

Compositional Variation of Muscovite as a Function of Metamorphic Grade and Assemblage in Metapelites from N. W. Maine

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Abstract. Chemical data are presented for 49 muscovites from high and low Al specimens collected from N. W. Maine at metamorphic grades ranging from the upper staurolite to the upper sillimanite zone. Data also are presented for two muscovite from St. Paul Island, two muscovites + three paragonites from Gassetts, Vermont, and one muscovite from an adamellite in N. W. Maine.

These data give further information on the effects of P, T, and bulk composition on muscovite composition. Specifically, temperature clearly influences the Na/Na + K ratio of muscovite in limiting assemblages but may not have much effect on the phengite content. Increase in pressure clearly does cause an increase in phengite content. Bulk composition (assemblage) has a very great effect on both Na/Na + K ratio and phengite content so that attempts to use either of these factors to monitor metamorphic grade should generally be done in the context of a limiting assemblage¹.

Introduction

Much data have been accumulated on muscovite from metamorphic rocks in recent years. Commonly attention has been focused on variations of muscovite composition as a function of metamorphic grade and bulk composition (i.e. assemblage). Much of the data have come from studies of specimens from the Swiss and Italian Alps, e.g. Graeser and Niggli (1967), Wenk (1970), Schwander *et al.* (1968), Sassi (1972) and Cipriani *et al.* (1971). The paper by Cipriani *et al.* is by far the most comprehensive available and includes virtually all important references to that time and gives a summary of all available analyzed muscovites from metamorphic rocks.

This paper is based on analyses of 49 muscovites from upper staurolite to upper sillimanite zone rocks and one from an adamellite in N. W. Maine. Also included are analyses of two muscovites from St. Paul Island (from the study of Phinney, 1963) and two muscovites and coexisting paragonite plus one single paragonite from Gassetts, Vermont. In the context of questions considered in Cipriani *et al.* (1971) and Schwander *et al.* (1968) the intent of this paper is as follows:

- (1) To show effects of metamorphic grade on muscovite composition.
- (2) To illustrate the effects of bulk composition (i.e. assemblage) on muscovite composition within a given grade.

¹ Limiting Assemblages being those in which the number of phases present equals the number of components required to describe the phases. Hence, phase compositions will be a function of the intensive parameters, Albee (1965).

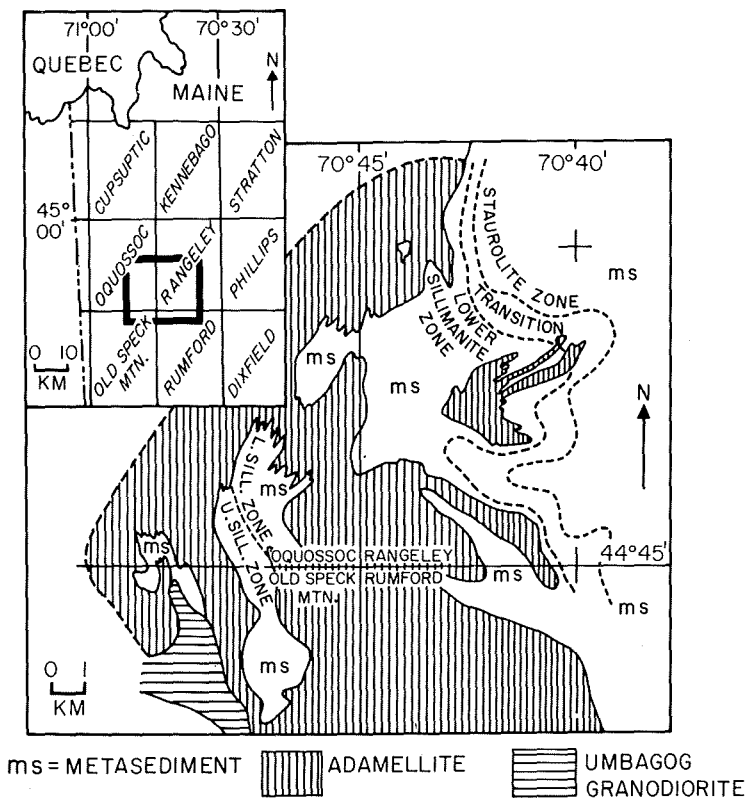


Fig. 1. Geologic map of portions of the Oquossoc, Old Speck Mtn., Rumford, and Rangeley quadrangles, Maine. Oquossoc (Guidotti, open file report, Maine Geol. Survey); Old Speck Mtn., Milton (1961); Rangeley, Moench (1971); Rumford, Moench (In Progress)

(3) To compare the data from N. W. Maine muscovites with those for samples from St. Paul Island and Gassetts, Vermont to see the effects of radically different pressure during genesis.

