

Significance of Tourmaline-Rich Rocks in the Grenville Complex of St. Lawrence County, New York

U.S. GEOLOGICAL SURVEY BULLETIN 1626-C



Chapter C

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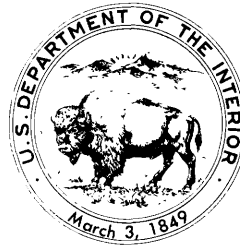
By C. ERVIN BROWN and ROBERT A. AYUSO

A study of tourmaline compositions
and their possible use as indices
of mineralization

U.S. GEOLOGICAL SURVEY BULLETIN 1626
CONTRIBUTIONS TO THE GEOLOGY OF MINERAL DEPOSITS

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CONTENTS

Abstract	C1
Introduction	C1
Purpose of study	C2
Stratigraphic and lithologic setting	C2
Petrography and summary of the bulk composition of tourmaline-bearing rocks	C3
Sampling	C7
Analytical procedure	C7
Results of compositional studies	C11
Tourmaline end members in nature	C11
Compositional variation of tourmaline in North Gouverneur area	C11
Calcium content of tourmalines	C20
Substitutions in hypothetical structural formula	C20
Mineralized areas and tourmaline compositions	C22
Discussion	C25
Comparison with the Balmat-Edwards district	C29
Tourmaline genesis in the North Gouverneur area	C29
Terranes showing most favorable implications for containing stratabound sulfide deposits	C31
References cited	C31

FIGURES

1. Index map of St. Lawrence County, N.Y. C2
2. Generalized geologic map showing sample localities for this study C4
3. Photographs of tourmalinite, brecciated tourmalinite, granular patches of
 black schorl, and schorl prisms C6
4. Photomicrographs of granoblastic tourmalinite and tourmaline-bearing
 rock C7
5. Distribution of selected elements in rocks from the North Gouverneur area,
 New York C8
6. $\text{TiO}_2\text{--FeO}/(\text{FeO} + \text{MgO})$ plot for tourmalines in lithologic units from North
 Gouverneur, N.Y., compared with those from Sullivan Mine, British
 Columbia C19
7. Ternary plot of $\text{Na}_2\text{O--MgO--FeO}$ for tourmaline from North Gouverneur,
 N.Y., and the Sullivan Mine, British Columbia C20
8. $\text{CaO--Al}_2\text{O}_3$ plot for tourmalines from North Gouverneur, N.Y., compared
 with those from the Sullivan Mine, British Columbia C21
9. $\text{Al}_2\text{O}_3\text{--FeO}/(\text{FeO} + \text{MgO})$ plot for tourmalines from North Gouverneur,
 N.Y. C22
10. $(\text{Na} + \text{Fe})\text{--Al}$ plot for tourmalines from North Gouverneur, N.Y., compared
 with those in rocks from Sullivan Mine, British Columbia C23
11. $(\text{Na} + \text{Ca})\text{--Al}$ plot for tourmalines from North Gouverneur, N.Y., compared
 with those in rocks from the Sullivan Mine, British Columbia C24
12. $(\text{Na} + \text{Mg} + \text{Al})\text{--Ca} + 2\text{Fe}$ plot for tourmalines from North Gouverneur area,
 New York, compared with those from Sullivan Mine, British Columbia C25
13. Ca--Na plot for tourmalines from North Gouverneur, N.Y., compared
 with those from Sullivan Mine, British Columbia C26
14. $\text{Al}_2\text{O}_3\text{--Na}_2\text{O}/(\text{Na}_2\text{O} + \text{CaO} + \text{K}_2\text{O})$ plot for tourmalines from North Gouverneur,
 N.Y., compared with those from the Sullivan Mine, British Columbia C27

TABLES

1. Summary of petrographic features of tourmaline-bearing rocks from the North Gouverneur area, New York **C10**
2. Electron microprobe analyses and structural formulas of tourmalines in the North Gouverneur area, New York **C12**
3. Chemical analyses of selected tourmaline-bearing rock types in the North Gouverneur area, New York **C17**
4. Semiquantitative spectrographic analyses of tourmaline-bearing rocks in the North Gouverneur area, New York **C17**
5. Semiquantitative spectrographic analyses of tourmaline in Beaver Creek area, New York **C18**

Significance of Tourmaline-Rich Rocks in the Grenville Complex of St. Lawrence County, New York

By C. Ervin Brown and Robert A. Ayuso

Abstract

Feldspathic quartzite and metapelite of Middle Proterozoic age north of Gouverneur, N.Y., contain abundant dravite-uvite (magnesian tourmaline). These rocks, more than 1,000 feet thick, are regionally metamorphosed to the upper amphibolite facies, are pyritic, and locally contain porphyroblastic scapolite. The rocks are underlain by talc-tremolite schist and a thick sequence of calcitic and dolomitic marble, and are overlain by quartz, calc-silicate carbonate rock, and a pyroxene scapolite unit. All rocks are part of the metasedimentary Grenville Complex.

Individual tourmaline-rich layers in the quartzite are as thick as 3 cm and contain as much as 50 percent fine-grained, brownish-green dravite. Subjacent talc schist contains fine-grained amber dravite, and silicated marble locally contains brown porphyroblasts of uvite. Gneissic granite, pegmatite, and migmatitic segregations cutting the tourmaline-bearing quartzite, gneisses, and schists are rich in black schorl.

Tourmalines from the tourmaline-rich quartzite and other metasedimentary and metaigneous rocks were analyzed by means of an electron microprobe. The compositions of tourmalines in the North Gouverneur area are clearly a function of the bulk composition of the rock. Tourmaline compositions in the quartzite have a wide range in $\text{FeO}/(\text{Fe}+\text{MgO})$ ratios, from 0.15 to 0.58, which are distinct from the ratios for tourmalines in granitic and pegmatitic rocks that range from 0.55 to 0.75. Tourmalines in the area have Na_2O contents that range from 0.85 to 4.25 weight percent. Aluminum in all tourmalines ranges widely from about 25 to 37 weight percent Al_2O_3 . The most heterogeneous tourmaline compositions are in the quartzites, although most compositions cluster in the dravite-uvite solid solution series. Substitutions involving Na, Ca, Mg, Fe, and Al result in the following compositional schemes: $\text{Na}+\text{Fe}=\text{Al}+\text{vacancy}$, and $\text{Na}=\text{Ca}$. Such coupled substitutions probably represent valid constraints for tourmalines from North Gouverneur.

The abundance of magnesian tourmaline in the metasedimentary rocks of the North Gouverneur area is comparable to that of tourmaline-rich rocks associated with sediment-hosted massive sulfide deposits—for example, Sullivan Mine, British Columbia. A recently described large lead-zinc deposit at Dugald River, Australia, is associated with tourmaline- and scapolite-bearing rocks similar in many respects to those north of Gouverneur. Compared with known tourmaline compositions in the Sullivan Mine area, tourmalines from North Gouverneur have comparable FeO and MgO values. We suggest that other tourmaline components

such as a low TiO_2 content, variable $\text{Na}/(\text{Na}+\text{Ca}+\text{K})$ ratios, and the antipathetic correlation of Na and Ca might also be useful in discriminating tourmalines associated with ore deposits.

Ore bodies at the nearby Balmat-Edwards mining district are in marbles associated with rocks of evaporitic origin. The boron- and scapolite-rich rocks under consideration in this study possibly also are related to an evaporite-producing environment. Similar rocks that have an evaporite origin are associated with stratabound sulfide deposits at many places.

INTRODUCTION

Tourmaline is a common accessory mineral in rocks of the Grenville Complex in the Grenville Lowlands of St. Lawrence County, N.Y. It occurs disseminated in a variety of metasedimentary rocks ranging from marble to feldspathic gneiss, and also in pegmatitic segregations and granites that intrude the tourmaline-bearing metasediments. Although tourmaline is normally present only as an accessory mineral, it is very abundant in certain rock units in belts mainly in the Richville 7.5-minute quadrangle (fig. 1). Cushing and Newland (1925, p. 24–25) called attention to rocks, particularly along the Oswegatchie River west and north of Richville, N.Y., that contain 35 to 50 percent tourmaline. Such rock units are lithostratigraphic sequences consisting of feldspathic quartzite and quartz-feldspar-mica granofels having layers that contain as much as 50 percent fine-grained tourmaline, and are classified as “tourmalinite.”

In 1966 Harold M. Bannerman (1972) mapped the belt of microcline-tourmaline-quartz gneiss along the Oswegatchie River (fig. 2), where at many outcrops the rocks are spectacularly brecciated in a zone of intense faulting. In an attempt to determine the significance of these tourmaline-rich rocks, and as an aid to future research, Bannerman had several chemical analyses made of rocks from this zone (Brown, 1980, analyses 45 through 71) and indicated the locations of tourmaline-rich rocks on his map of that area (Bannerman, 1972). The present study was in part instigated by his concern about the significance of these rocks, and by his encouragement that further research be done.

Subsequent geologic mapping by C. E. Brown in an area a few miles to the northwest of the Oswegatchie



Figure 1. Index map of St. Lawrence County, N.Y., showing location of the Richville 7.5-minute quadrangle.

River revealed a second belt of tourmaline-rich rocks between Beaver Creek and its South Fork (fig. 2). This area is mainly in the low-dipping upper limb of a detached recumbent antiform or nappe. Stratigraphic relations in the upper limb of this fold are not disrupted by faulting, and, because of the low dip, the stratigraphy is more easily deciphered than in the faulted area along the Oswegatchie River. Although separated by folds and faults, the stratigraphic column in the nappe is similar to that along the Oswegatchie River. The two principal differences are that (1) the sequence along the Oswegatchie at most places is in inverted order—believed by Brown to be structurally overturned—and (2) rocks in the nappe area were intruded by one large and many satellitic granite sheets, which transect the rock sequence at a low angle and which locally produced migmatite (Brown, 1969).

Purpose of Study

Studies of the Sullivan Mine area in British Columbia by Ethier and Campbell (1977), the Black Hawk Mine in Maine by Slack (1980), and the Elizabeth Mine in Vermont by Slack and Annis (1981) found that tourmaline-rich rocks are associated with—and probably genetically related to—the mineralized rock of stratabound sediment-hosted sulfide deposits. The tourmalines in these deposits are dravitic (and rarely uvitic)

in composition and are believed to be the result of boron and magnesium introduced syngenetically or diagenetically by hydrothermal solutions. Because of these relationships, Ethier and Campbell (1977) and Slack (1980, 1982) suggested the use of tourmaline as a prospecting guide for stratabound sulfide deposits.

Brown (1983) speculated that the anomalously abundant tourmaline in the vicinity of North Gouverneur indicated depositional conditions possibly related to massive base-metal sulfides. The study reported here was undertaken to determine the composition of tourmalines in the metasedimentary and metaigneous lithologies in the Grenville lowlands, and to compare these tourmaline compositions with those from mineralized districts for which chemical data are available. Unfortunately, there is a general lack of published chemical analyses of tourmalines associated with ore deposits, especially stratabound sulfides in evaporitic terranes. Our study documents the range in the composition of tourmaline found in rocks of different bulk compositions in a possible evaporitic setting, but because of the absence of published analyses of tourmaline in stratabound deposits within evaporitic terranes, our comparisons rely for the most part on tourmaline compositions from the Sullivan Mine, a turbidite-hosted massive sulfide deposit in British Columbia. These comparisons and the geologic setting of the study area allow speculation on the genesis of the tourmaline-rich rocks and their possible economic implications (Brown and Ayuso, 1982).

STRATIGRAPHIC AND LITHOLOGIC SETTING

The metasedimentary rocks of the Grenville Complex under consideration have undergone multiple folding, were regionally metamorphosed to the upper amphibolite facies, and contain sillimanite. Layering and foliation in them are essentially parallel to the lithologic boundaries and thus are believed to be nearly parallel to bedding at most places, although no primary sedimentary features have been recognized.

Because the stratigraphic section is more intact in the upper limb of the nappe north of North Gouverneur (fig. 2), the following description is of those rocks. The lowermost rock units constitute a thick sequence of marble labeled *mm* and *qm* (fig. 2) that is increasingly more siliceous and dolomitic upward, culminating in rocks that are now tremolitic quartzite and talc-tremolite schist containing minor amounts of anthophyllite (*tt*) (Brown, 1969). If essentially isochemical reactions during metamorphism are assumed (with loss of CO_2), the tremolitic rocks can be inferred to represent a protolith of quartz-rich dolomite and magnesite, an assemblage suggesting an evaporitic environment during deposition. Overlying these carbonate and cal-

cium-magnesium silicate lithologies are fine-grained clastic (noncarbonate) rocks that grade upward crudely from quartzite to biotite-muscovite-quartz schists (*qmt*). This upward fining gradation supports the assumption that the section here is not overturned. These rocks are oligoclase or microcline-rich quartzites interlayered with quartz-microcline-mica granofels, locally very micaceous. Porphyroblasts of muscovite, scapolite, and sillimanite are locally present in the micaceous rocks. Tourmaline occurs throughout, but is abundant only in the quartzite. The quartzite, present mainly in the lower part of the sequence, has layers as thick as several centimeters containing as much as 50 percent fine-grained stubby, subhedral tourmaline crystals less than 0.1 mm across (fig. 3A and 4A,B). Most rock types in this unit also contain pyrite, and many outcrops are intensely limonite-stained by the weathering of pyritic zones and local massive pyrite concentrations (Brown, 1983, loc. 57). Spectrographic analyses (C. E. Brown, 1969, unpub. data) of several rock specimens from this unit proved that some of the feldspathic rocks have anomalous amounts of barium probably occurring as a barium feldspar, although none has yet been identified (fig. 5).

Overlying unit *qmt* are quartz-rich, graphitic, dolomitic, and pyrrhotite-rich carbonate and calc-silicate rocks (*csq*). The contact with the underlying metapelites (*qmt*) is sharp and at places is marked by a 10-foot rusty feldspathic layer. At most places, this rock unit is very rusty from the breakdown of ubiquitous pyrrhotite. Also, it is altered extensively to light-green diopside and tremolite. The protolith probably was a quartzose ankeritic dolomite. The weathered rock surface and soil of *csq* are very distinctive because the outcrop surface is gritty, rusty, and hackly, and the soil is composed largely of limonite-stained granular quartz and diopside sand. Tourmaline is not a common accessory mineral in this unit, *csq*, and we have analyzed only one tourmaline-bearing sample from it (67-51, table 2).

The uppermost unit in the upper limb of the nappe is an extremely well layered paragneiss (*pda*) that occurs in a narrow isoclinal synform parallel and close to Beaver Creek (fig. 2). Thin layers of the paragneiss have a variety of mineral assemblages that contain mostly plagioclase or scapolite. Some of these assemblages consist of green diopside, scapolite, and quartz, and others contain microcline, tourmaline, quartz, and biotite (fig. 4B), or biotite microcline and quartz. All rock types include many minute grains of sphene. A medium-grained vitreous diopside- and sphene-bearing quartzite (Brown, 1980, analysis 41) forms prominent outcrop ridges on either limb of the synform and helps to outline the structure. Some layers also are rich in poikilitic tourmaline and biotite. Retrograde metamorphism has produced clotty areas of green hornblende and meionitic scapolite

in the diopside-rich granofels. The protoliths for this unit were probably interlayered shales, sandstones, and dolomitic muds.

A large outcrop area of granite (*hg*) is in the vicinity of Huckleberry Mountain (fig. 2). The granite has been folded with the metasediments and is foliated and locally lineated. Narrow strips of quartz, biotite, feldspar, and granofels of the tourmaline-bearing unit (*qmt*) are included in the granite area as fold keels or ridges. Along the eastern edge of the main granite body are similar infolded strips of the underlying quartz and silicate-rich dolomitic marble. The main granitic body and numerous smaller satellitic masses are tongue-shaped slabs that were intruded at a low angle to the lithologic contacts of the metasediments and subsequently folded with them. In the western part of the area, the slabs are at the contact between the main tourmaline-bearing unit (*qmt*) and marble (*qm*). Farther to the east, the granite sheets are lower in the sequence where they intrude the marble unit. The granite sheets transect the stratigraphic section at such a low angle that at any one outcrop the granite is essentially concordant.

Numerous thin granitic sheets intrude the tourmaline-bearing quartzite and metapelite (*qmt*) to the extent that the unit is locally migmatitic. Granite sheets around and to the north of Huckleberry Mountain contain abundant black tourmaline in grainy lineated patches (fig. 3C). The granites are rich in tourmaline only where they occur within or adjacent to the tourmaline-rich quartzites and metapelites (*qmt*). Small lenses of undeformed pegmatite also are common in the tourmaline-bearing quartzite and metapelite (*qmt*), all containing well-formed black tourmaline prisms mainly in their outer contact zone (fig. 3D).

PETROGRAPHY AND SUMMARY OF THE BULK COMPOSITION OF TOURMALINE-BEARING ROCKS

The most important petrographic characteristics of tourmalines in *qmt*, *pda*, and *hg* from the Gouverneur area are summarized in table 1. As seen in thin section, tourmalines from *qmt* are highly variable in color and in habit. Tourmaline grains forming distinct layers in this unit are typically green, subhedral, show patchy indistinct color zoning, and coexist with microcline or oligoclase and quartz. Additional petrographic variation is found in brecciated tourmaline-rich layers in unit *qmt* along the Oswegatchie River where brownish-yellow, subhedral tourmalines coexist with phlogopite, calcite, pyrite, and quartz. All tourmaline varieties in *qmt* generally show vague, indistinct color zoning from core to rim.

