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GEOGRAPHIC DISTRIBUTION OF CORRENSITE AND ASSOCIATED MINERALS IN SOUTHEASTERN ONTARIO

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Abstract

Abundant masses of pulverulent material are present in the veins and fractures of marble outcrops in the Grenville Province of southeastern Ontario. This material, collected in more than 40 outcrops, was air-dried and, after sieving and separation, subjected to X-ray-diffraction analysis on the <2 μm fraction. Corrensite, a regularly interstratified mineral composed of chlorite and an expandable mineral, here most frequently smectite but locally vermiculite, was identified in all samples. This long-spacing (29 Å) mineral is a product of the hydrothermal alteration of phlogopite. Corrensite and phlogopite are the most common constituents of the clay fraction and are locally associated with expandable minerals and chlorite. These two latter minerals are indicative of the process of hydrothermal alteration. Talc was also observed in some samples of the clay fraction, but its formation was related to local metamorphism. Corrensite abundance in the outcrops is not related to the calcite/dolomite ratio in the host rock, but is clearly associated with the intensity and frequency of tiny to large fractures that allowed the hydrothermal fluid medium to circulate.

Keywords: Grenville Province, marble outcrops, phlogopite alteration, corrensite, expandable minerals, chlorite, talc, ground preparation, Ontario.

Sommaire

De grandes quantités de matériel pulvérulent sont présentes dans les veinules et fissures des affleurements de marbre de la province du Grenville, dans le sud-est de l'Ontario. Ce matériau a été échantillonné dans plus de 40 affleurements et séché à l'air; après tamisage et séparation, la fraction <2 μm a été analysée par diffraction X. La corrensite, minéral interstratifié régulier composé de chlorite et d'un minéral gonflant, généralement la smectite ici mais parfois la vermiculite, a été identifiée dans tous les échantillons. Ce minéral à 29 Å serait un produit de l'altération hydrothermale de la phlogopite. Corrensite et phlogopite, les minéraux les plus fréquemment observés dans la fraction argileuse, sont ici et là associés à des minéraux gonflants et à la chlorite. Ces derniers minéraux témoignent de l'altération hydrothermale. Le talc a été aussi observé dans certains échantillons, mais sa formation résulterait plutôt de conditions métamorphiques locales. L'abondance de la corrensite dans les affleurements n'est pas liée au rapport de calcite à dolomite dans la roche encaissante, mais bien à l'intensité et à la fréquence du système de fractures qui a facilité la circulation des fluides.

Mots-clés: province du Grenville, affleurements de marbre, altération de la phlogopite, corrensite, minéraux gonflants, chlorite, talc, préparation du milieu, Ontario.

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Introduction

Corrensite is defined as a regular interstratification of chlorite with smectite or vermiculite, in which all components are trioctahedral minerals (Bailey *et al.* 1982). This mineral was thought to form in hypersaline sedimentary environments (Grim *et al.* 1960, Kopp & Fallis 1974), but compilations of the more recent literature have shown that corrensite occurs in a large variety of geological environments (Brigatti & Poppi 1984, Velde 1985). In the review on corrensite occurrences, Velde (1985) reported the following assemblages of three or more phyllosilicates: corrensite – chlorite – illite; corrensite – mixed layered illite – smectite – illite; corrensite – chlorite – illite – trioctahedral smectite; corrensite – chlorite – illite – dioctahedral smectite – talc.

Velde also reported that among the non-phyllosilicates associated with corrensite, dolomite frequently was observed. He concluded that corrensite forms in normal sedimentary rocks and weathering environments, during diagenesis and low-grade metamorphism, and by hydrothermal alteration. The frequent association of corrensite with chlorite led Brigatti & Poppi (1984) to propose, in their review, that corrensite is produced by degradation of chlorite, whereas Morrison & Parry (1986) reported the formation of a chlorite–smectite regularly interstratified mineral by chloritization of smectite. More recently, Meunier *et al.* (1988) reported the formation of corrensite by hydrothermal alteration of amphibole in dolomitic veinlets of a granite in France.

Corrensite, associated with phlogopite, was found in an outcrop of dolomite marble in southeastern Ontario (De Kimpe *et al.* 1987). Optical microscopy, X-ray-diffraction analyses, and field observations indicated that the interstratified mineral formed at the expense of the mica by hydrothermal alteration. Since this first report was prepared, additional field work and laboratory determinations were undertaken to further characterize the environmental conditions and clay-mineral assemblages prevailing in marble outcrops of southeastern Ontario.

