The Canadian Mineralogist
Vol. 41, pp. 1381-1396 (2003)

COMPOSITION OF BIOTITE FROM GRANITIC ROCKS OF THE CANADIAN APPALACHIAN OROGEN: A POTENTIAL TECTONOMAGMATIC INDICATOR?

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ABSTRACT

Biotite, the dominant ferromagnesian mineral in Paleozoic granitic rocks of the Canadian Appalachians, has been analyzed with an electron microprobe (wavelength dispersion) for major elements and by ^{57}Fe Mössbauer spectroscopy. We sampled a wide variety of rock types, ranging from gabbro, diorite, syenite to granite, but by far mostly granitic (*sensu lato*). The most pronounced variations are in total Al contents and Fe/(Fe + Mg) values. In the biotite quadrilateral (annite –siderophyllite – phlogopite – eastonite), biotite from A-type granites of the Humber and Avalon zones in Gaspé (Québec) and New Brunswick is characterized by low mean Al contents, ~1.15 atoms per formula unit (*apfu*), and variable Fe/(Fe + Mg) values in the range 0.4 to 0.9. In the granites of the Notre Dame arc of the Dunnage zone in Newfoundland, biotite has moderate mean values of Al (~1.40 *apfu*) and Fe/(Fe + Mg) (~0.58). In granites of the Gander zone of New Brunswick and Newfoundland, biotite has a mean Fe/(Fe + Mg) value of 0.6 and shows a pronounced trend of increasing total Al (1.05 to 1.75 *apfu*), confirming significant contributions of aluminous supracrustal material to the magmas, either by assimilation or anatexis. Finally, in granites of the Meguma zone, derived entirely from metasedimentary material, biotite exhibits a remarkable increase in total Al (1.30 to 2.00 *apfu*) and considerable iron-enrichment [Fe/(Fe + Mg) in the range 0.4 to 1], with compositions nearing the siderophyllite end-member. The biotite from most zones plots on or above the NNO buffer, indicating moderately oxidizing conditions, whereas that from the Meguma zone plots mainly between the QFM and NNO buffers, implying fairly reducing conditions during crystallization. Assuming a reasonable range of crystallization temperatures of 750 to 900°C, oxygen fugacities ranged from 10^{-10} to $10^{-16.9}$ bars during crystallization. The composition of biotite reflects primarily the nature of the host magmas. It cannot readily be used for tectonomagmatic characterization of these rocks without the aid of other types of data.

Keywords: biotite, chemical composition, granite, annite, phlogopite, Mössbauer spectroscopy, tectonic discrimination, Appalachian orogen, Canada.

SOMMAIRE

Des échantillons de biotite, minéral ferromagnésien dominant dans les roches granitiques des Appalaches canadiennes, ont été analysés avec une microsonde électronique et par spectrométrie de Mössbauer. Ces échantillons proviennent de roches plutoniques allant de gabbro, diorite, syénite jusqu'à granite, mais surtout granitiques (*sensu lato*). Les variations compositionnelles les plus prononcées se situent au niveau des teneurs en Al et du rapport Fe/(Fe + Mg). Dans le quadrilatère compositionnel de la biotite (annite – sidérophyllite – phlogopite – eastonite), la biotite des granites de type A des zones Humber et Avalon de la Gaspésie (Québec) et du Nouveau-Brunswick se caractérise par des teneurs en Al faibles (~1.15 atomes par formule unitaire, *apfu*), et par des valeurs Fe/(Fe + Mg) variant de 0.4 à 0.9. Dans les granites de l'arc Notre Dame de la zone Dunnage de Terre-Neuve, la biotite possède des valeurs modérées de Al (~1.40 *apfu*) et du rapport Fe/(Fe + Mg) (~0.58). Dans les granites de la zone Gander du Nouveau-Brunswick et de Terre-Neuve, la biotite possède un rapport Fe/(Fe + Mg) moyen de 0.6 et définit une lignée prononcée vers un enrichissement en Al total (1.05 à 1.75 *apfu*), confirmant une contribution importante de matériau supracrustal alumineux aux magmas, soit par assimilation ou encore par anatexie. Finalement, dans les granites de la zone de Meguma, ceux-ci dérivés entièrement de matériau métasédimentaire, la biotite démontre un enrichissement prononcé en Al total (1.30 à 2.00 *apfu*) et en fer [Fe/(Fe + Mg) de 0.4 à 1], avec des compositions qui se rapprochent du pôle sidérophyllite. La biotite de la majorité des zones se trouve sur ou au-dessus du tampon NNO, indiquant des conditions assez oxydantes, tandis

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que celle de la zone de Meguma se retrouve principalement entre les tampons QFM et NNO, indiquant des conditions beaucoup plus réductrices. En supposant une température de cristallisation de 750 à 900ºC, nous obtenons des pressions partielles d'oxygène de 10^{-10} à $10^{-16.9}$ bars pendant la cristallisation. La composition de la biotite des roches granitiques reflète surtout la nature du magma duquel les roches ont cristallisé. Cependant, nous ne croyons pas que cette composition puisse servir par elle-même à distinguer le contexte tectonomagmatique; l'apport d'autres types de données sera nécessaire.

(Traduit par la Rédaction)

Mots-clés: biotite, composition chimique, granite, annite, phlogopite, spectroscopie de Mössbauer, discrimination tectonique, orogène des Appalaches, Canada.

INTRODUCTION

Numerous studies have been published that demonstrate the use of biotite composition as a valuable guide to granite petrogenesis [*e.g.,* Dodge *et al.* (1969), Barrière & Cotton (1979), Neiva (1981), Speer (1981), Ague & Brimhall (1988), Finch *et al.* (1995)]. In addition, the classical experimental work of Wones & Eugster (1965) clearly established this mineral as a valuable indicator of redox conditions in granitic magmas. More recently, investigators have explored in considerable depth the geobarometric and geothermometric potential of this mineral (Patiño-Douce 1993, Patiño-Douce *et al.* 1993). In addition, the composition of biotite can be used in some instances as a tectonomagmatic indicator [*e.g.,* Nachit *et al.* (1985), Lalonde & Bernard (1993), Burkhard (1993), Abdel-Rahman (1994)].

Trioctahedral true micas of the annite–phlogopite, eastonite–siderophyllite solid solutions, *i.e.,* biotite, are found in nearly all types of granites and granitic rocks. By virtue of their crystal structure, these micas can accommodate most of the common elements present in granitic magmas. The following features make biotite a valuable probe of magma composition: 1) It is the most important reservoir of any excess aluminum in granites that do not have important amounts of garnet, cordierite, or the Al_2SiO_5 polymorphs; therefore, it directly reflects the peraluminosity of the host magma in such rocks, and, 2) it is the most readily available indicator of oxidation state. Magma peraluminosity and relative oxidation state have been the basis for the classification of granites into I and S types (Chappell & White 1974) and into those of the ilmenite and magnetite series (Ishihara 1977, 1981), respectively. Recently, granitic rocks of the different tectonostratigraphic zones of the Canadian Appalachian orogen (Fig. 1) have been the object of several detailed geological, geochemical and isotopic investigations aimed primarily at defining their source materials (*e.g.,* Whalen 1993). In addition, these investigators have addressed the topic of relationship between granitic magmatism and tectonism. The comprehensive dataset that exists for granites from the Canadian Appalachian provides an ideal framework with which to explore compositional relationships of biotite in granites of differing tectonic settings and to further elucidate the petrogenesis of these intrusive rocks.

In this study, we document by electron microprobe the chemical composition of over 159 biotite specimens taken from 76 different sites from the Canadian Appalachian orogen. Of these specimens, 54 were studied by Mössbauer spectroscopy to document the oxidation state of iron. In addition, results of 164 analyses of biotite taken from the literature have been included. This dataset constitutes the most complete one on igneous biotite ever undertaken in the Appalachian orogen.

SOURCES OF DATA, SPECIMENS AND ANALYTICAL METHODS

This study was greatly facilitated by the availability of a sizable collection of specimens of granitic rock with corresponding major, minor, trace element and isotopic whole-rock data from the Humber zone of Québec, the Dunnage, Gander and Avalon zones of New Brunswick, and the Dunnage zone of Newfoundland. The collection was amassed by one of us (JBW) in the course of regional field-oriented research at the Geological Survey of Canada. Specimens of granites from the Gander zone in Newfoundland were collected by the first two authors in August of 1995. Additional specimens were provided by Dr. A. Kerr of the Geological Survey of Newfoundland and Labrador, and by Professor D.B. Clarke of Dalhousie University. Some data were also taken from the literature (Table 1).