Lack of strong optical zoning and the dusty-green color in the tourmalines from *pda* are typical. In con-

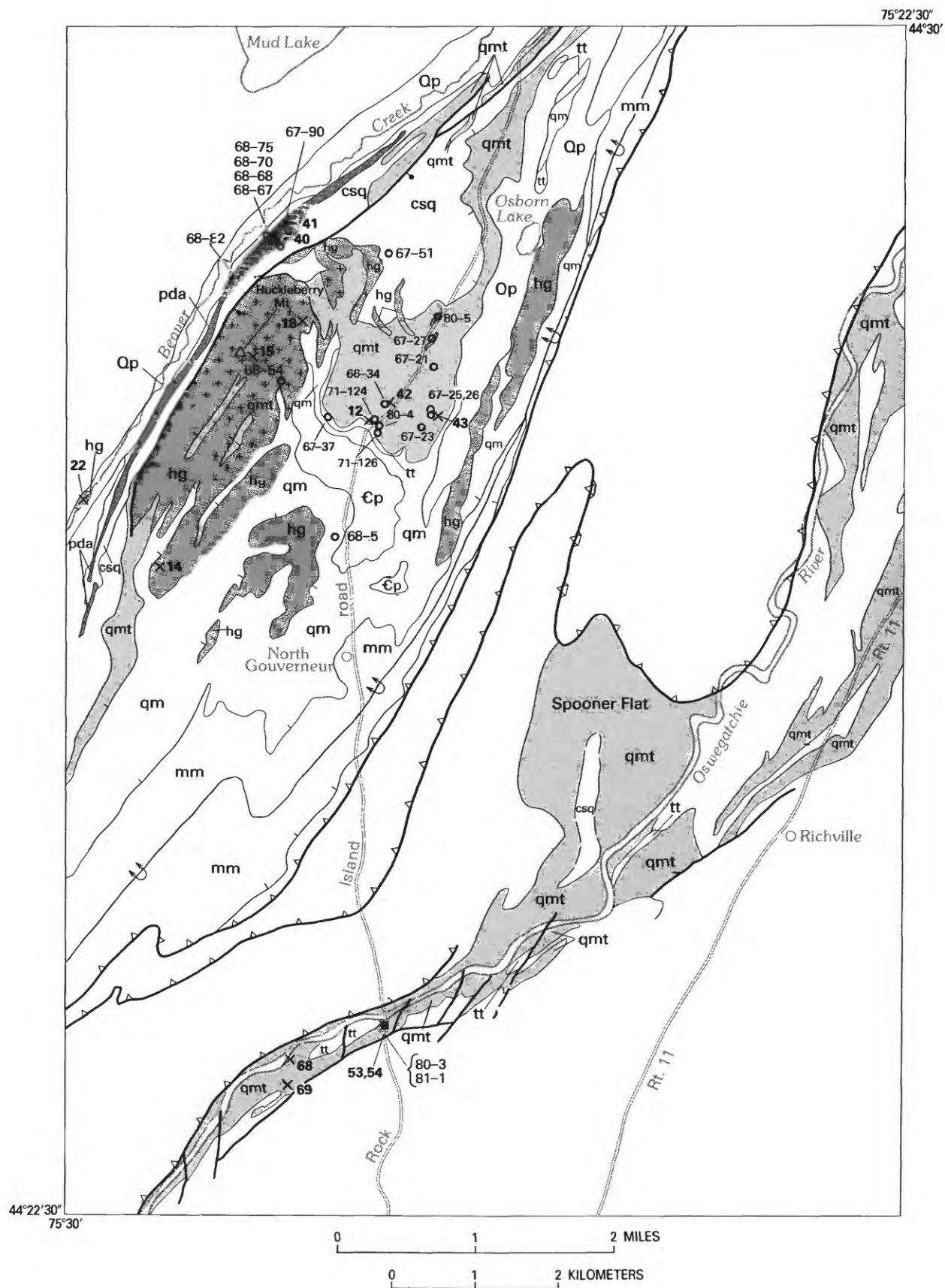


Figure 2. Generalized geologic map showing sample localities for this study. Geology along Oswegatchie River adapted from Bannerman (1972).

EXPLANATION

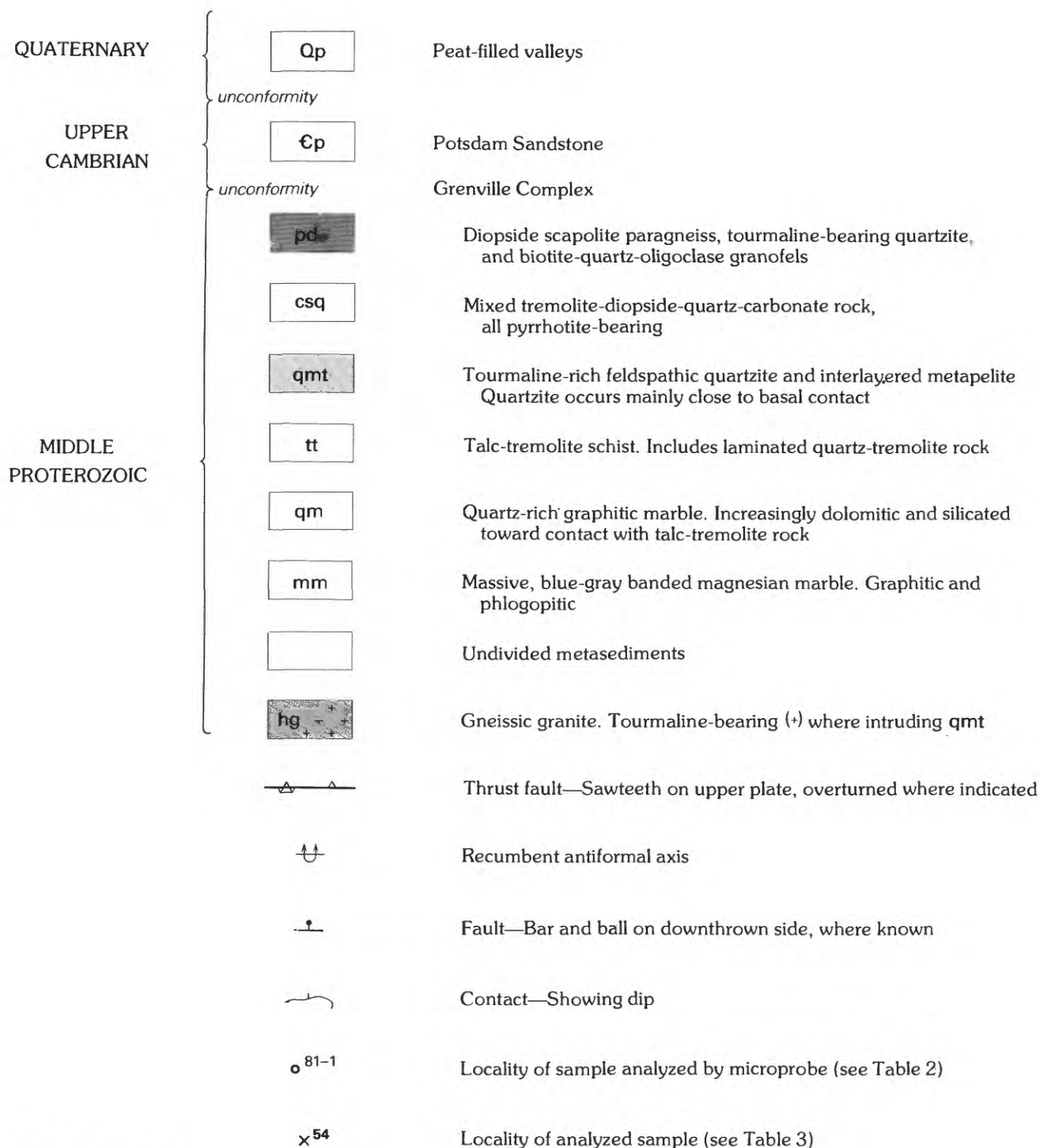


Figure 2. Continued.

trast to tourmalines from *qmt*, the tourmalines from *pda* are embayed and show resorption features. Also, tourmaline is characteristically present only in trace amounts in *pda*, except for sample 68–70 that has about 5 percent.

Tourmalines in granites (*hg*) have patchy zoning, are dark green as seen in thin section, and occur typically in patchy poikilitic grains. The streaky clots of tourmaline grains are commonly the principal dark mineral and are lineated weakly in the plane of foliation of the

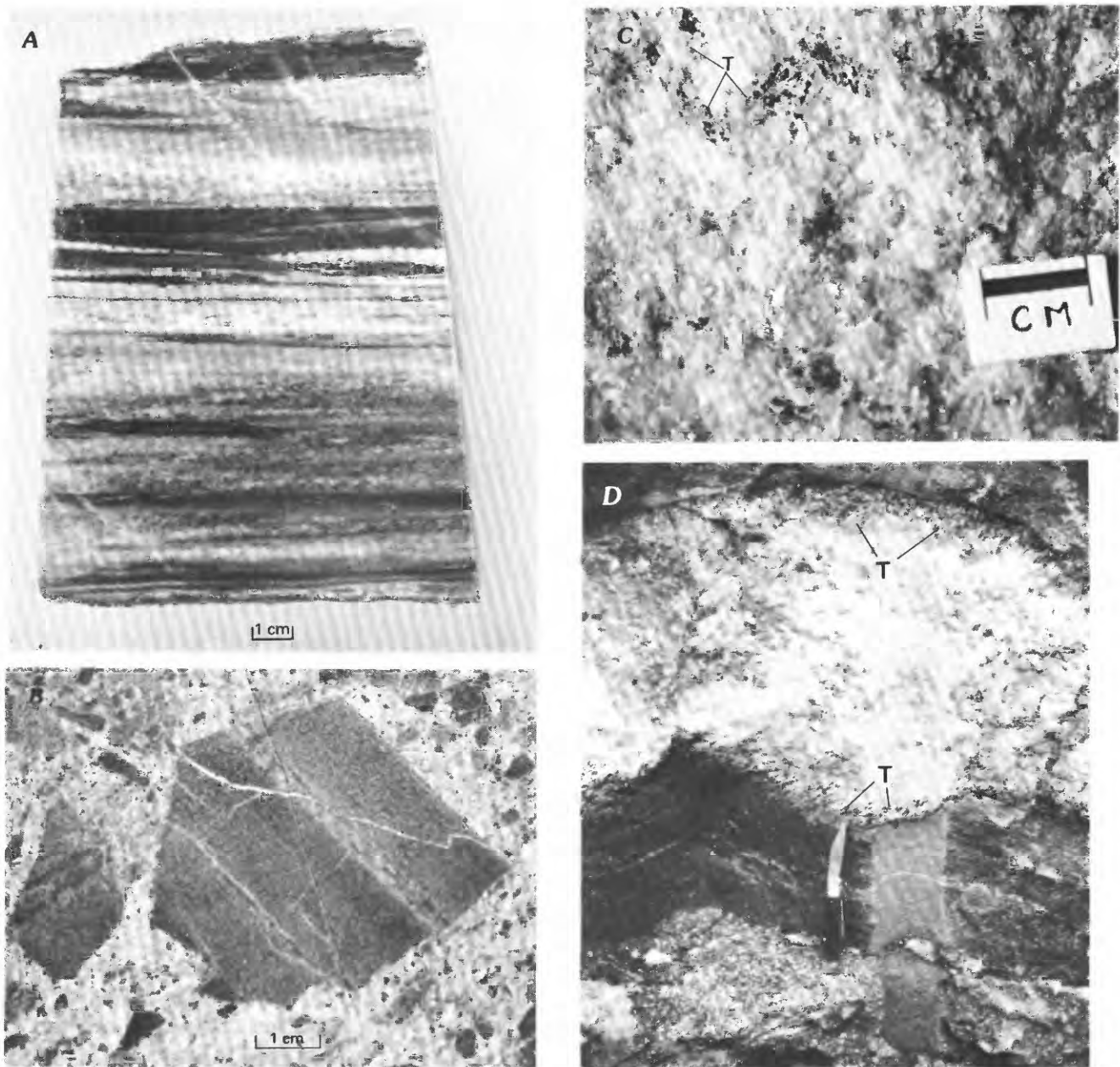


Figure 3. A. Polished slab of tourmalinite from lower part of unit *qmt* from along Rock Island Road close to sample locality 80-4. Dark layers are as much as 50 percent dravite. Light-colored layers are composed mainly of quartz, oligoclase, and biotite. B. Polished slab of brecciated tourmalinite from *qmt* from along Rock Island Road close to Oswegatchie River and sample locality 80-3. Matrix is composed of cataclasized *qmt* plus pyrite and minor calcite. C. Granular patches of black schorl (T) in granite sheet that intrudes *qmt*. Specimen collected from along Rock Island Road close to sample locality 67-27. D. Schorl prisms (T) around margin of small pegmatite lens in *qmt* at small road cut along Rock Island road close to sample locality 67-27. Note muscovite porphyroblasts in rock at bottom of view.

gneissic granite. Microcline, quartz, plagioclase, biotite, and muscovite coexist with the tourmalines, and the granite at places has accessory fluorite. Tourmaline in pegmatites occurs as well-formed black prisms arranged around wall-rock inclusions or along the border of the pegmatite mass (fig. 3D).

Composite tourmaline prisms (sample 68-5) in a quartzofeldspathic lens in *qm* have a dark-brown core, enveloped by indigo-blue rims. The surface between the

two tourmaline varieties is characterized by indentation, embayment, and resorption.

All textural features of the tourmalines from the North Gouverneur area, except sample 68-5, suggest that the tourmalines were generated during the metamorphic event that formed the rest of the mineral assemblage. Relic textures showing an earlier generation of tourmaline, subsequently transformed to other phases, are absent. Also lacking are features reminiscent

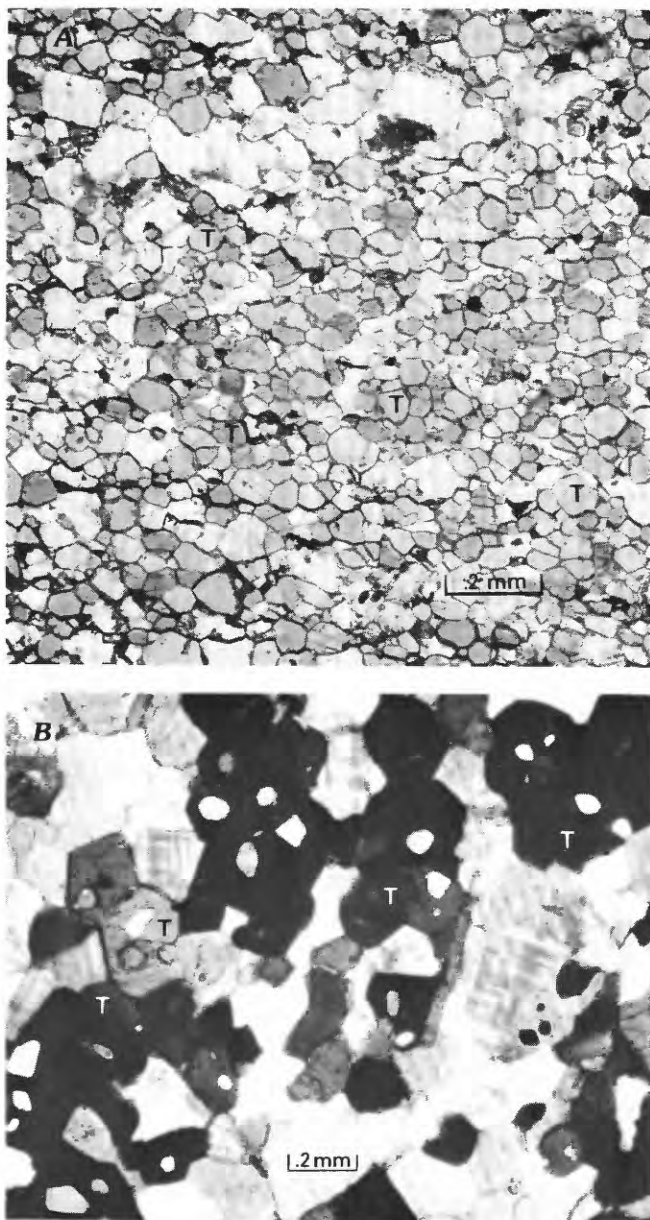


Figure 4. A. Photomicrograph of granoblastic tourmalinite from unit *qmt*. All gray grains are dravite. White grains are quartz or oligoclase. B. Photomicrograph of tourmaline-bearing rock from unit *pda*. Rock is microcline-tourmaline granofels having minor amounts of quartz and biotite. (T, Schorl prisms.)

of tourmalinization, whereby other minerals show partial replacement by tourmaline. Although poikiloblastic tourmaline is evident in some rock types, the textures indicate that tourmaline formed at the closing stages of the same event in which the surrounding phases formed.

Despite the relatively few bulk chemical analyses, it seems clear that the rock units in the North Gouverneur area have a wide range of compositions (tables 2, 3, and 4; fig. 5). Individual rock units, as exemplified by *qmt*, range widely from 47.2 to 80.0 weight percent SiO_2 , 6.9 to 17.2 weight percent Al_2O_3 , 0.50 to 14.0

weight percent CaO , and 0.28 to 9.3 weight percent K_2O . This suggests that significant compositional heterogeneity exists within individual rock units in a fairly restricted geographic area.

Figure 5 depicts some of the differences in the distribution of selected elements in the tourmaline-bearing rock units. For example, boron is generally higher in unit *qmt*, whereas barium seems to be subequal in units *qmt*, *pda*, and *hg*. Iron and titanium are higher in *pda* and *qmt* than in *hg* and *csq*, and magnesium decreases from *csq*, *pda*, *qmt*, to *hg*. Despite the relatively higher iron and magnesium contents in units *pda* and *qmt* compared with *hg*, the most iron-rich tourmalines are in the granite gneisses (*hg*).

SAMPLING

Twenty-three tourmaline-bearing specimens from several lithologic units in the North Gouverneur area (fig. 2) were selected from a collection of representative lithologic specimens collected during routine geologic mapping of the area. The three most important tourmaline-bearing rock types are the tourmaline-rich feldspathic quartzite (*qmt*), the diopside-scapolite paragneiss (*pda*), and the tourmaline-bearing granite gneiss (*hg*). Pegmatite and migmatite segregations within *qmt* and *pda* were also sampled, as were other lithologies in the vicinity such as talc-tremolite schist (*tt*), calcsilicate rock (*csq*), and a quartzofeldspathic lens in marble (*qm*).

Most of the samples came from a small area, about 10 square miles (26 km^2); therefore, we could determine tourmaline compositional variability resulting principally from differences in bulk composition of the host rocks. Rocks in *qmt* and *pda* have a wide range of lithologies; thus we expected that tourmaline within these units would be compositionally heterogeneous. In order to study the change in tourmaline composition across a lithologic unit, we selected closely spaced samples across *pda*. Tourmaline in pegmatites and migmatitic segregations were also studied for comparison with tourmalines in the nearby granitic bodies.

