

# Crustal mass transfer and index mineral growth in Barrow's garnet zone, northeast Scotland

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## ABSTRACT

The Barrovian metamorphic sequence of northeastern Scotland plays a critical role in our understanding of regional metamorphism at middle and lower crustal levels. The importance of regional temperature and pressure gradients in producing the sequence is well established, but the petrogenetic significance of mass transport of nonvolatile elements by metamorphic processes remains controversial. This paper focuses on the impact of fluid flow through fractures (now quartz veins) on the geochemical and petrologic evolution of metamorphosed clastic rocks in the northeastern part of Barrow's garnet zone, Scotland. The veins constitute ~13 vol% of the total rock mass and are generally surrounded by zones rich in plagioclase. A traverse along a typical metasedimentary bed cut by a vein reveals that, as one approaches the vein, (1) plagioclase porphyroblast size and An content increase, (2) the modal plagioclase/muscovite ratio increases, (3) bulk rock Na/K, Ca/K, Sr/Rb,  $(Na + Ca + K)/Al$ ,  $Mg/Fe^T$ , and  $(Mg + Fe^T)/Al$  increase, and (4) bulk rock Si/Ti decreases. The plagioclase-rich zones are interpreted to be alteration selvages that developed as a result of fluid infiltration through fractures; the alteration reaction destroyed muscovite and produced plagioclase. Furthermore, in some rocks the assemblage garnet + biotite + chlorite was stable in the most intensely altered areas directly adjacent to vein margins, whereas chloritoid + biotite + chlorite was stable in less-altered and unaltered areas. Geochemical relations suggest that the fluid flow was in a direction of increasing temperature and that the vein quartz was mostly derived from local wall rocks. It appears that open-system transport of nonvolatile elements can exert important, previously unrecognized controls on rock chemistry and the growth of common regional metamorphic mineral assemblages.

## INTRODUCTION

Metamorphic rocks make up, by volume, about half of the upper 30 km of the continents (cf. Percival et al., 1992; Wedepohl, 1995). Consequently, the amounts, length scales, and processes of element transport during metamorphism must be determined in order to understand the geochemical and petrological evolution of the crust. Barrovian-style regional metamorphism has affected major portions of mountain belts worldwide and is one of the most common styles preserved in the rock record. Barrow's landmark studies of the Highlands of northeastern Scotland established that mineral assemblages in clastic sedimentary rocks undergoing metamorphism change systematically at the regional scale in response to increasing metamorphic grade (Barrow, 1893, 1898, 1912). More recently, it has been shown that circulation of large quantities of  $H_2O-CO_2$  fluid is an integral part of Barrovian-style metamorphism of the middle, and lower crust (see Ferry, 1992; Skelton et al., 1995, and references therein). What remains poorly understood, however, is how the transport of nonvolatile rock-forming and trace elements during orogenesis and fluid flow influences mineral reactions and the geochemistry of metamorphic rocks. This paper focuses on the petrogenetic consequences of fluid circulation through a re-

gional network of fractures in the rocks studied by Barrow in his classic exposition of metamorphic index mineral zones (Barrow, 1893, 1898, 1912). The results strongly suggest that open-system transport of nonvolatile elements exerted important, previously unrecognized controls on rock chemistry and the growth of common regional metamorphic mineral assemblages.

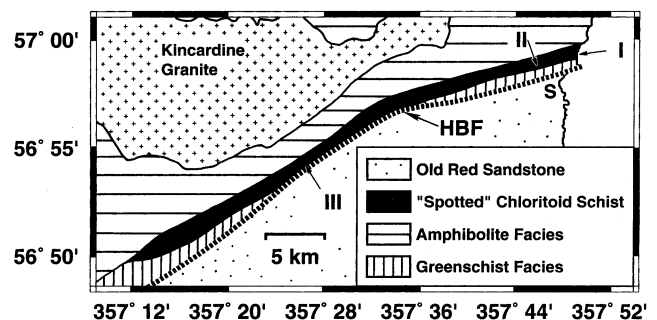
## GEOLOGIC RELATIONS

The role of nonvolatile element transport in the formation of the Barrovian metamorphic sequence of Scotland has long been debated. In 1967, Chinner advanced the far-reaching hypothesis that regional-scale, down-temperature mass transfer of potassium and water during prograde

