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Behavior of boron during contact metamorphism of calc-silicate rocks at Notch Peak, Utah

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ABSTRACT

B concentrations in the contact-metamorphosed Weeks Formation in the aureole of Notch Peak Stock, Utah, were determined in order to understand better the influence of metamorphic reactions on B behavior in light of the influence of B on the petrogenesis of granites. The Weeks Formation is a calcareous argillite consisting of calcite + quartz + muscovite + chlorite + plagioclase + dolomite where unmetamorphosed. With increasing grade, the phlogopite isograd is recognized by the reaction muscovite + dolomite + quartz + H_2O = phlogopite + chlorite + muscovite + CO_2, the diopside isograd by the reaction phlogopite + calcite + quartz = potassium feldspar + diopside + H_2O + CO_2 and the wollastonite isograd by the reaction calcite + quartz = wollastonite + CO_2. These reactions occurred in the vicinity of 450 °C and were largely driven by the influx of water from the stock.

B concentrations in samples from all metamorphic grades in the Weeks Formation were analyzed by prompt gamma neutron activation. Because B correlates well with Al in the unmetamorphosed rocks, the concentration of B is best expressed by the ratio $(1000 \times \mathrm{B})/\mathrm{Al}$. In the unmetamorphosed rocks, the average ratio is 1.55 ± 0.42; in the phlogopite zone, 1.04 ± 0.48; in the diopside zone, 0.16 ± 0.13; and in the wollastonite zone, 0.20 ± 0.11. There is a strong correlation between B concentrations and the disappearance of micas that contain the bulk of the B at low grades, whereas the minerals in the diopside zone have crystal structures that do not readily incorporate B. The released B was probably dissolved in the infiltrated magmatic fluid that later left the system. This study of B behavior in the Weeks Formation suggests that the persistence of B in metasediments may be controlled by mica-consuming calc-silicate reactions during metamorphism and the availability of evolved or infiltrated fluids for the removal of B.

INTRODUCTION

The behavior of B during metamorphism has been the subject of several recent studies (e.g., Manning and Pichavant, 1983; Sisson and Leeman, 1988) because of experimental evidence that the addition of B to granitic systems lowers the solidus and increases the solubility of water in melts (Pichavant, 1981; Benard et al., 1985), potentially leading to the formation of tourmaline granites. In addition, B may be an important indicator of marine sediment recycling into magmatic sources (Morris et al., 1988) because B is concentrated into clays and cherts from seawater (Harder, 1974; Truscott and Shaw, 1984). Ultimately, however, the incorporation of B into igneous rocks depends on processes occurring during heating of the crust. Therefore, understanding the effect of metamorphic reactions on B behavior is relevant to the petrogenesis of granites and the B geochemical cycle.

Only recently attention has been focused on B behavior during metamorphism. Truscott et al. (1986) documented low concentrations of B in granulites (on the average 2–4 ppm). It has been suggested that the low concentrations may be owing to B removal into melts during anatexis. Truscott and Percival (1988) observed progressive loss of B during metamorphism of the Quetico Belt, Ontario, with abrupt loss near the migmatite isograd coinciding with the loss of tourmaline. They also found that in the lower-grade rocks B is concentrated in micas, sillimanite, and minor tourmaline. Although high B concentrations have been documented in granulites, they have been ascribed to post-peak metamorphic hydrothermal