ANALYTICAL PROCEDURE

An automated, 3-channel, combined energy- and wave-length dispersive electron microprobe system at the U.S. Geological Survey, Reston, Va., was used for the partial chemical analyses of tourmalines from North Gouverneur (table 2). These analyses are necessarily incomplete because Fe_2O_3 , H_2O , B_2O_3 , and Li_2O are not determined by means of the microprobe. Although B_2O_3 and water determinations by means of wet-chemical methods are important in tourmaline studies, they require relatively large amounts of pure tourmaline separates. Because clean mineral separations are difficult to

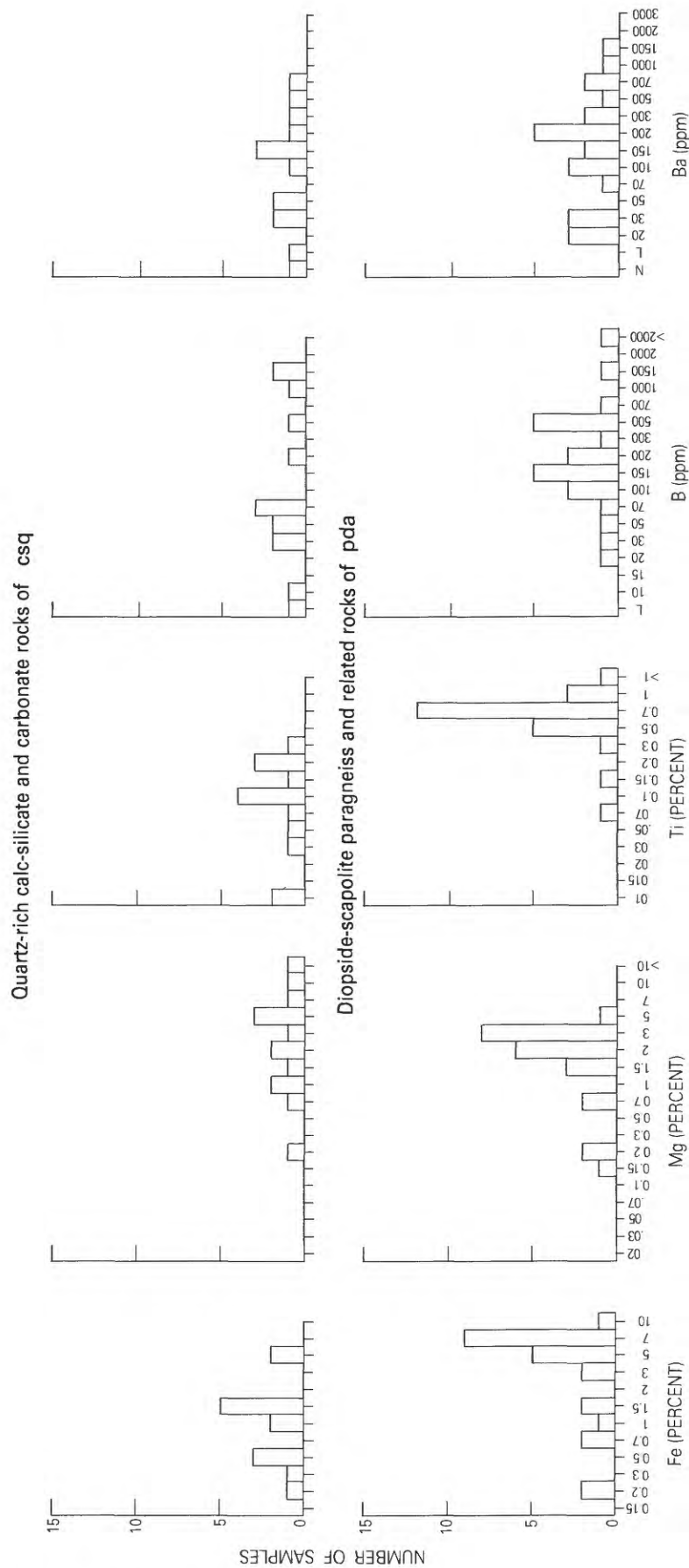


Figure 5. Distribution of selected elements in rocks from the North Gouverneur area. Semiquantitative spectrographic analysis by Jerry Matooka. L, detected, but below limit of determination; N, not detected.

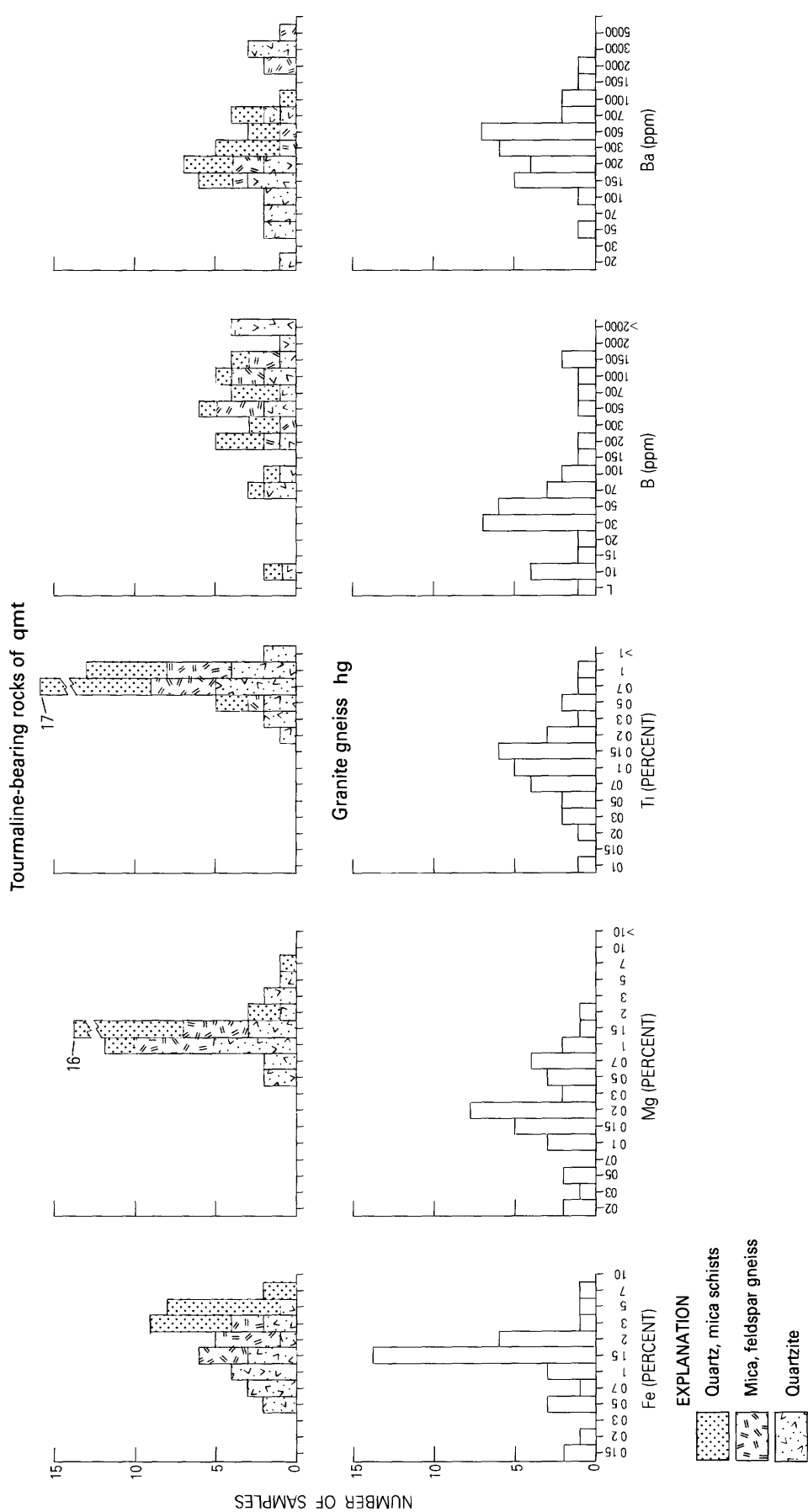


Figure 5. Continued.

Table 1. Summary of petrographic features of tourmaline-bearing rocks from the North Gouverneur area, New York [Abbreviations are as follows: py, pyroxene; am, amphibole; pl, plagioclase; mi, microcline; bi, biotite; sp, sphene; q, quartz; mu, muscovite; ph, phlogopite; sca, scapolite; cc, calcite; ch, chlorite after biotite; X, present; ---, not present]

Sample no.	Color	Habit	Zoning	Inclusions	Opaque oxides and sulfides	Assemblage	Tourmaline abundance (in percent)	Other
<i>pda</i>								
68-75	Brown green	Subhedral	---	Fluid	X	py, am, pl	< 1	-----
68-67	Brown green	Subhedral	---	Silicates	?	py, am, pl, bi, sp, q	1	Embayed
68-68	Brown green	Subhedral	---	-----	X	bi, pl	1	Embayed
68-70	Brown green	Anhedral	---	Silicate	X	q, am, sp, py		Poikilitic; same texture as diopside
<i>hg</i>								
68-54	Brown green	Anhedral	Patchy	Fluid, silicate	X	q, mi, bi, pl	5	Poikilitic
80-5	Dark green	Anhedral	Patchy	Fluid, silicate	?	mu, mi, pl, q	5	Fluid inclusions near core
67-27	Dark green	Subhedral	Patchy	Silicate	--	mi, q, mu	5	Poikilitic
<i>Pegmatite in qmt</i>								
71-124	Dark green	Anhedral	Patchy	Silicate		mi, pl, q	20	Poikilitic
67-23	Brown green	Subhedral	X	-----	?	pl, q, mu	5	Embayed, associated with quartz
<i>qmt</i>								
67-26	Green	Euhedral	X? Patchy	Silicate, fluid	--	bi, mi, q	10	In layers
67-21	Brownish yellow	Subhedral	X	-----	X	ph, cc, q	25	Random distribution
71-126	Brown green	Anhedral		Silicate	--	pl, sca, q, sp	5	Poikilitic intergrowth with scapolite
80-4	Yellow	Subhedral	X?	-----	X	mu, q, ch, pl?	25	Random distribution and in layers
80-3 (clast)	Brown green	Subhedral	--	Silicate, fluid	X	q, mi, pl, ch	20	Random distribution
80-3 (matrix)	Yellow	Euhedral	X	Silicate, fluid	X	ch, cc, q	5	Embayed, larger than tourmaline in clasts

obtain and equivalent data for tourmalines from massive sulfide deposits are unknown, such approach was deemed to be outside the scope of this study.

Tourmalines were carefully cleaned for semiquantitative spectrographic analyses (table 5), but most of the tourmaline included in the metasedimentary rocks was

too fine grained for hand cleaning, and thus the semi-quantitative analyses were mainly from porphyroblastic tourmaline or from crystals in pegmatitic segregations.

Fe₂O₃ is presumed to represent a major component in buergerite (Donnay and others, 1966), the ferric iron end member of the tourmaline group, and its abundance is assumed to be negligible in this region. Walenta and Dunn (1979) demonstrated that ferric iron is also an essential component in ferrodravite, the aluminum-poor iron analog of dravite. On the basis of new spectrographic analyses of tourmalines from Gouverneur (this study) together with previous investigations (Dunn and others, 1977), it seems reasonable to suggest that lithium-rich tourmalines (elbaïtes) are also volumetrically minor or nonexistent. Specialized geochemical environments exemplified by lithium-enriched pegmatites are known to be critical for the formation of elbaïtes (Deer and others, 1962). Such environments might exist in the pegmatites in the North Gouverneur area. However, because potentially favorable environments in this setting are very rare and because spectrographic analysis of tourmaline sample 71-124 from a pegmatite has more than 10 weight percent total iron and only 10 ppm lithium (table 5), we assumed that elbaïtes, if present in the Gouverneur area, are volumetrically negligible and therefore excluded discussion of them in this study.

Microprobe analyses were obtained on standard singly or doubly polished thin sections, carbon coated before analysis. Standards were chosen from a set of natural silicate minerals, with internal standardization checks made on compositionally well known silicates, including a San Luis Potosi tourmaline obtained from Brian Mason, Smithsonian Institution. Size of the electron beam ranged from 5 to 10 microns. Each grain was analyzed at a minimum of five spots, and each sample represents an average of at least five grains. The data were reduced on-line by means of the methods of Bence and Albee (1968). Compositional variations are shown on table 2. Structural formulas were calculated assuming a value of 3 boron atoms in the structural formula.

RESULTS OF COMPOSITIONAL STUDIES

Tourmaline End Members in Nature

Tourmaline is compositionally variable in natural environments because it forms under many geologic conditions. The theoretical formula may be stated as XY₃Z₆(BO₃)₃Si₆O₁₈(OH)₄, in which X = (Na, Ca, K); Y = (Mg, Fe, Mn, Li, Al); and Z = Al (Deer and others, 1962). Two solid solutions are the most common in nature: (1) from Fe-rich (schorl) to Mg-rich (dravite); and (2) from schorl to the Li-rich (elbaïte) tourmalines. Dunn and others (1977) documented another solid solution between dravite (Na > Ca) and uvite (Ca > Na), as

suggested earlier by Kunitz (1929). Fe³⁺-rich tourmalines are known as buergerite and ferri-dravite (Walenta and Dunn, 1979).

The Gouverneur area in New York contains tourmalines that have compositions mainly within the dravite-uvite solid solution series (Dunn and others, 1977). Our preliminary results, based on spectrographic analyses (table 5) also indicate that lithium-rich tourmalines probably do not occur here. Lithium content of tourmaline in the North Gouverneur area is restricted to values of less than 70 ppm, well below the composition of typical elbaïtes (>0.9 weight percent Li₂O according to Nemec, 1969).

Compositional Variation of Tourmaline in North Gouverneur Area

Microprobe analyses of tourmaline present other problems in addition to their incomplete nature. For example, Ethier and Campbell (1977) indicated that for the Sullivan Mine tourmalines, grain size and volatilization of boron and sodium during microprobe analysis were significant problems. For the North Gouverneur tourmalines, neither the grain size nor the loss of alkalis and boron was a major obstacle. More disturbing results, however, were found in the compositional range of SiO₂ (32.4 to 42.1 weight percent) and Al₂O₃ (24.9 to 36.5 weight percent) for these tourmalines (table 2). A tendency toward higher contents in SiO₂ and Al₂O₃ for the microprobe-analyzed tourmalines of the Sullivan Mine was also reported by Ethier and Campbell (1977), who cited previous determinations exemplifying similar analytical problems.

Several tourmaline samples from the North Gouverneur area showed anomalous microprobe compositions even after duplicate runs, use of different sets of microprobe standards, and a minimum of 25 spot analyses.

The average composition of tourmaline samples 71-126 (*qmt*), and 80-3(a) (*qmt*) are lower in SiO₂ (<33.8 weight percent) than the range of well-determined tourmaline compositions in Deer and others (1962) for schorl and dravites, and Dunn and others (1977) for uvites (<33.7 weight percent). Sample 80-4 (*qmt*) is enriched in SiO₂ (40.44 weight percent) and is similar to a uvite analysis given by Dunn and others (1977). The contents of Al₂O₃ in tourmaline samples 71-126 (*qmt*) and 71-124 (pegmatite in unit *qmt*) are lower than the minimum value (28.5 weight percent) in Deer and others (1962), but are within the published range for uvites (23.1 to 34.4 weight percent) shown by Dunn and others (1977).

The average tourmaline compositions presented in table 2 generally show reasonable variations for most oxides except for SiO₂ and Al₂O₃ in samples 80-4 (*qmt*),

Table 2. Electron microprobe analyses and structural formulas of tourmalines in the North Gouverneur area, New York
[Analytical data represents the average composition of traverses with the reproducibility as 2σ in parentheses. Cation proportions calculated assuming near the Oswegatchie River, south of North Gouverneur. Columns 8, 9, and 11 are from the matrix; column 10 is from a clast]

Column -----	1	2	3	4	5	6
Beaver Creek zone						
Lithologic unit-----	<i>qmt</i>	<i>qmt</i>	<i>qmt</i>	<i>qmt</i>	<i>qmt</i>	<i>qmt</i>
Sample number-----	66-34	67-21	67-25	67-26	80-4	71-126
Weight percent						
SiO ₂ -----	34.87 (0.90)	33.08 (0.27)	37.38 (0.39)	32.93 (0.08)	40.44 (3.76)	32.45 (0.25)
TiO ₂ -----	1.13 (.10)	.60 (.05)	.59 (.07)	.92 (.05)	1.70 (.23)	1.26 (.06)
Al ₂ O ₃ -----	35.00 (.26)	36.54 (.62)	35.07 (.37)	34.22 (.36)	30.90 (1.98)	27.86 (.06)
FeO ¹ -----	8.37 (.29)	2.23 (.18)	2.07 (.08)	5.03 (.17)	5.22 (.09)	9.85 (.10)
MnO -----	.05 (.06)	.01 (.02)	.02 (.02)	.03 (.06)	.07 (.04)	.10 (.02)
MgO -----	7.67 (.17)	9.92 (.28)	9.53 (.23)	8.91 (.11)	8.03 (.62)	9.58 (.11)
CaO -----	1.98 (.11)	1.49 (.13)	.39 (.22)	1.02 (.08)	1.20 (.37)	3.04 (.01)
Na ₂ O -----	1.53 (.05)	1.65 (.06)	2.81 (.19)	1.94 (.06)	2.14 (.13)	4.25 (.07)
K ₂ O -----	.17 (.04)	.11 (.02)	.07 (.02)	.20 (.06)	.06 (.36)	.06 (.01)
Cl -----	.02 (.02)	0	.03 (.01)	.03 (.04)	.02 (.01)	0
Total	90.79	85.63	87.96	85.23	89.78	88.45
Structural formula						
Si -----	5.493	5.345	5.857	5.436	6.211	5.533
B -----	3.000	3.000	3.000	3.000	3.000	3.000
Ti -----	.134	.073	.070	.114	.196	.162
Al -----	6.498	6.959	6.477	6.658	5.594	5.600
Fe -----	1.103	.301	.271	.694	.670	1.405
Mn -----	.007	.001	.003	.004	.009	.014
Mg -----	1.801	2.400	2.226	2.193	1.838	2.435
Ca -----	.334	.258	.065	.180	.198	.555
Na -----	.467	.517	.854	.621	.637	.413
K -----	.034	.023	.014	.042	.012	.013
Ratios						
FeO/(FeO + MgO)	0.522	0.184	0.178	0.361	0.394	0.507
Na ₂ O/ (Na ₂ O + CaO + K ₂ O)	.416	.508	.859	.614	.629	.578
Fe/(Fe + Mg + Mn)	.379	.111	.108	.240	.266	.365
Na/(Na + Ca + K)	.559	.648	.915	.737	.752	.421

80-3 (*qmt*), 68-68 (*pda*), and 68-54 (*hg*). Much of the variation, however, may result from the procedure of analyzing the same sample over a period of weeks. Typical variations for the other oxides are much lower, and no significant or consistent deviations from the mean were observed. It is likely, however, that samples showing substantial variation reflect different stages of tourmaline growth. In these samples, perhaps only the outermost zones may be in equilibrium with the coexisting minerals. Samples showing great compositional variability in the North Gouverneur area, however, generally have compositions that vary progressively from rim to core in individual grains.