Metamorphic Environment and Selection of N. W. Maine Specimens

Fig. 1 shows the location and general geology² of the area from which specimens were collected. Also shown are the several metamorphic isograds from Guidotti (1970A), (1970B). The metamorphic grades range from the upper part of the staurolite zone to the upper sillimanite zone as defined in Guidotti (1970B). The specific zones are:

group (a) Upper Staurolite Zone (U.St. zone): Characterized by the assemblage St + Bio + Gn + Chl³.

² Rangeley, Moench (1971); Rumford, Moench (in progress); Old Speck Mtn, Milton (1961); Oquossoc, Guidotti (unpublished).

³ Mu = Muscovite; Qz = quartz; Bio = biotite; St = staurolite; Si = sillimanite; Gn = garnet; Pl = plagioclase; Chl = chlorite Q; Pg = paragonite; Ky = kyanite; Ksp = K-feldspar.

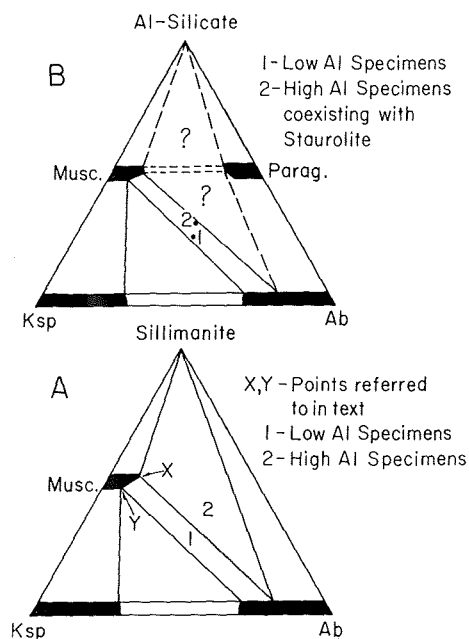


Fig. 2. Schematic AKNa projections (Thompson, 1961) for the Transition zone through Upper Sillimanite zone (A), and Upper Staurolite zone (B) of N.W. Maine

group (b) Transition zone (Tr. zone): Characterized by the assemblage $Si + St + Gn + Bio \pm Chl$.

group (c) Lower Sillimanite zone (Rangeley) (L. S. zone): Characterized by the assemblage $Si + St + Gn + Bio$.

group (d) Lower Sillimanite zone (Oquossoc) (L. S. zone): Characterized by the assemblage $Si + St + Gn + Bio$.

group (e) Upper Sillimanite zone (U. S. zone): Characterized by the assemblage $Si + Gn + Bio$.

From Fig. 1 it is evident that (c) and (d) are part of the same zone but (d) is in the higher grade portion. The Tr. zone is really only a "smearing" out of the isograd reaction separating the U. St. zone and L. S. zone. It is given the name Tr. zone because:

- (1) It has mineralogic aspects of both the U. St. and L. S. zones.
- (2) It is a mappable zone.

All of the indicated metamorphic grades correspond to moderately low pressures (i.e. below the aluminosilicate triple point) of ~ 3.5 Kb according to Guidotti (1970B).

Within each grade specimens for analysis of muscovite were chosen in two subgroups: (1) specimens with highly aluminous minerals (designated as high Al specimens) such as St or Si, and (2) specimens lacking such aluminous minerals (designated as low Al specimens). For groups (b), (c), (d), (e) this split involves specimens containing $Si + Mu + Pl$ vs specimens containing only $Mu + Pl$, (see