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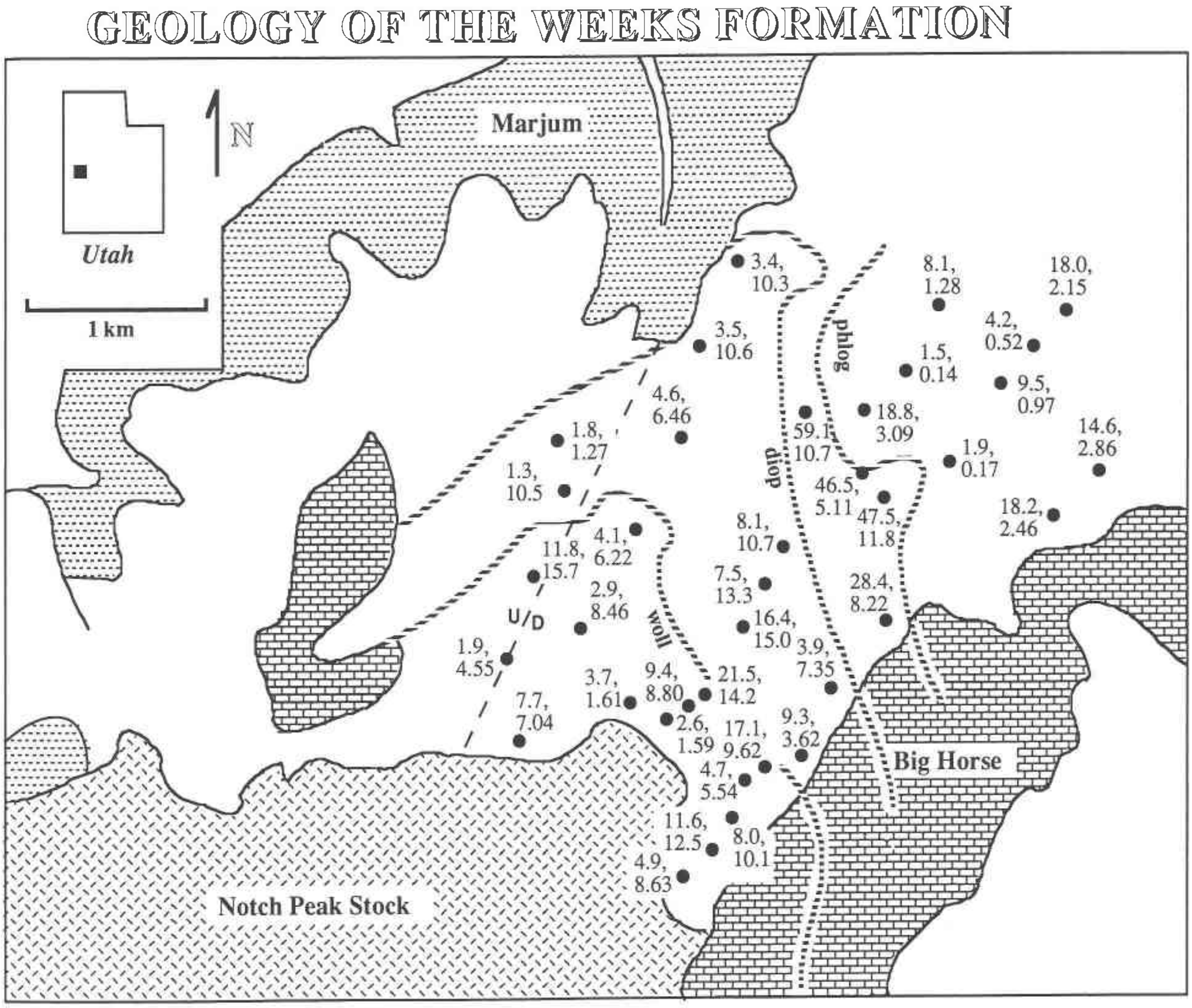

FIG. 1. Geological map of the Notch Peak contact-metamorphosed Weeks Formation with metamorphic isograds. B (ppm) and Al_2O_3 (wt%) concentrations are shown at each sample location.

alteration (Truscott et al., 1986; Ahmad and Wilson, 1981; Shaw et al., 1988). Sisson and coworkers (Sisson and Leeman, 1988; Sisson et al., 1988) investigated the role of B during migmatization of the Chugach metamorphic complex, Alaska. They observed a progressive loss of B in rocks from the amphibolite and granulite facies. The loss of B was attributed to tourmaline breakdown during metamorphism or concentration of B into tonalitic melts, which apparently formed in the migmatized core of the complex. Sisson and Leeman (1988) suggested that low B concentrations in the tonalites may be due to subsequent removal by fluids.