Although only weight-percent oxide variations

could be used for comparison of the tourmalines in the North Gouverneur area, a more useful method of study depends on the calculation of cation abundances in the hypothetical tourmaline formulas. These calculations, however, because of the incomplete chemical analyses obtained with the electron probe (for example, B₂O₃, H₂O), and because of the observed large variation in the two major oxides of SiO₂ and Al₂O₃, yield useful but probably only first-order approximations of the changes in the crystal chemistry of tourmalines in the North Gouverneur area. Despite these difficulties, the calculated cation proportions suggest general trends for the tourmalines of the area.

In addition to the changes in the ratios of

29 oxygen and 3 boron atoms in the structural formula. Columns 1–6 are from the area north of North Gouverneur; columns 7–11 are from

Column -----	7	8	9	10	11
South zone					
Lithologic unit-----	<i>qmt</i>	<i>qmt</i>	<i>qmt</i>	<i>qmt</i>	<i>qmt</i>
Sample number -----	81-1	80-3(a)	80-3(b)	80-3(c)	80-3(d)
Weight percent—Continued					
SiO ₂ -----	35.43 (0.68)	33.73 (0.71)	35.48 (0.66)	33.25 (0.65)	35.82 (1.95)
TiO ₂ -----	1.63 (.18)	1.04 (.20)	2.40 (.25)	1.40 (.08)	1.64 (.10)
Al ₂ O ₃ -----	28.61 (.55)	29.21 (1.20)	32.45 (.30)	27.99 (1.20)	30.52 (1.50)
FeO ¹ -----	10.14 (.63)	6.07 (.26)	2.17 (.15)	11.27 (.64)	.14 (.02)
MnO -----	.03 (.04)	.11 (.11)	.15 (.01)	.09 (.09)	.01 (.02)
MgO -----	9.11 (.10)	10.34 (.38)	12.50 (.98)	8.17 (.21)	12.29 (.40)
CaO -----	2.97 (.58)	4.04 (.44)	2.27 (.14)	3.04 (.19)	1.69 (.19)
Na ₂ O -----	1.48 (.37)	.85 (.21)	1.96 (.14)	1.23 (.07)	2.06 (.16)
K ₂ O -----	.23 (.03)	.14 (.07)	.11 (.04)	.28 (.04)	.03 (.03)
Cl -----	.02 (.01)	.03 (.02)	0	.06 (.04)	.02 (.02)
Total	89.65	85.56	89.49	86.78	84.22
Structural formula—Continued					
Si -----	5.757	5.639	5.578	5.638	5.836
B -----	3.000	3.000	3.000	3.000	3.000
Ti -----	.199	.131	.282	.179	.201
Al -----	5.479	5.756	5.972	5.594	5.861
Fe -----	1.378	.849	.283	1.598	.091
Mn -----	.004	.016	.020	.013	.001
Mg -----	2.207	2.577	2.909	2.065	2.985
Ca -----	.517	.724	.380	.552	.295
Na -----	.466	.275	.593	.404	.651
K -----	.048	.030	.022	.061	.006
Ratios—Continued					
FeO/(FeO + MgO)	0.527	0.370	0.148	0.580	0.011
Na ₂ O/ (Na ₂ O + CaO + K ₂ O)	.316	.169	.452	.270	.545
Fe/(Fe + Mg + Mn)	.384	.247	.088	.435	.006
Na/(Na + Ca + K)	.452	.267	.596	.397	.684

Na₂O/(Na₂O + CaO + K₂O) and FeO/(FeO + MgO), we discuss the change in the cation proportions for Na and Fe and suggest general compositional trends. The two oxide ratios generally, but only approximately, treat the occupancies of the X-sites (Na, Ca, K) and Y-sites (Mg, Fe, Mn, Li, Al). Besides ignoring the role of Al, these sample oxide ratios cannot be used, in contrast to cation proportions, for exploring the effect of coupled substitutions. Such substitutions have much broader value in explaining tourmaline compositions than the simple trends shown by oxide ratios.

A direct reflection of the variation in SiO₂ and Al₂O₃ contents of the tourmalines is given in the range of the calculated cation proportions. Although most

tourmaline samples have a value close to 6 for Si, some tourmalines have high Si values to a maximum of 6.514 (sample 80-5 in unit *hg*), and others have low Si values of about 5.436 (sample 67-26 in *qmt*). For comparison, the highest Si value given by Deer and others (1962) for dravite is 6.172, whereas the lowest is 5.758 for a schorl. Low Al values, less than needed to fill the Z-site, were also obtained in some tourmalines. Such contents were masked by the inherent analytical problems of the microprobe technique, but also probably include geological uncertainty surrounding the effect of other cations substituting for Al (for example, Fe²⁺, Fe³⁺, Mg, Ti, and so forth) in the Z-site.

Occupancy in the X-site (Na, Ca, K) appears to be

Table 2. Electron microprobe analyses and structural formulas of tourmalines in the North Gouverneur area, New York—Continued [Analytical data represents the average composition of traverses with the reproducibility as 2 σ in parentheses. Cation proportions calculated assuming near the Oswegatchie River, south of North Gouverneur. Columns 8, 9, and 11 are from the matrix; column 10 is from a clast]

Column -----	12		13		14		15		16	
Lithologic unit-----	<i>pda</i>		<i>pda</i>		<i>pda</i>		<i>pda</i>		<i>pda</i>	
Sample number-----	67-90		68-67		68-68		68-70		68-75	
Weight percent—Continued										
SiO ₂ -----	34.34	(0.34)	34.20	(0.23)	33.67	(1.23)	36.73	(0.08)	33.38	(0.13)
TiO ₂ -----	1.13	(.11)	1.51	(.22)	.78	(.08)	1.21	(.08)	.86	(.13)
Al ₂ O ₃ -----	24.88	(.74)	27.62	(.51)	32.64	(1.20)	26.84	(.42)	27.72	(.19)
FeO ¹ -----	12.36	(.19)	8.63	(.27)	7.71	(.68)	11.24	(.25)	9.64	(.21)
MnO -----	.04	(.04)	.06	(.05)	.04	(.06)	.07	(.02)	.04	(.08)
MgO -----	7.83	(.30)	10.24	(.13)	8.49	(.23)	8.85	(.07)	10.57	(.15)
CaO -----	2.22	(.10)	2.76	(.10)	1.89	(.15)	2.75	(.21)	2.97	(.13)
Na ₂ O -----	1.66	(.13)	1.44	(.06)	1.65	(.11)	1.48	(.14)	1.25	(.05)
K ₂ O -----	.12	(.03)	.14	(.10)	.20	(.09)	.12	(.02)	.15	(.03)
Cl -----	.02	(.02)	.04	(.05)	.02	(.03)	.03	(.02)	.02	(.03)
Total	84.6		86.64		87.09		89.32		86.60	
Structural formula—Continued										
Si -----	6.057		5.718		5.531		6.003		5.623	
B -----	3.000		3.000		3.000		3.000		3.000	
Ti -----	.150		.190		.096		.149		.109	
Al -----	5.173		5.442		6.319		5.170		5.503	
Fe -----	1.823		1.207		1.059		1.536		1.358	
Mn -----	.006		.009		.006		.010		.006	
Mg -----	2.059		2.552		2.079		2.156		2.654	
Ca -----	.420		.494		.333		.482		.536	
Na -----	.568		.467		.526		.469		.408	
K -----	.026		.030		.042		.025		.032	
Ratios—Continued										
FeO/(FeO + MgO)	0.612		0.457		0.476		0.559		0.477	
Na ₂ O/ (Na ₂ O + CaO + K ₂ O)	.415		.332		.441		.340		.286	
Fe/(Fe + Mg + Mn)	.469		.320		.337		.415		.338	
Na/(Na + Ca + K)	.560		.471		.584		.481		.418	

normal, and most tourmalines fall within the range of about 0.8 to 1.0. The lowest value for the total of Na + Ca + K is in sample 68-5 (in *qm*), specifically in the blue part of the composite tourmaline. The value for the total of Na + Ca + K is only 0.439 compared with 0.963 obtained on the brown core of the same tourmaline crystal.

The analyses in table 2 show that tourmalines from each unit (fig. 6) have distinct iron and magnesium compositions. The most notable characteristic is that tourmalines from the granitic gneiss (*hg*) and from the pegmatitic segregations within *qmt* and *pda* have FeO/(FeO + MgO) ratios of 0.55-0.75 that are generally

higher than those of tourmalines from the metasediments. Compared to the field of granite and pegmatite tourmalines from Portugal (Neiva, 1974; 1976), those from Gouverneur are higher in MgO and thus have lower FeO/(FeO + MgO) ratios.

The lowest FeO/(FeO + MgO) ratios in tourmalines are in unit *qmt*. They also have the most heterogeneous ratio of FeO/(FeO + MgO), from 0.01 to 0.58. Within *qmt*, tourmalines may be grouped according to whether they occur in the nappe structure north of North Gouverneur or along and southeast of the Oswegatchie River (fig. 2). The bulk of the data is from the northern area. Intrusions of granite and pegmatite

29 oxygen and 3 boron atoms in the structural formula. Columns 1-6 are from the area north of North Gouverneur; columns 7-11 are from

Column -----	17	18	19	20	21	22
	Granites			Pegmatites		
Lithologic unit -----	hg	hg	hg	ln pda	ln qmt	ln qmt
Sample number -----	67-27	68-54	80-5	68-82	71-124	67-23
Weight percent—Continued						
SiO ₂ -----	35.87 (0.27)	37.84 (3.80)	42.10 (0.27)	35.04 (0.23)	35.52 (0.22)	35.80 (0.73)
TiO ₂ -----	.71 (.05)	1.26 (.14)	.76 (.07)	2.85 (.05)	.93 (.14)	.96 (.04)
Al ₂ O ₃ -----	35.06 (.37)	25.67 (.91)	29.64 (.43)	26.76 (.04)	27.86 (.51)	31.83 (.40)
FeO ¹ -----	8.26 (.26)	14.71 (.35)	9.77 (.28)	12.80 (.01)	14.18 (.20)	8.75 (.75)
MnO -----	.04 (.03)	.22 (.07)	.15 (.02)	.08 (.01)	0	.03 (.07)
MgO -----	6.86 (.23)	4.74 (.37)	4.91 (.31)	8.60 (.06)	7.47 (.25)	6.86 (.41)
CaO -----	.88 (.12)	1.82 (.25)	1.00 (.15)	1.58 (.03)	2.55 (.10)	.98 (.06)
Na ₂ O -----	2.35 (.06)	1.90 (.21)	2.06 (.07)	2.29 (.03)	1.77 (.05)	2.12 (.05)
K ₂ O -----	.12 (.03)	.15 (.06)	.12 (.02)	.14 (.03)	.15 (.10)	.12 (.01)
Cl -----	.04 (.01)	.02 (.02)	.01 (.01)	.02 (.01)	.04 (.03)	0
Total	90.19	88.53	90.52	90.16	90.47	87.45
Structural formula—Continued						
Si -----	5.659	6.318	6.514	5.750	5.829	5.846
B -----	3.000	3.000	3.000	3.000	3.000	3.000
Ti -----	.084	.158	.088	.352	.115	.118
Al -----	6.520	5.051	5.405	5.175	5.388	6.129
Fe -----	1.090	2.054	1.264	1.757	1.946	1.195
Mn -----	.005	.031	.020	.011	0	.004
Mg -----	1.613	1.230	1.132	2.104	1.827	1.671
Ca -----	.149	.236	.166	.278	.448	.172
Na -----	.705	.615	.618	.729	.564	.671
K -----	.204	.032	.024	.029	.030	.025
Ratios—Continued						
FeO/(FeO + MgO)	0.546	0.756	0.666	0.598	0.655	0.561
Na ₂ O/ (Na ₂ O + CaO + K ₂ O)	.701	.491	.648	.571	.396	.658
Fe/(Fe + Mg + Mn)	.403	.620	.523	.454	.516	.416
Na/(Na + Ca + K)	.803	.632	.765	.704	.541	.773

characterize this area, whereas postmetamorphic fault-related brecciation and recrystallization typifies the belt of *qmt* found near the Oswegatchie River.

Tourmaline in two samples (80-3 and 81-1; table 2) from a large roadcut in unit *qmt* along Rock Island Road, south of the Oswegatchie River, is consistently different in compositional trends from tourmalines to the north in this unit. The two samples may themselves be distinguished because sample 80-3 is from a greenish-gray reduced breccia having a pyritic matrix and sample 81-1 (fig. 3B) is an oxidized (hematite-stained) equivalent of sample 80-3. Field observations suggest that the pyrite formed after brecciation.

Sample 80-3 contains at least two generations of tourmaline enclosed in matrix and in clasts, and these are distinguished both optically and chemically. Tourmalines in the matrix of sample 80-3 are the most compositionally diverse found in this study. Their FeO/(FeO + MgO) ratios range from about 0.01 to 0.37, and contrast with tourmaline in the clasts that have ratios of about 0.58 (fig. 6). Matrix tourmalines in sample 80-3 contacting pyrite are depleted in iron (FeO/FeO + MgO = 0.11), whereas matrix tourmalines in the same immediate area but not in contact with pyrite have higher FeO/(FeO + MgO) ratios (0.15 to 0.58). The composition of the tourmaline in sample

Table 2. Electron microprobe analyses and structural formulas of tourmalines in the North Gouverneur area, New York—Continued [Analytical data represents the average composition of traverses with the reproducibility as 2 σ in parentheses. Cation proportions calculated assuming 29 oxygen and 3 boron atoms in the structural formula. Columns 1–6 are from the area north of North Gouverneur; columns 7–11 are from near the Oswegatchie River, south of North Gouverneur. Columns 8, 9, and 11 are from the matrix; column 10 is from a clast]

Column	23	24	25	26
Quartzofeldspathic lens in <i>qmt</i>				
Lithologic unit	Brown tourmaline	Blue tourmaline	<i>csq</i>	<i>tt</i>
Sample number	68–5	68–5	67–51	67–37
Weight percent—Continued				
SiO ₂	36.89 (0.36)	34.91 (0.25)	37.24 (0.29)	37.73 (0.47)
TiO ₂	.37 (.13)	.13 (.05)	.23 (.03)	1.77 (.08)
Al ₂ O ₃	30.98 (.76)	36.91 (.45)	28.90 (.13)	30.05 (.10)
FeO ¹	.92 (.20)	13.41 (.35)	2.34 (.34)	.69 (.19)
MnO	.03 (.04)	0	.02 (.02)	.01 (.01)
MgO	12.64 (.30)	.01 (.03)	12.86 (.25)	12.52 (.13)
CaO	2.28 (.15)	.14 (.15)	3.55 (.03)	1.26 (.13)
Na ₂ O	1.81 (.14)	1.29 (.06)	.98 (.04)	2.31 (.02)
K ₂ O	.05 (.02)	0	.04 (.01)	.02 (.03)
Cl	.02 (.02)	.07 (.02)	0	.03 (.03)
Total	85.99	86.87	86.16	86.39
Structural formula—Continued				
Si	5.907	5.775	6.016	5.997
B	3.000	3.000	3.000	3.000
Ti	.045	.016	.028	.212
Al	5.847	7.201	5.503	5.630
Fe	.123	1.856	.316	.092
Mn	.004	0	.003	.001
Mg	3.017	.002	3.097	2.966
Ca	.391	.025	.614	.215
Na	.562	.414	.251	.712
K	.010	0	.008	.004
Ratios—Continued				
FeO/(FeO + MgO)	0.068	0.999	0.154	0.052
Na ₂ O/ (Na ₂ O + CaO + K ₂ O)	.437	.902	.214	.643
Fe/(Fe + Mg + Mn)	.039	.999	.093	.030
Na/(Na + Ca + K)	.584	.943	.288	.765

¹Total iron as FeO.

81–1 (oxidized breccia) has a ratio of FeO/(FeO + MgO) of about 0.53 and falls well within the range defined by *qmt* to the north (fig. 6). Compositionally, tourmalines from 81–1 and from the clasts in sample 80–3 (c) have ratios that are more similar (0.53 compared with 0.58) than the ratio of the matrix tourmaline in sample 80–3 (a,b,d).

In contrast to the tourmalines in unit *qmt* north of North Gouverneur, those from near the Oswegatchie River define a broad trend of decreasing TiO₂ with increasing FeO/(FeO + MgO) ratio (fig. 6). This trend is opposite to that of unit *qmt* tourmalines from north of North Gouverneur, which show increasing TiO₂ at higher FeO/(FeO + MgO) ratios. Despite the numerous

granites and pegmatites north of North Gouverneur (fig. 2), tourmalines in metasediments there do not show gradational compositional features arising from proximity to the igneous rocks. Also, because of the large range in TiO₂ content (from 0.58 to 2.40 weight percent) and FeO/(FeO + MgO) ratios, these oxides cannot be used to distinguish samples in *qmt*. Possible exceptions are two samples of matrix tourmalines in sample 80–3 (b, d; table 2) that appear to have both a low FeO/(FeO + MgO) ratio (0.011–0.148) and high TiO₂ content (>1.6 weight percent).

