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Pressure, temperature and metamorphic zonation studies of pelitic schists in the Merrimack Synclinorium, south-central New Hampshire

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Abstract

Metamorphic isograds in pelitic rocks in nine 15′ quadrangles in south-central New Hampshire are divisible into the upper 5 of Tracy's (1975) high-grade zones: (2) the staurolite–sillimanite zone, (3) the sillimanite zone, (4) the sillimanite–alkali feldspar–muscovite zone, (5) the sillimanite–alkali feldspar zone, (6) and the sillimanite–alkali feldspar–cordierite zone. These zones replace the single alkali feldspar–sillimanite zone originally mapped by Thompson and Norton (1968). Biotite–garnet geothermometry and plagioclase composition studies confirm the zonal pattern deduced from the mineralogical relations.

Three stages of Acadian metamorphism are recognized. An early M(1) andalusite metamorphism is evidenced by sillimanite pseudomorphs after andalusite and flecky gneisses occurring near the contacts of the Devonian Kinsman Series plutons. This early metamorphism has been strongly overprinted by M(2) metamorphism, which locally attained cordierite–alkali feldspar grade and produced the dominant isograd pattern. M(2) metamorphism cross cuts Devonian Kinsman and Spaulding Series plutons in this region. M(3) locally retrograded the M(2) terrane, and produced a lower temperature decussate muscovite.

Introduction

Geothermometry, geobarometry, and metamorphic zonation studies are effective methods for determining the metamorphic history of a region. This paper uses these methods to determine the nature of metamorphism within nine, 15′ quadrangles located along the Merrimack Synclinorium in south-central New Hampshire.

Metamorphic rocks in southern New Hampshire were previously regarded as second-sillimanite grade (Thompson and Norton, 1968). Based on this assumption, models were presented to account for the widespread distribution of high-grade rocks in this region (Hamilton and Myers, 1967; Moench and Zartman, 1976). Both of these models suggest that plutonic rocks are the cause of metamorphism. However, the relationship between igneous and metamorphic rocks in southern New Hampshire is more complex. We present results showing that the dominant metamorphism is unrelated to the plutonic rocks and, in addition, that a single second sillimanite isograd does not describe the metamorphism for southern New Hampshire.

Regional geology

This study concentrates on the Mt. Monadnock, Peterborough, Lovewell Mountain, Hillsboro, Sunapee, Mt. Kearsarge, Penacook, Cardigan, and Holderness quadrangles. These quadrangles are located along the axis and western limb of the Merrimack Synclinorium in southern New Hampshire (Fig. 1).

The metasediments in this region range in age from Ordovician to Devonian in age (Lyons, 1979). Only the pelitic members of the formations that crop out in this region were examined. These rocks have been intensely deformed by at least three phases of folding (Englund, 1974; Nielson, 1974). The earliest stage of folding produced nappes with northeast or, locally, northwest trending axes. The nappes were later folded into broad open folds about northwest to west trending axes. Finally, the entire package was folded into tight folds with northeast trending axes, producing the dominant structural grain observed in the Merrimack Synclinorium. All of the folding is presumed to be Acadian in age (Lyons *et al.*, 1982).

The metasediments are intruded by syntectonic and post-tectonic plutons belonging to the New Hampshire Plutonic Series. Four members of the series are present: the Kinsman, Bethlehem, Spaulding, and the Concord suites. The Kinsman, Bethlehem, and the Spaulding

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plutons are early to late syntectonic intrusives. They range in age from 402 to 393 m.y., respectively (Lyons and Livingston, 1977). The Kinsman and Bethlehem suites are commonly strongly foliated, reflecting intrusion during early stages of folding. They range in composition from tonalites to granites. The Spaulding suite is less foliated and ranges from granite to gabbro in composition. The Spaulding plutons were intruded during the waning stages of the Acadian orogeny. All three of these suites occur as sheet-like plutons, no greater than 2.5 km thick (Nielson *et al.*, 1976). The Concord suite is considerably younger than the other three members with ages ranging from 327 to 275 m.y. (Lyons *et al.*, 1982). The Concord suite is typically a two-mica granite, and shows only a weak flow foliation.

A three stage sequence of metamorphism is recognized in south-central New Hampshire (Chamberlain, 1981; Chamberlain and Lyons, 1981). The earliest period of metamorphism, M(1), was a widespread, andalusite producing event, recognized by pseudomorphs of sillimanite after chiastolite. The second stage of metamorphism, M(2), was high-grade, and produced the dominant metamorphic assemblages now present in southern New Hampshire. This period of metamorphism is described in this paper. The third stage of metamorphism, M(3), was a local retrogression of these high-grade assemblages, and is recognized by chlorite replacing biotite and garnet, and the occurrence of a low temperature, decussate muscovite.

Methods

Compositions of 265 minerals from 102 polished thin sections were determined using a three spectrometer Materials Analysis Corporation automated electron microprobe at Massachusetts Institute of Technology. All analyses were collected using a 15 kV accelerating potential and a 0.03 μA beam current. Analytical data were reduced using the Bence and Albee (1968) procedure. Natural and synthetic silicates were used as standards.

All mineral analyses given in Tables 1 through 5 represent averages of 2 to 4 analyses collected on the same grain. Individual analysis points for a given sample were collected within 2 mm of each other. Each garnet core composition used in this study represents an average of 3 analyses taken within the center of the grain. Similarly, each garnet rim composition used in this study represents an average of 3 analyses collected within 50 microns of the edge of the garnet grain.

Metamorphic zones

The metamorphic zones mapped in this study follow the scheme outlined by Tracy (1975). Based on discontinuous and continuous reactions in rocks of pelitic composition, Tracy (1975) divided the metamorphic terrane in central Massachusetts into six zones. Each zone has its own characteristic mineral assemblage. The following is a brief discussion of each of these zones.