metamorphism destabilized Al-rich minerals like chloritoid in favor of K-rich micaceous assemblages in most of the greenschist facies rocks of northeastern Scotland. Several later workers (e.g., Atherton, 1977) argued that Chinner's petrological observations could be explained equally well by isochemical metamorphism. Chinner (1967) and Yardley and Baltatzis (1985) suggested that some alkali mass transfer may have also occurred during low-temperature, retrograde metamorphism.

This study centers on the belt (40 km long, 0.5–1.5 km wide) of "spotted" chloritoid schists that crops out in the northeastern part of the Dalradian Supergroup (Fig. 1). The schists are visually distinctive and have drawn the attention of

**Figure 1. Simplified geologic map showing metamorphic rocks of Dalradian Supergroup (peak metamorphism ~520–490 Ma; cf. Harte et al., 1984, and references therein), post-peak metamorphic granite, and Old Red Sandstone. Boundaries of "spotted" chloritoid schist belt are approximate. Study areas I, II, and III near Red Man, Ury House, and Drumtochty Castle, respectively. S = Stonehaven; HBF = Highland Boundary fault. Geologic relations after Barrow (1912), Chinner (1967), Atherton (1977), and Harte et al. (1984).**



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many workers. For example, Barrow began his classic 1898 (p. 149) paper as follows: "In the neighbourhood of Drumtochty, near Fordun, I found numerous fragments of a green schist, which are characterized by white or yellowish spots." Chinner (1967, p. 271) stated that "a persistent and puzzling feature of a large proportion of the chloritoid-schists is the presence of ovoid 'spots' . . ." and that "it is difficult to resist the temptation to believe that these porphyroblasts [the spots] have some genetic significance . . ." The spots are ellipsoidal plagioclase porphyroblasts that underwent varying degrees of replacement by complex pseudomorph assemblages, including kaolinite, sericite, carbonates, and albite, during retrograde metamorphism (cf. Yardley and Baltatzis, 1985). Quartz veins are abundant throughout the spotted schist belt and occur mainly in boudin necks and along lithologic contacts. Linear measurement traverses (cf. Ague, 1994) across the superbly exposed coastal outcrops of study area I (Fig. 1) indicate that the veins constitute ~13 vol% of the rock mass and have a geometric mean width of ~3 cm. A key relationship discovered in this study is that the plagioclase spots become larger and more abundant as one approaches the veins; some vein margins are composed almost entirely of plagioclase (Figs. 2 and 3). The long axes of the spots lie in the plane of the steeply dipping cleavage that characterizes the metamorphic rocks adjacent to the Highland Boundary fault (Highland Border Steep belt; cf. Harte et al., 1984). In many samples, the plagioclase spots contain numerous inclusions of chloritoid crystals, the preferred orientation (long axes of chloritoids lying in the steep cleavage plane) of which appears identical to that of matrix chloritoid grains. These relations demonstrate that the spots are greenschist facies metamorphic phenomena that grew over the steep cleavage and over preexisting chloritoid crystals. Because garnet appears at slightly lower metamorphic grade than chloritoid in northeastern Scotland (cf. Droop and Harte, 1995), it is highly probable that the spots grew under garnet zone pressure-temperature (*P-T*) conditions.

## METHODS

Determinations of rock and mineral chemistry are given in Appendixes 1 and 2<sup>1</sup>. Metamorphic *P*, *T*, and H<sub>2</sub>O activity (*a*<sub>H<sub>2</sub>O</sub>; activity denoted by *a*) were estimated using the TWEEQU program of Berman (1991). The activity model for chlorite was taken from Powell (1978); the activity models recommended by Berman (1991) were used for all other phases. *P-T* conditions were estimated by calculating the three possible equilibria among the components muscovite, annite, phlogopite, pyrope,