Regional Geology

The Grenville Province (Fig. 1) is characterized by a variety of rocks of medium to high metamorphic grade, predominantly of Helikian (1600–850 Ma) age

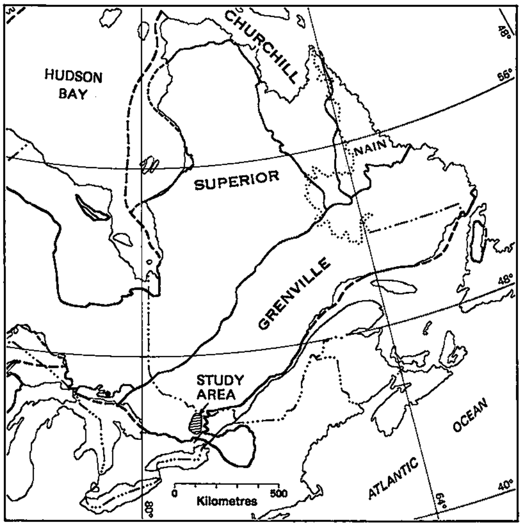

FIG. 1. Location of the study area in the Grenville Province, eastern Canada.

(Wynne-Edwards 1964, Stockwell *et al.* 1970). Metamorphosed carbonate rocks are widespread within a 31,000-km^2 area of the Central Metasedimentary Belt of the Province (Kretz 1980). Geologically, this area is very complex because of the profound effects of metamorphism, igneous intrusion, metasomatism and deformation that accompanied the Grenville orogeny (Hewitt 1956). High-grade metamorphic terranes had suffered deep burial or were in close contact with intrusive bodies, whereas other parts showed a lower-grade metamorphism, with preservation of original structures and textures in sedimentary, volcanic and intrusive rocks. Intrusion of granitic and mafic magmas resulted in active contact metamorphism in addition to the regional metamorphism in the general area. Using coexisting mineral assemblages within the siliceous carbonate rocks of a study area overlapping the present one, Ewert (1977) established zones of metamorphic grades that increase toward the plutonic masses. Ewert also used the calcite–dolomite geothermometer to determine a temperature range from less than 350°C in the low-grade regions to over 670°C in the high-grade ones. Some marbles occur as massive bodies, but others are considerably fractured, possibly as a result of "ground preparation," which may take place in several ways. Ground preparation is a change that makes country rocks more receptive to, or more reactive with, fluids by increasing their permeability, causing a favorable chemical change, or inducing brittleness (Park & MacDiarmid 1964). These authors give several examples that were applicable in this study area. Dolomitization of calcite induces a change in unit-cell size, which makes the rock brittle. Addition of silica to carbonate rocks, either as a form of SiO_2 or as silicates, is a frequent mode of chemical ground-preparation. Recrystallization of the country rock adjacent to intrusive bodies commonly increases permeability and brittleness. Ground preparation permits the circulation of hydrothermal solutions. Primary magmatic gases are acid and show high reactivity (Barth 1962). Therefore, if the contact rock is basic, especially limestone, the acid gases are absorbed and react effectively to produce a variety of reaction products. Pyroxene and garnet, with low Fe contents, or hematite if the Fe content is high, are commonly formed and commonly are accompanied by phlogopite (Barth 1962). The mica can also be formed during low-pressure metamorphism of siliceous dolomitic marble (Skippen 1974), according to the reaction: 3 dolomite + K-feldspar + H_2O → phlogopite + 3 calcite + 3 CO_2. Phlogopite is widespread in the study area but it is most abundant in the southern part because of an increasing K-feldspar content in the marbles southward (Ewert 1977). The phlogopite occurs as tiny crystals (1–2 mm) throughout the rocks or as large crystals (up to 5 cm in diameter) in veins and fractures. The phlogopite invariably has a typical micaceous cleavage and a pseudohexagonal morphology.