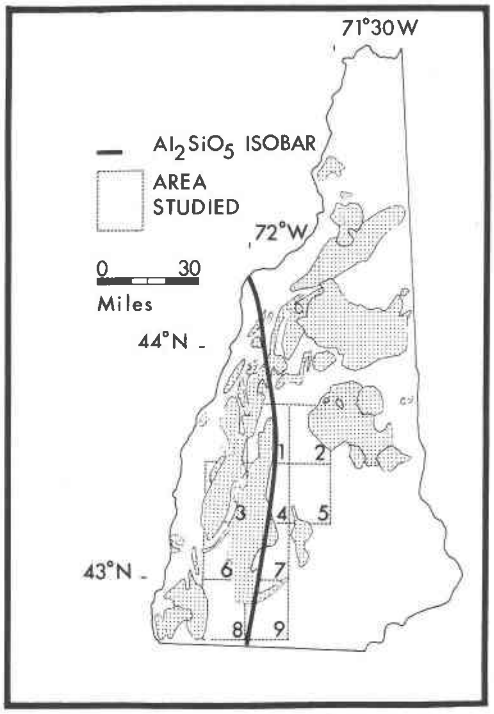

Fig. 1. General geological map of New Hampshire showing the major plutonic and metamorphic features (modified from Thompson and Norton, 1968). The quadrangles studied are: 1) Cardigan, 2) Holderness, 3) Sunapee, 4) Mt. Kearsarge, 5) Penacook, 6) Lovewell Mountain, 7) Hillsboro, 8) Mt. Monadnock, 9) Peterborough. The heavy line shows the approximate trace of the isobaric surface corresponding to the kyanite-sillimanite-andalusite triple point as shown by Thompson and Norton (1968). Andalusite is found east of the isobar and kyanite west of the isobar.

Zone 1

The characteristic mineral assemblage for this zone is kyanite–muscovite–staurolite–garnet–biotite–quartz. We did not find this mineral assemblage in south-central New Hampshire. However, kyanite–staurolite bearing rocks are found just north of this study area (Rumble, 1973), and are also found to the south-west in the Bronson Hill Anticlinorium (Tracy and Robinson, 1980).

Zone 2

The lowest grade mineral assemblage present in south-central New Hampshire belongs to the lower sillimanite zone (Zone 2). In this zone kyanite is displaced by sillimanite, resulting in the characteristic mineral assemblage: sillimanite–muscovite–staurolite–garnet–biotite–quartz.

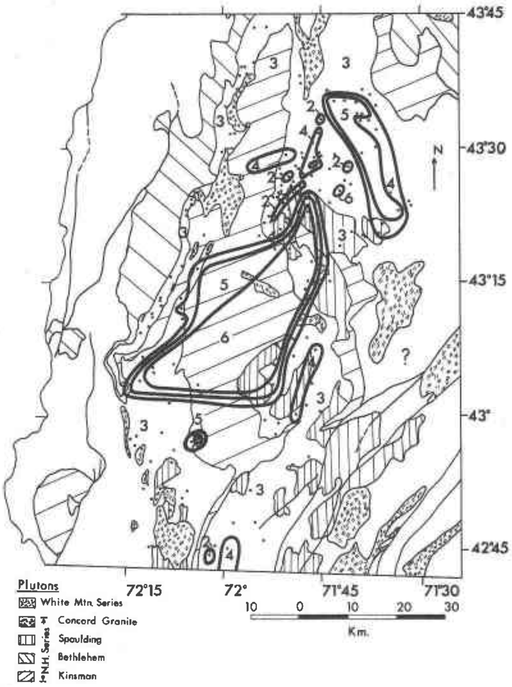

Fig. 2. Metamorphic map of south-central New Hampshire. The metamorphic assemblages mapped are: (2) sillimanite–staurolite, (3) sillimanite–muscovite, (4) sillimanite–muscovite–alkali feldspar, (5) sillimanite–alkali feldspar, (6) cordierite–sillimanite–alkali feldspar.

Zone 3

The vast majority of pelitic rocks in this region belong to the upper sillimanite zone (Zone 3). This zone is marked by the complete disappearance of staurolite in the Zone 2 assemblage and the occurrence of the new assemblage: sillimanite–muscovite–biotite–quartz–garnet–plagioclase. Based on Tracy's (1978) work in central Massachusetts, we presume staurolite is eliminated by the reaction:

staurolite + Na-muscovite + quartz
= sillimanite + biotite + K-richer muscovite
+ albite + garnet + water, (1)

originally proposed by Guidotti (1970).

Zone 4

The second-sillimanite zone (Zone 4) is identified by the first appearance of the pair alkali feldspar–sillimanite in rocks of pelitic composition. The characteristic assemblage of this zone is sillimanite–muscovite–alkali feldspar–biotite–garnet–plagioclase–quartz. Based on studies of the second-sillimanite isograd in central Massachusetts (Tracy, 1978) and in Maine (Evans and Guidotti, 1966), we presume that the presence of alkali feldspar–sillimanite in the pelitic rocks in New Hampshire is the result of the discontinuous reaction:

muscovite + quartz + albite
= alkali feldspar + sillimanite + water. (2)

In the system A–K–Na–Ca reaction 2 becomes the continuous reaction:

muscovite + plagioclase + quartz
= alkali feldspar + Ca-richer plagioclase + sillimanite
+ water (Tracy, 1978). (3)

Zone 5

Zone 5 is mapped on the complete disappearance of muscovite in the Zone 4 assemblage resulting in the new assemblage: sillimanite–alkali feldspar–biotite–garnet–plagioclase–quartz. Zone 5 represents the completion of reaction 2 or 3.