In order to understand the factors that influence the behavior of B during metamorphism of sediments that do not contain tourmaline or another borosilicate mineral, we determined B concentrations in the contact-metamorphosed Upper Cambrian Weeks Formation, a calcareous argillite metamorphosed by the Notch Peak Stock, Utah. We used the highly accurate prompt gamma neutron activation method. Our understanding of the Notch Peak contact metamorphic aureole (Hover-Granath et al., 1983; Nabelek et al., 1984; Labotka et al., 1988a, 1988b) provides a foundation for investigating the influence of mineral reactions on B behavior during the metamorphism of the sediments. The results have implications for understanding the generation of B-rich magmas from calcareous sediments and the B geochemical cycle.

GEOLOGY OF THE WEEKS FORMATION

The Weeks Formation is one of three exposed sedimentary units that were intruded 145 Ma by the Notch Peak Stock in the House Range, west-central Utah (Nabelek et al., 1986, 1988a) (Fig. 1) at a depth of 4–6 km. The metamorphism of the overlying Big Horse Member of the Orr Formation was studied in detail by Hover-

Granath et al. (1983), Nabelek et al. (1984), and Labotka et al. (1988a, 1988b).

The Weeks Formation is a dark, fine-grained, thin-to-medium bedded calcareous shale, but its upper unit becomes progressively more argillaceous. The middle unit contains disseminated graphite. The regional dip of the sedimentary rocks is about 10° to the southwest, except in the vicinity of the intrusion, where the dip becomes somewhat steeper. The prograde metamorphism of the Weeks Formation is recognized by three isograds. Incipient metamorphism is defined by the appearance of phlogopite in calcite + quartz + chlorite-containing rocks by the reaction

$$3 \text{ muscovite} + 18 \text{ dolomite} + 5 \text{ quartz} + 16 \text{ H}_2\text{O} = 3 \text{ phlogopite} + 2 \text{ chlorite} + 18 \text{ calcite} + 18 \text{ CO}_2. \quad (1)$$

The principal assemblage in the phlogopite zone is calcite + quartz + phlogopite + plagioclase. Muscovite may also have been removed by the reaction

$$\text{muscovite} + \text{calcite} + 2 \text{ quartz} = \text{potassium feldspar} + \text{anorthite} + \text{H}_2\text{O} + \text{CO}_2. \quad (2)$$

However, the exact temperature relationship of this reaction to reaction 1 is unknown. The diopside isograd is marked by the reaction

$$\text{phlogopite} + 3 \text{ calcite} + 6 \text{ quartz} = \text{potassium feldspar} + 3 \text{ diopside} + \text{H}_2\text{O} + 3 \text{ CO}_2. \quad (3)$$

This isograd also marks the highest grade at which micas are present in these calc-silicate rocks. The highest metamorphic grade is manifested by the wollastonite-forming reaction

$$\text{calcite} + \text{quartz} = \text{wollastonite} + \text{CO}_2. \quad (4)$$

This reaction was the major decarbonation reaction in the rocks. The wollastonite zone is characterized by the assemblage calcite + wollastonite + diopside + vesuvianite + potassium feldspar + plagioclase. Vesuvianite appears to have formed by the reaction of scapolite with diopside, wollastonite, and calcite. Scapolite, which is widespread in the Weeks Formation, first appears at the diopside isograd, probably as a result of reaction between plagioclase, calcite, and NaCl in the coexisting fluid. Grossular occurs only occasionally, and titanite is a minor phase.