MATERIALS AND METHODS

The present study area (Fig. 2) covered approximately 6,000 km^2 (about 125 km long and 50 km wide) and extended from Ottawa to Verona, north of Kingston. Rock samples and associated material in the veins and fractures were collected from more than 40 marble outcrops, and at each site the approximate intensity of ground preparation was rated as none to intense by the frequency of occurrence of fractures. Samples were air-dried and passed through a 2-mm sieve (De Kimpe *et al.* 1987). Subsamples were dispersed in H_2O using ultrasonic vibration at a frequency of 40 kHz. Sand was collected on a 53-μm sieve and air-dried. Clay was separated from the silt by sedimentation and centrifugation, and recovered by freeze-drying. Rock samples were crushed and ground to pass a 53-μm sieve in order to establish the ratio of calcite to dolomite.

Mineralogical analyses were carried out by X-ray diffraction, infrared absorption spectroscopy, and optical microscopy. The clay samples were prepared as preferentially oriented specimens by allowing a suspension of 30 mg of Mg-saturated clay in 1 mL H_2O to dry on a 25 × 30 mm glass slide. Slides were prepared in duplicate, one for glycerol solvation (Miles & De Kimpe 1985), and one for heat treatment at 550°C. Another slide was prepared with K-saturated clay as explained above, and was examined at 25°C and after heat treatment at 550°C. Analyses were performed on an automated Scintag Pad V

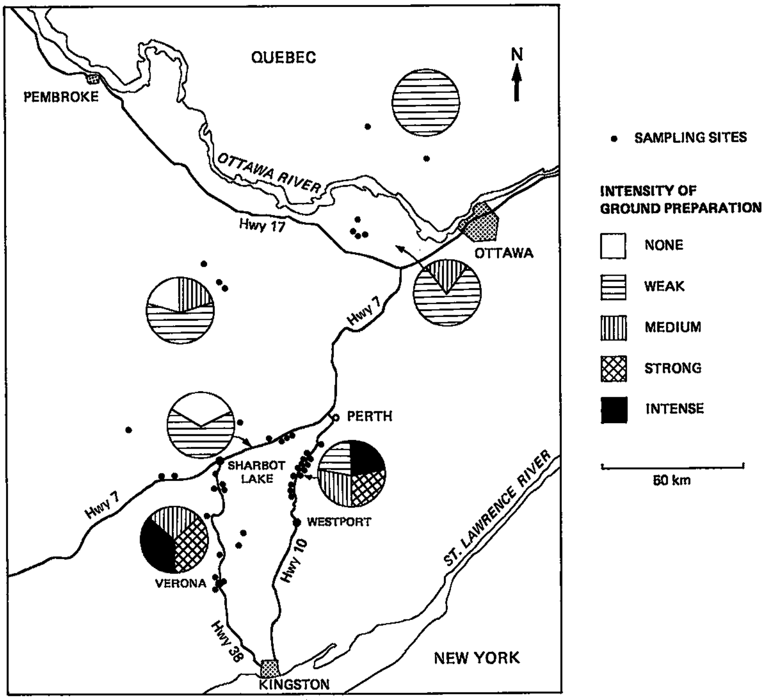

FIG. 2. Sampling sites and intensity of ground preparation in the study area.

diffractometer with Co$K\alpha$ radiation using a graphite monochromator. For the rock samples, specimens were prepared using 30 mg of ground material on a 25 × 30 mm slide, and the analyses were performed with a Philips diffraction unit, also using Co$K\alpha$ radiation. The heights of the most intense peaks (3.035 Å for calcite and 2.89 Å for dolomite) were used to calculate the relative ratio of calcite to dolomite in the rocks. The relative intensities of the 10-Å peak of phlogopite and the presence of talc, identified by peaks at 9.34 and 3.13 Å, were also recorded. Additional X-ray-diffraction analysis was performed by means of a Gandolfi camera on single grains separated from the rock samples under an optical microscope. Undisturbed samples were examined under the microscope to characterize mineral assemblages. For the analysis by infrared absorption spectroscopy, selected samples were prepared as KBr pellets and analyzed with a Beckman IR 4250 infrared spectrophotometer.