Zone 6

The cordierite–alkali feldspar zone (Zone 6) is based on the appearance of cordierite in the assemblage: quartz–biotite–garnet–sillimanite–plagioclase–cordierite–alkali feldspar. We presume that cordierite is formed from the discontinuous reaction:

biotite + sillimanite + quartz
= garnet + cordierite + alkali feldspar + water, (4)

based on the work of Tracy *et al.* (1976) in central Massachusetts. This assemblage occurs at the maximum level of metamorphism in Massachusetts (Barker, 1962; Tracy *et al.*, 1976), and in southern New Hampshire (this study).

Metamorphic map

Figure 2 is a detailed isograd map of south-central New Hampshire. Instead of a single second-sillimanite isograd for this area, which was shown by Thompson and Norton (1968), metamorphic grade ranges from the lower sillimanite zone (Zone 2) to the cordierite–alkali feldspar zone (Zone 6). The predominant grade is upper sillimanite zone (Zone 3).

The dominant metamorphic feature in this region is a northeast trending metamorphic ridge of second-sillimanite zone and cordierite–alkali feldspar zone rocks extending from the Lovewell Mountain quadrangle to the northeast section of the Mt. Kearsarge quadrangle (Fig. 2). Another smaller metamorphic ridge of second-sillimanite grade rocks is present northeast of the larger ridge, in the Holderness and Penacook quadrangles. We are unsure of the exact nature of this second ridge because of a relative paucity of samples in this area. In some areas the isograds are complex. In the northern part of the Mt. Kearsarge quadrangle, for example, metamorphic grade ranges from the lower sillimanite zone to the cordierite–alkali feldspar zone over a few kilometers.

The most important aspect shown by this metamorphic map is that the isograds crosscut the Kinsman and Spaulding Series plutons. It has long been debated whether the presence of sillimanite, garnet, and cordierite in some of the Kinsman Series plutons was due to metamorphism (Barker, 1961; Thompson *et al.*, 1968) or to a magmatic origin (Clark, 1972; Lyons and Morse, 1970). We suggest that cordierite, garnet, and sillimanite in these plutons were formed during metamorphism and that isograds can be mapped within plutons. Evidence for this is: (1) garnet and cordierite bearing assemblages within the plutons occur only in areas adjacent to high-grade regions in the pelitic rocks; and (2) garnet–biotite pairs within the Kinsman and Spaulding plutons give temperatures that are consistent with the temperature trends in the adjacent pelitic rocks. Based on this evidence, we have mapped the Zone 4 and Zone 6 isograds within the plutons. Zone 4 is mapped on the first appearance of garnet and Zone 6 is mapped on the presence of cordierite and garnet bearing assemblages within plutons.

Mineral composition

There is a systematic variation in the composition of plagioclase (Tracy, 1978) and muscovite (Guidotti, 1973) with increasing metamorphic grade in Massachusetts and Maine, respectively. We find a similar variation in the composition of plagioclase with increasing metamorphic grade in southern New Hampshire. Muscovite composition, however, does not appear to vary systematically with metamorphic grade in this region.

Plagioclase

The anorthite content in plagioclase in a limiting assemblage should be a function of metamorphic grade. Theoretically, at the second-sillimanite isograd the anorthite content of plagioclase in the assemblage quartz–plagioclase–muscovite–alkali feldspar–sillimanite will increase as reaction 3 progresses (Tracy, 1978).

Studies of the variation of the plagioclase composition at the second-sillimanite isograd, however, have not yielded consistent results. Evans and Guidotti (1966) reported that the anorthite content of plagioclase did not vary systematically across the second-sillimanite isograd in Maine. In contrast, an increase of anorthite content with metamorphic grade is clearly demonstrated in Massachusetts (Tracy, 1978). Our results (Fig. 3) also show anorthite contents rising from an average of An_{20} in the sillimanite zone (Zone 3) to An_{30} in the cordierite–alkali feldspar zone (Zone 6). This spread of anorthite contents corresponds to a temperature interval from 550 to 700°C, using the garnet–biotite calibration of Ferry and Spear (1978). In the sillimanite–muscovite–alkali feldspar assemblages (Zone 4) there is a spread of anorthite content of about 10 percent, following the chemical trends predicted by reaction 3. Plagioclase isopleths follow the general trend of isograds, demonstrating an increase in metamorphic grade toward the central portion of this region (Fig. 3). Some representative microprobe analyses of plagioclase feldspars from Zone 3 through Zone 6 in south-central New Hampshire are given in Table 1.

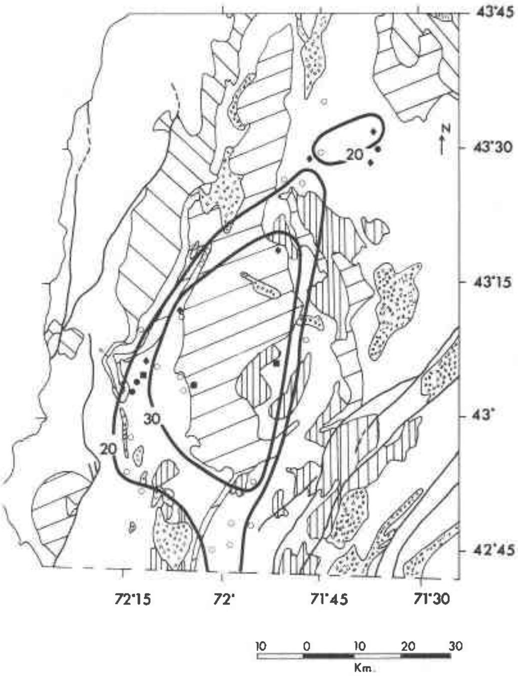

Fig. 3. Plagioclase isopleth map. The numbers correspond to equal anorthite content in plagioclase. Open circles represent quartz–sillimanite–muscovite assemblages. Diamonds are for quartz–sillimanite–muscovite–alkali feldspar assemblages. Solid circles represent quartz–sillimanite–alkali feldspar assemblages. Squares are for quartz–sillimanite–alkali feldspar–cordierite assemblages.