The maximum temperature of about 600 °C occurred at the contact with the stock and was less than 400 °C at the phlogopite isograd. However, all the metamorphic reactions probably occurred below or near 450 °C as a result of fluid infiltration (Labotka et al., 1988a). At these temperatures and a pressure of 2 kbar, the diopside and wollastonite zone assemblages require fluid compositions with $X(\text{CO}_2) < 0.05$ (Labotka et al., 1988a), suggesting infiltration of water-rich fluids. The change of whole rock $\delta^{18}\text{O}$ values from around 18‰ in the unmetamorphosed Weeks Formation toward the stock's value of 9.6‰ in the wollastonite zone suggests that the stock was the source of the fluid (Nabelek et al., 1984, 1988a).

ANALYTICAL TECHNIQUES

Concentrations of B were determined by prompt gamma neutron activation analysis (PGNAA) at the 10-MW Missouri University Research Reactor (MURR), following the method of Hanna et al. (1981). PGNAA is a very accurate method for analyzing elements, such as H, B, Cd, Sm and Gd, that emit prompt γ-rays during irradiation. MURR utilizes a horizontal beam port with a thermal neutron flux of 5×10^8 neutrons $\text{cm}^{-2}\ \text{s}^{-1}$ at the sample position. The sample is sealed in a teflon bag and placed at a 45° angle directly in front of the neutron beam. The emitted γ-rays were counted with a Nuclear Data ND66 analyzer system that employs a 1.85-keV resolution capacity and 19%-efficiency Ge(Li) detector mounted at an angle of 90° from the axis of the beam and a distance of 75 cm from the sample.

The 477.6-keV prompt γ-ray of B is the result of the reaction $^{10}\text{B}(n, \alpha)^7\text{Li}^* \rightarrow {}^7\text{Li} + \gamma$. It is Doppler-broadened owing to the recoil of ^7Li following α-particle release and lies between 469 keV and 489 keV. B concentrations were obtained by comparing counts per second per milligram of a sample to counts per second of a pure liquid B standard of known quantity and concentration that was dried on filter paper and packed in a teflon bag.

For samples that do not have Na, a simple count integral is used to measure accurately the B concentration. However, Na is a common cause of interference in the B peak region, with its strongest peak at 472.3 keV (Fig. 2). In order to determine the B concentration with confidence and accuracy, the Na contribution to the peak region must be factored out. Two methods of compensating for the Na interference have been used in the past. Gladney et al. (1976) used a graphical interpolation method to subtract the Na peak. However, there are no guidelines for the interpolation, and, because of the very high neutron-capture cross section of B compared to Na, even the smallest error in the interpolation will cause a large degree of uncertainty in the final values. Failey (1979) suggested using a known ratio of the sum of the 874-keV and 894-keV Na peaks to that of the 472-keV peak to subtract the Na interference. However, the area above the background occupied by the 874-keV and 894-keV peaks is very small and therefore difficult to measure accurately. Thus, any error in the measurement of these peaks will translate to error in the measurement of the 472-keV peak.

Two methods recently developed prove to be more reliable and accurate. The first of these is known as the middle method because it uses a collection of data from only the flat portion of the B peak (Curtis et al., 1980). The second method, developed at MURR, uses the total right-hand portion of the B peak from the 476-keV energy level. This method usually gives slightly better results because a larger area of the B peak is integrated. The

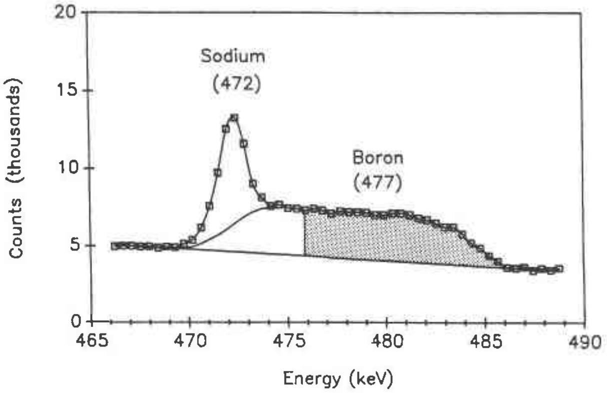

Fig. 2. A Doppler-broadened B peak showing Na interference. To determine B concentrations, only the shaded right-hand region is integrated and compared to the right-hand region of a pure B standard.

middle method may be more reliable on samples bearing significant amounts of Co owing to a small Co peak at 484 keV. In most geologic samples, however, the Co peak does not exceed background. The results from the measurement of B using the right-hand method on five replicates of several quality-control, geological standards from the National Bureau of Standards are in Table 1. On samples containing more than 10-ppm B, 1-σ errors are better than 3% but are 30–50% near the detection limit of 0.1 ppm.