Minerals were analyzed by a combination of wavelength-dispersion X-ray spectrometry with the electron microprobe and by Mössbauer spectroscopy. Representative Mössbauer spectra of biotite are shown in Figures 2A and 2B. Their interpretation is discussed in Appendix 1. Whole-rock compositions were determined by X-ray fluorescence spectroscopy. Analytical methods are described in Appendix 1.

The location, geochronology and a brief description of the petrography of all the plutonic rocks used in this study are summarized in Appendices 2a and 2b, available from the Depository of Unpublished Data of CISTI, National Research Council of Canada (cisti.info @nrc.ca).

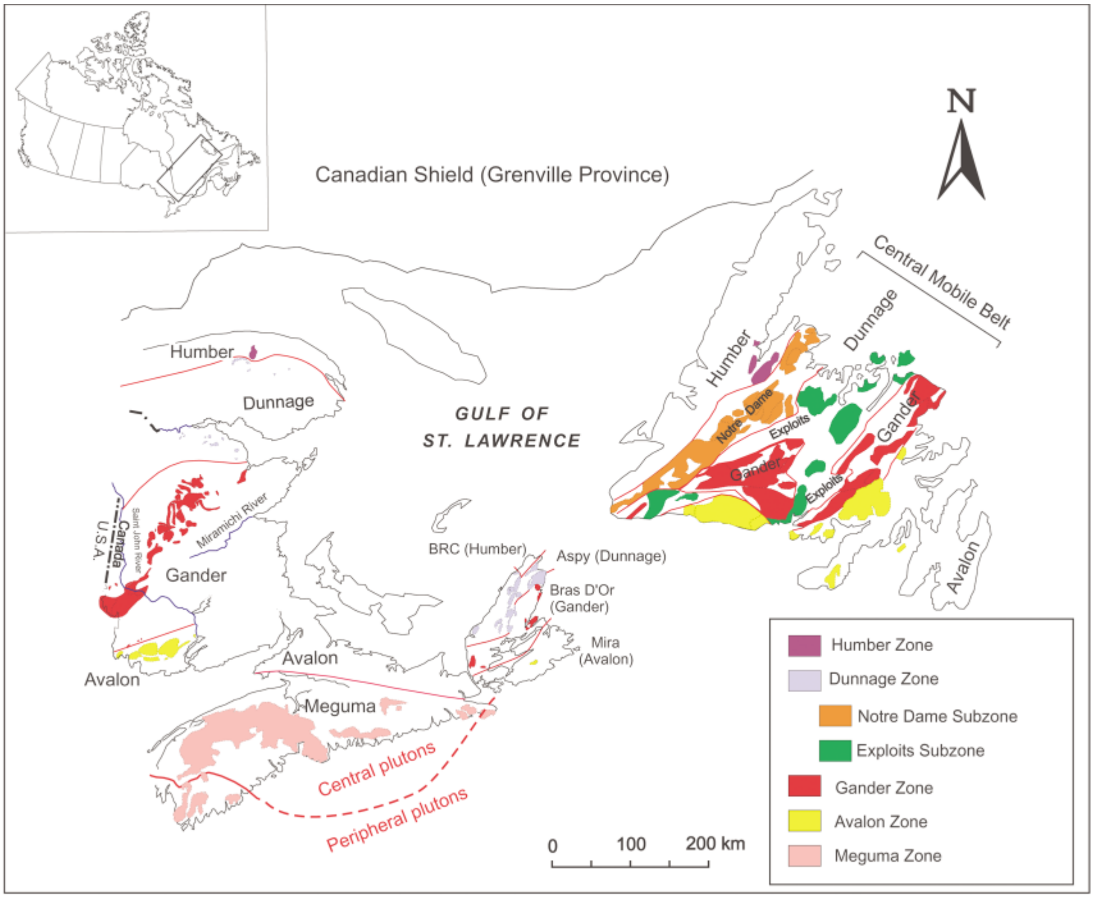

FIG. 1. Distribution of Paleozoic granitic rocks in tectonic zones of the Canadian Appalachians. Compiled from Whalen (1993), Tate & Clarke (1997), and Kerr (1997).

PLUTONISM IN THE CANADIAN APPALACHIAN OROGEN

The Canadian Appalachian orogen is generally interpreted in terms of the opening and closing of the Late Precambrian – Early Paleozoic Iapetus ocean. The timing for the opening of the Iapetus is well constrained to the Late Precambrian, whereas closure was at some time during the Middle Ordovician to Early Devonian interval. The orogen (Fig. 1) comprises five main tectonostratigraphic zones (Williams 1979, Williams *et al.* 1988). From west to east, these are: 1) the Humber zone, the western margin of the Iapetus ocean or the ancient North American continental margin, composed of supracrustal and ultramafic allochthons, 2) the Dunnage zone, composed of allochthonous oceanic remnants of Iapetus, 3) the Gander zone, a metasedimentary belt, possibly an opposing Gondwanan continental margin, 4) the Avalon zone, a Late Proterozoic orogenic province with a thin Paleozoic cover sequence, probably a fragment of Gondwanaland accreted during the Pan-African cycle and, finally, 5) the Meguma zone, the easternmost lithotectonic terrane in the Appalachian orogen, situated south of the Avalon zone of northern Nova Scotia and composed largely of Paleozoic rocks (Williams 1979) and believed to be an exotic terrane (Schenk 1971).

In New Brunswick, the Dunnage zone is mainly obscured by younger sequences. In Newfoundland, where it is well exposed, it has been subdivided into the Notre Dame arc and the Exploits subzone, each of which has distinct lithological, faunal and evolutionary characteristics (Williams *et al.* 1988). The southern part of the Notre Dame arc (or subzone), a region underlain mainly by high-grade ortho- and paragneisses, was redefined as the Dashwoods subzone by Piasecki *et al.* (1990).

Plutonic rocks comprise about one quarter of all exposed rocks in the Canadian Appalachians and are found in all five tectonostratigraphic zones mentioned above (Currie 1995). These rocks are granitic in the broad sense, meaning that for the most part they are members of the tonalite to alkali-feldspar granite suite. Much of this granitic plutonism occurred during Middle Ordovician to Early Devonian times (Whalen 1993). Major tectonic models of granite genesis for Newfoundland (Kerr 1997, Kerr *et al.* 1992) and New Brunswick (Whalen 1993, Whalen *et al.* 1997) associate plutonism to post-collisional lithospheric delamination. Details of the geological, petrological, geochemical and isotopic characteristics of these suites and plutons have been discussed elsewhere (Whalen 1993, Whalen *et al.* 1997, Kerr 1997, Clarke *et al.* 1997); a brief review is given here, however.

TABLE 1. DATA ON BIOTITE COMPOSITION IN GRANITIC ROCKS OF THE APPALACHIAN OROGEN, INCLUDED IN THIS STUDY FROM OTHER SOURCES

zone	number of samples	locality	source
Meguma zone, Nova Scotia	24	South Mountain batholith	Stallard (1975)
	8	South Mountain batholith	Allan (1980)
	14	South Mountain batholith	Maillet (1984)
	10	South Mountain batholith	Ding (1995)
	4	South Mountain batholith	D.B. Clarke (unpubl. data)
	12	Musquodoboit batholith	MacDonald (1981)
	12	Plutons of Canso and Forest Hill area	Hill (1991)
	29	Port Mouton pluton	Douma (1988)
	23	Halfway Cove – Queensport pluton	Ham (1988)
Humber zone, Québec	25	McGerrigle complex	Wallace (1988)
Dunnage zone, Newfoundland	6	Topsail igneous complex	J.B. Whalen (unpubl. data)
Gander zone, Newfoundland	18	Granitic rocks of the Bay d'Espoir region	Bennet (1990)

Plutonism in the Humber zone in Québec is sparse and essentially limited to the Devonian McGerrigle complex, which is composed of metaluminous amphibole-bearing syenite, monzonite, granite, gabbro and associated hybrid rocks that truncate deformation fabrics in the allochthons. According to Whalen (1993), these rocks were derived from igneous or reworked mantle-derived (infracrustal) protoliths.