Muscovite

The phengite, titanium, and paragonite content of muscovite in a limiting assemblage is a function of metamorphic grade. Going from the lower sillimanite zone to the sillimanite–alkali feldspar zone in Maine the paragonite content of muscovite decreases, and the titanium content increases (Cheney and Guidotti, 1973; Evans and Guidotti, 1966). In addition, the phengite composition may also be controlled by temperature and pressure (Cipriani *et al.*, 1971). These authors suggest that the phengite composition in muscovite increases with an increase in pressure and decreases with an increase in temperature. In contrast to these results, however, we have found no systematic compositional change in muscovite with metamorphic grade in south-central New Hampshire. We believe that this is due, in part, to the presence of two distinct populations of muscovite.

One group of muscovites are lepidoblastic with a restricted range of paragonite contents (7.9 to 12.2%), and a mean paragonite content of 9.8% (samples Mo-25-80 and Ho-12-80 in Table 2). These muscovites have titanium concentrations greater than 0.5 wt.%. The other group of muscovites are decussate with a wider range of

Table 1. Representative microprobe analyses of plagioclase from Zone 3 through Zone 6 in south-central New Hampshire

Sample	Mo-25-80	Ho-12-80	LM-4-67	LM-3-67
Zone	3	4	5	6
SiO_2	64.40	63.04	62.82	59.42
Al_2O_3	22.86	23.32	24.62	26.08
FeO	0.06	0.02	0.05	0.11
CaO	4.04	4.59	5.81	7.77
Na_2O	9.35	8.74	8.28	7.84
K_2O	0.13	0.14	0.12	0.15
Total	100.84	99.86	101.69	101.01
Formula based on 8 oxygens.				
Si	2.819	2.789	2.738	2.630
Al	1.180	1.217	1.265	1.361
Fe	0.002	0.001	0.002	0.004
Ca	0.190	0.218	0.271	0.368
Na	0.793	0.750	0.699	0.642
K	0.007	0.008	0.007	0.008
Mol % An	19.2	22.3	27.7	36.2

For further analyses and modal information see Chamberlain (1981).

paragonite contents (8.2 to 19.6%), and a mean paragonite content of 14.5% (samples LM-8-66 and Hi-46-72 in Table 2). The titanium concentration of the decussate muscovite is generally less than 0.5 wt.%. There is no systematic difference in the phengite composition between these two types of muscovite.

The lower average paragonite content and the higher titanium content, as well as the lepidoblastic nature of the first group of muscovites suggests they formed at higher temperatures and under greater stress than the decussate muscovites. Based on this textural and compositional evidence we believe that the lepidoblastic muscovites represent early, prograde muscovites formed near the peak of metamorphism and that the decussate muscovites formed later during rehydration and cooling after the peak of metamorphism.

Table 2. Representative microprobe analyses of muscovites from south-central New Hampshire

Sample	LM-8-66	Hi-46-72	Ho-12-80	Mo-25-80
Zone	3	3	4	3
SiO_2	48.80	47.27	46.98	46.59
Al_2O_3	35.05	37.95	35.16	35.57
TiO_2	0.49	0.15	0.68	0.91
FeO	1.00	1.04	1.33	1.07
MnO	0.01	0.00	0.00	0.00
MgO	0.88	0.60	0.54	0.46
CaO	0.02	0.04	0.30	0.00
Na_2O	1.08	1.37	0.45	0.73
K_2O	9.43	8.49	10.51	10.09
Total	96.76	96.92	95.94	95.43
Formula based on 22 oxygens.				
Si	6.332	6.102	6.206	6.168
Al	5.362	5.775	5.476	5.552
Ti	0.048	0.014	0.067	0.091
Fe	0.109	0.113	0.147	0.119
Mn	0.001	0.000	0.000	0.000
Mg	0.169	0.116	0.107	0.091
Ca	0.003	0.005	0.042	0.000
Na	0.271	0.342	0.114	0.187
K	1.562	1.399	1.772	1.704
% Na/(Na+K)	14.8	19.6	6.0	9.9

For further analyses and modal information see Chamberlain (1981).

The inability to demonstrate a systematic change in muscovite composition with metamorphic grade in south-central New Hampshire may be due, in part, to the difficulty in some instances of clearly distinguishing prograde from retrograde muscovites. Because there is some compositional and textural overlap between these two groups of muscovites it is not always clear what generation of muscovite is present.

Conditions of metamorphism

The compositions of minerals involved in exchange reactions are a function of temperature, pressure, bulk composition, and the fluid phase. Many of these exchange reactions have been calibrated so that temperatures and pressures during the metamorphism in southern New Hampshire can be estimated.

Temperature

Metamorphic temperatures in pelitic rocks from south-central New Hampshire have been calculated using garnet–biotite, garnet–plagioclase, garnet–cordierite, plagioclase–muscovite, and plagioclase–alkali feldspar geothermometers (Chamberlain, 1981). In general, all five geothermometers show the same regional trends in metamorphic temperatures across southern New Hampshire (Chamberlain, 1981). However, because garnet–biotite is by far the most common geothermometer available in the pelitic schists in this region we have used this geothermometer to create a metamorphic temperature map. We feel that the garnet–biotite temperatures are a good reflection of the metamorphic conditions in southern New Hampshire since there is generally good agreement between garnet–biotite temperatures and temperatures calculated using the four, previously mentioned geothermometers (Chamberlain, 1981).