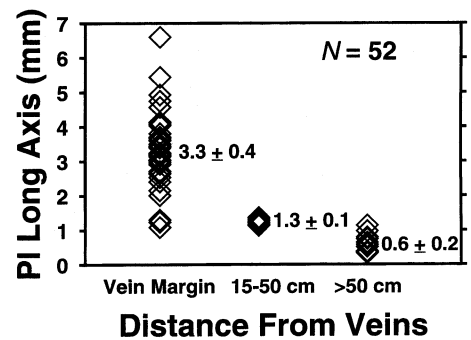
almandine, anorthite, grossular, and quartz. I used 21 equilibria involving these, components and clinocllore, albite, and paragonite to estimate *a*<sub>H<sub>2</sub>O</sub>. The *a*<sub>HCl</sub>/*a*<sub>H<sub>2</sub>O</sub> of the fluid was estimated from the biotite composition following the methods of Zhu and Sverjensky (1992). Fluid speciation calculations were done for the system SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub>-FeO-MgO-CaO-Na<sub>2</sub>O-K<sub>2</sub>O-H<sub>2</sub>O-HCl using the following: (1) the thermodynamic data of Berman (1991) for minerals; (2) the thermodynamic data of Johnson et al. (1992) and Sverjensky et al. (1991) for aqueous species; (3) the activity models summarized above for solids; and (4) the extended Debye-Hückel activity coefficient relation for charged aqueous species. Equilibrium between the fluid and clinocllore, muscovite, albite, anorthite, grossular, almandine, and quartz components was assumed.

## RESULTS

A representative garnet zone sample from study area I (JAB-101L) that contains fresh, nonretrograde plagioclase spots was selected for detailed study. Primary sedimentary layering of pelite, semipelite, and psammite layers is present in the study sample, and a quartz vein, which occupies a boudin neck, cuts across the layers at one end of the sample (Fig. 3). The vein contains minor pyrite and chlorite. Near the quartz vein (Figs. 3B and 4) the rock contains quartz + plagioclase + chlorite + biotite + muscovite + garnet (garnet is almandine rich; rim composition is Alm<sub>68.7</sub>Prp<sub>4.9</sub>Grs<sub>10.0</sub>Sps<sub>16.3</sub>). Accessory phases include rutile and pyrite. Farther from the vein, chloritoid becomes part of the assemblage and garnet disappears.

A comprehensive mineralogical and bulk chemical traverse along a typical semipelitic bed provides new and unique constraints on mass transfer and mineralogical evolution (Figs. 3A and 4). Plagioclase spots clearly increase in size and abundance as distance from the vein decreases (Figs. 3B and 4). The increase in modal plagioclase is matched by a sympathetic decrease in muscovite, leading to a substantial increase in the plagioclase/muscovite ratio toward the vein (Fig. 4). Bulk rock Na/K and Ca/K also increase toward the vein (by a factor of ~10), consistent with the modal plagioclase/muscovite relations (Fig. 4). Bulk chemical Si/Ti decreases, whereas Sr/Rb, (Na + Ca + K)/Al, Mg/Fe<sup>T</sup> (Fe<sup>T</sup> = total Fe), and (Mg + Fe<sup>T</sup>)/Al increase toward the vein (Fig. 4). The mean An content of the plagioclase increases from ~An<sub>30-35</sub> far removed (about 10 cm) from the vein to ~An<sub>40-45</sub> adjacent to the vein.

The paragenesis in the plagioclase-rich vein margin was used for estimation of *P*, *T*, and fluid chemistry. The *P-T* estimate is 0.38 GPa and 535 °C, consistent with previous estimates for "peak" metamorphic conditions for area I (cf. Harte et al., 1984; Droop and Harte, 1995). The estimated *a*<sub>H<sub>2</sub>O</sub> and *a*<sub>HCl</sub>/*a*<sub>H<sub>2</sub>O</sub> at 0.38 GPa and 535 °C are 0.9 and 4.2 × 10<sup>-4</sup>, respectively. Fluid