Plutonic rocks in the Dunnage zone include granite, granodiorite, tonalite, diorite and gabbro. In New Brunswick, these rocks are Siluro-Devonian in age, whereas in Newfoundland, they are Early to Middle Ordovician. On this basis of geochemical and isotopic

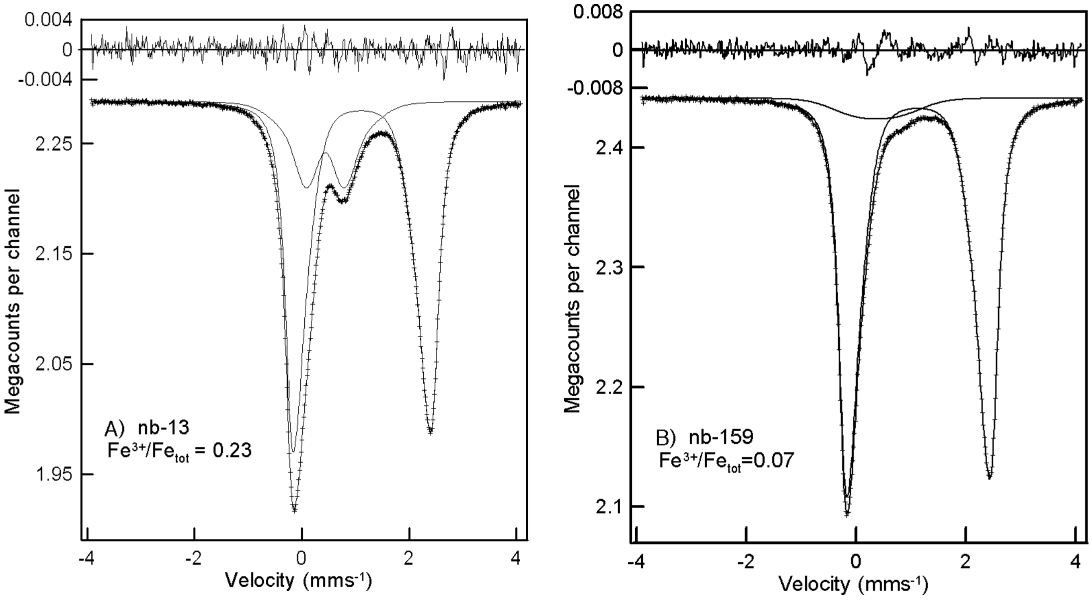

FIG. 2. Representative Mössbauer spectra of biotite from Appalachian granites. Both spectra were acquired at room temperature; see Appendix 1 for more details on the analytical methods. The curve joining the data points represents the fitting model. Other curves show the contributions of $^{VI}Fe^{2+}$ and $^{VI}Fe^{3+}$. The difference spectrum is show at the top with an exaggerated abscissa. A) Specimen nb–13, Pokiok batholith, Gander zone in New Brunswick. B) Specimen nb–159, Lost Lake pluton, Gander zone in New Brunswick.

signatures, Whalen *et al.* (1997) proposed that these rocks originated from both mantle- and slab-derived magmas that were modified through assimilation – fractional crystallization (AFC) processes in the lower crust, and also from partial melts of solidified, previously hybridized rocks of the lower crust.

The Gander zone is the site of voluminous granitic plutonism in the Canadian Appalachian orogen. Ages span the Middle Ordovician, Silurian and Devonian periods. Here, the rocks are predominantly biotite granite *sensu stricto* that in some cases are K-feldspar megacrystic or muscovite-bearing. More calcic and quartz-poor rocks (*i.e.,* granodiorite, tonalite, and gabbro) are of minor importance. Geochemical and isotopic studies of these rocks indicate that they are derived from a combination of old Precambrian basement, Paleozoic cover rocks and some mantle-derived material (Kerr *et al.* 1995).

Within the Avalon zone, plutonism is limited to the Siluro-Devonian St. George batholith in southern New Brunswick, emplaced near the tectonic junction with the Gander zone. This batholith is composed of metaluminous, biotite or biotite–amphibole granite and granodiorite derived from I-type or mantle-derived sources (Whalen 1993). Some granites of this suite are A-type and peralkaline.

Approximately one third of the Meguma zone is composed of Devonian granitic plutons that are best classified as ilmenite-series or S-type granites (Clarke & Muecke 1985). Two types are recognized by their spatial, genetic and chemical characteristics (Fig. 1) (Clarke *et al.* 1997); 1) the central plutons, slightly younger (372 Ma) and composed of strongly peraluminous garnet-, cordierite- and even andalusite-bearing monzogranite or granodiorite and, 2) the peripheral plutons, slightly older (378 Ma) and composed of more calcic, hornblende- or biotite-bearing tonalite, granodiorite and monzogranite. Mafic rocks are minor.

BIOTITE IN THE PLUTONIC ROCKS

In this manuscript, we use the term "biotite" in the sense of the IMA classification on micas (Rieder *et al.* 1998), *i.e.*, as a series name for true trioctahedral micas between, or close to, the annite [$KFe^{2+}{}_3AlSi_3O_{10}(OH)_2$] – phlogopite [$KMg_3AlSi_3O_{10}(OH)_2$] and siderophyllite [$KFe^{2+}AlAl_2Si_2O_{10}(OH)_2$] – eastonite [$KMg_2AlAl_2Si_2O_{10}(OH)_2$] series. Biotite is the principal mafic mineral in the granitic rocks of the Canadian Appalachian orogen. It occurs as subhedral to euhedral flakes that vary from 0.5 to 5 mm in diameter. In the majority of rocks, the textures are consistent with a primary magmatic origin. In the older deformed units, the biotite defines a planar fabric and is interpreted as a recrystallized phase. Biotite from the rocks of the Gander zone in Newfoundland and New Brunswick displays an interesting contrast of pleochroism. In those from Newfoundland, biotite is dark brown to greenish brown pleochroic, whereas in those from New Brunswick, it is more reddish, suggesting higher total Fe contents, since Ti values for both zones are identical (Lalonde & Bernard 1993). Chlorite commonly is present as a product of alteration of the biotite. It occurs interleaved with the mica along the {001} cleavage and in some cases may completely replace it. Prehnite, pumpellyite and grossular–andradite garnet have also been observed as lenticular patches along {001}. In all cases, analyzing the fresh biotite between the altered zones was easily done with the electron microprobe.

COMPOSITION OF THE BIOTITE

The 323 micas analyzed from granitic rocks of the Canadian Appalachians are all trioctahedral true micas ($2.51 < M$ site < 2.99 atoms per formula unit) and, for the most part, annite. Twelve representative compositions based on $O_{10}(OH, F,Cl)_2$ are presented in Table 2, and the compositional features of all the micas studied are summarized in Table 3.

Stallard (1975) determined the Fe^{3+}/Fe value of biotite by conventional wet-chemical methods in a study of biotite from the South Mountain batholith. He obtained Fe^{3+}/Fe values ranging from 0.06 to 0.37 for 18 specimens. Our spread of Fe^{3+}/Fe values, from 0.09 to 0.19, obtained by Mössbauer spectroscopy on eight specimens of this batholith and two specimens of the Port Mouton pluton, is considerably smaller. In fact, our Mössbauer determinations of the Fe^{3+}/Fe value of all 54 mica specimens from our suite of granitic rocks of the entire Canadian Appalachians, whether oxidized or reduced, range only from 0.05 to 0.23. This comparison demonstrates the poor precision of conventional wet-chemical determinations, which are subject in most laboratories to high experimental errors, and also overestimate ferric iron contents because of oxidation of Fe^{2+} during the acid decomposition.

PERALUMINOSITY OF BIOTITE AND HOST ROCKS

Biotite is perhaps the most common mineralogical sink for excess aluminum in granitic rocks in general. This is the case for our Appalachian specimens, as demonstrated in a plot of the peraluminosity index (A/CNK) of biotite *versus* that of the corresponding whole-rock (Fig. 3). In this plot, biotite falls above the 1:1 line, indicating clearly its higher A/CNK value relative to that of its host rock. Only two data points on the diagram have biotite A/CNK values that are lower than those of the corresponding whole-rock. For these two specimens, which are biotite–muscovite granites from the Gander zone in New Brunswick, the whole-rock peraluminosity must be attributed, at least in part, to muscovite. There is also a fairly strong linear correlation between the biotite and the whole-rock A/CNK values. A small group of points at low whole-rock A/CNK values falls off this trend. These points correspond to gabbro and other mafic rock units, in which amphibole and pyroxene are important mafic phases.