RESULTS AND DISCUSSION

Phyllosilicate assemblages and quality of the corrensite

The seven phyllosilicate assemblages identified in the <2 μm fraction, and their frequency of occurrence, are shown in Table 1. X-ray-diffraction patterns of glycerol-solvated specimens (Fig. 3) showed the major lines of the minerals present in these assemblages. The presence of talc, identified in 12 samples mainly north of Highway 7, is related to the pressure and fluid-phase composition during metamorphism (Ewert 1977). Talc is generally absent not only at lower pressures, but also at higher pressures if the fluid phase is extremely CO_2-rich; formation of such a fluid may be the result of phlogopite formation in K-feldspar-bearing dolomitic marbles (Skippen 1974). Talc did not appear to play any role in corrensite formation; therefore, it is not justifiable to extend the list of mineral assemblages to include this phyllosilicate. Graphite was identified in several samples by X-ray diffraction using a Gandolfi camera (De Kimpe *et al.* 1987).

Forty-five percent of the samples contain corrensite alone (assemblage 1) or in association with phlogopite (assemblage 2). This difference in association correlates with the nature of the sampling site: samples collected in veins with large phlogopite crystals generally separate into two distinct size-fractions; corrensite is concentrated in the <5 μm fraction, and phlogopite, in the coarser one. In contrast, samples collected from cavities containing a large amount of small-size phlogopite crystals have both corrensite and phlogopite in the <2 μm fraction.

TABLE 1. PHYLLOSILICATE MINERAL ASSEMBLAGES

Minerals	Occurrences	Frequency
(1) corrensite	9	22.5
(2) corrensite-phlogopite	9	22.5
(3) corrensite-phlogopite-vermiculite	4	10
(4) corrensite-phlogopite-smectite	2	5
(5) corrensite-phlogopite-vermiculite-smectite	4	10
(6) corrensite-phlogopite-chlorite	5	12.5
(7) corrensite-phlogopite-chlorite-expanding mineral	7	17.5

Corrensite quality, *i.e.*, the regularity of the interstratification of the chlorite and smectite layers, was assessed by calculating the coefficient of variation (*CV*) for the higher-order spacings of the *d*(001) reflection, as recommended by Bailey *et al.* (1982). Accurate peak-positions were obtained by slow step-scanning (10-s counts at each 0.02° 2θ) from 1 to 100° 2θ. This type of analysis was performed for 25 samples that provided an X-ray-diffraction pattern with a large number of higher-order basal reflections. The *CV* calculation was generally based on more than 12 orders. The values obtained range from 0.19 to 0.78; only one sample exceeds the 0.75 upper limit proposed by these authors for regular interstratifications. The average *CV* value is 0.47 ± 0.17, which indicates well-ordered interstratification.

Corrensite generally corresponds to the mineral described earlier by De Kimpe *et al.* (1987) at Sharbot Lake. In two samples, however, the expandable mineral component behaved under glycerol solvation and heat treatment more like vermiculite than smectite. The significance of this observation is discussed below.

Relation between corrensite and geological features in the study area

Host-rock calcite/dolomite ratio. The calcite/dolomite peak-intensity ratio varies widely from