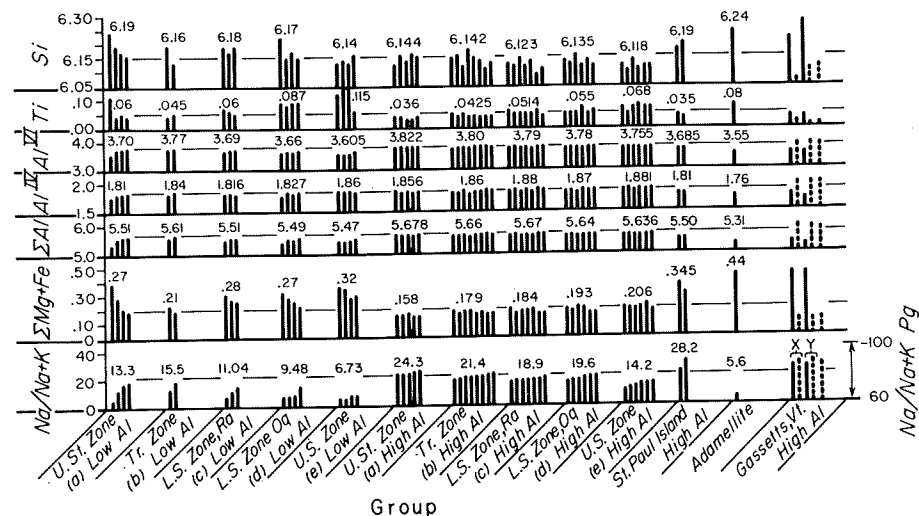


Fig. 3. Summary of muscovite analytical data based on formula proportions. Each bar represents one specimen; broken bars for paragonite. Numbers over groups of bars give average values. X, Y are coexisting pairs of muscovite and paragonite. Specimens in order of increasing Na/Na + K. Groups (a)–(e) as in text

Fig. 2A). Specimens from group (a) all contain the assemblage Mu + Pl on an AKNa projection. Fig. 2B shows the likely configuration of tie lines for the AKNa system in the U. St. zone. The high Al specimens from group (a) all coexist with staurolite. Hence it is reasonable to expect them to plot near the upper boundary of the Mu + Pl field. Chemical data discussed below strongly support this suggestion.

Analytical Data

The analytical results are presented according to groups (a–e) in Tables 1A–1J⁴. Tables 1A–1E contain the results for muscovites from Al specimens and Tables 1F–1J for low Al specimens. Table 1K presents the analyses of muscovite from St. Paul Island, muscovite and paragonite from Gassetts, Vermont, and muscovite from an adamellite in N.W. Maine. Fig. 3 summarizes the important aspects of these tables and will be used extensively in the following sections of this paper. Table 1 presents representative analyses from Tables 1A–1K.

Analyses were carried out on an ARL electron probe using the Bence and Albee (1968) oxide procedure. Operating conditions were set so as to maximize counting ratio and minimize sample damage. The *anhydrous* analytical totals are clumped between 95 and 96% so that addition of 4 to 5% H₂O brings the analytical total to 100%. Formulas were calculated on a basis of 22 anhydrous oxygen. Comparison with several analyses done on the same muscovite samples (all from high Al specimens) by the Cal Tech electron probe [reported in Guidotti (1970B)] and (in prep) shows a very good correlation of values. However, for elements present at low levels (e.g. Fe, Mg, Ti) one needs to use a single set of analyses to see

⁴ Tables 1A–1K are on file with ASIS/NAPS c/o Microfiche Publications, 305 E. 46th St., New York, N.Y. 10017. Please order NAPS document No. 02151 remitting in advance \$ 1.50 for microfiche or \$ 5.00 for photocopies.

Table 1. Representative analyses of muscovite from high Al specimens (without*) and low Al specimens (*)