DETERMINATION OF OXYGEN FUGACITY

In Figure 4, the composition of biotite from the granitic rocks of the Canadian Appalachians is plotted in the Fe^{2+} – Fe^{3+} – Mg diagram of Wones & Eugster (1965), along with the three common oxygen fugacity buffers quartz – fayalite – magnetite (QFM), nickel – nickel oxide (NNO) and hematite – magnetite (HM). In general, the majority of compositions plot on or above the NNO buffer, and a few fall on or above the QFM buffer.

Figure 4A shows that biotite from the Humber–Dunnage zone falls between the NNO and HM buffers. The few compositions that plot near the HM line, indicating fairly oxidizing conditions, come from samples that have coexisting magnetite and ilmenite. Biotite from the Dunnage zone in Newfoundland plots mainly between the NNO and HM lines, but with some compositions falling between the NNO and QFM buffers (Fig. 4B). In general, these compositions define general trends of increasing Fe^{3+} with evolution (as defined by increasing total iron), suggesting late oxidation in these suites. Samples from the Dunnage zone coexist with an oxide assemblage of magnetite – ilmenite. Magnetite or ilmenite was only rarely seen as single oxide phases in the rocks of this zone. Other opaque minerals that may accompany magnetite and ilmenite are chalcopyrite, pyrite and hematite. Titanite, as a primary mineral, is uncommon, but it does occur in some samples of the Dunnage zone.

The majority of biotite compositions from the Gander zone in New Brunswick also fall on or above the NNO buffer (Fig. 4C). However, biotite compositions from the Gander zone in Newfoundland define an elongate cluster that falls mainly on the NNO buffer (Fig. 4D). In the rocks of these two zones, the main opaque phases are magnetite + ilmenite ± chalcopyrite ± pyrite ± hematite. The magnetite to ilmenite ratio varies from rock to rock. Titanite as a primary accessory mineral occurs mainly in hornblende-bearing granites.

In the Meguma zone, biotite compositions plot between the QFM and NNO buffers (Fig. 4E), like those of ilmenite-series granites in Japan (Ishihara 1977), indicating fairly reducing conditions. The oxide mineral in the Meguma granites is ilmenite.

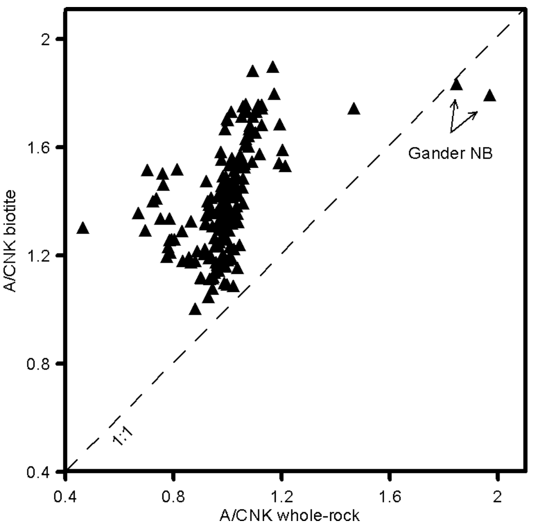

FIG. 3. Plot of A/CNK ratio [molar $Al_2O_3/(CaO + Na_2O + K_2O)$] of biotite *versus* whole-rock values for all specimens studied. All biotite specimens, except for two samples from biotite–muscovite granite of Gander zone in New Brunswick, are considerably more aluminous than their corresponding whole-rock values.

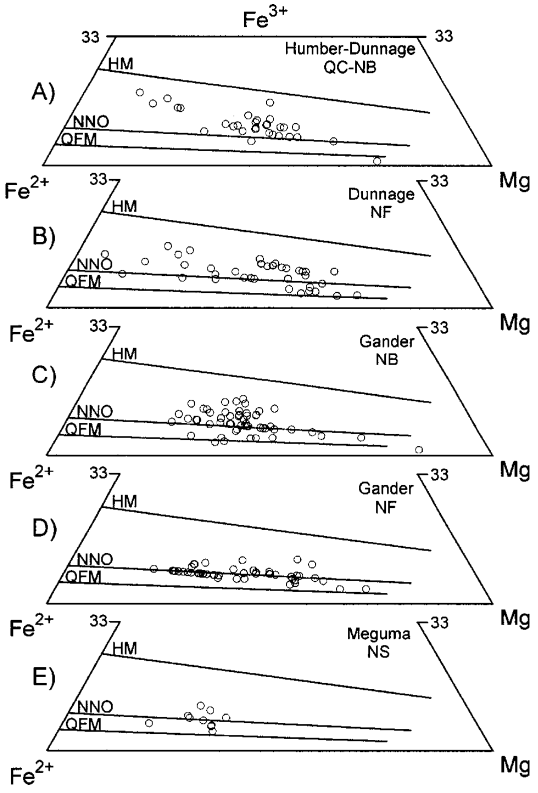

FIG. 4. Composition of biotite from the granitic rocks of the different tectonic zones expressed in the Fe^{2+}–Fe^{3+}–Mg diagram of Wones & Eugster (1965), along with the three common $f(O_2)$ (oxygen fugacity) buffers: quartz – fayalite – magnetite (QFM), nickel – nickel oxide (NNO), and hematite–magnetite (HM).

A qualitative evaluation of oxygen fugacity can be made from the Fe/(Fe + Mg) value of biotite by using the calibrated curve of Wones & Eugster (1965) in $f(O_2)$–T space for biotite + K-feldspar + magnetite equilibrium (Fig. 5). These three phases coexist in all the granitic rocks studied in this report, except for those from the Meguma zone, where magnetite is absent. Assuming a reasonable range of crystallization temperatures of 750° to 900°C for these rocks, estimated $f(O_2)$ values span a range of 10^{-10} to $10^{-16.9}$ bars. Values for individual tectonic zones are presented in Table 4.

TABLE 4. ESTIMATED VALUES OF OXYGEN FUGACITY DURING THE CRYSTALLIZATION OF GRANITIC ROCKS IN THE CANADIAN APPALACHIANS, BASED ON BIOTITE COMPOSITION

zone	province	Fe/(Fe + Mg)	$f(O_2)$ bars
Humber–Dunnage	QC–NB	0.41–0.60	$10^{-12} - 10^{-15}$
Dunnage	NF	0.40–0.84	$10^{-11.6} - 10^{-16.7}$
Gander	NB	0.53–0.76	$10^{-13.7} - 10^{-16.4}$
Gander	NF	0.49–0.60	$10^{-13.2} - 10^{-14.7}$
Avalon	NB	0.42–0.91	$10^{-12} - 10^{-16.8}$
Meguma	NS	0.37–0.99	$10^{-10} - 10^{-16.9}$

Symbols: QB: Québec, NB: New Brunswick, NS: Nova Scotia, NF: Newfoundland.

COMPOSITION OF BIOTITE IN THE ANNITE – SIDEROPHYLLITE – PHLOGOPITE – EASTONITE QUADRILATERAL

The annite – siderophyllite – phlogopite – eastonite quadrilateral (ASPE) is commonly used to illustrate the total Al and Fe/(Fe + Mg) compositional relationships of trioctahedral micas from igneous rock suites (Speer 1984). This diagram is particularly powerful since these

