

Evidence for major mass transfer and volume strain during regional metamorphism of pelites

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ABSTRACT

Systematic examination of published sedimentary and metamorphosed pelite analyses has revealed evidence of significant mass transfer and volume strain during regional metamorphism. Statistical analysis of the data shows that Barrovian zone metamorphism of pelitic schist generally causes increases in the whole-rock concentrations of the low-solubility elements Ti and Al. The observed increases in Ti and Al contents as functions of metamorphic grade are almost certainly due to residual enrichment caused by the removal of other more soluble species. Application of mass-balance principles to the pelite compositional trends indicates that the average pelite may lose as much as 30% of its mass and volume during progressive metamorphism from subgreenschist to amphibolite facies conditions. The bulk of the lost mass is silica, not volatiles. In addition, other elements, particularly Ca, Na, and K, appear to be highly mobile in deep-crustal metamorphism. Contrary to conventional interpretations, it is concluded that the regional metamorphism of pelites is not an isochemical process.

INTRODUCTION

Except for the loss of volatiles and the movement of certain trace elements, the regional metamorphism of aluminous shale is generally regarded as an isochemical process (Shaw, 1956; Butler, 1965; Ronov et al., 1977; Yardley, 1977, 1986; Ferry, 1982, 1988; Haack et al., 1984; Wood and Walther, 1986). In other words, for transport distances greater than typical hand-sample scale, the major rock-forming elements such as Si are thought to be virtually immobile during metamorphism (see Wood and Walther, 1986). This view was firmly established in the petrologic literature by Shaw (1956), who reached his conclusions on the basis of the geochemistry of the Littleton Formation of New Hampshire and also on comparative study of published sedimentary and metamorphic rock analyses.

It has been well documented that large-scale fluid flow may occur during regional metamorphism (Rye et al., 1976; Rumble et al., 1982; Chamberlain and Rumble, 1988; Ferry, 1988; Wickham and Taylor, 1990). It is important that the metamorphic fluids may transport mass, and therefore the potential for metasomatism during regional metamorphism is great (see Fyfe et al., 1978; Ferry and Dipple, 1991). For example, Ferry (1983) and Tracy et al. (1983) found negative correlations between volumetric fluid to rock ratios and alkali contents in metacarbonates. This suggests that the fluid phase may be an important agent of mass transfer in metacarbonate systems. In spite of these significant advances, major element metasomatism in pelitic rocks has been relatively little studied, and significant mass transfer has yet to be well documented.

In view of the widespread fracture-controlled and pervasive fluid flow now known to occur in dynamic metamorphic systems, the time is right to investigate further the geochemistry of pelitic schists with the goal of detecting evidence of mass transfer. Toward this end, I first reexamined statistically the bulk chemistry of the Littleton Formation as described by Shaw (1956) and considered a new compilation of published sedimentary and metamorphic rock analyses in order to unravel trends in pelite compositions as a function of metamorphic grade. I then undertook a mass-

balance analysis of metasomatic effects and associated volumetric strains. Throughout the discussion here, particular attention is focused on the geochemistry of Al and Ti because, of all the major elements in pelites, Al and Ti have the lowest solubilities in the low-CO₂ fluids typical of pelitic sequences (see Ragnarsdottir and Walther, 1984; O'Hara and Blackburn, 1989). Therefore, Al and Ti are the best choices for establishing a geochemical "reference frame" in which the mass-balance relations of other more mobile species can be assessed. Nonetheless, if appropriate complexing agents such as carbonate species are present, even Al and Ti may be mobile to some degree (Anderson and Burnham, 1983; Kerrick, 1990; Philippot and Selverstone, 1991). Other low-solubility elements, such as Zr and Cr, could also be used for a geochemical reference frame, but these elements were not determined in most of the analyses considered herein.

LITTLETON FORMATION—A REEXAMINATION

In his 1956 study, Shaw concluded that aside from volatile loss and certain trace element changes, the Littleton Formation underwent only minor metasomatism involving addition of Na and Ca during metamorphism. These results notwithstanding, a statistical reexamination of the bulk-chemical data indicates that the geochemistry of the Littleton is probably more complex than previously thought. This is especially clear if we consider the behavior of Al₂O₃ and TiO₂. The oxide averages for Shaw's (1956) low-grade and medium- and high-grade pelites are shown in Table 1. It is important to point out that the statistical comparisons of oxide concentrations described below are done with the analyses normalized on an H₂O- and CO₂-free basis so that changes in percent of oxide as a function of metamorphic grade reflect factors other than loss of volatile species.