The garnet–biotite geothermometer is based on Fe–Mg distribution between garnet and biotite. There are three calibrations for this geothermometer, one of which is experimental (Ferry and Spear, 1978) and two of which are semi-empirical (Thompson, 1976; Goldman and Albee, 1977). The Ferry and Spear (1978) calibration is based solely on a binary Fe–Mg system, whereas the Goldman and Albee (1977) calibration also takes into account minor amounts of Ca and Mn substitution in the garnet. Because the garnets examined in this study are best described as almandine–pyrope garnets with only small amounts of spessartine and grossular components (Table 3), and because the Goldman and Albee (1977) calibration may over correct for Mn and Ca substitution in garnet (Ferry, 1980), we feel that the Ferry and Spear (1978) calibration best approximates the metamorphic temperatures. We have, however, calculated temperatures using all three calibrations for comparison. In this study, temperatures calculated using the calibration of Ferry and Spear (1978) are generally 40° higher than temperatures calculated using Thompson's (1976) calibration and 100° higher than temperatures determined from Goldmn and Albee's (1977) calibration (Table 3). Temperatures determined from all calibrations, however, show the same regional trends (Chamberlain, 1981).

The application of the garnet-biotite geothermometer to this region is complicated by compositional zoning in garnets. Typically, the garnets have Mg-rich cores and Mn and Fe-rich rims. We attribute this zoning pattern to retrograde effects. At about the sillimanite zone self-diffusion of cations in almandine garnet appears to be sufficiently rapid to eliminate compositional zoning acquired at lower grades of metamorphism (Yardley, 1977). The zoning pattern seen in the high-grade garnets is probably a result of retrograde alteration during cooling after the peak of metamorphism (Amit, 1976; Richardson, 1975).

Table 3. Representative microprobe analyses of garnets and biotites from south-central New Hampshire

Sample	Garnet		Biotite	Garnet		Biotite
	Core	Rim		Core	Rim	
	Ho-7-80		Ho--7-80	LM-40-66		LM-40-66
SiO_2	38.09	37.92	36.46	36.07	37.92	33.89
Al_2O_3	22.13	21.78	18.01	21.77	21.37	18.66
TiO_2	0.00	0.01	3.07	0.02	0.03	3.51
FeO	33.50	34.17	14.47	35.28	35.61	19.45
MnO	0.56	0.68	0.00	2.19	2.41	0.01
MgO	6.34	5.75	13.14	3.61	3.33	9.41
CaO	0.58	0.28	0.02	0.99	0.92	0.02
Na_2O	-	-	0.13	-	-	0.11
K_2O	-	-	9.34	-	-	9.21
Total	101.2	100.59	94.64	99.93	101.59	94.29
Garnet formulae are based on 12 oxygens.						
Biotite formulae are based on 22 oxygens.						
Si	2.970	2.986	5.452	2.915	3.004	5.235
Al	2.035	2.022	3.173	2.074	1.997	3.403
Ti	0.000	0.001	0.345	0.001	0.002	0.408
Fe	2.185	2.250	1.808	2.385	2.359	2.513
Mn	0.037	0.045	0.000	0.150	0.162	0.001
Mg	0.736	0.674	2.926	0.435	0.393	2.166
Ca	0.048	0.024	0.003	0.086	0.078	0.004
Na	-	-	0.036	-	-	0.034
K	-	-	1.781	-	-	1.816
Temperature:						
FS	630	587		636	604	
GA	553	518		515	495	
T	600	570		600	578	

FS = Ferry and Spear (1978); GA = Goldman and Albee (1977);
T = Thompson (1976)
For further analyses see Chamberlain (1981).

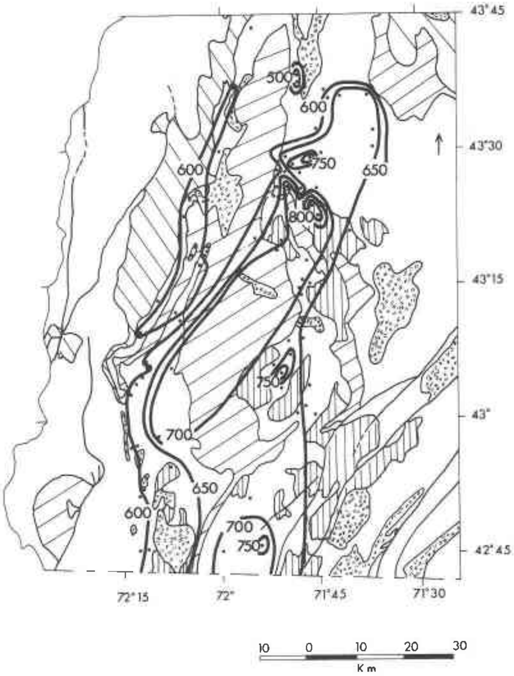

Fig. 4. Metamorphic temperature map of south-central New Hampshire. Temperatures were calculated using the garnet-biotite calibration given by Ferry and Spear (1978). Temperatures are in degrees centigrade.

It is likely that biotite also reequilibrated at lower temperatures during cooling. At first glance, it would appear that using garnet-core compositions and matrix biotite compositions to calculate temperatures would give erroneous results. However, reasonably accurate, prograde temperatures can be obtained from garnets that have undergone retrograde reactions (Tracy *et al.*, 1976; Robinson and Tracy, 1982). As shown by these authors, in biotite-rich rocks retrograde reactions will cause negligible changes in matrix biotite composition while the garnet-rim composition changes greatly. In biotite-rich rocks, therefore, reasonable estimates of maximum prograde temperatures can be made using garnet-core composition and matrix biotite composition. In all of our samples selected for geothermometry, biotite is far more abundant than garnet. Furthermore, we have avoided using samples where secondary chlorite is replacing biotite and thus altering the composition further.