	A	A*	B	B*	C	C*	D	D*
	Ra-C50-66	Ra-a31-66	Ra-b48-66	Ra-C30-66	Ra-b93-66	Ra-a90-66	O-J-67	O-J-16'
FeO	0.68	1.27	0.68	0.88	0.67	1.01	0.83	1.22
MnO	0.005	0.02	0.01	0.02	0.004	0.02	0.02	0.03
MgO	0.42	0.77	0.57	0.66	0.52	0.78	0.48	0.73
SiO ₂	46.69	46.51	46.44	46.96	46.20	46.38	46.43	46.30
Al ₂ O ₃	36.74	35.22	36.58	35.87	36.47	35.33	36.38	35.27
K ₂ O	8.44	9.88	8.86	9.94	9.11	9.75	9.33	10.33
BaO	0.17	0.26	0.26	0.20	0.21	0.61	0.17	0.43
Na ₂ O	1.90	0.93	1.58	0.94	1.33	0.82	1.37	0.60
TiO ₂	0.38	0.42	0.47	0.44	0.46	0.59	0.49	0.84
H ₂ O ^a	4.63	4.78	4.62	4.15	5.10	4.75	4.56	4.31
Formula based on 22 oxygens								
Si ^{IV}	6.14	6.19	6.12	6.19	6.12	6.17	6.13	6.15
Al ^{IV}	1.86	1.81	1.88	1.81	1.88	1.83	1.87	1.85
Al ^{VI}	3.83	3.71	3.80	3.76	3.81	3.71	3.79	3.67
Fe	0.07	0.14	0.08	0.10	0.07	0.11	0.09	0.14
Mg	0.08	0.15	0.11	0.13	0.10	0.16	0.10	0.15
Mn	0.001	0.002	0.002	0.003	0.000	0.002	0.002	0.003
Ti	0.04	0.04	0.05	0.04	0.05	0.06	0.05	0.08
Σ	4.021	4.042	4.042	4.033	4.03	4.042	4.032	4.043
K	1.42	1.68	1.49	1.67	1.54	1.66	1.57	1.75
Na	0.49	0.24	0.40	0.24	0.34	0.21	0.35	0.15
Ba	0.01	0.01	0.01	0.01	0.01	0.03	0.01	0.02
Σ	1.92	1.93	1.90	1.92	1.89	1.90	1.93	1.92
ΣAl	5.69	5.52	5.68	5.57	5.69	5.54	5.66	5.52
ΣMg + Fe	0.15	0.29	0.19	0.23	0.17	0.27	0.19	0.29
Na/Na + K	25.6	12.5	21.1	12.55	18.10	11.2	18.4	7.90

A = Upper Staurolite Zone; B = Transition Zone; C = Lower Sillimanite Zone, Rangeley;
D = Lower Sillimanite Zone, Oquossoc

^a H₂O required to sum oxides to 100%.

most clearly some of the trends discussed below. The same is true for attempts to monitor small changes of major cations. Of particular interest though, is the observation that when the Cal Tech analyses are viewed separately (covering the same metamorphic grades as in this study), trends emerge which are similar to those based upon analyses in this study. In view of these comments, analyses of specimens O-C-13, O-J-88, O-C-35, O-C-38 are especially worth comparing, Table 2, because they were performed on the same mineral separate on both the Cal. Tech. and U. Wisc. electron probes. Both were analyzed by the Bence and Albee (1968) procedure; however, different standards were employed.

Discussion of Analytical Results (1): High Al Specimens

It is evident from Fig. 2A and B that muscovite in the high Al specimens in groups (b), (c), (d), and (e) will have compositions dependent only on the meta-

Table 2. Comparison of muscovite analyses from this study (A), with results reported in Guidotti (1970b), (B)

	(A) O-C-13	(B) O-C-13	(A) O-J-88	(B) O-J-88	(A) O-C-35	(B) O-C-35	(A) O-C-38	(B) O-C-38
FeO	0.83	0.90	0.86	0.92	0.81	0.83	0.87	0.81
MnO	0.03	0.01	0.02	—	0.01	—	0.01	0.01
MgO	0.54	0.54	0.43	0.50	0.43	0.47	0.59	0.58
CaO	—	—	—	—	0.00	—	0.00	—
SiO ₂	45.92	45.58	46.22	45.83	46.37	45.82	46.78	46.62
Al ₂ O ₃	36.19	36.50	36.28	36.59	36.18	36.97	36.17	37.71
K ₂ O	9.89	9.79	9.40	9.18	8.92	8.91	9.09	9.17
BaO	0.35	n. d.	0.23	n. d.	0.19	n. d.	0.21	n. d.
Na ₂ O	0.94	0.94	1.21	1.05	1.60	1.52	1.39	1.37
TiO ₂	0.53 ^a	0.69	0.66	0.71	0.52	0.51	0.49	0.50
H ₂ O ^b	4.84	5.05	4.75	5.22	5.04	4.97	4.46	3.23

^a Considerable scatter of probe counts.

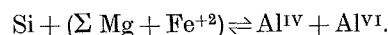
^b Added to bring Sum to 100%.

n. d. = Not determined.

morphic conditions. This is also approximately true for muscovite from high Al specimens in group (a).