Although the concentrations of Al₂O₃ and TiO₂ appear to increase with metamorphic grade, Shaw (1956) concluded, on the basis of Student's *t* tests, that when devolatilization was taken into account the two groups did not have significantly different average Al₂O₃ or TiO₂ contents. However, given the relative immobility of Ti and Al in pelitic sequences, there are other statistical hypotheses bearing directly on the problem of metasomatism that can be addressed with these data. In particular, we can test the null hypothesis that the differences in Al₂O₃ and TiO₂ are due to chance against the alternative hypothesis that the medium- and high-grade rocks have greater mean Al₂O₃ and TiO₂ contents than the low-grade rocks. This involves using Student's *t* test with appropriate one-sided significances (see Mendenhall and Sincich, 1988).

The results of the tests are that mean Al₂O₃ and TiO₂ concentrations are significantly greater in the medium- and high-grade rocks relative to their low-grade counterparts, at probability levels of 96.9% and 97.5%, respectively. These results rest on the assumptions that (1) the samples were selected from populations with approximately normal relative frequency distributions and common variances, (2) the samples were randomly selected in an independent manner, and (3) the average compositions of the low-grade and medium- and high-grade groups were the same prior to metamorphism. According to Shaw (1956), there are no compelling reasons to reject these assumptions. Furthermore, Phillips (1988) argued that positive correlations between Al and Ti are unlikely in aluminous sediments, and this is consistent with compositional trends in

Note: Additional material for this article is Supplementary Data 9123, available on request from the GSA Documents Secretary (see footnote 1).

modern turbidite muds (see McLennan et al., 1990). Thus, in contrast to Shaw (1956), I conclude that average Al_2O_3 and TiO_2 concentrations probably increased relative to the sum total of other nonvolatile oxides during metamorphism of the Littleton Formation. It is worthwhile to note that the results of applying new, albeit controversial, types of multivariate statistical tests designed specifically for compositional data by Aitchison (1986) also support this conclusion. Implications for metasomatism are presented in more detail below.

NEW COMPILATION OF LITERATURE DATA

In the course of his comparative analysis of pelite chemistry, Shaw (1956) compiled a bulk-composition data base that has been immensely valuable to many geochemical and petrologic studies. Nonetheless, it is important for the purposes of this study to construct a new data base, for several reasons. Most important is that more analyses have become available that can be used to place further bounds on pelitic compositions as a function of metamorphic grade. In addition, this study is restricted to the products of regional metamorphism through the classical Barrovian index-mineral zones.

Every effort has been made to use only those rocks with definite pelitic protoliths whose metamorphic grade could be well established. The rocks range in age from Archean to Cenozoic and they come from sedimentary basins and metamorphic belts worldwide. Owing to the inevitable large range in initial bulk compositions and differences in metamorphic history, the analyses have been grouped only broadly in terms of (1) shale and "slate," (2) greenschist facies (chlorite, biotite, and garnet zones), and (3) amphibolite facies (staurolite, staurolite-kyanite and staurolite-andalusite, and sillimanite zones). The data base includes both wet chemical and X-ray fluorescence (XRF) analyses.