Al concentrations were obtained by inductively coupled plasma-emission spectroscopy on powdered samples dissolved in lithium metaborate flux. The details of the analytical procedure are in McCall et al. (1990).

The ratios of O isotopes in calcite from the Weeks Formation were analyzed using conventional extraction techniques and a Finnigan Mat Delta-E mass spectrometer at the University of Missouri. The $\delta^{18}O$ values are reported relative to SMOW. The O isotopic composition was determined on CO_2 that was liberated by reaction with 100% phosphoric acid for 12 h at 25 °C. Replicate determinations on NBS-19 (TS limestone) during the course of analyses yielded $\delta^{18}O$ value of 28.52 ± 0.19‰.

RESULTS

Inasmuch as the Weeks Formation is a multicomponent sedimentary mixture, the concentrations of B, as those of other elements, vary substantially at any given metamorphic grade (Fig. 1). Therefore, to be able to determine the effect of metamorphism on B, the B concentrations were normalized to Al. This normalization seems appropriate, as most B in the samples very likely resides in Al-containing micaceous minerals, the correlation coefficient between B and Al in the unmetamorphosed rocks is high at 0.947, and Al is thought to be relatively immobile in hydrothermal fluids. Furthermore, this normalization is necessary because of the substantial loss of mass from the highest-grade rocks as CO_2. Whereas we would have liked to have determined B concentrations in micas, the unmetamorphosed and phlogopite-zone rocks are too fine grained to obtain clean mica separates. Nevertheless, a strong indication that B indeed resides in micas is in the positive correlation among the B concentrations and the proportions of muscovite and phlogopite in the unmetamorphosed and phlogopite zone rocks, respectively (Fig. 3). Moreover, that the correlation passes very near the origin implies that the amount of B in other minerals is insignificant. The mica proportions were determined by mass-balance using whole-rock major-element and mineral-chemistry data, following Labotka et al. (1984). Note that the phlogopite-zone rocks not only contain overall higher concentrations of B than the unmetamorphosed rocks, but also of Al (Fig. 1), which is an indication of some stratigraphically related differences in the bulk composition of the sediments.

TABLE 1. Concentrations of boron in ppm for several geological standard reference materials

Reference material	% Counts (Na/B)	This work	Literature mean*
SRM-278 (Obsidian rock)	46.0	25.2 ± 0.6	25.1 ± 0.2
SRM-1633a (Fly ash)	7.6	39.2 ± 0.5	39.7 ± 1.3
SRM-1632a (Bituminous coal)	3.7	58.7 ± 0.7	53 ± 2
SRM-1635 (Subbituminous coal)	1.2	110 ± 2	115 ± 17

* Gladney et al. (1984).

There is a drop in normalized B concentrations at the diopside isograd (Fig. 4). The diopside- and wollastonite-zone rocks have nearly ten times less normalized B than the unmetamorphosed and phlogopite-zone rocks. In addition, the spread in the normalized B concentrations is smaller in the former than the latter. Two samples from the stock have low B concentrations (3.65 and 3.94 ppm) and (1000 × B)/Al ratios (0.50 and 0.62).