Consideration of Fig. 3 and Tables 1A to 1E (especially noting the formula proportions) shows the following as functions of increased metamorphic grade:

- (1) The sum of Mg + Fe increases as also does Ti⁴⁺.
- (2) Na decreases and K increases; so that the ratio Na/(Na + K) decreases.
- (3) There is a possible but rather uncertain suggestion that Al, especially in the octahedral sites, decreases. There is also a similar suggestion for Si^{IV}.
- (4) It is clear that the increase of Σ Mg + Fe⁵ is not fully balanced by increase of Si and decrease of Al—that is an increase of the phengite content via:



(5) Instead, some of the decrease of Al^{IV} that should result from increase of Σ (Mg + Fe) in the octahedral sites, seems to be offset by (Ti⁴⁺) requiring substitution of (Al⁺³)^{IV} for (Si⁺⁴)^{IV}. This is also suggested by the apparent increase of Al^{IV} as grade increases, despite a decrease of Σ Al. [Note also that Σ (Mg + Fe) — Ti \approx Si in excess of (6)]. Moreover, the decrease of Al^{VI} is numerically only slightly less than the increase of Σ [(Mg + Fe + Ti)], if comparison is made between the average values in the U. St. zone and U. S. zone.

(6) Despite the small, systematic compositional variations discussed above, the overriding feature is that within a given group of samples, Al, Mg, and Fe are quite constant, due to the fact that the muscovite occurs in a limiting assemblage, Albee (1965). From group to group there is no large variation of these cations as to be expected from the tie-line geometry shown in Fig. 2A. However Na/(Na + K) does undergo a significant change due to the lateral migration of point X on Fig. 2A.

⁵ Fe in the Maine specimens is assumed to be primarily Fe⁺⁺ because: (1) No magnetite or hematite is present in these rocks. (2) Most specimens contain graphite, thereby indicating low *f*O₂ conditions.

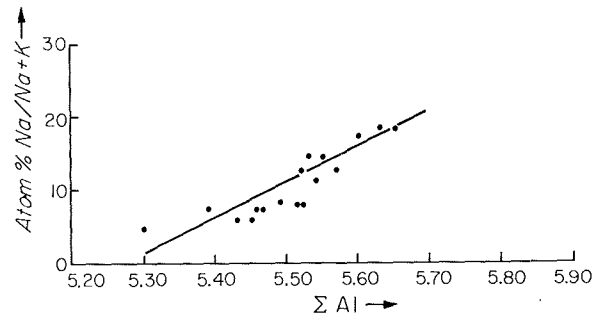


Fig. 4. Na/(Na + K) vs $(Al^{IV} + Al^{VI})$ in low Al specimens, N. W. Maine

Discussion of Analytical Results (2): Comparison of Low Al and High Al Specimens within a Given Grade

In contrast with the muscovites from high Al specimens, those from low Al specimens show the following:

(1) Commonly there is a larger variation of ΣAl but ΣAl is always \leq that in the muscovite from high Al specimens.

(2) Generally there is some increase of Si^{IV} but it is not large and probably not enough to account for all of the observed decrease of ΣAl .

(3) Much of the decrease in Al shows up in the octahedral positions and seems to relate to significant increases in $\Sigma (Mg + Fe + Ti)$.

(4) Not all specimens show (1)–(3) but a spectrum exists. It would appear that to varying degrees, from specimen to specimen, increase of $\Sigma (Mg + Fe)$ is balanced by increase of Ti^{+4} and/or phengite content as discussed above. Fig. 3 suggests that increase of $\Sigma (Mg + Fe)$ in the low Al specimens of group (a) is balanced by increase of phengite content. In contrast, in group (d), the increase of $\Sigma (Mg + Fe)$ appears to be balanced also in part by increase of Ti^{+4} .

(5) Fig. 4 shows a quite clear relation between the ratio Na/(Na + K) and ΣAl and clearly implies the sloped shape shown between points X and Y for the muscovite phase region on Fig. 2A. Moreover this supports the earlier suggestion that the muscovite from the high Al specimens of group (a), is close to that appropriate to the tie line forming the upper boundary of the Mu + Pl field.

Two important considerations relating to the muscovites from the Mu + Pl field are:

(a) The wide variation of Al, Si, Mg, Fe, and Ti are fully expected because the assemblage is not a limiting one.

(b) In this assemblage, bulk composition, e.g. Al_2O_3 or Na_2O has a great effect on the Na/(Na + K) ratio of muscovite. Hence, this ratio is an indicator of metamorphic grade only in limiting assemblages.

