepidolite-subtype pegmatites and involves schorl – elbaite (– "fluor-elbaite") (Fig. 15, trend 2) and elbaite (– "fluor-elbaite") – rossmanite (trend 3) substitutions (although rossmanite itself is absent). In tourmaline from the Separation Rapids pegmatites (and Tanco), schorl–dravite substitution is encountered in the most primitive samples (most of the Fe-suite pegmatites), and this is interpreted as a pegmatite – host-rock interaction signature. With increased fractionation, schorl – elbaite (– "fluor-elbaite") then elbaite (– "fluor-elbaite") – rossmanite substitutions follow in the Mn-suite pegmatites. Tanco is the only petalite-subtype pegmatite examined so far in which foitite–schorl substitution is clearly established (trend 1) and in which extreme fractionation led to rossmanite crystallization.

Late replacement-induced crystallization of (Fe-rich) foitite in the Separation Rapids pluton and Mn-suite pegmatites is more likely due to a hydrothermal event that occurred long after the pegmatites consolidated.

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TABLE 5. SUMMARY OF PEGMATITE – HOST-ROCK INTERACTIONS AND SUBSTITUTION SCHEMES OPERATING IN TOURMALINE FROM PETALITE-SUBTYPE GRANITIC PEGMATITES

	1	2	3	4	5	6	7	8	9	10	11
Pakeagama Lake area											
Pakeagama pluton srl with <0.2 *apfu* Mg	••	•••					••••	•			
Pakeagama pluton replacement compositions foi	•••	••		•••••				•••••			
NW pegmatite srl–elb to elb–srl		•••••							••••		
SE pegmatite qtz-rich wall zone elb-srl with < 1.5 *apfu* Fe		•••••	•						••••	•	•
SE pegmatite qtz-spd core, elb		•••••	•						•••	••	•
Primitive magmatic veins 98-SS21B, 99-PKL12, srl	••	•••						•	••••		
Magmatic veins elb-srl		•••••	•						•••	••	
Pollucite-bearing vein 99-SS128, elb-srl		•••••	••						•••	••	
Albitized veinlets 98-FWB37, elb		•••••	••						•••	••	
Metasomatic veins, elb-srl	•••••								••••	•	
Exocontact, banded iron-formation, 99-FWB15S srl-drv	•••••						•••••				
Selvedge, 99-FWB24, drv	•••••				•		•••	•			
Separation Lake area											
Group 1 Fe-suite pegmatites srl-drv	••••	•					••••	•			
Group 2 Fe-suite pegmatites srl-elb		•••••									
Pegmatite 93-265 Mn-suite elb	•	••••	•						•••••		••
Marko's pegmatite Mn-suite srl to elb-srl to elb	•	••••	•						•••	••	•••
Marko's pegmatite near BIF contact, srl	•••••										
Replacement veinlets in Mn-suite, 93-265-97-1B. foitite-srl to srl	•••••			•••••				•••••			
Selvedge, 97-71, srl-drv	•••••					•	••••				
Tanco, Manitoba (data from Selway *et al.* 2000 a, b)											
Unit 10, srl-elb	••	•••				•		••	••		
Unit 20, srl-elb	•	••••	••					••	•••		•
Unit 30, elb		•••••	•						•••••		•
Unit 40-50, elb-rsm		•••••	•••••						•	••••	•••
Unit 60, foi-srl to srl		•••••						•••••			
Exocontact, fer-srl to srl-drv	•••••					•	••••				
Uruba, Minas Gerais, Brazil (data from Quéméneur *et al.* 1993)											
Pegmatite, elb-rsm		•••••							•	••••	
Exocontact, srl-drv	•••••						•••••				

Relative importance is indicated by the number of dots. Symbols used: drv dravite, elb elbaite, fer feruvite, foi foitite, qtz quartz, rsm rossmanite, spd spodumene, srl schorl. Column headings: 1 Pegmatite – host-rock signature, 2 Magmatic signature, 3 Late-stage magmatic Ca–F complexing, 4 Late hydrothermal event, 5 Schorl – Uvite, 6 Schorl – Feruvite, 7 Schorl – Dravite, 8 Foitite – Schorl, 9 Schorl – Elbaite, 10 Elbaite – Rossmanite, 11 "Oxy-Mn-Foitite" ("Mn-foitite").

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