This compilation differs from that of Shaw (1956) in several important respects. First, as noted above, only regional metamorphic rocks have been considered. This was done in order to exclude the uncertain metasomatic effects of hydrothermal fluid circulation around cooling plutons in contact-metamorphic environments. Second, un lithified materials such as residual weathering clays have not been considered. Third, high-grade rocks that may have been affected or generated by partial melting events, such as migmatites, granitic gneisses, and charnockites, have been excluded. Fourth, in an effort to eliminate sandstones and quartzites from the data base, analyses were not used if bulk Al_2O_3 was less than 12 wt%. This value was chosen because it is the approximate maximum Al_2O_3 content of rocks generally classified as sandstones or psammites (compare with Pettijohn, 1975, chapter 7). Fifth, calcareous rocks such as marls were filtered from the data set on the basis of whole-rock CaO, rather than CO_2 . CaO content was used for the following reasons: (1) CO_2 has not been determined in a large number of the analyses, and (2) CO_2 is lost during metamorphism, and it is therefore not a reliable indicator of original carbonate content. Because the vast majority of rocks described as "pelitic" have CaO less than 6 wt%, this value was used as an upper limit on CaO content in this study (cf. Pettijohn, 1975, chapter 8). Finally, the size of the data base has been increased, so that the total number of analyses ($N = 351$) is more than double that used by Shaw (1956). Whereas this data set certainly does not include every analysis of sedimentary and metamorphosed pelites in the literature, it is, I hope, a statistically valid sample.¹

Average compositions for the various rock groups are given in Table 2. Although some of the analyses represent composites of more than one sample, each analysis was given equal weight in the averages. As discussed above, all statistical comparisons of oxide concentrations between different rock groups are made using Student's *t* tests (see Mendenhall and Sincich, 1988), the average compositions shown in Table 2 being normalized on a volatile- and loss on ignition (LOI)-free basis.

¹A list of references from which the data base was compiled, GSA Supplementary Data 9123, is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301.

Although some of the trends shown in Table 2 are well known, such as decreases in weight percents of volatiles and in $\text{Fe}^{3+}/\text{Fe}^{2+}$ with increasing grade, important additional geochemical systematics are evident that have not been described in detail in the past. For example, SiO_2 concentrations decrease as metamorphic grade increases. Average SiO_2 contents for the greenschist and amphibolite facies differ from the average SiO_2 content of shale and slate at probability levels exceeding 99%. In contrast, TiO_2 , Al_2O_3 , total iron as Fe_2O_3 , MgO, and MnO all tend to increase as a function of metamorphic intensity. Average concentrations of these oxides in the amphibolite facies group are all significantly different, at probability levels greater than 98%, from the corresponding concentrations in shale and slate. In contrast, mean weight percent of CaO, Na_2O , and K_2O remain essentially unchanged as metamorphic grade increases, when loss of volatiles is taken into account. In summary, it appears from the average compositional data that the regional metamorphism of pelitic rocks is *not* an isochemical process. Although the changes in oxide concentrations may seem to be relatively small, they are in fact indicative of significant metasomatism during metamorphism.

MASS-BALANCE ANALYSIS OF METASOMATISM

Whereas oxide weight percent comparisons may be useful in assessing general chemical differences between different rock groups, they provide no quantitative information about mass transfer. Therefore, I now turn to a mass-balance analysis of the bulk-chemical data. This analysis requires two fundamental conditions: (1) the average sedimentary rock composition (protolith) must represent that of the metamorphosed rocks prior to metamorphism, and (2) an immobile reference element must be identified. In this study, there are no compelling reasons to conclude that the first condition is not satisfied. As noted above, low-solubility elements such as Al and Ti can be used to satisfy the second condition, at least to a first approximation.

The most basic quantities that can be computed are changes in mass from an initial state to some final state. If we denote an immobile reference species with the subscript *i*, then the percentage change in total mass based on the reference element *i*, here defined as T_i , is simply

$$T_i \equiv \left[\frac{\text{final mass} - \text{initial mass}}{\text{initial mass}} \right] \times 100 = \left[\frac{C_i^o}{C_i^f} - 1 \right] \times 100, \quad (1)$$

where C_i^o and C_i^f are the concentrations of *i* in the initial and final states, respectively. Following Brimhall et al. (1988), we can extend this formulation to describe the percentage changes in the mass of a specific mobile species *j*, here denoted as τ_j^f :

$$\tau_j^f = \left[\left(\frac{C_j^o}{C_j^f} \right) \left(\frac{C_j}{C_j^o} \right) - 1 \right] \times 100, \quad (2)$$

where C_j^o and C_j^f are the concentrations of *j* in the initial and final states, respectively. Negative values of T_i and τ_j^f indicate mass loss, whereas positive values correspond to mass gain. For example, a τ_j^f value of +100% means that the mass of element *j* has doubled in going from an initial to a final state. We proceed by first using equations 1 and 2 in conjunction with the Littleton Formation data, and then continue with the average compositional systematics shown in Table 2.