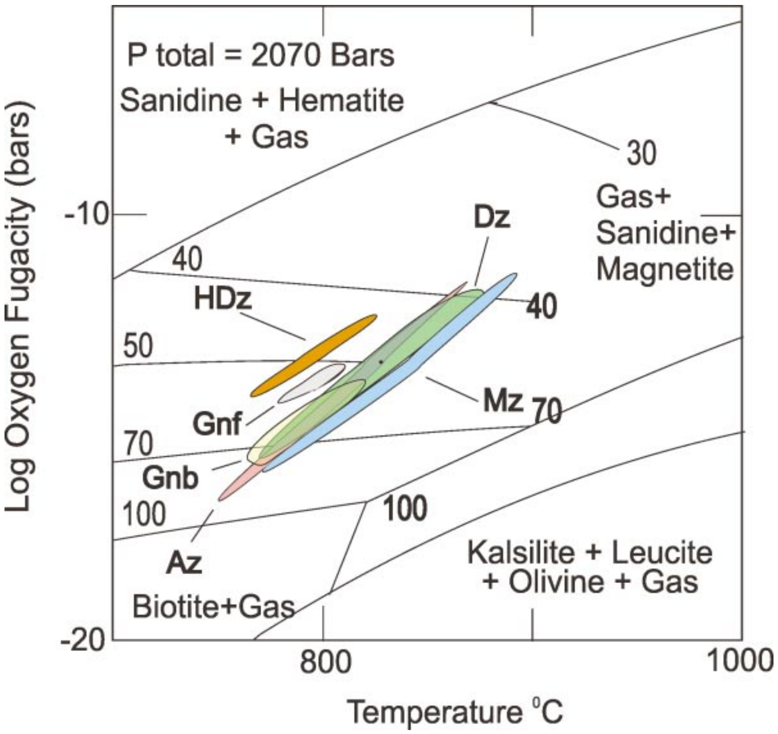

FIG. 5. Log $f(O_2)$ – T diagram for the biotite + sanidine + magnetite + gas equilibrium at P_{total} = 2070 bars (Wones & Eugster 1965). Illustrated are Fe/(Fe + Mg) isopleths. Labeled fields represent the range of oxygen fugacity for samples from the different tectonic zones in the Canadian Appalachians, assuming an equilibrium temperature of 750–900°C. Symbols as follows: Az: Avalon zone, Gnb: Gander zone, New Brunswick, Gnf: Gander zone, Newfoundland, HDz: Humber–Dunnage zone, New Brunswick and Québec, Dz: Dunnage zone, Newfoundland, Mz: Meguma zone. This diagram represents the central portion of the original diagram of Wones & Eugster (their Fig. 4), enlarged to better illustrate the fields defined by the biotite from the different tectonic zones.

TABLE 2. REPRESENTATIVE COMPOSITIONS AND STRUCTURAL FORMULAE OF BIOTITE FROM CANADIAN APPALACHIAN GRANITES

Zone	Avalon(NB)		Dunnage(NF)		Gander(NF)		Gander(NB)		Humber-Dunnage (NB-QC)		Meguma(NS)	
Specimen	nb-180	nb-206	nf-78	nf-49	ak-166	ak-291	nb-29	nb-166	nb-53	mg-44	ns-4	306-2
[1]***Mean of***	3	5	3	4	2	5	7	6	4	7	5	5
Whole-rock SiO_2 value	74.10	67.17	73.60	68.15	75.19	68.46	72.35	64.45	74.95	66.09		72.28
Component oxides (wt.%)												
SiO_2	34.95	37.23	33.76	35.64	37.65	36.12	34.57	34.65	34.83	37.38	34.91	34.72
TiO_2	3.45	4.20	2.23	4.74	2.22	2.32	2.01	2.90	3.69	3.39	2.20	3.43
Al_2O_3	12.59	13.17	17.59	14.33	17.52	14.92	18.10	19.36	12.82	11.93	18.50	19.57
Fe_2O_3	4.64	3.99	3.30	3.03	3.83	3.21	5.03	1.75	5.98	4.10	3.03	3.79
FeO	27.95	15.30	19.85	15.43	15.70	16.36	16.10	18.40	24.50	20.83	21.22	18.98
MnO	0.70	0.33	0.71	0.43	1.84	0.54	0.95	0.21	0.78	0.25	0.90	0.34
MgO	3.52	12.90	8.67	12.70	6.65	12.19	9.46	8.59	5.06	8.74	6.04	5.72
CaO	0	0.03	0.03	0.81	0.05	0.04	0.01	0	0.04	0.01	0.00	0.01
Na_2O	0.07	0.10	0.07	0.17	0.05	0.06	0.07	0.22	0.18	0.07	0.05	0.15
K_2O	9.37	9.93	9.70	8.48	9.31	9.96	9.94	9.66	9.18	9.79	9.79	10.10
[2]H_2O	3.28	3.66	3.66	3.86	3.35	3.68	3.68	3.75	2.83	2.95	3.88	3.69
F	0.79	0.65	0.43	0.23	1.23	0.53	0.53	0.40	1.87	1.75	0.05	0.51
Cl	0.39	0.17	0.04	0.04	0.02	0.04	0.01	0.01	0.32	0.42	0.00	0.06
O≡F,Cl	0.42	0.31	0.19	0.11	0.52	0.23	0.22	0.17	0.86	0.83	0.02	0.22
Total	101.28	101.35	99.85	99.78	99.90	99.73	99.73	99.73	101.22	100.78	100.54	100.84
Mineral formulae based on $O_{10}(OH,F,Cl)_2$												
Si^{4+}	2.789	2.780	2.613	2.689	2.865	2.750	2.635	2.636	2.749	2.884	2.684	2.638
Al^{3+}	1.184	1.159	1.387	1.274	1.135	1.250	1.365	1.364	1.192	1.085	1.316	1.362
Fe^{3+}	0.027	0.061		0.037					0.059	0.031		
ΣT	4	4	4	4	4	4	4	4	4	4	4	4
Al^{3+}			0.218		0.436	0.089	0.261	0.372			0.360	0.391
Ti^{4+}	0.207	0.236	0.130	0.269	0.127	0.133	0.115	0.166	0.219	0.197	0.127	0.196
Fe^{3+}	0.252	0.163	0.192	0.135	0.219	0.184	0.289	0.100	0.296	0.207	0.175	0.217
Fe^{2+}	1.865	0.955	1.285	0.973	0.999	1.042	1.026	1.170	1.617	1.344	1.364	1.206
Mn^{2+}	0.047	0.021	0.047	0.027	0.119	0.035	0.061	0.014	0.052	0.016	0.059	0.022
Mg^{2+}	0.419	1.436	1.000	1.428	0.754	1.383	1.075	0.974	0.595	1.005	0.692	0.648
ΣM	2.790	2.811	2.872	2.832	2.654	2.866	2.827	2.796	2.779	2.769	2.777	2.680
Ca	0	0.002	0.002	0.065	0.004	0.003	0.001	0	0.003	0.001	0.000	0.001
Na	0.011	0.014	0.011	0.025	0.007	0.009	0.010	0.032	0.028	0.010	0.007	0.022
K	0.954	0.946	0.958	0.816	0.904	0.967	0.918	0.937	0.924	0.964	0.960	0.979
ΣI	0.965	0.962	0.971	0.906	0.915	0.979	0.929	0.969	0.955	0.975	0.967	1.002
OH	1.748	1.825	1.889	1.940	1.701	1.867	1.871	1.902	1.490	1.518	1.988	1.870
F	0.199	0.153	0.105	0.055	0.296	0.128	0.128	0.096	0.467	0.427	0.012	0.123
Cl	0.053	0.022	0.005	0.005	0.003	0.005	0.001	0.001	0.043	0.055	0.000	0.007
Fe/(Fe+Mg)	0.789	0.309	0.050	0.436	0.618	0.470	0.550	0.566	0.763	0.607	0.690	0.687
$Fe^{3+}/(Fe_{tot})$	0.119	0.146	0.130	0.122	0.180	0.150	0.220	0.079	0.155	0.133	0.114	0.152

[1] Number of determinations included in the mean.

[2] H_2O calculated assuming that (OH+F+Cl) = 2.

TABLE 3. SUMMARY OF COMPOSITIONAL FEATURES OF BIOTITE FROM CANADIAN APPALACHIAN GRANITES

zone	no. of samples*	*M*-site occupancy	ΣAl	$^{VI}Al^{3+}$	$^{VI}Ti^{4+}$	Fe/(Fe + Mg)	Fe^{3+}/Fe
Humber-Dunnage NB - QC	30	2.68-2.86 (2.79)	1.01-1.26 (1.15)	0.00-0.18 (0.01)	0.20-0.30 (0.22)	0.19-0.44 (0.29)	0.05-0.13 (0.08)
Dunnage, NF	38	2.71-2.92 (2.81)	1.19-1.60 (1.40)	0.00-0.28 (0.13)	0.10-0.28 (0.18)	0.19-0.44 (0.29)	0.05-0.12 (0.08)
Gander, NB	55	2.69-2.89 (2.79)	1.05-1.78 (1.47)	0.00-0.45 (0.18)	0.09-0.27 (0.18)	0.21-0.38 (0.32)	0.03-0.12 (0.08)
Gander, NF	50	2.65-2.91 (2.81)	1.18-1.77 (1.50)	0.00-0.50 (0.24)	0.08-0.24 (0.15)	0.18-0.40 (0.30)	0.05-0.11 (0.07)
Avalon, NB	18	2.71-2.91 (2.81)	1.05-1.35 (1.18)	0.00-0.12 (0.02)	0.15-0.26 (0.21)	0.21-0.46 (0.32)	not determined
Meguma, NS	132	2.46-2.99 (2.75)	1.32-2.01 (1.72)	0.18-0.78 (0.43)	0.02-0.27 (0.18)	0.19-0.50 (0.35)	0.04-0.10 (0.06)

* Number of determinations included in mean. *M*-site occupancy, ΣAl, $^{VI}Al^{3+}$ and $^{VI}Ti^{4+}$ are expressed in atoms per formula unit. Values in parentheses are means. Symbols: QB: Québec, NB: New Brunswick, NS: Nova Scotia, NF: Newfoundland.

two variables are indicators of peraluminosity and redox state, respectively, of the rock hosting the mica. The composition of biotite from all the zones in this study is plotted in the ASPE quadrilateral in Figure 6.