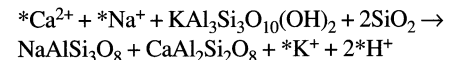


**Figure 2.** Plagioclase (PI) porphyroblast "spot" long axis as function of distance measured perpendicular to vein-wall-rock contacts. Long axis dimensions determined on 52 samples from study areas I, II, and III. Mean long axis length (mm) ±95% confidence also shown.

speciation calculations indicate that the total Cl molality compatible with the estimated *a*<sub>HCl</sub>/*a*<sub>H<sub>2</sub>O</sub> is about 0.65 *m*. Varying *a*<sub>H<sub>2</sub>O</sub> between 0.1 and 1.0 in the calculations has a negligible impact on this result. For *a*<sub>H<sub>2</sub>O</sub> = 0.9 and total Cl molality = 0.65 *m*, the following total molalities were computed: Na = 0.50 *m*, K = 0.13 *m*, Ca = 9.2 × 10<sup>-3</sup> *m*, Mg = 9.0 × 10<sup>-4</sup> *m*, and Fe = 2.0 × 10<sup>-5</sup> *m*. The calculated pH = 4.6 (neutral pH at these *P-T* conditions is 4.8).

## DISCUSSION

The most likely explanation for the observed mineralogical and chemical systematics (cf. Fig. 4) is that the plagioclase-rich zones are alteration selvages that developed as a result of mass transfer associated with vein-forming processes (cf. Ague, 1994). Fluid must have been present during alteration because solid-state diffusion rates at greenschist facies conditions are too slow to have produced the observed metasomatic changes. Fracture formation will tend to increase significantly the permeability of typical metamorphic rocks. Consequently, the veins probably represent areas where permeability was created (through fracturing) and fluid flow was focused during deformation. The increases in bulk Na/K, Ca/K and (Na + Ca + K)/Al toward the vein leave little doubt that reactions of the general form:



played a key role in the alteration (asterisks denote components in the aqueous phase required for mass balance; precise speciation is not implied). Moreover, the assemblage garnet + biotite + chlorite was stable in the heavily altered areas directly adjacent to the vein, whereas chloritoid + biotite + chlorite was stable in less-altered and unaltered areas. The transition to the less-aluminous, garnet-bearing assemblage was probably a result of the increases in bulk rock (Na + Ca + K)/Al and (Mg + Fe<sup>T</sup>)/Al that occurred near the vein margin (Fig. 4). The results presented herein lead to the surprising

<sup>1</sup>GSA Data Repository item 9704, Appendixes 1 and 2, is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301. E-mail: editing@geosociety.org.

conclusion that greenschist facies mass transfer of nonvolatile elements produced the bulk compositions necessary for garnet growth in at least some of the rocks within the northeastern part of Barrow's garnet zone. The role of mass transfer in other parts of the garnet zone remains to be established.

For the sample investigated, selvages are 2–3 cm wide in psammite layers, 6–8 cm wide in semipelite layers, and 10–15 cm wide in pelite layers (Figs. 3 and 4). Consequently, the relative ease of grain-scale solute migration through the various rock types probably increased in the order psammite, semipelite, pelite. These relations are consistent with those expected for metamorphic rocks (Chamberlain and Conrad, 1991) and suggest that the transfer of mass to and from the vein through the rock matrix was largely parallel to the layering (and schistosity). Several mechanisms for driving the mass transfer are examined below.

Regional infiltration of highly sodic fluids that are grossly out of chemical equilibrium with their wall rocks can produce voluminous Na ± Ca alteration (Battles and Barton, 1995). Possible fluids include (1) seawater, (2) saline formation waters, (3) brines derived from evaporates (or metaevaporites), and (4) saline and alkaline surface waters formed in arid environments. The depth of metamorphism (~14 km) argues against infiltration of chemically unmodified seawater, formation water, or other near-surface waters. The Cl contents of evaporite-derived brines are far in excess of that of the metamorphic alteration fluid (cf. Battles and Barton, 1995). Moreover, carbonate units (potential hosts for evaporate beds) are exceedingly rare throughout the chloritoid schist belt. The nearest major metacarbonate sequence is exposed ~5 km to the north of the belt. On balance, it is unlikely that infiltration of sodic fluids caused the metasomatism.