Tourmalines in *pda* from samples that are closely spaced across a stratigraphic sequence show much less variation than those from *qmt*. The FeO/(FeO + MgO)

Table 3. Chemical analyses of selected tourmaline-bearing rock types in the North Gouverneur area, New York, in weight percent

[n.d., not determined. See Brown (1980) for additional information about these analyses. Sample locality shown on figure 2 of this report]

Rock unit -----	Granite (hg)					pda		qmt					
Chemical analysis no. ¹ ----	12	14	15	18	22	40	41	42	43	53	54	68	69
Rapid rock analysis													
SiO ₂ -----	65.7	71.7	71.6	71.3	72.2	57.4	88.3	80.0	77.2	63.4	60.9	47.2	61.0
Al ₂ O ₃ -----	16.8	14.7	14.8	15.0	14.7	10.9	3.0	6.9	11.7	15.6	17.2	10.0	17.0
Fe ₂ O ₃ -----	1.0	.10	.91	.49	.73	1.4	.64	1.5	.65	3.2	3.1	4.6	3.7
FeO -----	1.3	1.0	.72	.92	.24	2.7	.60	3.3	.32	1.2	.61	.51	.49
MgO -----	.78	.50	.30	.30	.40	7.6	.90	2.7	1.3	3.6	2.2	4.6	2.5
CaO -----	2.3	1.0	1.3	1.0	.90	11.9	1.9	.50	1.5	.78	1.6	14.0	.58
Na ₂ O -----	3.8	3.7	3.5	3.7	2.4	3.3	1.1	.20	4.4	.63	.57	.50	.70
K ₂ O -----	6.7	5.6	5.8	5.8	6.2	2.6	.11	2.6	1.0	8.0	9.1	.28	9.3
H ₂ O ⁺ -----	.50	.90	.05	.96	.56	.38	.57	.34	.54	1.6	1.0	1.4	.91
H ₂ O ⁻ -----	.05	.05	.63	.04	.17	.17	.07	.11	.06	.08	.04	.08	.07
TiO ₂ -----	.42	.26	.24	.21	.21	1.2	1.0	.71	.93	1.2	1.2	1.1	1.2
P ₂ O ₅ -----	.19	.18	.14	.16	.21	.31	.10	.13	.17	.14	.12	.16	.10
MnO -----	.05	.08	.02	.07	.00	.10	.05	.05	.05	.04	.06	.24	.06
CO ₂ -----	.11	<.05	.05	<.05	.03	<.05	.78	<.05	.06	<.05	1.5	9.7	<.05
Aqua regia solution as SO ₃ -----	.21	n.d.	n.d.	n.d.	n.d.	n.d.	.24	n.d.	n.d.	n.d.	n.d.	5.1	n.d.
Wet chemical analysis													
B ₂ O ₃ -----	n.d.	n.d.	n.d.	n.d.	.58	n.d.	0.32	0.38	0.38	1.38	1.78	3.47	2.31
Cl -----	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.03	n.d.	n.d.
F -----	0.14	.068	.039	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	0.83	n.d.	n.d.
Sum -----	100.05	99.9	100.1	100.0	99.5	100.0	99.7	99.5	100.2	100.9	101.8	102.9	100.0

¹These numbers identify sample locality on figure 2 and are from Brown (1980), where more information about analyses can be found.

Table 4. Semiquantitative spectrographic analyses of tourmaline-bearing rocks in the North Gouverneur area, New York, in parts per million

[L, detected, but below limit of determination; N, not detected at limit of determination. See Brown (1980) for more information about samples. Sample locality shown on figure 2 of this report]

Lithologic unit -----	Granite (hg)					pda		qmt					
Analysis number ¹ ----	12	14	15	18	22	40	41	42	43	53	54	68	
Ag -----	L	L	N	L	N	L	L	L	L	N	N	L	
B -----	50	50	N	500	3,000	700	500	1,000	500	20,000	20,000	30,000	
Ba -----	1,000	700	50	500	300	500	30	300	200	700	1,000	100	
Be -----	7	7	N	10	1.5	3	N	1	3	N	N	N	
Co -----	N	N	N	N	N	15	N	10	N	20	15	30	
Cr -----	3	N	N	N	2	50	30	20	50	150	150	100	
Cu -----	200	5	10	3	3	5	15	5	5	5	10	5	
La -----	150	100	N	70	70	100	N	N	N	50	N	N	
Mo -----	N	N	N	N	N	N	N	N	N	N	N	3	
Nb -----	15	10	N	7	L5	10	10	10	10	10	L5	N	
Ni -----	N	N	N	N	L10	L	N	L	L	N	5	70	
Pb -----	30	20	N	30	20	N	N	N	N	10	20	7	
Sc -----	7	5	N	5	N	10	3	7	7	20	20	20	
Sn -----	20	N	N	N	N	N	N	N	N	N	N	N	
Sr -----	1,000	200	1,000	150	70	700	10	30	30	150	200	300	
V -----	30	15	7	10	10	70	50	70	70	150	150	100	
Y -----	50	30	N	30	30	70	20	20	15	30	15	30	
Zr -----	700	200	N	150	100	200	150	150	150	200	200	150	
Ce -----	500	500	N	200	100	300	N	N	N	N	N	N	
Ga -----	20	20	N	20	15	10	5	10	15	20	20	20	
Yb -----	5	3	N	3	1.5	7	2	2	1.5	3	1.5	3	

¹These numbers identify sample locality on figure 2 and are from Brown (1980).

Table 5. Semiquantitative spectrographic analyses of tourmaline in Beaver Creek area, New York

[Results are reported as midpoints of interval data 1.0, 0.7, 0.5, 0.3, 0.2, 0.15, 0.1 (and multiples of 10). The precision of reported value is approximately plus or minus one bracket at 68 percent or two brackets at 96 percent confidence. Symbols used are G, greater than 10 percent or value shown; N, not detected at limit of determination or below value shown. Numbers in parenthesis indicate lower limit of detection. Looked for but not detected in all samples at lower limit of detection (in parenthesis): Ag (0.5), As (1,000), Au (20), Be (1.5), Bi (10), Cd (50), La (50), Mo (3), Nb (10), Pd (2), Pt (50), Sb (200), Sn (10), Te (2,000), U (500), W (100), Zn (300), K (7,000), P (2,000), Ce (200), Ge (10), Hf (100), In (10), Re (50), Ta (500), Th (200), Tl (50), Yb (1)). Analyst, Nancy M. Conklin]

Sample number	¹ EB-80-1	² EB-71-35	³ EB-80-2	⁴ EB-71-124	⁵ EB-77-7	⁶ EB-69-99	⁷ EB-69-98
Percent							
Si	G	G	G	G	G	G	G
Al	G	G	G	G	G	G	G
Na	0.7	0.7	0.7	1.5	0.3	1.	1.5
Fe	1.5	.1	.7	G	.3	3.	3.
Mg	7.	10.	10.	5	G	7.	7.
Ca	3.	3.	3.	2.	3.	2.	1.
Ti	.3	.15	.3	.7	.3	.3	.150
Parts per million							
Mn(1)	50,000	70	150	700	5	150	70
B (20)	G	G	G	G	G	G	G
Ba(3)	30	3	N3	3	N3	3	7
Co(5)	N	N	N	15	N	N	N
Cr(1)	30	700	70	300	30	70	N
Cu(1)	30	15	7	7	2	3	N
Ni(5)	5	3	3	10	3	3	N
Pb(10)	15	N	N	N	N	N	N
Sc(5)	N	N	15	30	N	70	N
Sr(5)	700	200	300	300	200	700	700
V (7)	N	1,500	150	700	70	1,000	N
Zr(10)	N	N	N	30	N	N	N
Ga(5)	30	30	70	70	30	100	70
⁸ Cs(10)	N10	N10	N10	N10	N10	N10	N10
⁸ Li(1)	20	20	50	10	10	70	70
⁸ Rb(2)	N2	N2	N2	N2	N2	2	3

¹Wine-red tourmaline grains that occur with manganese-bearing tremolite (hexagonite) at Arnold Talc Mine, and Wight Mine near Fowler, N.Y. Described by Ayuso and Brown (1984).

²Pale-green tourmaline grains in coarse calcitic marble that includes much phlogopite and has crystals and irregular masses of scapolite as much as 3 inches across. Small quarry west of Rock Island Road 800 feet north of Beaver Creek Crossing.

³Light-brown tourmaline prisms in pegmatitic clots in coarse-grained marble. Locality 3, Brown (1983).

⁴Black tourmaline in pegmatite within tourmaline-rich rock. Locality shown on figure 2.

⁵Amber tourmaline in white phlogopitic marble. Locality 96, Brown (1983).

⁶Coarse black crystalline masses with quartz. Locality 29, Brown (1983).

⁷Pale-blue tourmaline that encrusts perthite crystals in small pegmatite. Locality 28, Brown (1983).

⁸Analyzed by means of a special method for alkalis that significantly lowers sensitivities for Na and Li and makes possible the determination of Cs and Rb.

ratios range from 0.476 to 0.612 (fig. 6), and samples collected near the *csq* contact showed no compositional change. In contrast to the trend in *qmt* tourmalines, those from *pda* occur in rocks having abundant sphene, and show no gradient in TiO₂ abundance relative to the FeO/(FeO + MgO) ratio.

Analyses of tourmalines in a few samples from the other stratigraphic units, *csq*, *tt*, and a quartzofeldspathic lens in *qm* show that their compositions have a lower FeO/(FeO + MgO) ratio than the bulk of the data showed for *qmt* and *pda* (fig. 6). Samples 67-37

from unit *tt* also has a high TiO₂ content (1.77 weight percent) and plots within the field defined by *qmt* tourmalines from near the Oswegatchie River.

Because of the overlap in tourmaline compositions for FeO/(FeO + MgO) ratios and in TiO₂ content, the host rock for tourmalines cannot be clearly distinguished. It is, however, possible to distinguish the tourmalines in the granite and pegmatites because they have high FeO/(FeO + MgO) ratios and because they are similar to tourmalines in aplites and granites (Neiva, 1977).

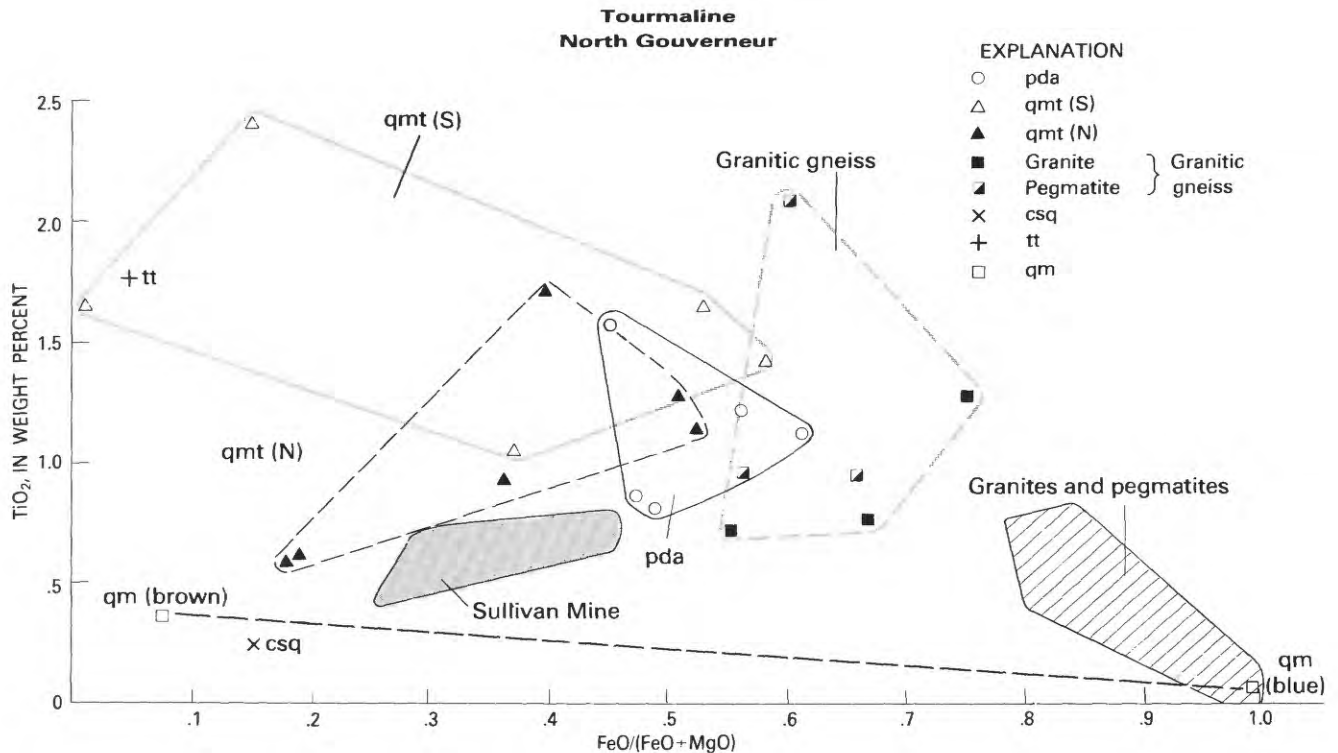


Figure 6. TiO_2 - $\text{FeO}/(\text{FeO}+\text{MgO})$ plot for tourmalines in lithologic units from North Gouverneur, N.Y., compared with those from Sullivan Mine, British Columbia (Ethier and Campbell, 1977), and Portuguese granites and pegmatites (Neiva, 1974). Tie line connects the compositions of the composite tourmaline in the marble unit (qm).

Slack (1982) used a plot of the oxides FeO - MgO - Na_2O to show the compositions of tourmalines from selected stratabound sulfide deposits. The North Gouverneur tourmalines are also shown on the ternary plot of Na_2O - MgO - FeO (fig. 7) and suggest the following: (1) All tourmalines, regardless of lithology, plot in a trend subparallel to the join MgO - FeO . Most of the MgO -rich tourmalines in each lithologic unit plot slightly above the MgO - FeO join; this probably suggests a trend toward higher Na_2O contents with higher MgO . (2) Granites and pegmatites have tourmaline compositions plotting closest to the FeO - Na_2O join, in accordance to the field defined by lithium-poor tourmaline from granites and pegmatites. Tourmalines from granites from North Gouverneur are, however, more magnesian than tourmaline normally found in such environments. (3) Tourmalines from *pda* as a group are more magnesian and less sodic than those from granites and pegmatites. (4) The *qmt* tourmalines are heterogeneous in FeO , MgO , and Na_2O . Tourmalines from near the Oswegatchie River plot closer to the MgO - FeO sideline than tourmalines from north of North Gouverneur. (5) Tourmalines from *tt*, *csq*, and the brown tourmaline from *qm* are magnesian with a moderate range in Na_2O content.

Tourmalines in this study show a strong trend subparallel to the MgO - FeO sideline, despite the wide

range of lithologic types and mineral assemblages that they represent. Because of the fairly constant Na_2O content in these tourmalines, the FeO - MgO - Na_2O plot emphasizes the range in FeO and MgO contents in the tourmaline. Control on the Na_2O content of tourmaline might be a reflection of the equilibrium assemblage, particularly with respect to another phase that has varying ratios of $\text{Na}_2\text{O}/\text{CaO}$ (for example, plagioclase). Compared with the average compositions compiled by Slack (1982) of tourmalines that are intergrown with sulfide minerals, the North Gouverneur tourmalines overlap the field of tourmaline associated with stratabound sulfide deposits, despite their known coexistence with volumetrically minor sulfide concentrations. In fact, the area occupied by tourmalines intergrown with sulfides in the FeO - MgO - Na_2O plot and that of tourmaline not immediately associated with sulfides but obtained from the same vicinity show similar ranges in FeO and MgO at roughly the same Na_2O content. A reasonable conclusion from this comparison is that Na_2O content of tourmalines plotted on the ternary FeO - MgO - Na_2O does not distinguish between tourmalines abundantly intergrown with sulfides and those without sulfides.

The amount of aluminum in the North Gouverneur tourmalines cannot be used to distinguish the different host lithologies. Alumina (Al_2O_3) ranges widely

Tourmaline North Gouverneur

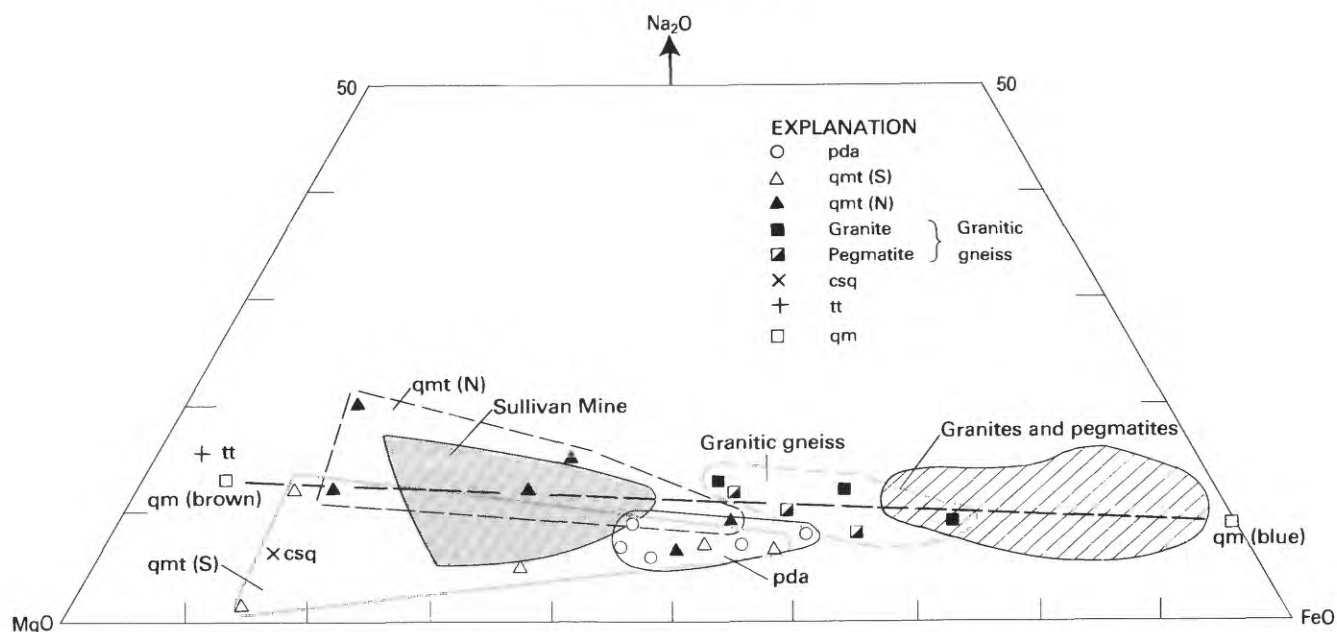


Figure 7. Ternary plot of Na₂O-MgO-FeO for tourmaline from North Gouverneur, N.Y., the Sullivan Mine, British Columbia (Ethier and Campbell, 1977), and the field of granites and pegmatites (Neiva, 1974).

from 24.8 to 36.54 weight percent. Combined with the distribution of CaO, the fact that all tourmalines show slight decreases in CaO as Al₂O₃ increases (fig. 8) suggests that higher alkali contents might be found in the more aluminous tourmalines. Tourmalines from granites and pegmatites from North Gouverneur tend to be the lowest in CaO content for a given range in Al₂O₃, especially for Al₂O₃ < 30 weight percent. Within individual rock units from North Gouverneur, the highest Al₂O₃ content in tourmalines might be found in those with the lowest FeO/(FeO + MgO) ratio (fig. 9). However, the broad and general decrease in Al₂O₃ as FeO/(FeO + MgO) ratio decreases, suggested in the metasedimentary units, is much less evident in tourmalines from the granites and pegmatites. Tourmalines from north of North Gouverneur are generally higher in Al₂O₃ at a given FeO/(FeO + MgO) ratio than tourmalines from near the Oswegatchie River.