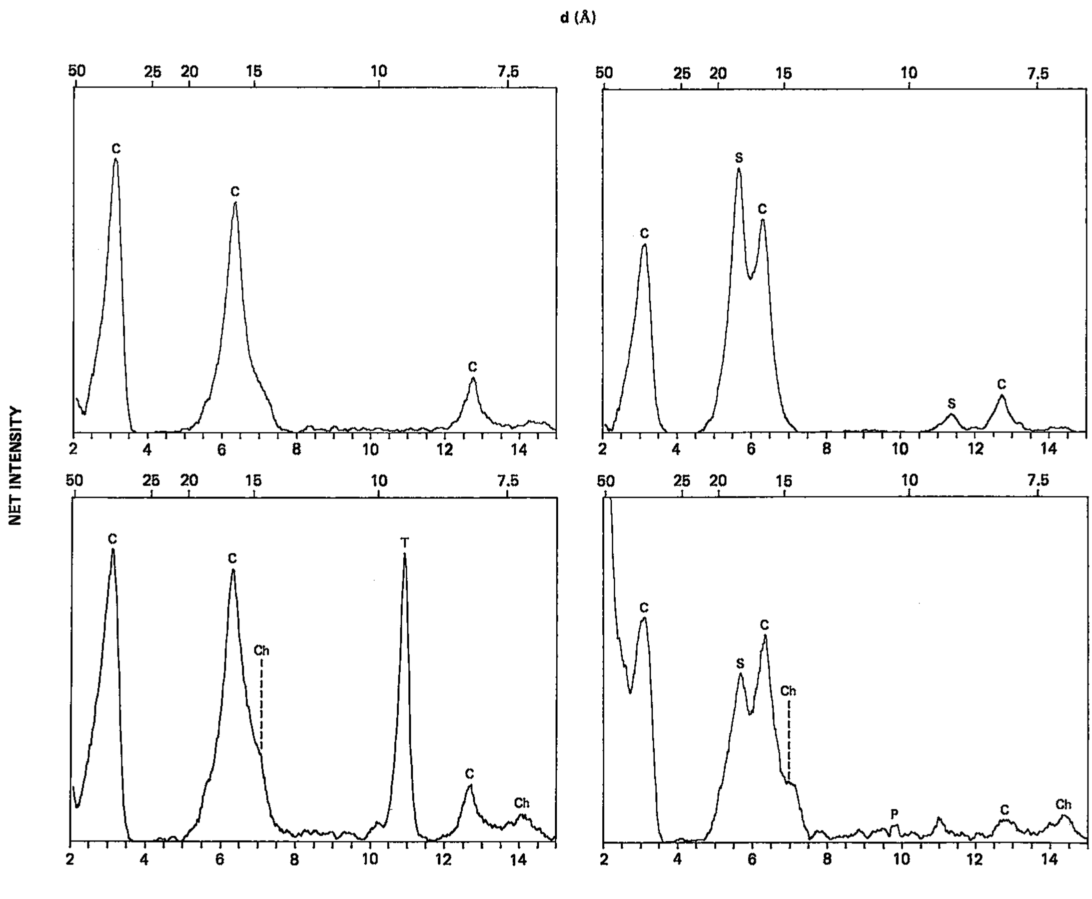

FIG. 3. X-ray-diffraction patterns of glycerol-solvated samples presenting some typical assemblages: (1) pure corrensite C, (2) corrensite + smectite S, (3) corrensite + chlorite Ch + talc T, (4) corrensite + smectite + chlorite + phlogopite P.

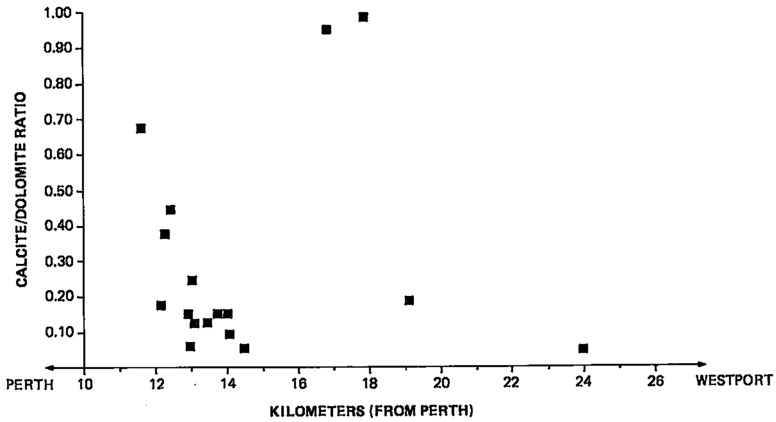

FIG. 4. Ratio of calcite to dolomite in marble outcrops along Highway 10 between Perth and Westport.

sample to sample, and it is difficult to show a trend. The very short-range variability of the ratio is well illustrated by the measurements made on 17 rock samples collected over a distance of 12 km between Perth and Westport (Fig. 4). In most samples, the ratio is less than 0.50, but it exceeds 0.60 in three outcrops. Similar high and low values alternate along Highway 38 from Sharbot Lake to Verona. Ewert (1977) reported that the dolomite content in the marbles generally increases from the Carleton Place area to the Tichborne–Westport area. He used the molar concentration of Mg in calcite to establish the highest temperature reached during metamorphism for a number of field stations: where calcite coexists with dolomite, the maximum amount of magnesium possible in the calcite structure is related to temperature and is relatively independent of pressure (Goldsmith & Newton 1969). This analysis, of great interest to geologists, has less application for the present study, and therefore was not undertaken.