Another possibility is that the metasomatism was driven by fluid flow along a  $T$  gradient. For example, Orville (1962) discussed how flow of Cl-bearing fluid up a  $T$  gradient (up- $T$  flow) will tend to deplete typical quartzofeldspathic rocks in K and enrich them in Na. By comparing observed metasomatic effects to those predicted by experiments or mass-transfer calculations, one can, in principle, determine time-integrated fluid flux and flow direction (cf. Orville, 1962; Dipple and Ferry, 1992). The determinations require values for the partial derivatives:  $(\partial X_i/\partial T)_P$ , where  $X_i$  is the mole fraction of species  $i$  in the fluid. The chemical effects of flow along  $P$  gradients are typically negligible and are ignored here. Fluid speciation calculations using  $P = 0.4$  GPa,  $a_{\text{H}_2\text{O}} = 0.9$ , and total Cl molality = 0.65  $m$  yield the following  $(\partial X_i/\partial T)_P$  values, expressed as  $^{\circ}\text{C}^{-1}$ , for the  $T$  range ~525–550  $^{\circ}\text{C}$ : (1) Na =  $2.4 \times 10^{-5}$ ,\* (2) K =  $2.9 \times 10^{-5}$ , (3) Ca =  $-2.3 \times 10^{-6}$ , (4) Mg =  $-1.7 \times 10^{-7}$ , and (5) Fe =  $-6.4 \times 10^{-9}$ . Negative values indicate that mass is removed from the fluid and deposited in the rock with increasing  $T$ ; positive values indicate the opposite sense of

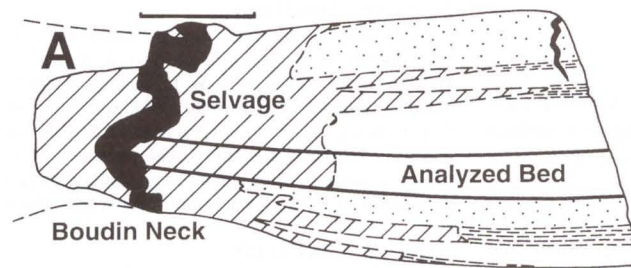


Figure 3. A: Sketch of sample JAB-101L; view perpendicular to boudin axis and relict bedding. Quartz vein shown in black, approximate extent of macroscopically observable selvage denoted by diagonal rule. Protolith rock types: stipple = psammite; unpatterned = semipelite; short dashes = pelite. Margins of semipelite bed analyzed in detail (see Fig. 4) denoted by heavy black lines. Penetrative schistosity is subparallel to bedding. Scale bar = 4 cm. B: Photograph illustrating plagioclase-rich selvage adjacent to vein (vein at left; ovoid, gray "spots" are plagioclase porphyroblasts). Round, pink crystals are garnets (arrow). Garnets occur in all layers adjacent to vein but are largest in pelitic and semipelitic layers. Scale in centimetres.

mass exchange. The calculations predict that up- $T$  flow will increase rock Na/K, Ca/K, and Mg/Fe<sup>T</sup>; these increases are all qualitatively consistent with the selvage metasomatism (Fig. 4). Thus, it is concluded that the metasomatism is best explained as a result of regional fluid flow in a direction of increasing  $T$ .

Following Dipple and Ferry (1992), the time-integrated fluid flux was estimated from the observed Na and K metasomatism because (1) the thermodynamic properties of Na-K aqueous chloride solutions are well known and (2) Na and K species dominate the solute budget. The  $T$  gradient in the direction of flow,  $dT/dz$ , was set to 25  $^{\circ}\text{C km}^{-1}$ ; changing  $dT/dz$  changes the flux estimate by the factor  $25/(dT/dz)$  ( $z$  is the distance coordinate). Using the model of Dipple and Ferry (1992), which assumes advection-dominated transport and local fluid-rock equilibrium, the calculated time-integrated flux required to produce the observed change in Na/K at the vein-wall-rock contact (Fig. 4) is  $\sim 3 \times 10^6 \text{ cm}^3 \text{ fluid cm}^{-2}$ . This result is subject to significant uncertainties, but suggests, nonetheless, that metasomatism required infiltration of large volumes of aqueous fluid.