Calcium Content of Tourmalines

The CaO content in tourmalines from the North Gouverneur area ranges from 0.39 to 4.04 weight percent (table 2) and cannot be correlated with the FeO/(FeO + MgO) ratio. Samples from *qmt* are the most chemically heterogeneous tourmalines as they span compositions from near end-member dravite (12.29 weight percent MgO)—compared with dravite from Gouverneur which has 13.67 weight percent MgO (cited

in Deer and others, 1962)—to uvite (4.04 weight percent CaO). Most tourmalines from *qmt* generally have, however, less than 3.0 weight percent CaO (except sample 80-3(a) with 4.04 weight percent). Tourmaline samples from the breccia near the Oswegatchie River are for the most part indistinguishable from the rest of the *qmt* samples on the basis of their CaO and MgO contents.

In *pda*, most tourmalines range from 1.89 to 2.97 percent CaO (table 2, fig. 8). The few analyses of magnesian tourmalines from *tt*, *csq*, and the brown tourmaline in *qm* suggest that, with the exception of *csq*, these tourmalines are low in CaO content (table 2, fig. 8).

Substitutions in Hypothetical Structural Formula

Although the microprobe analyses are incomplete, consideration of such hypothetical formulae is useful in indicating possible crystal chemical substitutions operating during crystallization of tourmaline. Such substitutions suggest that elemental chemical exchanges within individual lithologic units were generally adjusted in a regular manner to changing conditions during tourmaline growth.

The change in Na and Fe for Al in the Gouverneur tourmalines suggests that Na and Fe are generally but broadly inversely correlated with Al (fig. 10). The most sodic and iron-rich tourmalines are in the granites and pegmatites, whereas tourmalines most impoverished in

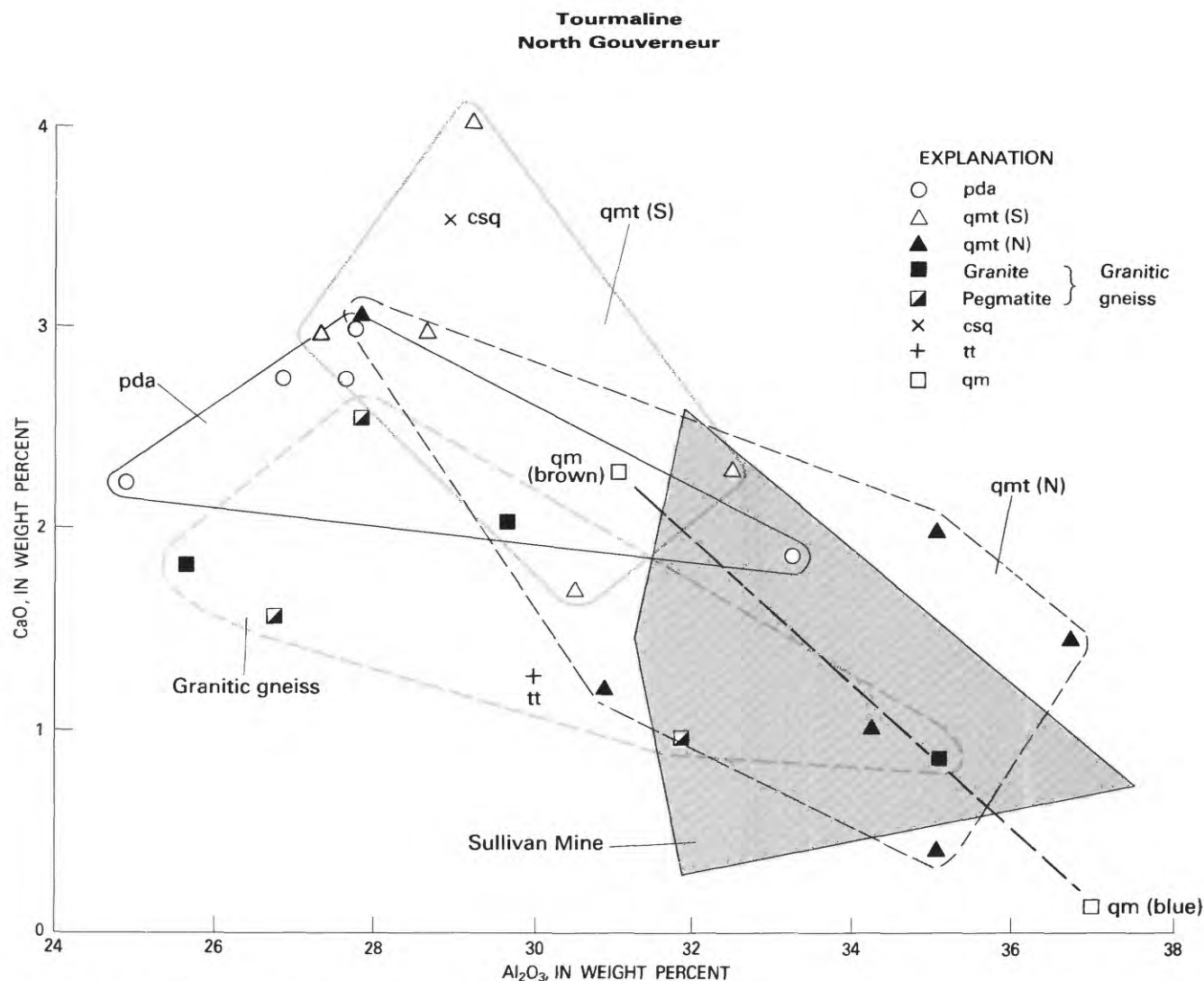


Figure 8. CaO-Al₂O₃ plot for tourmalines from North Gouverneur, N.Y., compared with those from the Sullivan Mine, British Columbia.

these components are in *qmt*, *tt*, and *csq*. The overall pattern is in basic accordance with a change from schorl-like tourmalines in granite and pegmatites toward tourmaline compositions in the uvite-dravite series occurring in metasediments, especially in *qmt*. Tourmalines from the Sullivan deposit are distinct from this trend as they follow no consistent pattern. In contrast to the apparent Na+Fe=Al correlation (alkali-defect substitution), neither the Sullivan or the North Gouverneur tourmalines show a correlation in Na+Mg=Al as found by Schreyer and Abraham (1976) for dravites from an evaporite deposit.

The change between Na and Ca for total Al may occur on a regional basis, and it is shown by the slight decrease in Na+Ca as Al increases (fig. 11). However, no meaningful distinctions are evident between rock units. The most regular and gradual correlation is present in

pda, whereas the most scattered is in the granites and pegmatites. For comparison, tourmalines from Sullivan are aluminous and plot toward lower values of Na+Ca as defined by the North Gouverneur tourmalines.

The general variation between Na+Mg+Al and Ca+2Fe might be one of the mechanisms controlling the progressive change in tourmaline composition in the North Gouverneur area. In this exchange, Na+Mg+Al are inversely correlated with Ca+2Fe²⁺ (fig. 12). Tourmaline from north and south of the Oswegatchie River is not distinguishable on this plot. Sullivan Mine tourmalines seem to have compositions similar to tourmalines in the North Gouverneur area. Tourmalines from the granitic gneiss unit are generally higher in the sum of Ca+2Fe compared with other tourmalines in the metasediments. The composite brown-blue tourmaline spans almost the entire range

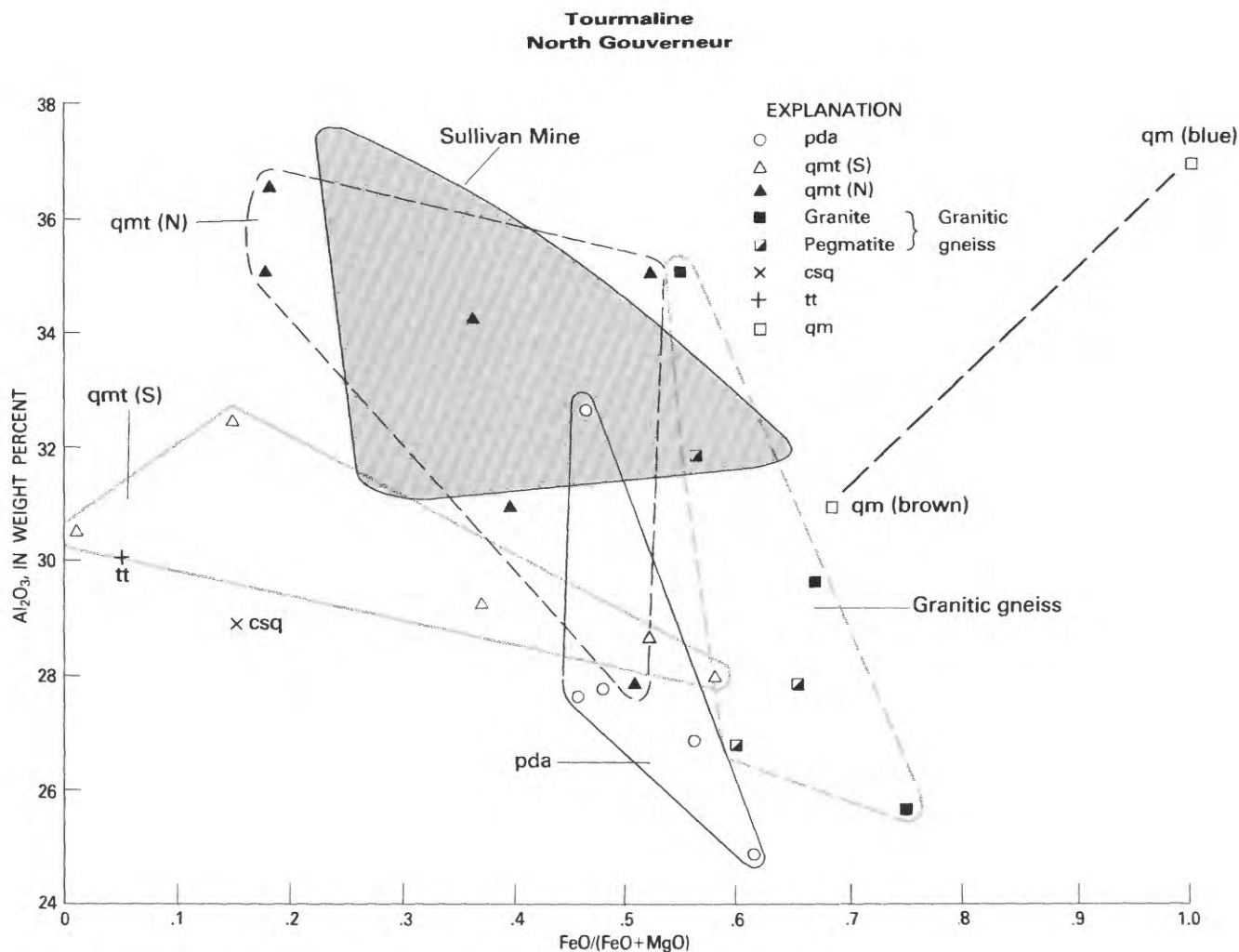


Figure 9. Al₂O₃-FeO/(FeO+MgO) plot for tourmalines from North Gouverneur, N.Y.

shown by tourmalines from the metasedimentary rocks. Tourmalines from unit *pda* in the North Gouverneur area have a smooth variation in composition. On a regional basis, however, tourmaline from North Gouverneur shows substantial compositional overlap. As expected, tourmalines from North Gouverneur granite and pegmatite are somewhat higher in iron (more schorl rich) than those in the samples from unit *qmt* that belong to the dravite-uvite series.

The substitution of Na for Ca (fig. 13) in the Z site could serve as an indicator of similar crystallization conditions among different groups of tourmalines. However, the dispersal of values of tourmalines from North Gouverneur cannot allow appropriate comparison with Sullivan tourmalines. Contents of less than 2.5 weight percent CaO and higher than 1.2 weight percent Na₂O encompass the compositional range of Sullivan, but most of the North Gouverneur samples also show this range. Clearly, the variation in sodium and calcium con-

tents is not a definitive test of similarity between North Gouverneur and Sullivan tourmalines.

Mineralized Areas and Tourmaline Compositions

Although anomalous tourmaline concentrations, which have compositions near the dravite end member, exist in unit *qmt*, the range in FeO/(FeO + MgO) ratios of tourmaline in a geographically restricted area suggests that this ratio is insufficiently specific for selection of the most favorable area for potential mineralization. The use of other major and trace elements in conjunction with the FeO/(FeO + MgO) ratio may prove to be more useful indicators of tourmalines associated with mineral deposits. Tourmalines in the Sullivan deposit are characterized by large variations in the Na₂O/(Na₂O + CaO + K₂O) ratio, in contrast to tourmalines from the enclosing Aldridge metasediments and nearby granites. Similar dispersals in compositional

**Tourmaline
North Gouverneur**

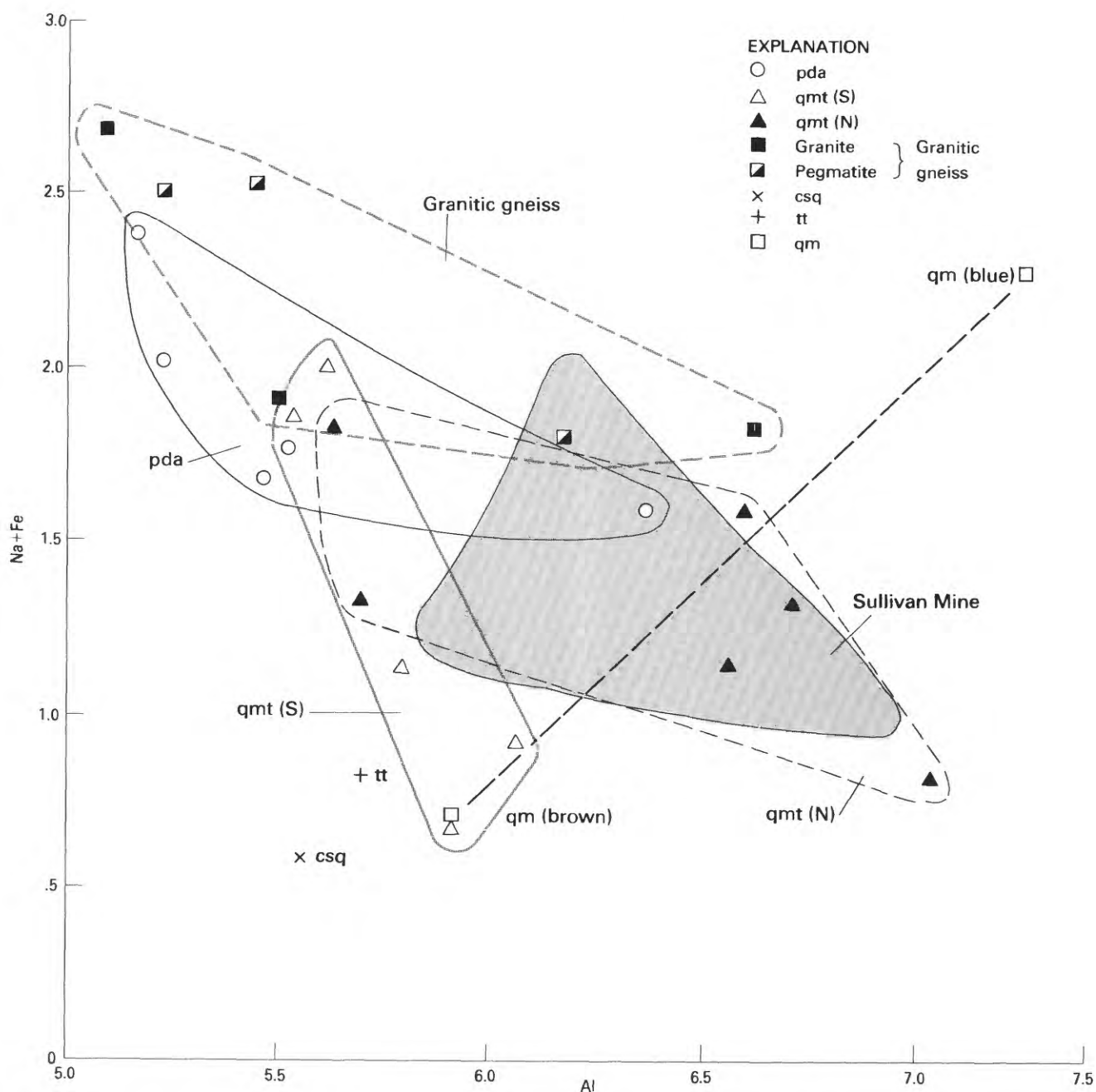


Figure 10. (Na+Fe)-Al plot in number of ions per formula unit for tourmalines from North Gouverneur, N.Y., compared with those in rocks from the Sullivan Mine, British Columbia.

variations are found in *qmt* tourmalines relative to those from *pda* and *hg* (fig. 14). The variations in tourmalines from *qmt* are also in the same range as those in the Sullivan Mine. In addition, tourmalines from the Sullivan Mine are low in CaO (generally less than 2.00 weight percent). An idealized profile of tourmalines

from mineralized areas should also note the prevailing low contents of TiO_2 (<0.1 weight percent) for the Sullivan Mine (fig. 6). Chlorine and oxides such as K_2O and MnO do not appear to have interpretable trends either in the North Gouverneur area or in the Sullivan ore body.