Biotite from rocks in the Humber–Dunnage zone (Fig. 6A) exhibits a fairly wide range of Fe/(Fe + Mg) values, all at low total Al contents, reaching up to ~1.25 Al atoms per formula unit (*apfu*). Biotite from the Dunnage zone defines a field with a wide range of Fe/(Fe + Mg) values and a moderate range of total Al contents (Fig. 6B). In this field, no trend is recognizable with regards to rock type, but those from Fogo Island (Newfoundland) have high Fe/(Fe + Mg) values and low Al-contents.

The biotite samples from the Gander zone in New Brunswick (Fig. 6C) define a field with a relatively nar-

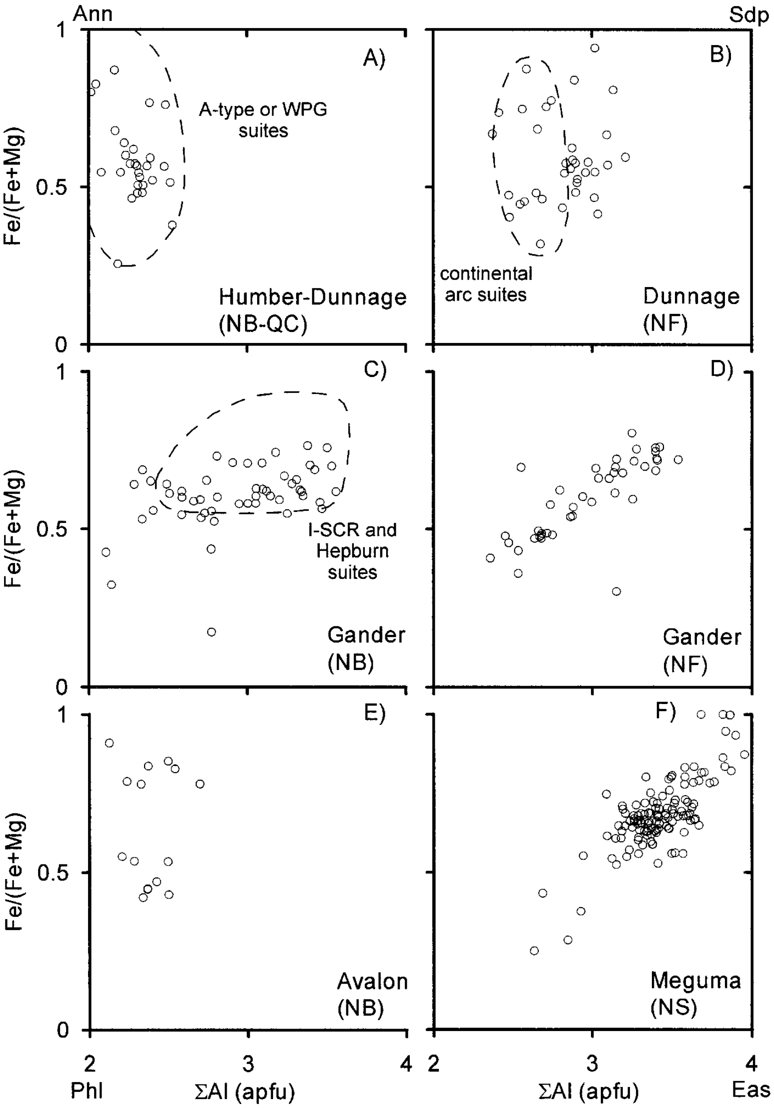

FIG. 6. Composition of biotite from Canadian Appalachian granites expressed in the Fe/(Fe + Mg) *versus* ΣAl diagram, also known as the annite (Ann) – siderophyllite (Sdp) – phlogopite (Phl) – eastonite (Eas) quadrilateral. Fe: total iron, *i.e.*, $Fe^{2+} + Fe^{3+}$. Dashed lines on some diagrams indicate the compositional fields of suites documented in the literature (see text for details).

row range of Fe/(Fe + Mg) values with a large range of total aluminum values, from 1.05 to 1.78 *apfu*. Biotite from the Newfoundland Gander zone shows a positive correlation between Fe/(Fe + Mg) values and total Al in the ASPE quadrilateral (Fig. 6D). In these rocks, a general trend of increasing biotite Fe/(Fe + Mg) values and Al contents was observed with increasing SiO_2 content of the host rocks.

Biotite in the Avalon zone of New Brunswick defines a field that, similar to the Humber–Dunnage group, has a fairly wide range of Fe/(Fe + Mg) values with a narrow zone of low total Al contents (Fig. 6E). Finally, the trend defined by biotite in Meguma-zone rocks, like that for the Gander zone in Newfoundland, is characterized by increasing Fe/(Fe + Mg) values and Al contents in the ASPE quadrilateral, with some compositions approaching the siderophyllite end-member (Fig. 6F).

DISCUSSION

In this study, granites from the Canadian Appalachians were grouped according to their tectonostratigraphic zones because of the direct correlation that exists between the zonal distribution of these rocks and their geochemical and isotopic characteristics. These major tectonostratigraphic associations or zones of granitic rocks were clearly distinguished by the composition of their biotite in the ASPE quadrilateral plots (Fig. 6).

In his compilation of biotite compositions of plutonic rocks, Abdel-Rahman (1994) discriminated among: 1) anorogenic extension-related peralkaline granites, 2) calc-alkaline I-type orogenic suites and 3) peraluminous rocks including S-type granites. Several useful diagrams were introduced to discriminate the composition of biotite from these different tectonic settings in terms of its FeO, MgO, and Al_2O_3 contents. Of these diagrams, we will use the ternary FeO*–MgO–Al_2O_3 plot, probably the most powerful discriminant since, much like the ASPE quadrilateral, it reflects variations in redox state and peraluminosity.

Below, we compare the compositional fields or trends of biotite from Appalachian granites with those of well-documented granitic suites from other belts and through syllogistic reasoning, attempt to infer both the petrogenetic and tectonomagmatic characteristics of their host rocks.

Biotite compositions from the Humber–Dunnage suites, shown in the ASPE quadrilateral in Figure 6A, compare closely with those of documented A-type or WPG suites: Ascutney Mountain igneous complex (Schneiderman 1991), Klokken gabbro-syenite complex (Parsons 1981, Parsons *et al.* 1991), Mboutou gabbro–syenite complex (Parsons *et al.* 1986), Baie-des-Moutons syenite complex (Lalonde & Martin 1983), Topsail batholith in Newfoundland (J.B. Whalen, pers. commun.), Mont Saint-Hilaire alkaline intrusion (Lalonde *et al.* 1996). Both groups are characterized by low total Al contents, in some cases even being Al-deficient, with a wide range of Fe/(Fe + Mg) values that extend from phlogopite [Fe/(Fe + Mg) < 0.5] to nearly pure end-member annite [Fe/(Fe + Mg) $\simeq$ 1].

In the FeO*–MgO–Al_2O_3 plot of Abdel-Rahman (1994) (Fig. 7A) biotite compositions from the Humber–Dunnage suites plot mainly in the A-type field. This observation is consistent with the host whole-rock geochemical signature which, in all common discrimination diagrams, plot mainly in the A-type field (or within-plate granite field) (Whalen 1993).

Compositions of biotite from the Dunnage zone in Newfoundland compare well in the ASPE quadrilateral with those of documented continental arc suites (see Fig. 6B) such as the Coastal batholith of Peru (Mason 1985), the Bishop suite of the Wopmay orogen (Lalonde & Bernard 1993), and the magnetite-series granites of Japan (Czamanske *et al.* 1981). Compositions of biotite from the Dunnage zone in Newfoundland show a range of Fe/(Fe + Mg) values that is nearly identical to that defined by continental arc suites, but with greater variations in total Al contents, suggesting a greater contribution from metasedimentary material to the Newfoundland magmas.