The overall effect of these zoning patterns, however, is small. Temperatures determined using garnet core compositions are generally only 30° higher than temperatures determined from garnet rim compositions (Table 3). In addition, we see the same regional variations in temperature using both garnet core and garnet rim compositions (Chamberlain, 1981).

The metamorphic temperature map for this region (Fig. 4) was constructed using garnet core compositions on 86 samples and the temperature–pressure expression given by Ferry and Spear (1978). A pressure of 3.8 kbar was used for samples located east of the triple point isobar projection, and a 4.0 kbar pressure was used for samples west of the isobar. An error in these pressure estimates will not significantly affect the temperature, as a pressure difference of 2.0 kbar used in the calculation causes only a 10°C difference in the calculated temperatures. The calculated temperatures range from 450 to 800°C across southern New Hampshire. Isotherms show the same general trends observed with isograds (Fig. 4). A ridge of high temperature extends in a northeast direction from the Lovewell Mountain quadrangle to the northern part of the Mt. Kearsarge quadrangle. Like isograds, isotherms can be traced from the pelites through the Kinsman and Spaulding plutons.

In general, an increase in the grade of metamorphic assemblage is associated with an increase in metamorphic temperature (compare Figs. 2 and 4). However, there are some instances where apparently lower grade mineral assemblages give garnet–biotite temperatures well above the expected temperature for that assemblage. The persistence of lower grade mineral assemblages into higher grade zones is not uncommon in Maine (Guidotti, 1974; Evans and Guidotti, 1966) and Massachusetts (Tracy, 1975). We suggest that the difference between temperatures and mineral assemblages observed in our study is probably the result of local variations in activity of water during metamorphism, of bulk chemical effects, or of error in the temperature estimation.

Pressure

Compositions of coexisting garnet and plagioclase in an assemblage with quartz and sillimanite provide an estimate of pressure (Ghent, 1976). Pressures were calculated for eight samples using the equation given by Ghent (1976). The temperatures necessary for the calculation were deduced from garnet–biotite pairs in the same thin section. Calculation of pressure also requires values of the activity coefficients for the grossular and anorthite components of garnet and plagioclase. A value of 1.28 was used for the activity coefficient of anorthite in plagioclase, based on the work of Orville (1972). The activity coefficient for the grossular component in garnet was determined using a regular symmetric solution model for the binary solution of grossular–almandine (Ghent, 1976), and a interaction parameter of 1000 cal/mole from Ganguly and Kennedy (1974). The grossular component in garnet was corrected for ferric iron following the scheme of Ghent *et al.* (1979). The resolution of this geobarometer is approximately 1.5 kbar.

Table 4. Garnet–plagioclase pressures calculated using the equation of Ghent (1976)

Sample	Plagioclase X_{An}	Garnet X_{Ca}	$T^{o}C$	Pressure Kbar
LM-41-66	0.29	0.021	632	4.1
Su-80-66	0.27	0.025	585	4.6
Su-13-66	0.27	0.030	591	5.3
Mo-25-80	0.19	0.015	611	4.3
Mo-34-80	0.23	0.025	625	5.7
Mo-28-80	0.27	0.027	590	4.8
Hi-131-72	0.39	0.035	646	5.1
Hi-46-72	0.18	0.024	614	6.6

For garnet X_{Ca} = Ca/(Mg+Fe+Mn+Ca)
For plagioclase X_{An} = Ca/(Ca+Na)

Pressures calculated from coexisting garnet–plagioclase compositions are given in Table 4. The calculated pressures are geologically reasonable and vary between 4.1 and 6.6 kbar. However, the pressure estimates do not reflect their relative location to the triple point isobar projection (Thompson and Norton, 1968) in this region (Fig. 1). In addition, almost all of the pressure estimates fall between the two experimentally determined triple-point pressures of 3.8 kbar (Holdaway, 1971) and 5.5 kbar (Richardson *et al.*, 1969). Several explanations are possible: (1) we are seeing the effects of the inaccuracy of the garnet–plagioclase geobarometer (±1.5 kbar); (2) the location of the triple point projection in southern New Hampshire is incorrect or; (3) the garnet–plagioclase pressures indicate a later stage of metamorphism.

It is unlikely that the mapped location of the triple point isobar in southern New Hampshire is grossly in error, for the triple-point assemblage is found within this study area (Lyons, 1979) and just north of this region (Rumble, 1973). Furthermore, we know of no occurrences of kyanite east of the triple point isobar or andalusite west of the triple point isobar in southern New Hampshire. Whether the difference between the garnet–plagioclase pressures and the triple point pressure reflect inaccuracies in the garnet–plagioclase geobarometer or a later stage of metamorphism, however, is unclear. A late-stage pressure increase during metamorphism has been suggested by Tracy and Robinson (1980). Additional work is needed to see if the pressure difference observed in New Hampshire is real.

Fluid composition

The composition of the fluid phase present during metamorphism may be an important variable in controlling the mineral assemblage. For instance, if P_{H_2O} was found to vary independent of total pressure and temperature during metamorphism, the areas with high-grade mineral assemblages might show a low fugacity of water. A recent study in Maine (Cheney and Guidotti, 1979) suggests that alkali feldspar–sillimanite assemblages have lower fugacities of water than the adjacent lower-grade sillimanite assemblages. These authors suggest that the presence of alkali feldspar–sillimanite in these rocks is partly controlled by the fugacity of water. The stability range of a mineral assemblage may also be affected by the presence of additional gases in the fluid phase. For example, in graphitic pelites the metamorphic fluid contains the gaseous species CH_4, CO, CO_2, O_2, and H_2 (French, 1966). The presence of these gases reduces P_{H_2O} and affects the stability range of the mineral assemblage. Thus, it is possible that graphitic pelites may contain higher-grade assemblages than adjacent non-graphitic pelites.