As shown in Figure 4, Si/Ti decreases significantly in the vein selvage. Under the reasonable assumption that Ti was less mobile than Si (cf. Ague, 1994), the Si/Ti systematics (Fig. 4) indicate that Si was lost from the selvage. The most likely scenario is that silica diffused from the rocks into cracks (now represented by quartz veins) during deformation and fluid flow (cf. Ague, 1994). In addition, the up- $T$  flow could have dissolved some silica from the wall rocks, although the time-integrated fluid flux was insuf-

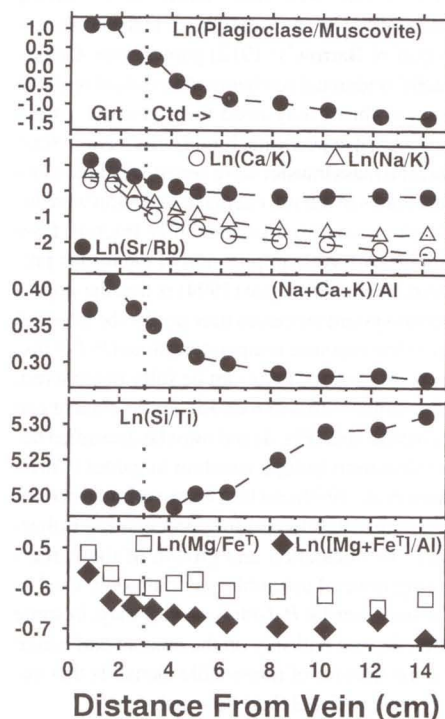


Figure 4. Mineralogical and bulk-rock geochemical profiles across typical metamorphosed semipelite bed (see Fig. 3A). Vein-wall-rock contact is at  $X = 0$  cm; rock at  $X > -10$  cm appears to have been little affected by vein-related mass transfer. Bulk-rock chemical ratios computed on molar basis. Garnet + biotite + chlorite present to left of vertical dotted line; chloritoid + biotite + chlorite present to right. Ctd = chloritoid; Grt = garnet; Fe<sup>T</sup> = total iron; Ln = natural log.

\*Note: Na value should be:  $-2.4 \times 10^{-5}$

ficient to cause massive silica dissolution (cf. Dipple and Ferry, 1992).

The veins constitute ~13% of the rock mass and have a mean width of ~3 cm. A conservative estimate for the mean selvage width throughout the spotted chloritoid schist belt is 6 cm (3 cm on either side of a vein). These estimates imply that nearly 40 vol% of the rock mass is made up of veins and their selvages. Therefore, nonvolatile element mass transfer appears to have affected a substantial portion of the total rock volume.

The sources and regional pathways for the fluids will be addressed by future studies. One possibility is that near horizontal up-*T* flow occurred as part of a giant (10–100 km flow path) metamorphic hydrothermal system analogous to the type envisioned by Ferry (1992). Another possibility is that near vertical up-*T* flow occurred along an inverted geothermal gradient in the hanging wall above a long-lived, regional overthrust. The Highland Boundary fault (Fig. 1) is interpreted to be such a thrust in some tectonic models, but the relationships between fault movement and metamorphism remain unclear (cf. Harte et al., 1984).

## CONCLUSIONS

The results of this study strongly suggest that chemical and mineralogic alteration occurred adjacent to fractures (now quartz veins) during greenschist facies metamorphism in the northeastern part of Barrow's (1912) garnet zone. Consequently, traditional isochemical models of regional metamorphism may need to be revised. In the study area, it appears that brittle deformation, fluid flow, and mass transfer were genetically linked because zones of intense chemical alteration were localized around high permeability fracture flow conduits. A further implication of the results presented here and in Ague (1994) is that the impact of open-system processes may need to be assessed before the pressure-temperature-time (*P-T-t*) histories of orogenic belts can be fully deciphered. For example, shifts in rock Rb and Sr content can be considerable (Fig. 4) and must be quantified before strontium isotope zonation in garnet (Christensen et al., 1989) can be used to quantify rates of garnet growth and tectonic processes. Furthermore, the nucleation and growth of the garnet-bearing mineral assemblages commonly used to determine crustal *P-T-t* trajectories may, in some rocks, depend critically on the amount and nature of mass transfer of nonvolatile elements that occurs along regional fluid flow paths.