In the discrimination diagrams of Abdel-Rahman (1994), biotite from the Dunnage rocks in Newfoundland falls principally in the calc-alkaline I-type field, with only a few data points in the peraluminous and alkaline A-type fields (Fig. 7B). Those data in the alkaline field belong to the Fogo Island suite, which is bimodal, suggestive of a rift-type environment.

Biotite from the Gander zone in New Brunswick defines a nearly horizontal trend of increasing total Al at relatively constant Fe/(Fe + Mg) in the ASPE quadrilateral (Fig. 6C). This trend has been frequently documented in the literature, *e.g.,* in the I–SCR (strongly contaminated and reduced I-type) granites of the California batholiths (Ague & Brimhall 1988) and the Hepburn intrusive suite of Wopmay orogen (Lalonde & Bernard 1993). The field defined by biotite from the Gander zone in New Brunswick overlaps considerably that of the combined Hepburn and I–SCR fields, but is located slightly below these, at lower Fe/(Fe + Mg) values, suggesting that the Gander granites in New Brunswick evolved under slightly more oxidizing conditions (Fig. 6C). This is consistent with the coexistence of magnetite and ilmenite in these rocks, in contrast to the sole occurrence of ilmenite in the majority of Hepburn rocks in Wopmay orogen and I–SCR rocks in California.

In the FeO*–MgO–Al_2O_3 diagram of Abdel-Rahman (1994), biotite compositions from rocks of the Gander zone in New Brunswick are distributed between the calc-alkaline and peraluminous fields (Fig. 7C); compositions of biotite from the mafic to intermediate rocks fall principally in the C (calc-alkaline) field, whereas those from the peraluminous granites plot in the P (peraluminous) field. Within the Gander zone in

New Brunswick, all Ordovician granites have within-plate whole-rock geochemical signatures, whereas younger Siluro-Devonian granitic rocks have mixed volcanic-arc to within-plate signatures (Whalen 1993). Although biotite compositions from this zone appear to reflect a mixed derivation, they do not include a ten-

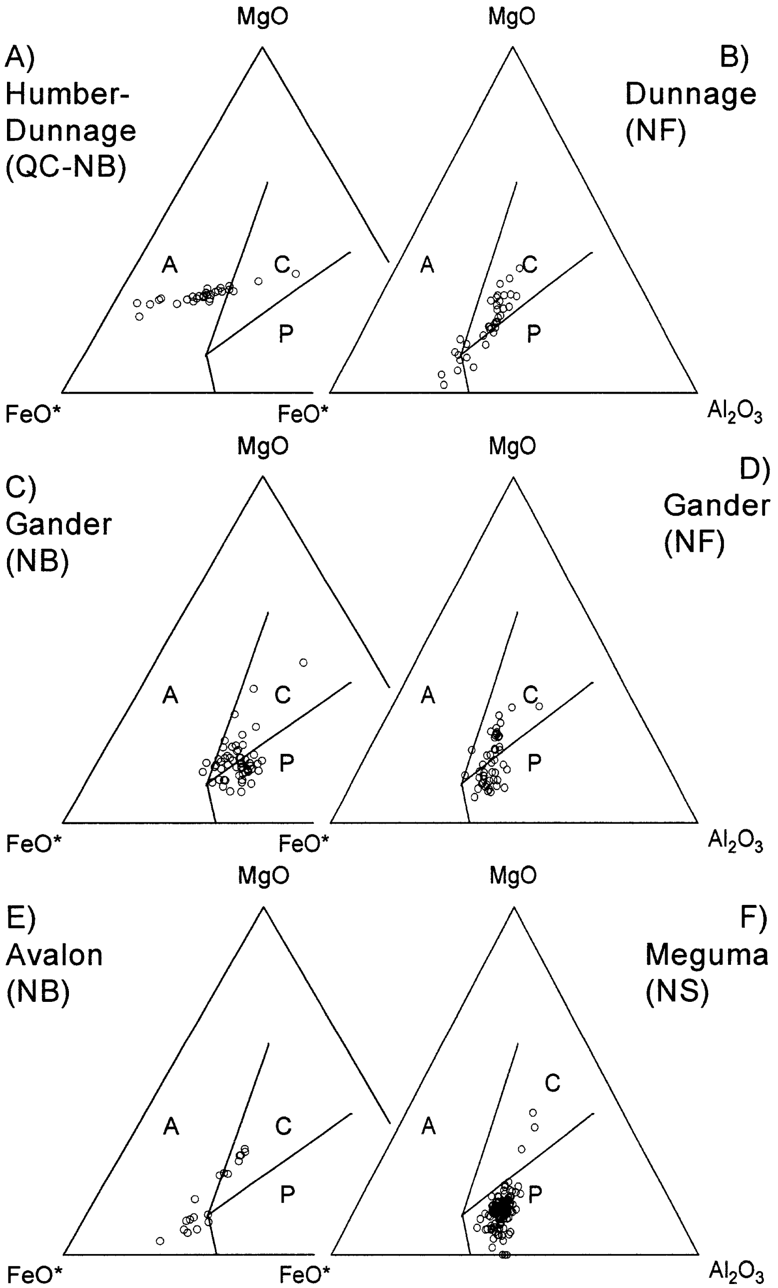

FIG. 7. Composition of biotite from Canadian Appalachian granites expressed in the discrimination diagrams of Abdel-Rahman (1994). A: alkaline, C: calc-alkaline, and P: peraluminous granite fields. FeO* = [FeO + (0.89981*Fe_2O_3)].

dency to compositions typical of anorogenic or A-type granitic rocks.

In the ASPE quadrilateral, micas from the Gander zone of Newfoundland show, in contrast to those from that zone in New Brunswick, a progressive increase of Fe/(Fe + Mg) and total Al values from mafic to felsic (or evolved) rocks (Fig. 6D). This trend is also observed in the Japanese ilmenite-series granites (Czamanske *et al.* 1981), as well as the hybrid and calc-alkaline Hercynian granites of northern Portugal (de Albuquerque 1973, Neiva 1981, Konings *et al.* 1988). This trend, with increasing Fe/(Fe + Mg) values and Al contents, is interpreted as reflecting conditions of low oxygen fugacity (at or below the Ni–NiO buffer) caused by significant contributions of metasedimentry material to the magmas, either by assimilation or anatexis. In Abdel-Rahman's triangular plot, biotite samples from the Newfoundland Gander zone fall in the peraluminous and calc-alkaline fields (Fig. 7D) and define a somewhat more linear trend than the cluster defined by the New Brunswick ones.

All Siluro-Devonian granites from the Avalon zone of New Brunswick represent different phases of the St. George batholith. According to Whalen (1993), this batholith includes plutons that can be easily classified as either A- or I-type, as well as intrusions that display only some A-type characteristics. In the discrimination diagram of Abdel-Rahman (1994), the composition of biotite from these rocks falls in the calc-alkaline I-type and within-plate A-type fields (Fig. 7E). Biotite compositions derived mainly from amphibole – biotite granites fall in the field of calc-alkaline, I-type rocks, and those derived mostly from biotite granites plot in the field of alkaline A-type rocks, consistent with their tectonomagmatic characteristics.

According to Clarke *et al.* (1997), the central plutons of the Meguma suite are derived entirely from crustal material and probably owe their origin to crustal thickening associated with accretion of the Meguma zone to the Avalon zone. In contrast, peripheral plutons are of mixed derivation (sub-Meguma-Group source rocks and mantle-derived mafic magmas) and probably owe their origin to intrusion of subduction-related mafic magmas into the middle to lower crust prior to final emplacement of the Meguma zone. In the ASPE quadrilateral (Fig. 6F), biotite compositions from the peripheral and central plutons of the Meguma zone show a trend defined by a pronounced increase in both Fe/(Fe + Mg) values and total Al values. This trends extends to the siderophyllite apex of the diagram. At first sight, this trend appears quite similar to that of biotite from the Gander zone of Newfoundland, but the Gander trend has a lower range of Fe/(Fe + Mg) values and Al contents, implying more oxidizing conditions and a lower contribution of crust than in the Meguma zone. This inference is further confirmed by the occurrence of magnetite, ilmenite and titanite (in variable proportions) in the majority of Gander rocks in Newfoundland. In the Meguma zone, ilmenite is the common oxide phase, whereas magnetite and titanite are usually absent. Not surprisingly, on the discrimination diagrams of Abdel-Rahman (1994), the biotite in the Meguma rocks plots almost exclusively in the peraluminous field (Fig. 7F).