Ground preparation and its relation to the calcite/dolomite ratio. Five classes of ground preparation were used to describe rock fracturing in the carbonate outcrops: none, weak, medium, strong, intense. Sixty-nine observations throughout the study area are schematically summarized in Figure 2. Outcrops with no ground preparation were the exception. The number of marble occurrences and their degree of ground preparation generally increased from north to south in approaching the major intrusive bodies.

Ground preparation is not clearly related to the calcite/dolomite ratio, as demonstrated by the range of values associated with intensity of ground preparation (Table 2). Therefore, dolomitization of calcite was probably not a major factor responsible for rock fracturing. Magnesium was likely present in the limestone before burial and diagenesis (R. Kretz, oral comm. 1987), so that there was little, if any, rock fracturing due to cell adjustment during recrystallization. Magnesium could also have been set in the carbonate by metasomatism or during metasomatism accompanying metamorphism. Pneumatolytic or hydrothermal alteration of phlogopite may have also contributed to rock fracturing, as is discussed in the next section.

Corrensite formation. The assemblages of phyllosilicate minerals reported in Table 1 indicate the nature of the minerals in the marbles. The net X-ray-diffraction intensity of the major peaks of each mineral was used to determine approximately the relative abundance of the phyllosilicates in the assemblages. This hypothesis seemed acceptable on the basis that all phyllosilicates would adopt a preferred orientation upon sedimentation on a glass slide. Twenty-six of the 40 samples containing corrensite also have corrensite as the most abundant mineral. Vermiculite or smectite (or both) are more abundant than corrensite in 7 samples, phlogopite is higher than corrensite in 3 samples, chlorite in 2 samples, chlorite + phlogopite and chlorite + expandable minerals in 1 sample each.

These observations do not contradict the conclusions of De Kimpe *et al.* (1987), wherein corrensite was reported as the unique phyllosilicate identified in the <2 μm fraction of the St. George Lake marble except for some phlogopite in a few samples.

TABLE 2. GROUND PREPARATION AND CALCITE/DOLOMITE RATIO IN THE OUTCROPS

ground preparation	number of observations	range of the calcite/dolomite ratio min.	max.	mean
none	4	0.04	0.96	0.59 ± 0.42
weak	17	0.02	1.00	0.62 ± 0.41
medium	15	0.01	0.95	0.34 ± 0.37
strong	13	0.15	1.0	0.41 ± 0.35
intense	20	0.06	0.56	0.25 ± 0.27

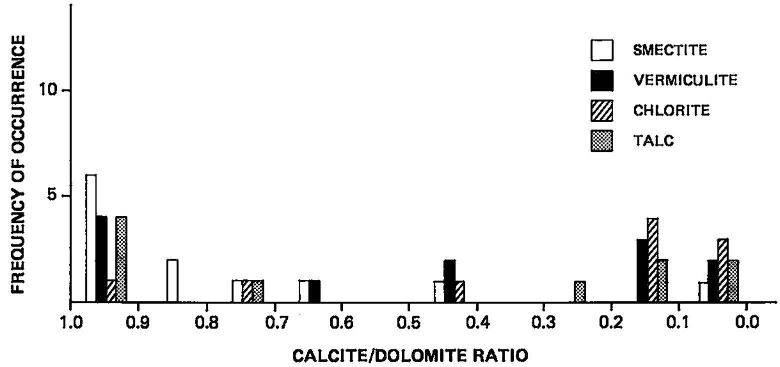

FIG. 5. Frequency of occurrence of phyllosilicates associated with corrensite, as a function of the ratio of calcite to dolomite.

That report showed optical microscope observations on the replacement of phlogopite by corrensite. The present regional study on the occurrence and formation of corrensite at the expense of phlogopite by hydrothermal alteration provides a more complete description of the mechanism of recrystallization.