DISCUSSION

The decrease in the normalized B concentrations at the diopside isograd suggests that reactions occurring at the isograd are responsible for the sharp decrease. As suggested by Figure 3, muscovite and phlogopite appear to contain the bulk of B in the unmetamorphosed and phlogopite-zone rocks, respectively. Therefore, the most plausible explanation for the drop is that the loss of B is related to the breakdown of micas by reactions 1, 2, or 3 because minerals formed at the diopside isograd have crystal structures that cannot accommodate large concentrations of B. For example, it has been found that in B-rich granitic rocks, muscovite contains nearly ten times the amount of B as coexisting feldspars and about five times the amount found in coexisting biotite (Shearer and Papike, 1986; Rockhold et al., 1987), whereas biotite generally contains the highest concentrations in granulites (Truscott et al., 1986). Groat et al. (1989) reported B_2O_3 concentrations of as much as 2.5 wt% in vesuvianites

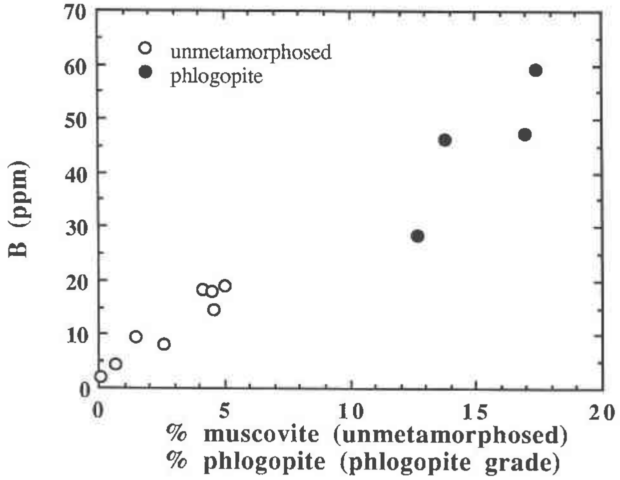

Fig. 3. Plot of total B concentrations against the weight proportions of micas in the unmetamorphosed and phlogopite-grade rocks.

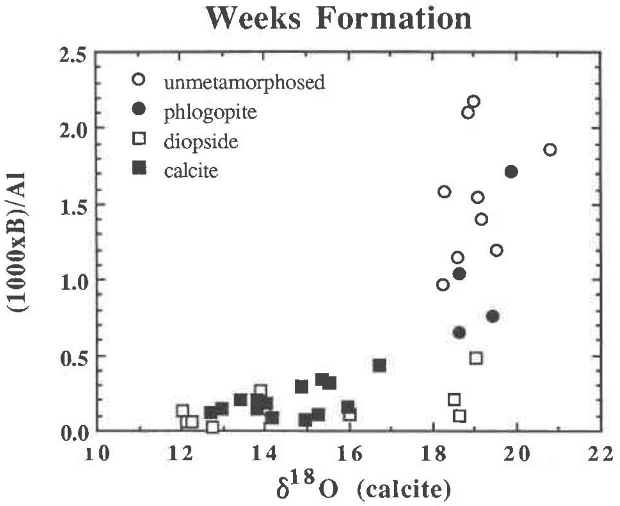

Fig. 5. Plot of (1000 × B)/Al ratios vs. calcite $\delta^{18}O$ values in the Weeks Formation.

from several localities. However, vesuvianite does not appear until the wollastonite isograd in the Weeks Formation. Since the rocks lost most B at the diopside isograd, there was little B available for incorporation into vesuvianite.

The B released during the breakdown of micas was probably dissolved into the infiltrated magmatic fluid that later left the system. B is thought to be highly soluble in hydrothermal fluids, as indicated by experimentally determined fluid-melt partition coefficients that are generally in excess of one in systems containing several weight percent B_2O_3 (Pichavant, 1981; London et al., 1988) and by the observed mobility of B in fluids emanating from pegmatitic intrusions (Shearer et al., 1984). The fluid emanating from the Notch Peak Stock was probably undersaturated in B with respect to the minerals in the Weeks Formation, given the low B concentrations in the stock. Any significant dissolved B in the magmatic fluid would probably be reflected in elevated B concentrations in vesuvianite-containing rocks in the wollastonite zone, which is not the case.