CONCLUSIONS

In this study, we show that the most pronounced differences in biotite from Canadian Appalachians granitic rocks occur in total Al contents and in Fe/(Fe + Mg) values, two parameters used to construct the annite – siderophyllite – phlogopite – eastonite quadrilateral of trioctahedral micas. Both parameters are sensitive indicators of conditions that prevailed in the host magmas. Specific conclusions of our study are summarized below:

1. Biotite compositions from A-type granites of the Humber and Avalon zones, whether reduced or oxidized, are distinctly depleted in aluminum and enriched in total iron. A few samples of plutonic rocks from Fogo Island (Newfoundland Dunnage zone), attributed to extensional tectonic environments, show the same features. Biotite compositions from I-type units have the same Al contents as A-type rocks, but being moderately enriched in Mg, they plot in the calc-alkaline I-type field in the discrimination diagrams. These compositional features of biotite are consistent with the nature of their host rocks.
2. Granites from the Dunnage zone in Newfoundland (Notre Dame arc) are interpreted as having formed within a subduction-related environment. They contain biotite that is compositionally comparable with biotite from California batholiths, which are associated with a typical continental magmatic arc. Compared to biotite compositions from other continental arc suites, such as the Coastal batholith of Peru, the Bishop intrusive suite of Wopmay orogen of the Northwest Territories of Canada, and the magnetite-series granites of Japan, biotite from Notre Dame arc granites has moderate Al contents, suggestive of significant contributions from aluminous metasedimentary material during its petrogenesis; this is consistent with the available petrological and geochemical data.
3. Granites from the Gander zone of New Brunswick and Newfoundland, derived from a mixture of supracrustal and mantle-derived source components, contain biotite that shows a pronounced trend of increasing total Al, confirming significant contributions to the magmas from aluminous supracrustal material, either by assimilation or anatexis.
4. The peripheral plutons of the Meguma zone, derived from a mixture of supracrustal and mantle-derived mafic magma, have biotite compositions that resemble those of the granites of the Gander zone from Newfoundland. In contrast, biotite compositions from the central plutons, considered to be derived entirely from crustal material, show a remarkable increase in both total Al and Fe, approaching the siderophyllite end-

member. Biotite from the central plutons is generally associated with muscovite, and possibly with garnet, cordierite and andalusite.

These examples show that the composition of biotite can be a discriminating tool in identifying the nature of granite protoliths. However, can it also be used in an interpretation of tectonic settings?

Appalachian Siluro-Devonian granites include peralkaline A-type, metaluminous to slightly peraluminous I-type, and peraluminous S-type granites whose major- and trace-element compositions are comparable to those of granite suites from classical within-plate, convergent and collisional environments, respectively. Whalen (1993), Whalen *et al.* (1994) and Kerr (1997) have suggested that these various types of granite could be the product of a major thermal anomaly supplied by upwelling of the asthenospheric mantle following post-collisional lithospheric delamination. If this is the case, then neither the major-element composition of the biotite nor the whole-rock compositions should be used to interpret the tectonic origin of these granites, since a single tectonic process would have been responsible for their generation. Alternatively, in a recent comprehensive synthesis on Appalachian tectonic evolution, van Staal *et al.* (1998) concluded that accretion of the Notre Dame arc to Laurentia, followed by the Exploits arc (and equivalent arcs) occurred from 475 to 455 Ma, followed by diachronous collision of Laurentia with Avalon during the Silurian, and Avalon–Meguma collision during the Devonian. Within this tectonic framework, Siluro-Devonian granites of the Humber and Dunnage zones were emplaced in a post-collisional setting, whereas granite emplacement into the Gander, Avalon and Meguma zones was, at least in part, synchronous with proximal collisional events. Generation of Siluro-Devonian granites within the easternmost Appalachian zones could, therefore, be directly tied to various convergent margin and collisional processes, including subduction, back-arc extension and overthickening of the crust.

Our results show that the major-element composition of biotite can serve as one tool among others (*e.g.,* Lameyre & Bowden 1982, Pitcher 1983, Pearce *et al.* 1984, Batchelor & Bowden 1985, Maniar & Piccoli 1989) for the classification and characterization of granites and for understanding their petrogenesis, but that none of these methods is infallible or should be employed in isolation. We therefore conclude, like Abdel-Rahman (1994), that the composition of biotite can be a useful tectonomagmatic indicator if coupled with other parameters, such as major- and trace-element data on the whole rocks, isotopic geochemical data, field and other geological constraints.

ACKNOWLEDGEMENTS

We are grateful to Dr. Denis G. Rancourt, University of Ottawa, for access to the Mössbauer laboratory, Dr. A. Kerr, Geological Survey of Newfoundland and Labrador, and Dr. D.B. Clarke, Dalhousie University, for providing whole-rock specimens. Financial support from the Natural Sciences and Engineering Research Council of Canada by means of grants to AEL is acknowledged. The first author also acknowledges the financial support of the Ministry of Culture and Higher Education of Iran. We thank A. Patiño-Douce, D. Kontak, R.F. Martin and R. Berman for their constructive reviews. This paper is contribution number 2003004 of the Geological Survey of Canada.

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Received March 17, 2002, revised manuscript accepted October 17, 2003.

APPENDIX 1. ANALYTICAL METHODS

Minerals analyzed in the course of this study were done so by wavelength-dispersion X-ray spectrometry for 11 elements (K, Na, Ca, Mg, Mn, Fe, Al, Ti, Si, F, and Cl) using the JEOL 8900 Superprobe of the McGill University Microprobe Laboratory. Typical operating conditions were 15 kV and 20 nA. Results of all 323 analyses are given in Appendix 3 (available from the Depository of Unpublished Data of CISTI, National Research Council of Canada, Ottawa, Ontario K1A 0S2, cisti.info@nrc.ca). Analytical results from numerous other sources were included, however. In the great majority of these cases, these determinations were also done by wavelength-dispersion X-ray spectrometry with the electron microprobe.

The oxidation state of iron in 54 specimens of mica was determined by transmission ^{57}Fe Mössbauer spectroscopy (MS). All specimens were separated from crushed rock by combined heavy liquid and magnetic separation methods. In all cases, purity was verified under the binocular microscope to be greater than 95%. For those samples not measured by MS, FeO and Fe_2O_3 values were extrapolated from those of similar petrographic units that were measured. All spectra were obtained at room temperature (RT, 22°C) using a ^{57}Co rhodium matrix source on a velocity range of ±4 mm/s with a constant-acceleration transducer. Data were collected on 1024 channels, which covered twice the Doppler velocity range. Spectra were calibrated with a ^{57}Fe-enriched iron foil, both before and after each experiment. All positions are reported with respect to this calibration spectrum, *i.e.,* with respect to the center shift (CS) of metallic α-Fe at RT. All spectra were folded to give a flat background (BG). The preparation of absorbers and the methods of spectral analysis used are identical to those described by Lalonde *et al.* (1996).

All the RT spectra of biotite exhibit remarkable similarity and have three main absorption peaks centered at ~–0.1, +1.0 and +2.3 mm/s, which respectively correspond to 1) the low-energy lines of both octahedral Fe^{2+} and octahedral Fe^{3+} quadrupole doublets, 2) the high-energy lines of octahedral Fe^{3+} quadrupole doublets, and 3) the high-energy lines of octahedral Fe^{2+} quadrupole doublets. None of these spectra show a shoulder at ~0.4 mm/s corresponding to the high energy-lines of quadrupole doublets of Fe^{3+} in tetrahedral sites (Rancourt *et al.* 1992).

Representative Mössbauer spectra of biotite samples are shown in Figures 2A and 2B, and all 54 spectra are displayed in Appendix 4 (available from the Depository of Unpublished Data, CISTI, National Research Council of Canada, cisti.info@nrc.ca). In all spectra, the contributions of octahedral Fe^{2+} were assumed to be sums of three Gaussian components. The contributions of octahedral Fe^{3+} were assumed to be composed of one or two Gaussian components depending on the structure of the spectra. Fitting parameters for the best fits of all spectra are given in Appendix 5 (also available from the Depository of Unpublished Data, CISTI). Results show a considerable range of Fe^{3+}/Fe values, from 0.05 to 0.23.

For biotite samples for which corresponding whole-rock compositional data were not already available, the concentration of whole-rock major, minor and trace elements was determined by sequential wavelength-dispersion X-ray fluorescence spectroscopy (XRF) with a Philips PW2400 automated spectrometer. These analyses were performed on glass discs of fused rock samples. The results are tabulated in Appendix 6 (also available from the Depository of Unpublished Data).