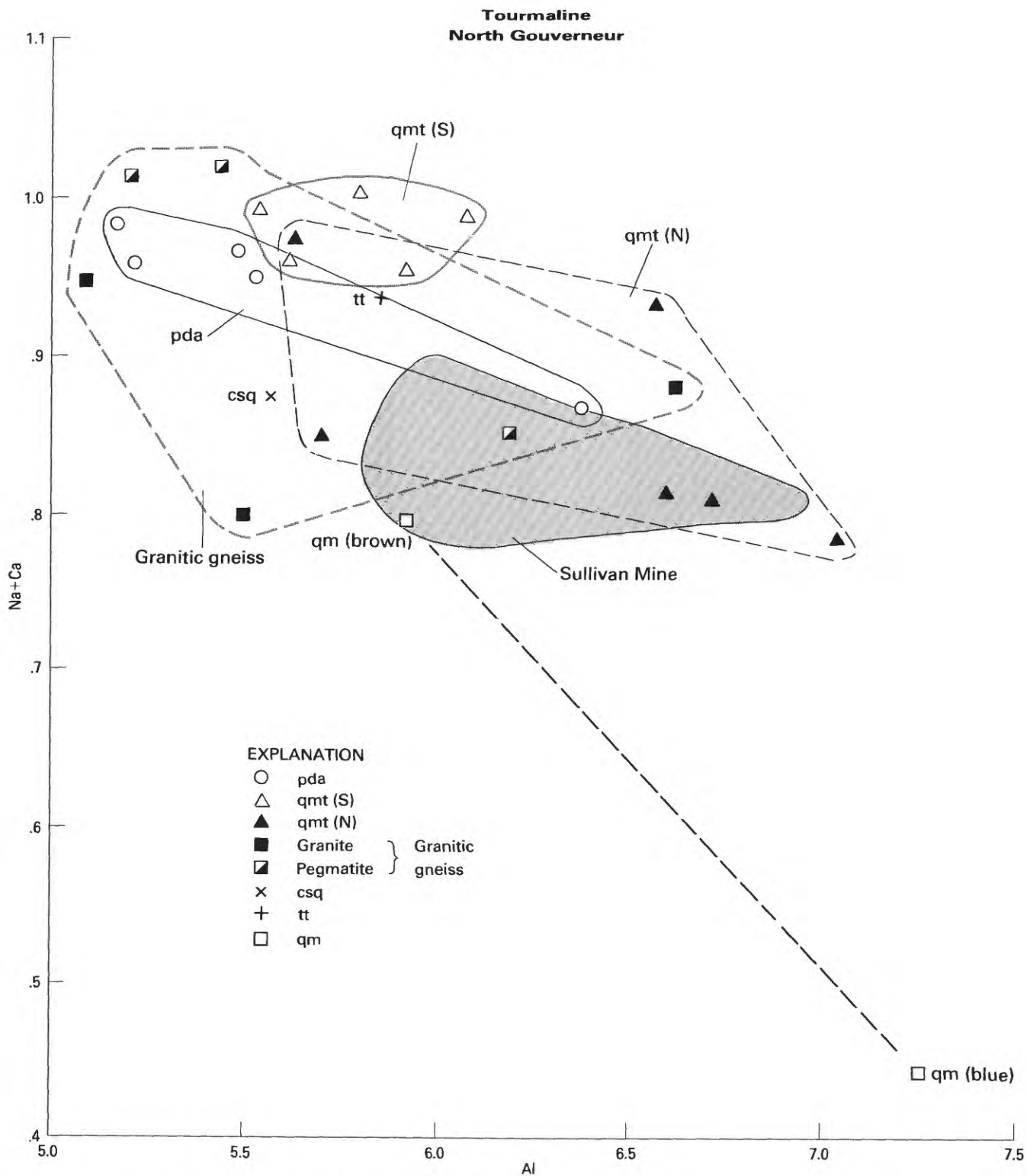


Figure 11. (Na+Ca)-Al plot in number of ions per formula unit for tourmalines from North Gouverneur, N.Y., compared with those in rocks from the Sullivan Mine, British Columbia.

Tourmaline North Gouverneur

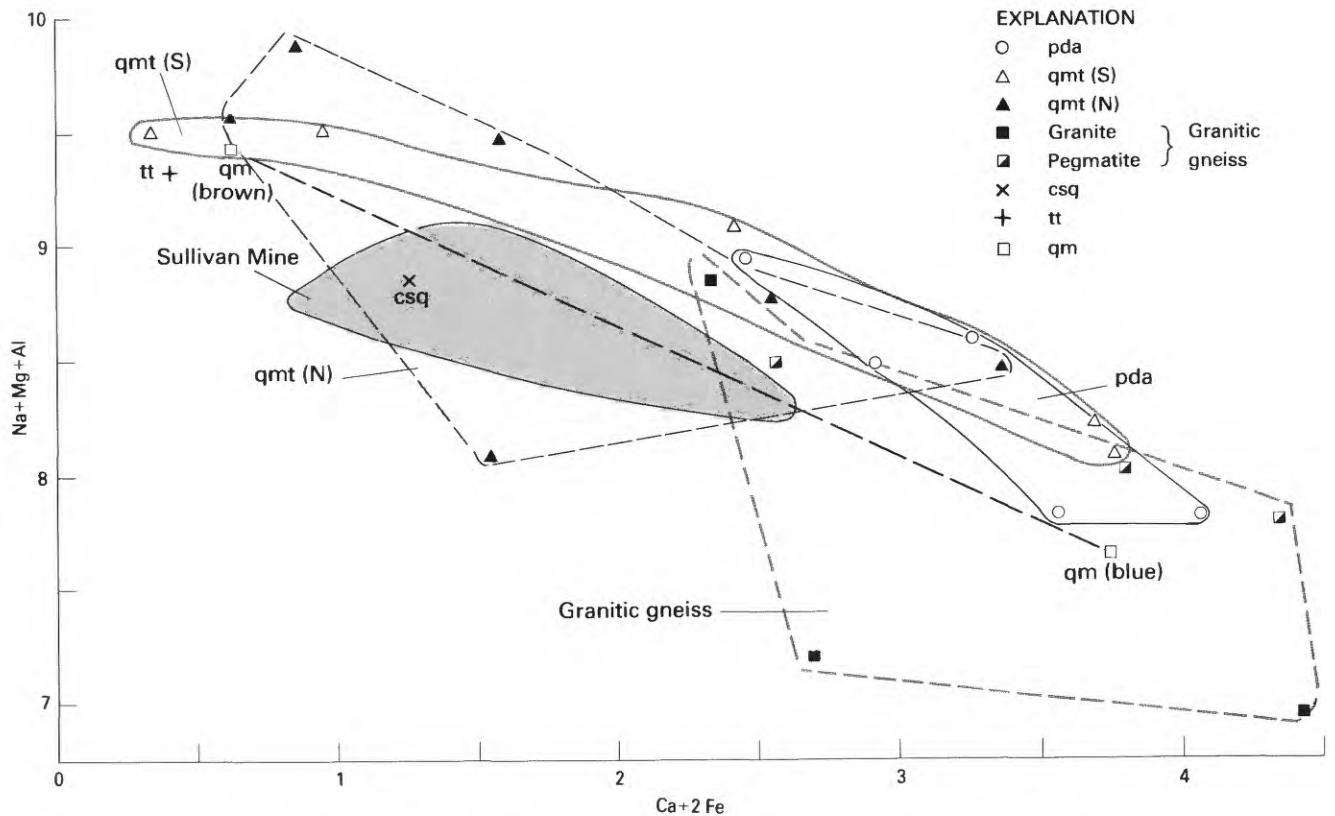


Figure 12. (Na+Mg+Al)-Ca+2Fe plot in number of ions per formula unit for tourmalines from North Gouverneur area, New York, compared with those from the Sullivan Mine, British Columbia.

In tourmalines from the Sullivan Mine, Na_2O and CaO show a well-defined antipathetic correlation (Ethier and Campbell, 1977). A similar change in tourmalines from the North Gouverneur area is in agreement with regular substitution of Ca for Na in the Z site in tourmaline from all the rock units (fig. 14). Tourmalines from potentially mineralized areas, or from different units in the North Gouverneur area, cannot be distinguished on this basis.

In contrast to tourmalines from Sullivan, those from units *hg* and *qmt* show a broad increase in Al_2O_3 with higher $\text{Na}_2\text{O}/(\text{Na}_2\text{O} + \text{CaO} + \text{K}_2\text{O})$ ratios (fig. 14). Sullivan tourmalines, on the other hand, generally show variable ratios of $\text{Na}_2\text{O}/(\text{Na}_2\text{O} + \text{CaO} + \text{K}_2\text{O})$ at about the same content of Al_2O_3 . Tourmalines from lithologic units within the North Gouverneur area, especially *qmt*, *pda*, and *hg*, show significant compositional overlap. Those from granites and pegmatites and *qmt* (North of the Oswegatchie River) show a broad positive correlation between Al_2O_3 and $\text{Na}_2\text{O}/(\text{Na}_2\text{O} + \text{CaO} + \text{K}_2\text{O})$ ratios.

The trend toward decreasing Al_2O_3 as $\text{FeO}/(\text{FeO} + \text{MgO})$ ratio increases (fig. 9), evident for individual units in the North Gouverneur tourmalines, is in contrast with the variable $\text{FeO}/(\text{FeO} + \text{MgO})$ values at constant values of Al_2O_3 in the tourmalines from Sullivan. On this basis, tourmalines from North Gouverneur and those from the Sullivan deposit are significantly different.

DISCUSSION

The geological premise for using tourmaline as a prospecting tool is that the processes of tourmalinization and ore formation are temporally and spatially related. Although the absence of plutonic bodies is a strong indication of nonigneous origin, anomalous tourmaline concentrations may, however, arise from other processes. According to Slack (1982), other processes that might lead to tourmaline concentrations include authigenic, detrital, regional metasomatic,

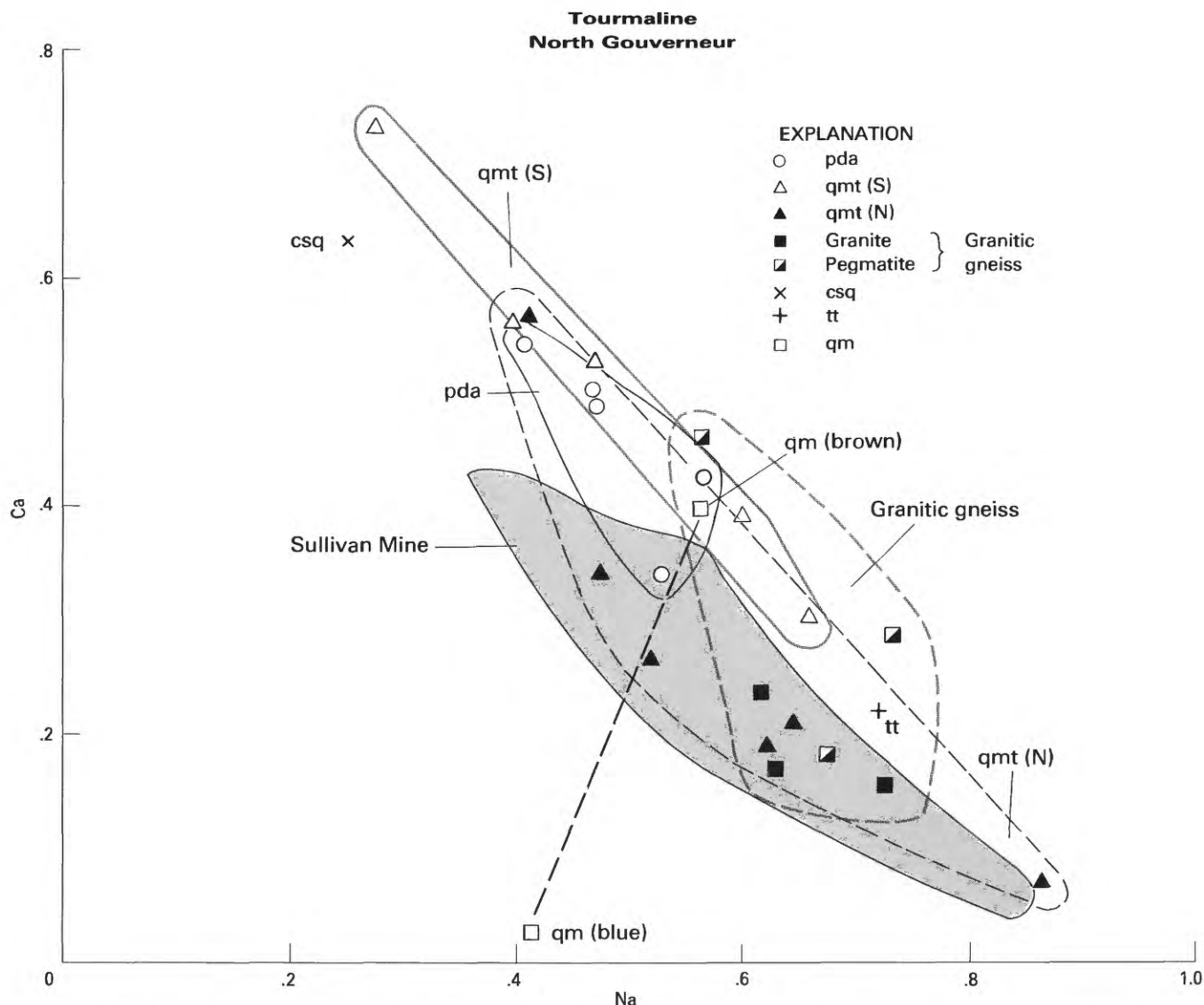


Figure 13. Ca-Na plot in number of ions per formula unit for tourmalines from North Gouverneur, N.Y., compared with those from the Sullivan Mine, British Columbia.

evaporitic-sabkha, and submarine hydrothermal. This follows because tourmaline has a wide stability field ranging from typical sedimentary environments (Krynine, 1946) to metamorphic conditions even at high pressure (Smith, 1971). Ethier and Campbell (1977) further suggested that pH ranges may be more important in determining tourmaline stability than pressure-temperature conditions. Because of this extensive stability field for tourmaline, to correlate ore deposition with tourmaline occurrence seems risky unless additional geologic information, especially modal abundance, is also taken into account. Thus, a close association between ore and tourmaline occurrence does not necessarily imply genetic equivalency. However, the anomalous occurrence of tourmaline in great abundance—that is, more than 25 percent of a rock—commonly in the

vicinity of sulfide deposits is compelling evidence of a genetic association. In addition to the anomalous concentration of tourmaline, a magnesian composition (dravite) was documented by Ethier and Campbell (1977) and Slack (1982) to characterize tourmalines intergrown with sulfides. However, Mg-rich dravites were also characteristic of tourmalines not directly associated with sulfides as depicted in Slack's compilation of selected stratabound deposits. Although Mg-rich dravites may occur without associated sulfides, the presence of anomalously large concentrations of tourmaline might still suggest proximity to stratabound sulfide deposits.

In the North Gouverneur area, tourmalinite layers in *qmt* and the ubiquitous presence of disseminated tourmaline in other rock units identify them as

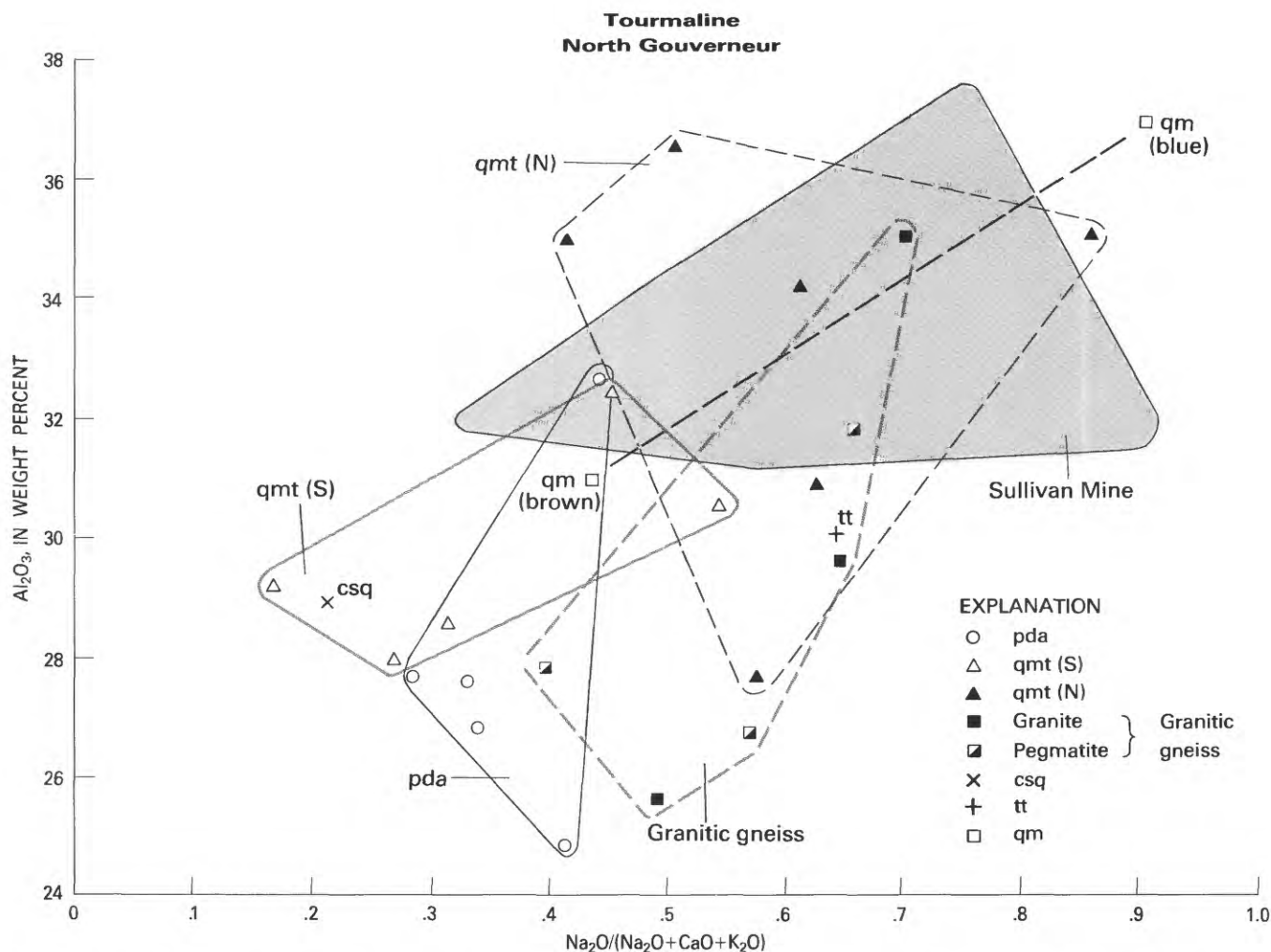


Figure 14. Al_2O_3 - $\text{Na}_2\text{O}/(\text{Na}_2\text{O}+\text{CaO}+\text{K}_2\text{O})$ plot for tourmalines from North Gouverneur, N.Y., compared with those from the Sullivan Mine, British Columbia.

anomalously abundant in tourmaline. The next step from an exploration standpoint is to compare the occurrence of tourmaline and tourmaline compositions near North Gouverneur with occurrences that are associated with major sulfide deposits. A good point of reference for such a comparison is the Sullivan ore body, a stratabound sediment-hosted deposit formed within a turbidite sequence of Middle Proterozoic age (Freeze, 1966; Hamilton and others, 1982). The abundance of carbonate rocks in the sequence under *qmt* also indicates that the rocks around North Gouverneur are similar to those near the deposits in northeastern Australia, especially at Dugald River. The deposit at Dugald River (Whitcher, 1975) is a large stratabound zinc-lead-silver lode in a local graphitic shale unit within a large area of scapolite-rich calc-silicate rocks. Scapolite-bearing calc-silicate rocks also are present in the North Gouverneur area in unit *pda*. They might represent impure lime-rich silts. In the Dugald River

deposit, parts of the carbonaceous shales commonly have 2,000 ppm boron and, locally, as much as 5,000 ppm boron. Unfortunately, tourmaline compositions in the Australian deposits are unknown, and thus our comparison with them is incomplete.