Discussion of Analytical Results (3): Comparison of N.W. Maine Results with Those from St. Paul Island and Gassetts, Vermont

The specimens from St. Paul Island contain the assemblage St + Gn + Bio and are put in the staurolite zone by Phinney (1963). This would be the U. St. zone in

the terminology of this paper as SP. 28 and SP 51 are from just downgrade of the establishment of the Ky + Bio join according to Phinney. However, from the assemblage data given they could actually be isofacial with the rocks Phinney describes with the assemblage Ky + St + Bio. It is clear that they formed under conditions *near* those required to establish the join Ky + Bio just as the USt. zone and Tr zone in this study involve conditions for establishment of the Si + Bio join. Hence the St. Paul specimens are high-pressure equivalents (also somewhat higher temperature) of group (a) and (b) specimens. Because SP28 and SP51 contain staurolite, it is likely, (for reasons discussed above), that the muscovite from them lies near the upper boundary of the Mu + Pl. Field.

The specimens from Gassetts, Vermont have Mu + Pg + Ky + St + Gn + Bio + Chl and hence are approximately the high-pressure equivalents of the Tr. zone in N.W. Maine. They probably involve the tie line flip of Chl + St. to (aluminosilicate) Ky + Bio.

One can estimate (from the various phase diagrams of the aluminosilicates) that the specimens from St. Paul Island and Gassetts formed at pressures at least several kilobars greater than those in N.W. Maine and probably at somewhat higher temperatures. From Fig. 2A and 2B it is evident that muscovite from the assemblage Ky + Mu + Pg should be as Al rich as that from the assemblage Si + Mu + Pl and possibly it should be even more Al-rich.

Considering the tie-line geometry and bulk compositions one would expect similar Al, Fe, Mg, and Si in muscovites from St. Paul Island, Gassetts, Vermont, and the high Al specimens from group (a) and (b) of N.W. Maine. Fig. 3 shows that this is not the case, as specimens from St. Paul Island and Gassetts have significantly greater phengite contents. (Note also that Ti⁺⁴ is similar in all). In the light of work by Velde (1965), Ernst (1963) and numerous others the most reasonable explanation is that high pressure has facilitated higher phengite content in the St. Paul and Gassetts specimens. Indeed, pressure is the only parameter which is significantly different between the contrasted muscovite groups. A point of interest is the radically different phengite contents in coexisting muscovite and paragonite and its implications for tie line slopes in Fig. 2B. The explanation for this radical difference in phengite content between Mu and Pg is not obvious.

Discussion of Analytical Results (4): Muscovite from Adamellite in N.W. Maine

Only one muscovite from an adamellite has been analyzed (Fig. 3). It coexists with Pl + Ksp. To be noted, this muscovite is by far the most phengitic and K-rich of all the N.W. Maine samples. This is certainly expected in terms of the tie-line geometry of Fig. 2A, e.g. note point Y. Based upon the discussions in Guidotti (1970A, B), muscovite from the adamellite has probably formed at a pressure similar to the metapelites and at a temperature only 100°C or so greater than the metapelites from the U.S. zone. Hence it can be reasonably compared with the metamorphic muscovites. The main point to be noted is the extreme effect of difference in assemblage (bulk composition) (Si + Mu + Pl vs Ksp + Mu + Pl) on the muscovite composition.

Conclusions

Several conclusions emerge from the above considerations.

(1) Any use of $\text{Na}/(\text{Na} + \text{K})$ as a monitor of metamorphic grade should be done in terms of specific assemblages and these should be *specific limiting assemblages*.

(2) The same is true for attempts to use phengite content as an indicator of metamorphic grade. Temperature may have a relatively small influence on phengite content but pressure clearly has a large influence.

(3) For relatively low-pressure metamorphism, phengite content may not vary much as a function of grade from U. St. zone to $\text{Ksp} + \text{Si}$ zone (see Evans and Guidotti (1966) for data on the latter). Moreover, as developed in the discussion of analytical procedures, even the minor variation between analyses carried out in different laboratories may be sufficient to mask these small changes in phengite content. Alternatively, any proposed "small variation" in phengite content based upon muscovite analyses carried out in different laboratories should be viewed with some degree of skepticism.

(4) The observations and conclusions in this paper are clearly in general agreement with those presented in Cipriani *et al.* (1971) and Sassi (1972). However, the closer control on assemblage maintained in this study, enables a somewhat more refined interpretation of some of the factors influencing the composition of muscovite.

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