The composition of muscovite and plagioclase in an assemblage with quartz and sillimanite can be used to determine the activity of water (Cheney and Guidotti, 1979). Activities of water were calculated for eleven samples using equation 14 of Cheney and Guidotti (1979). Independent temperature and pressure estimates necessary for the calculations are from garnet–biotite pairs in the same thin section, and from the locations of samples relative to the triple point isobar projection. Errors introduced by using estimated pressures in the calculation are small. A 2.0 kbar difference in pressure will give differences in the calculated activities of only ±0.1. The activity of paragonite in muscovite was estimated from mixing parameters of Chatterjee and Froese (1975), and corrected for Ti, Mn, Mg, and octahedral Fe following the method of Ferry (1976). The calculated activities are estimated to be accurate to ±0.2 when errors of ±25°C in the garnet–biotite geothermometer are propagated through equation 14 of Chatterjee and Froese (1975) (Ferry, 1980). This error is further compounded by errors in estimating the mixing parameters and the composition of muscovite and plagioclase. The calculated activities, therefore, are relative rather than absolute numbers.

The general lack of prograde muscovite in sillimanite–alkali feldspar bearing assemblages makes the comparison of fluid compositions in this assemblage with lower-grade assemblages difficult. The activity of water for one sillimanite–alkali feldspar–muscovite assemblage, however, is not significantly different than most of the water activities for the lower-grade sillimanite bearing assemblages (Table 5). In most cases the calculated activities fall within a narrow range, suggesting that the activity of water varied little during metamorphism. Furthermore, if we use Ferry's (1980) error estimation of X_{H_2O} (see below), in most cases P_{H_2O} does not significantly differ from P_{Total}. More data are needed to confirm these trends.

Because of uncertainties in determining temperature using garnet–biotite geothermometry and in estimating muscovite and plagioclase compositions, it is clear that X_{H_2O} values estimated in this manner are somewhat inaccurate. A recent study which used muscovite and plagioclase equilibria to determine X_{H_2O} in a lower pressure metamorphic terrane also found the method to be inaccurate (Ferry, 1980). Ferry (1980) suggests that his X_{H_2O} values are uncertain to ±0.3 when errors in mineral composition, as well as errors in estimating temperature are taken into account. Using the ±0.3 as an error bar, Ferry (1980) concludes: (1) that in most cases P_{H_2O} was less than P_{Total} and; (2) that P_{H_2O} was variable during metamorphism. Although our results differ from Ferry's we are uncertain whether this is due to differing styles of metamorphism or inaccuracies in the method of estimating X_{H_2O}.

Table 5. Activities of water for some pelitic schists in south-central New Hampshire. The calculations to determine the activity of water are based on the muscovite and plagioclase compositions. All calculations were made using the equation of Cheney and Guidotti (1979). Fugacities and pressures are in bars.

Sample	Musc. X_{Pg}	Plag. X_{Ab}	f_{H_2O}	a_{H_2O}	T°C	P	Zone
LM-3-66	0.149	0.639	2468	1.1	615	4000	4
LM-8-66	0.148	0.645	1410	0.69	581	4000	3
LM-12-67	0.122	0.669	2422	1.0	630	4000	3
LM-26-66	0.196	0.879	1709	0.79	610	4000	3
Su-13-66	0.163	0.725	1551	0.74	591	4000	3
Su-3e-66	0.148	0.707	1446	0.69	589	4000	3
Mo-28-80	0.120	0.724	1264	0.60	590	4000	3
Mo-25-80	0.099	0.801	1327	0.59	611	4000	3
Mo-34-80	0.116	0.767	1877	0.86	625	4000	3
Hi-46-72	0.196	0.825	1825	0.87	613	3800	3
MK-55-74	0.206	0.899	2969	1.3	645	3800	3

For muscovite: X_{Pg} = Na/(Na+K)
For plagioclase: X_{Ab} = Na/(Na+Ca)

The composition of the fluid phase in a graphitic system was determined following the method reviewed by Eugster (1977). This method requires the independent estimation of pressure and temperature, the knowledge of fugacity for one of the gaseous components, and the assumption that P_{Total} equals P_{Fluid}. The important reactions involved in this system are:

$$C + O_2 = CO_2 \tag{5}$$

$$CO + 1/2\ O_2 = CO_2 \tag{6}$$

$$H_2 + 1/2\ O_2 = H_2O \tag{7}$$

$$CH_4 + 2\ O_2 = CO_2 + 2\ H_2O \tag{8}$$

(Eugster and Skippen, 1967)
The equilibrium constants for each of these reactions were taken from the data of Ohmoto and Kerrick (1977). The fugacity coefficients for CH_4, O_2, and H_2 are from Ryzhenko and Volkov (1971). The fugacity coefficients for H_2O and CO_2 are from Burnham *et al.* (1969) and Zharikov *et al.* (1977), respectively. Calculations were made using a pressure estimate of 4.0 kilobars and a temperature from garnet–biotite pairs in the same thin section. The fugacity of water is taken from Table 5.

The calculated gas fugacities in five graphitic schists suggest a water-dominant metamorphic fluid (Table 6). These calculations indicate that methane and carbon dioxide are the only other gases that are volumetrically significant. The low oxygen fugacity calculated is consistent with the total lack of magnetite in the pelites in southern New Hampshire,

There is no correlation of higher-grade mineral assemblages with graphitic schists in this region. The isograds and isotherms crosscut graphitic units. At least on a regional scale the fluid composition has apparently had little to do with the distribution of mineral assemblages.