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# SUPPLEMENTAL MATERIAL: ROCK AND MINERAL ANALYSES

## APPENDIX 1. XRF ANALYSES OF JAB-101L SEMI-PELITE LAYER

X (cm)	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	MgO	MnO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	Rb	Sr	LOI	Total
0.80	73.3	0.539	11.0	6.15	1.86	0.05	1.48	1.11	0.84	0.07	41	137	2.55	98.95
1.70	73.8	0.544	11.2	5.95	1.71	0.04	1.60	1.20	1.06	0.08	56	152	2.30	99.48
2.55	73.6	0.542	11.4	6.05	1.68	0.04	1.14	0.94	1.54	0.07	67	121	2.75	99.75
3.35	73.2	0.544	11.1	5.85	1.62	0.04	0.81	0.73	1.79	0.08	71	102	2.75	98.51
4.20	74.5	0.556	11.1	5.74	1.61	0.04	0.62	0.59	1.89	0.08	73	88	2.90	99.63
5.05	74.3	0.544	11.3	5.79	1.64	0.05	0.54	0.51	1.98	0.08	76	77	2.85	99.58
6.30	74.2	0.542	11.3	5.77	1.59	0.05	0.44	0.42	2.10	0.08	77	73	2.65	99.14
8.30	75.4	0.526	11.2	5.67	1.56	0.05	0.37	0.32	2.15	0.08	78	65	2.45	99.78
10.20	76.2	0.511	10.9	5.51	1.50	0.05	0.32	0.29	2.12	0.08	72	61	2.50	99.98
12.30	75.3	0.503	10.5	5.48	1.45	0.05	0.26	0.25	2.14	0.08	70	61	2.50	98.51
14.20	76.9	0.505	10.6	5.18	1.40	0.05	0.22	0.26	2.12	0.08	72	60	2.35	99.67

*Notes* : Rb and Sr concentrations in ppm, all other concentrations in wt%. All Fe as Fe<sub>2</sub>O<sub>3</sub>. LOI = loss on ignition. X is distance from vein margin to center of analyzed rock section. X-ray fluorescence (XRF) analyses done by X-ray Assay Laboratories, Don Mills, Ontario.

## APPENDIX 2. JAB-101L MINERAL ANALYSES

	Plagioclase	Garnet*	Chlorite	Muscovite	Biotite
Si	2.609	3.000	2.589	3.151	2.708
Al <sup>IV</sup>	1.393	–	1.411	0.849	1.292
Al <sup>VI</sup>	–	2.022	1.457	1.867	0.524
Ti	N.D.	0.003	0.005	0.008	0.104
Fe	0.002	2.035	2.841	0.093	1.463
Mg	N.D.	0.146	1.639	0.078	0.751
Mn	N.D.	0.482	0.027	<0.001	0.008
Ca	0.387	0.297	N.D.	N.D.	N.D.
Ba	N.D.	N.D.	0.001	0.006	0.004
Na	0.604	N.D.	0.001	0.078	0.020
K	0.004	N.D.	0.003	0.785	0.831
F	N.D.	N.D.	B.D.	0.020	0.081
Cl	N.D.	N.D.	0.001	0.001	0.006
Wt% Sum	99.66	99.02	88.03	95.04	95.05

*Notes* : All Fe as FeO. N.D.=not determined. B.D.=below detection. Structural formulas for plagioclase, garnet, chlorite, muscovite, and biotite based on 8, 12, 14, 11, and 11 oxygens, respectively. Compositions determined using the JEOL JXA-8600 electron microprobe at Yale University, employing wavelength dispersive spectrometers, natural and synthetic standards, off-peak and fluorescence-corrected mean atomic number background corrections, and  $\phi(\rho z)$  matrix corrections.

\*Rim composition.