Metamorphic conditions or ultrametamorphic processes that result in boron metasomatism are two mechanisms likely to result in the formation of tourmaline. But, transport of boron-rich fluids during regional metamorphism from *qmt* into the thin granitic sheets is not supported by the different compositional trend and by the compositional variations evident in *qmt* compared with those in granite tourmalines. However, the appearance of locally migmatitic areas in *qmt* suggests that the felsic sheets may represent partial melts of the tourmaline-rich metasedimentary rocks. As documented by Chorlton and Martin (1978) and Pichavant (1981), boron significantly lowers the liquidus and solidus of granitic melts. This suggests that partial

melting in *qmt* might have been localized by the presence of abundant tourmaline resulting in boron-rich felsic melts. From such felsic liquids, it is conceivable that tourmaline precipitated in equilibrium with silicate phases and silicate liquid. Tourmaline in these felsic sheets might thus reflect igneous crystallization in contrast to the tourmaline in the metasediments that reflects metamorphic reactions. Thus, it may be suggested that the two rock types (*qmt* and the granite sheets) reflect different processes leading to tourmaline formation. Although granitic plutons are common in the North Gouverneur area, compositions of tourmaline from the metasediments can be distinguished from those in the igneous bodies.

The occurrence of such diverse tourmaline compositions indicates that bulk composition and the presence of a felsic liquid leading to igneous crystallization are responsible for the tourmaline in the granite sheets. Where tourmaline compositions show regional trends, tourmalines from igneous bodies and from metasediments have different ranges in composition.

Although compositions of tourmalines in the pegmatites in *qmt* (samples 71–124, 67–23) and *pda* (sample 68–82) are similar to those of the hosts, the granite and pegmatite tourmaline suites clearly differ from the tourmaline fields of *qmt* and *pda*. We favor the conclusion that, in the North Gouverneur area, tourmalines from igneous and metasedimentary units reflect the bulk composition of the host rocks. The implication is that the tourmalines in metasedimentary rocks are not genetically related to the igneous plutons.

In the North Gouverneur area, tourmalines from single lithologic units show significant compositional variation. For example, tourmalines from both areas of *qmt* have $\text{FeO}/(\text{FeO} + \text{MgO})$ ratios that range widely from 0 to 0.58 (fig. 6), whereas a smaller but nevertheless substantial spread is also shown by tourmalines in *pda* (0.46–0.62). Tourmaline compositions in pegmatites and migmatitic segregations in *qmt* and *pda* tend to be higher in $\text{FeO}/(\text{FeO} + \text{MgO})$ ratios than those in the host metasediments and have values more characteristic of the granite gneiss tourmalines (0.56–0.75).

Tourmalines from *qmt* are as magnesian as tourmalines from the Sullivan Mine, but they also extend to more iron-rich compositions. Preliminary analyses of tourmalines from the calcsilicate-bearing rock (*csq*), talc-tremolite schist (*tt*), and the brown tourmaline from the quartzofeldspathic lens in marble (*qm*) show compositions similar to those from the Sullivan Mine. In contrast, tourmalines from *pda*, from the pegmatites in *qmt* and *pda*, the gneissic granite *hg*, and the blue tourmaline in the quartzofeldspathic lens in marble (*qm*) are all richer in iron than tourmalines in known mineralized areas elsewhere.

At first glance, the composition of tourmaline in *qmt* together with its anomalous abundance suggests that this unit and nearby units may be favorable for containing stratabound sulfide deposits. Because of the large compositional spread, tourmaline compositions from this unit apparently reflect primary lithological heterogeneity, local metasomatic or hydrothermal processes, or the integration of several tourmaline-forming events in the area. Detailed geologic mapping shows that lithologic diversity alone in this unit may control the spread in tourmaline compositions rather than any subsequent metamorphic or hydrothermal events. Because our samples came from a restricted geographic area, the chances of regional differences in pressure and temperature were minimized. Ethier and Campbell (1977) recognized such a variation of tourmaline compositions at the Sullivan Mine, although in their samples they determined a more narrow range in $\text{FeO}/(\text{FeO} + \text{MgO})$ ratio (0.23–0.32) than the range documented at North Gouverneur.

In the course of typical metamorphic reconstitution of mineral assemblages during and after tourmaline crystallization, the $\text{FeO}/(\text{FeO} + \text{MgO})$ ratios in tourmaline may show significant departures from initial compositions obtained from the premetamorphic fluids. Continuous readjustment in the FeO content of tourmaline depending on the coexisting assemblage (bulk composition) and changing intensity of metamorphism suggests that the measured $\text{FeO}/(\text{FeO} + \text{MgO})$ ratio may represent the integration of processes during metamorphism. If the $\text{FeO}/(\text{FeO} + \text{MgO})$ ratio is continuously readjusted, the compositions of earlier formed tourmalines (perhaps in equilibrium with sulfides) are irrevocably modified during subsequent events. This suggests that $\text{FeO}/(\text{FeO} + \text{MgO})$ ratios might be inadequate to completely characterize the composition of tourmalines deposited from primary fluids, especially because Bachinski (1976) demonstrated the enrichment of iron in sulfides coexisting with silicates as the grade of metamorphism increased. Ethier and Campbell (1977) suggested that tourmaline compositional variation in the Sullivan Mine was controlled by bulk composition and by solution pathways.

Bulk composition differences in the rock units of the North Gouverneur area are the predominant controls on the composition of tourmaline. Because of the apparent variability in the protolith of unit *qmt*, it seems reasonable to expect similar variations in the composition of tourmaline even without subsequent re-equilibration. Despite the well-known large stability field for tourmaline, apparently the compositional heterogeneity reflects mainly the differences in bulk compositions within the units. The effect of local bulk composition was also recognized by Ethier and Campbell (1977, p. 2355) in the Sullivan ore body in the Aldridge Formation of British Columbia. By restricting

the area sampled in this study, we may assume that metamorphic conditions have been similar throughout. Thus, it seems unreasonable to suggest that the variability in tourmaline composition reflects reequilibration during a later tourmalinizing process. Furthermore, subsequent equilibration would strive toward compositional homogeneity rather than accentuate the differences. We conclude by suggesting that, for most samples, tourmaline compositions in the North Gouverneur area reflect the critical control of bulk composition of the rock.

Early crystallization of tourmaline is suggested by the lack of relics (as in partially recrystallized or reacted grains) and by the absence of tourmaline replacements in the North Gouverneur area. However, two exceptions are evident: (1) In sample 80-3, two tourmaline types, one in clasts and the other in the matrix, probably did not form simultaneously. The unit, where sampled, is brecciated in a fault zone, which may have served as a conduit for hydrothermal fluids. Some re-equilibration of the primary tourmaline could be expected, particularly in the matrix because of the presence of late-forming pyrite. As a result of recrystallization and coexistence with an iron sink (pyrite), the composition of tourmaline dramatically changed to the highly magnesian and variable character. (2) In sample 68-5, tourmaline from a quartzo-feldspathic lens in the marble unit shows that, during growth of the core of the grain, the composition was close to the dravite end member. As fluid conditions changed, the resulting blue tourmaline enveloping the brown interior became rich in iron and extremely poor in magnesium. The indented surface between the two compositional varieties suggests an interval of nondeposition or leaching of tourmaline before overgrowth of the blue tourmaline.

Comparison with the Balmat-Edwards District

Speculation about stratabound massive sulfide mineralization associated with boron and magnesian minerals in the tourmaline-rich rocks of the North Gouverneur area should be compared with mineralization in the well-known nearby Balmat-Edwards zinc district, 15 miles (24 km) to the southeast. The rocks of the two areas have many similarities, but also striking differences.

The ore bodies at Balmat occur in a thick heterogeneous sequence of interlayered pure and quartz- or calc-silicate-bearing dolomitic marbles, described by Brown and Engel (1956). The ore consists of sphalerite, pyrite, and minor galena that occur in several of the marble units. Interlayered in the mineralized zones are two significant stratigraphic markers that bear important genetic implications. One contains much

anhydrite, serpentine, talc, and diopside and is recognized as a metamorphosed evaporite (Lea and Dill, 1968, p. 27). The other marker is talc-tremolite schist. The protolith of the schist, if isochemical reactions with loss of CO₂ are considered, possibly was a siliceous dolomite containing magnesite—this also suggests an evaporitic origin. Ore minerals are believed to have formed syngenetically (Solomon, 1963; Dill and deLorraine, 1978), and their association with evaporitic sediments suggests a related origin.

In the North Gouverneur area, dolomitic and silicated marbles in *qm* and also the talc-tremolite schist (*tt*) are similar to some rocks of the Balmat-Edwards district. In addition, occurrences of stratabound sphalerite are also known in the marble unit (Brown, 1970; 1983). However, in contrast to the ubiquitous occurrence of tourmaline either as abundant accessory minerals or as tourmalinites in the sequence near North Gouverneur, the metasediments of the Balmat-Edwards district contain insignificant amounts of tourmaline (Brown and Engel, 1956).^{1,2} Another important difference is that in the Gouverneur-North Gouverneur area the marbles are nearly all graphitic and phlogopitic, whereas those in the mining district (except for Brown and Engel's (1956) units 7 and 15) do not have notable phlogopite or graphite.

Probably the most significant difference between the two areas is the scarcity of tourmaline in Balmat-Edwards rock units, and thus an absence of significant boron in the environment of deposition. Thus, the mineralization in carbonate rocks at Balmat and the implied mineralization in the tourmaline-rich clastic rocks of the North Gouverneur-Richville area are not closely related—if at all—because of the special conditions necessary to produce an excess of boron in the rocks.

Tourmaline Genesis in the North Gouverneur Area

Tourmaline is ubiquitously disseminated in rocks of the North Gouverneur area where it has concentrations as high as 50 modal percent in thin tourmalinite layers in feldspathic quartzites. Similar layers that have 35 percent tourmaline in the Aldridge Formation of British Columbia were estimated by Ethier and Campbell (1977, p. 2359) to contain 12,000 ppm boron. Such concentrations, according to them, are anomalously

¹Fronde! and Baum (1974, p. 178) cited Brown and Engel (1956) and Lea and Dill (1968) for noting boron-rich rocks in their reports on the Balmat district. A thorough search of these reports and personal knowledge of the area proved this citation to be erroneous.

²The authors have reported on a rare occurrence of manganese-rich tourmaline in the talc-tremolite schist in the Fowler talc belt of the mining district. However, the quantity of tourmaline there is miniscule (Ayuso and Brown, 1984).

high. Their studies of the rocks close to the Sullivan ore body suggest that boron was “* * * introduced at or near the depositional surface * * *,” thus discounting the effect of boron transport by residual magmatic fluids after sedimentary lithification. According to Ethier and Campbell (1977), metal- and boron-rich fluids used the same pathways (collapse breccias) upward to the sea floor; consequently, the direct precipitation of anomalous concentrations of tourmaline is closely associated with sulfide deposition. No evidence of evaporitic conditions was observed by them (1977, p. 2359). In contrast to the turbidite sequence prevailing at Sullivan, tourmalines from North Gouverneur are more clearly associated with rocks formed under evaporitic conditions (Brown, 1969, Moine and others, 1981, p. 411).

The Dugald River lode in Australia occurs in graphitic shales and slates containing cordierite, andalusite, and accessory tourmaline (Whitcher, 1975). Evaporitic rocks are both higher and lower in the section, and these are represented by scapolite granulites and argillaceous limestone, respectively. As seen from figure 2, unit *qmt* in the North Gouverneur area also is enclosed by carbonate and calc-silicate rocks above and below. Mineralization at Dugald River consists of pyrrhotite, pyrite, and sphalerite in roughly subequal amounts, subordinate galena, and traces of chalcopyrite, pyrrargyrite, tetrahedrite, and arsenopyrite (Whitcher, 1975). Although rocks of unit *qmt* are pyrite rich in places, no substantial concentrations of other sulfide minerals have been found, but an occurrence of chalcopyrite has been noted (Brown, 1983, loc. 53). Despite the lack of exposed significant sulfides here, the analogy to Dugald River appears especially applicable.

Whitcher (1975) proposed, as an ore-producing model, “chemical or biochemical precipitation of metal sulfides” in the Dugald River lode in a downwarped basin, dominated by carbonate rocks. Formation in a sabkha environment of stratiform metalliferous sediments was advanced by Renfro (1974) to account for deposits such as the Kupferschiefer (Germany and Poland) and the Roan (Zambia, Zimbabwe, and Zaire). Dugald River and this group of deposits may well have similar origins. Tourmaline occurs in anomalous concentrations closely associated to mineralized areas in the Zambian Copper belt (Mendelsohn, 1961; Garlick, 1967), especially near the Chibiluma deposits. Abundant tourmaline occurs as stumpy crystals below the sulfide-rich horizons at Chibiluma and persists as fine-grained tourmaline in the sulfide-rich beds (Fleischer and others, 1976). Unfortunately, the role of tourmalinization in these environments has not been properly addressed. Because of the lack of definitive data, we cannot assume that, as was true in the Sullivan deposit, tourmaline genesis and mineralization took place simultaneously. This is because, in evaporitic environments,

an additional boron source is available, namely from borate minerals (Rankama and Sahama, 1950), in contrast to the external source for boron formed in turbidite environments (Sullivan deposit), where boron- and metal-rich brines were introduced concurrently.

At least two possibilities may be envisioned to explain tourmaline origin in the North Gouverneur area: (1) tourmalines were generated by diagenesis or regional metamorphism of borate minerals, and (2) tourmalinites and accessory tourmaline result from submarine fumarolic activity.

Because of the possible primary abundance of borate minerals within an evaporitic section, tourmalines in the Gouverneur area might have formed as a result of normal diagenetic or metamorphic reactions involving minerals already enriched in boron during a regional event, and need not be associated with metal-rich brines. Presence of tourmalinite in nonevaporitic areas, on the other hand, is consistent with the idea that conformable and anomalous concentrations of quartz and tourmaline (tourmalinite) reflect precipitation from oozes at the sea floor (Ethier and Campbell, 1977, p. 2359). Or alternatively, in evaporitic terranes, because of the high solubility of alkali borates, boron may have existed in solution in basin pore fluids until crystallization as tourmaline during diagenesis or metamorphism.

The origin of the tourmalinites is unexplained by direct reconstitution of borate minerals (borate to tourmaline) as in alternative 1, unless the original lithology consisted of a mix of silica ooze and borates. Anomalous concentrations of tourmaline and quartz in tourmalinites from nonevaporitic terranes are more conventionally explained by deposition at the sea floor having an external boron source, although Abraham and others (1972) suggested that the tourmalinites may represent volcanic exhalations or highly saline environments. Field observations such as thickness and abundance of tourmalinite beds, as well as the evaporitic geological features in the North Gouverneur area, seem to exclude direct recrystallization of borate minerals and metasomatic quartz-tourmaline “sweat-outs,” but allow the crystallization of tourmaline from boron-rich basinal brines. Remobilization by pore fluids and concentration of dispersed borates in the section to account for the origin of tourmalinite are generally in agreement with the conformable attitude of tourmalinites, their thickness, and their random distribution. Thus, because of the strong possibility of borate minerals intercalated within the section, tourmaline growth in the North Gouverneur area may be a result of remobilization of borates by basinal fluids during diagenesis and metamorphism. Because the tourmalinite layers are more abundant in some units (for example, *qmt*), bulk composition as in the typical boron-rich (borate-rich) layers of the protolith might have controlled the concentration

of tourmaline. Within these anomalous boron-rich units, tourmaline fixed the B_2O_3 in a manner similar to that proposed by Kulke (1976) for fixation of B_2O_3 by tourmaline in a metaevaporite environment. Kulke and Schreyer (1973) also documented the occurrence of tourmaline (dravite) in metaevaporites containing lapis lazuli in silicate marbles that enclose concordant beds of lime silicate and amphibolite units.

Because of the difference in geologic setting between the North Gouverneur area and the Sullivan Mine, we discount the obvious connection of tourmaline crystallization to fumarolic activity as in alternative 2. We emphasize that, as originally proposed by Ethier and Campbell (1977) and Slack (1982), the anomalous concentration of tourmaline remains the central issue to be explained for the potential use of tourmaline as an exploration guide. However, although magnesian tourmalines seem to be the norm in known mineralized areas, the composition of tourmaline may be significantly readjusted as a result of recrystallization. Thus, the composition of tourmalines closely associated with sulfides does not necessarily represent the equilibrium attained for the observed assemblage of tourmalines and sulfides.

Terranes Showing Most Favorable Implications for Containing Stratabound Sulfide Deposits

Occurrences of abundant magnesian tourmaline in metasedimentary rocks have been proposed as a prospecting guide. An objective of this study of tourmaline compositions was to determine whether anomalously abundant tourmaline in rocks in the North Gouverneur area was chemically similar to tourmalinites associated with stratabound sulfide deposits, such as at Sullivan, British Columbia.

Lithologies in rock unit *qmt*, particularly those for samples 67-21, 67-25, and 67-26 (fig. 2), show the strongest chemical similarities to the tourmalines at Sullivan in that they are dravitic, have low TiO_2 and FeO , high Al_2O_3 , low $FeO/(FeO + MgO)$ and highly variable $Na_2O/(Na_2O + CaO + K_2O)$ ratios. Similar tourmaline-rich lithologies occur throughout the area of *qmt* north of North Gouverneur. Also, Bannerman (1972) showed many other localities of tourmaline-rich rocks in the zone along the Oswegatchie River that were not adequately appraised in this study (fig. 2). However, careful study of the stratigraphic section in the vicinity of North Gouverneur reveals that its depositional origin is distinctly different from that of the rocks at Sullivan, British Columbia, and the ore-producing model there does not apply here because these rocks have a strong mineralogic affinity to rocks of an evaporitic origin. At many places such as Dugald River, in northeastern

Australia, rocks formed in an evaporitic environment contain huge stratabound sulfide deposits. Many of these also are rich in tourmaline, but no chemical data are available for comparison. Nevertheless, these evaporite-associated deposits widen the range of lithologic varieties that could be mineralized—namely, graphitic schists that are included within scapolite- and calc-silicate-rich rocks. Numerous occurrences of such rock have been observed in the belt of rock within 2 miles northwest of Beaver Creek, shown by Buddington (1934, Hammond sheet) as crystalline limestone containing linear lenses of white quartzite. These rocks are tourmaline-bearing, graphitic, and are associated with diopside-scapolite-rich rocks. These are all similar to rock types that are associated with Dugald River-type deposits in Australia. Further appraisal of these for sulfide deposits requires geophysical or geochemical techniques.

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