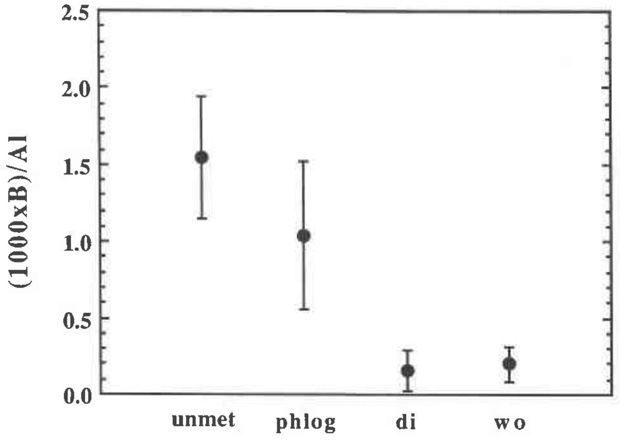

Fig. 4. Plot of B concentrations normalized to Al at each metamorphic grade. The error-bars are one-σ standard deviations of the means.

It may be argued that the loss of B from the diopside and wollastonite zones is not related to the breakdown of micas but, instead, is simply due to dissolution in the infiltrated fluid. A plot of (1000 × B)/Al against $\delta^{18}O$ values in calcite (Fig. 5) suggests that this is not the case. The approach of $\delta^{18}O$ values toward the stock's value of 9.6‰ is to a large degree a measure of the amount of magmatic fluid with which a given sample interacted (Nabelek et al., 1984, 1988b). Although the diopside and wollastonite-zone rocks have on the average lower $\delta^{18}O$ values than the unmetamorphosed and phlogopite-grade rock, there is no simple correlation between (1000 × B)/Al and $\delta^{18}O$ in the high-grade rocks. Instead, the (1000 × B)/Al ratio is small and essentially invariable, whereas there is a wide range in $\delta^{18}O$ values. Furthermore, several samples from the diopside zone have the same $\delta^{18}O$ values as their premetamorphic precursors, yet have very small (1000 × B)/Al ratios. This suggests that fluid infiltration was not the primary cause of the loss of B, although fluids were clearly necessary to dissolve B released from micas during metamorphic reactions.

An important feature of reactions 1, 2, and 3 is that they involve carbonate minerals. In the Weeks Formation, carbonate minerals generally predominate modally over micas. As a result, micas were completely consumed during the reactions. Hence, the carbonate minerals were indirectly responsible for the loss of B from the high-grade rocks in the Weeks Formation. In carbonate-poor metapelites where most micas are not eliminated by prograde reaction until the onset of granulite facies metamorphism, B may be more likely to persist into high metamorphic grades.

CONCLUSIONS

The strong dependence of B concentrations on mica-consuming metamorphic reactions evident in the Weeks Formation indicates that the reactions controlled the removal of B from rocks during metamorphism. The sharp decrease in B concentrations at the diopside isograd where all the micas were consumed indicates that rocks which do not contain micas, borosilicate mineral, or a mineral capable of holding significant B at high metamorphic grades will not retain high concentrations of B. The loss of B from such rocks may be further enhanced by the influx of fluids which drive metamorphic reactions in regional and contact metamorphic terrains (e.g., Ferry, 1984; Tracy et al., 1983; Labotka et al., 1984, 1988a).

This study suggests that the bulk composition of metasediments may control the behavior of B during metamorphism. In carbonate-poor metapelites, mica-consuming reactions do not occur until the onset of the second sillimanite isograd. At high pressures, such reactions may lead to melting and, owing to the high solubility of B in granitic melts, to B-rich granites. In the Weeks Formation, the micas were consumed at relatively low temperatures (≤450 °C) because of the presence of carbonate minerals. Therefore, the implication of this study is that calcareous pelites may lose much B prior to anatexis owing to the occurrence of calc-silicate, mica-consuming reactions during metamorphism. The persistence of B in sedimentary rocks until anatexis is controlled in part by the relative proportion of micas and carbonate minerals.

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