Cause and timing of metamorphism

We recognize a three-stage sequence of metamorphism in south-central New Hampshire. The earliest period of metamorphism M(1) was an andalusite-producing event. M(1) metamorphism was widespread, for sillimanite pseudomorphs after andalusite are seen in this region, as well as in the Bronson Hill Anticlinorium (Thompson *et al.*, 1968) and in central Massachusetts (Tracy and Robinson, 1980). The early andalusite terrane was later metamorphosed to higher-grade assemblages. This second stage of metamorphism M(2) ranged from sillimanite grade to cordierite–alkali feldspar grade, producing the dominant isograd pattern now observed. Pressures and temperatures during M(2) metamorphism were 4.5±1.5 kbar and 450 to 800±25°C, based on garnet–plagioclase geobarometry and garnet–biotite geothermometry. The third stage of metamorphism M(3) locally retrograded the earlier M(2) asemblages. M(3) metamorphism is recognized by chlorite replacing garnet and biotite, and the presence of a lower temperature, decussate muscovite.

Some constraints can be placed on cause and timing of these three stages of metamorphism based on regional geologic relations, textural evidence, and the distribution of mineral assemblages. The earliest period of metamorphism is poorly understood because the M(1) textures were nearly completely destroyed by the later M(2) event. There is evidence, however, suggesting that the Kinsman and Bethlehem Series plutons, which are synkinematic, provided heat for this metamorphism. Nielson (1974) has shown that flecky gneisses in New Hampshire occur near the margins of the Kinsman Series plutons, suggesting contact metamorphism. In addition, mica pseudomorphs after chiastolite are found at the base of the Bellows Falls pluton (Thompson *et al.*, 1968). The second stage of metamorphism must have occurred after 393 m.y. and before cessation of Acadian tectonism. M(2) isograds and isotherms crosscut 393 m.y. old Spaulding plutons and M(2) sillimanite and biotite lie in the plane schistosity. The cause of M(2) metamorphism is uncertain, but it is unlikely that igneous activity was the heat source. Isograds and isotherms are unrelated spatially to later plutons, and the gravity profile for this region (Nielson *et al.*, 1976) does not indicate plutons beneath the high-grade areas. Furthermore, it is difficult to demonstrate that M(2) is related to deformation.

Table 6. Partial pressures of gas species in upper sillimanite zone (Zone 3), graphite bearing, pelitic schists. Fugacities were calculated for a total pressure of 4000 bars and the garnet–biotite temperature. All fugacities are in bars. Temperatures are in degrees centigrade.

Sample	f_{H_2O}	f_{CO_2}	f_{CH_4}	f_{H_2}	f_{CO}	f_{O_2}	T°C
Mo-25-80	1327	109	108	14.2	4.0	4.0×10^{-22}	611
Mo-28-80	1264	96	97	11.6	2.1	7.4×10^{-21}	590
Mo-34-80	1677	542	550	35.4	10.9	2.5×10^{-20}	625
Hi-46-72	1825	514	520	31.7	9.1	1.2×10^{-20}	613
LM-8-66	1410	154	155	13.7	3.2	3.1×10^{-21}	581

The metamorphic highs in this region show no relation to known structures. Tracy and Robinson (1980), however, suggest that M(2) metamorphism in central Massachusetts is caused by backfolding in the Merrimack Synclinorium following nappe emplacement. They cite a late stage pressure increase as evidence. It is possible that sillimanite pseudomorphs after andalusite and garnet–plagioclase pressure estimates also give evidence for this late stage pressure increase in southern New Hampshire. Because garnet–plagioclase geobarometry is imprecise and because the sillimanite pseudomorphs may be due to either a pressure or temperature increase this conclusion is uncertain.

We suggest that M(2) metamorphism may be the result of rapid overthickening in the Merrimack Synclinorium and thermal reequilibration following the Acadian orogeny (Chamberlain and Lyons, 1981). England (1978) and Oxburgh and England (1980) have shown that rapid overthickening can be responsible for the dominant metamorphism seen in the eastern Alps. These authors suggest that peak metamorphic temperatures are reached after overthickening. It is possible that a similar situation existed in the Merrimack Synclinorium at the end of the Acadian orogeny. Following nappe emplacement and refolding in this region the depressed isotherms may have risen through the overthickened pile and thermally metamorphosed the prexisting metamorphic terrane.

The cause of M(3) is unclear. M(3) retrograde mineral assemblages are unrelated to post-tectonic plutons and are not structurally controlled. It is possible that M(3) metamorphism is merely a continuation of M(2) metamorphism, and that M(3) represents the rehydration and recrystallization of M(2) assemblages during cooling following the peak of metamorphism.

The polymetamorphic history of south-central New Hampshire is interestingly similar to that of northern Maine.

Holdaway *et al.* (1982) present evidence for four stages of metamorphism occuring within the transition zone from regional to a contact metamorphic terrane in Maine. According to these authors, in Maine, M(1) metamorphism is a regional, low-grade, chlorite-producing event; M(2) is a regional, andalusite–staurolite grade metamorphism; M(3) is a contact to regional, staurolite, sillimanite, sillimanite–alkali feldspar metamorphism and M(4) is a local, contact metamorphism producing andalusite and staurolite-bearing assemblages. There are two basic similarities between this metamorphic history and that recognized in southern New Hampshire. First, the transition from an early, andalusite grade metamorphism to a higher-grade sillimanite-producing metamorphism occurs in each region. Second, a pressure increase during this transition is , in each case, a possibility (Holdaway *et al.*, 1982). There are, however, several distinct differences between the two histories. In Maine, for example, the andalusite-producing event, M(2), is regional and unrelated to plutons and the sillimanite-producing metamorphism, M(3), is in part contact in origin; whereas in southern New Hampshire the andalusite-producing event, our M(1), may be contact in origin and the sillimanite producing event, our M(2), is clearly regional. Also, there appears to be no late contact metamorphism in southern New Hampshire similar to M(4) in Maine. Clearly, futher work addressing this issue is needed to construct a complete metamorphic framework for the New England region.

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