Formation of corrensite at the expense of phlogopite implies the following steps: a) loss of interlayer K^+ from phlogopite; b) formation of an expanding mineral with hydrated interlayer cations; c) chloritization of alternate layers of the expanding mineral. It is part of the sequence phlogopite → vermiculite → smectite → corrensite → chlorite, where a relatively anhydrous mineral, phlogopite, is transformed to more hydrated ones. In the Mg-rich environment of dolomitic rocks (April 1980), the hydrothermal reaction proceeds to the right. This was shown by microscope observations (De Kimpe *et al.* 1987). The different phyllosilicates indicated above were observed in the mineral assemblages (Table 1). Expandable minerals (assemblages 3–5), the precursors of corrensite (steps a, b), were identified in 25% of the occurrences, vermiculite and smectite being present in 20 and 15% of the samples, respectively. Preferential chloritization of smectite rather than vermiculite is demonstrated by the fact that 38 corrensite samples contain smectite as the expandable mineral component, and two only have a vermiculite component. The latter situation possibly occurs when the rate of chloritization of the expandable mineral exceeds that of vermiculite transformation to smectite. Assemblages 3–5 are interpreted to be those in which corrensite formation has not yet reached equilibrium because chloritization of alternate layers is incomplete. Excess chloritization of the expandable mineral, indicated by the presence of chlorite (assemblage 6, Table 1) is found in 12.5% of the samples, and this low percentage rules out chlorite as a precursor of corrensite in the study area, as was suggested elsewhere (Brigatti & Poppi 1984). Direct formation of chlorite at the expense of phlogopite is also ruled out because an additional brucitic layer cannot be inserted until the mica interlayer space has been expanded through the removal of the interlayer K^+ (steps a, b). Finally, coexistence of expandable minerals and chlorite in addition to corrensite and phlogopite (assemblage 7, Table 1) is found in 17.5% of the samples. This indicates that chloritization likely proceeds along preferential planes, in a manner similar to that proposed by Jackson *et al.* (1952) for the formation of expandable minerals at the expense of mica. The coefficient of variation calculated for corrensite samples from assemblage 7 is 0.51, not significantly greater than the average value. This excludes the possibility that corrensite is composed of aggregates of the fundamental components chlorite and smectite (Nadeau *et al.* 1984).

Although assemblages 3–7 (Table 1) do not show any geographic trends and may be more a consequence of the intensity of hydrothermal alteration, the nature of the associated minerals is, to some extent, related to the calcite/dolomite ratio in the rocks (Fig. 5). Smectite occurs more frequently where the calcite/dolomite ratio is high, whereas the contrary is found for chlorite. Vermiculite occurrences seem to be independent of the ratio.

At this stage of the regional investigation of occurrence and formation of corrensite, it is not possible to determine whether ground preparation is a cause or a consequence of the phlogopite transformation to corrensite. As mentioned earlier, phlogopite is commonly observed in veins and fracture planes. A high intensity of ground preparation before the hydrothermal events would be favorable to the movement of hydrothermal fluids throughout the marbles and, from this point of view, it could be considered a direct cause of corrensite formation. On the other hand, formation of an expandable mineral at the expense of phlogopite implies a 40% increase along the *c* dimension of the crystal. This would exert a high pressure in the host rock, possibly sufficient to fracture it. Intense ground-preparation generally correlates with the abundance of corrensite in the outcrops.

There is no direct clue for the age of corrensite formation in southeastern Ontario. Ewert (1977) discussed the nature of the fluid phase, CO_2- or H_2O-rich, involved at various stages during metamorphism. That corrensite formation occurred in a post-metamorphic period is suggested by the following observations: (1) hydrothermal alteration involved a large percentage of the phlogopite crystals that were in contact with the hydrothermal fluid. However, large crystals still present in the powdery corrensite mass clearly demonstrate that the hydrothermal alteration was a later event. (2) The temperature during metamorphism in the study area was established to range from 350° for the low-grade regions to 670°C in the high-grade regions (Ewert 1977). Such temperatures would be too high to stabilize a hydrated mineral such as corrensite. Corrensite appears under normal diagenetic conditions at temperatures of approximately 90°C to 100°C, and at 200°C in low-pressure environments (Kübler 1973). (3) At two sites, corrensite is found in non-carbonate rocks. The site near Carp, west of Ottawa, is of special interest as corrensite developed at the expense of phlogopite present in an igneous rock that intruded dolomite. This again suggests a more recent period than 950 Ma for the hydrothermal alteration.

Conclusions

Hydrothermal alteration of phlogopite to corrensite was a common process in the marble outcrops of southeastern Ontario. The coexistence of vermiculite, smectite and chlorite along with corrensite and phlogopite is representative of various stages of the transformation sequence of the relatively anhydrous mineral to hydrated ones. The conditions of formation are incompatible with the range of temperatures and pressures associated with the metamorphism in the area, and strongly suggest that formation of corrensite occurred during a later stage of hydrothermal activity.

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