Comparison of the low-grade and medium- to high-grade Littleton rocks yields mean $T_{Ti} = -17\% \pm 7.4\%$ and $T_{Al} = -12\% \pm 4.5\%$ (1 σ standard error uncertainties). If we assume that Ti and Al provide a geochemical reference frame, these results indicate that metamorphism of the Littleton was accompanied by mass loss. It is surprising that the bulk of this mass (~75%–80%) was silica, rather than volatiles. The mean values of $\tau_{Ti}^{Si} = -20\% \pm 7.4\%$ and $\tau_{Al}^{Si} = -16\% \pm 4.9\%$ reflect substantial silica removal during metamorphism. In addition, because the concentrations of Fe, Mg, and K do not increase with metamorphic intensity whereas those of the reference elements do, it is probable that Fe, Mg, and K were also lost during metamorphism. In contrast, Ca and Na appear to have been added

TABLE 1. AVERAGE COMPOSITION OF LITTLETON ROCKS, FROM SHAW (1956)

	Low-grade	Medium- and high-grade
SiO ₂	62.58 ±1.243	60.26 ±1.215
TiO ₂	0.87 ±0.070	1.05 ±0.044
Al ₂ O ₃	18.09 ±0.633	20.64 ±0.776
Fe ₂ O ₃	1.60 ±0.295	1.41 ±0.270
FeO	5.07 ±0.319	5.49 ±0.421
MgO	2.18 ±0.368	1.93 ±0.103
CaO	0.16 ±0.040	0.52 ±0.087
Na ₂ O	0.81 ±0.171	1.38 ±0.175
K ₂ O	3.68 ±0.207	3.72 ±0.153
H ₂ O	4.18 ±0.115	2.80 ±0.222
CO ₂	0.03 ±0.006	0.03 ±0.009

Note: Uncertainties are given as one standard error of the mean value. The low-grade rocks are shale and slate, whereas the medium- and high-grade rocks are amphibolite facies pelite. There were 7 samples for low-grade group and 11 samples for medium- and high-grade group.

TABLE 2. AVERAGE COMPOSITIONS OF PELITIC ROCKS

	Shale and slate	Greenschist	Amphibolite
SiO ₂	60.34 ±0.379 (105)	58.33 ±0.760 (85)	56.25 ±0.436 (161)
TiO ₂	0.76 ±0.021 (101)	0.90 ±0.024 (80)	1.05 ±0.019 (161)
Al ₂ O ₃	17.05 ±0.229 (105)	18.63 ±0.376 (85)	20.18 ±0.261 (161)
Fe ₂ O ₃ *	7.37 ±0.199 (105)	8.06 ±0.295 (85)	9.31 ±0.153 (161)
MgO	2.69 ±0.105 (105)	3.01 ±0.154 (85)	3.23 ±0.088 (161)
MnO	0.09 ±0.008 (73)	0.29 ±0.075 (79)	0.18 ±0.015 (160)
CaO	1.45 ±0.144 (104)	1.50 ±0.146 (85)	1.54 ±0.089 (161)
Na ₂ O	1.55 ±0.100 (105)	1.59 ±0.094 (85)	1.80 ±0.062 (161)
K ₂ O	3.64 ±0.109 (105)	3.98 ±0.150 (85)	4.02 ±0.092 (161)
P ₂ O ₅	0.14 ±0.009 (87)	0.18 ±0.028 (77)	0.19 ±0.008 (156)
H ₂ O†	4.25 ±0.144 (102)	2.94 ±0.196 (34)	3.02 ±0.094 (106)
CO ₂	1.05 ±0.173 (71)	0.32 ±0.108 (14)	0.18 ±0.044 (40)
LOI‡	5.54 ±0.559 (15)	4.18 ±0.371 (50)	1.56 ±0.125 (55)
Volatiles**	4.8 ±0.21 (104)	3.7 ±0.24 (82)	2.5 ±0.09 (161)
Fe ³⁺ /Fe ²⁺	0.66 ±0.07 (85)	0.42 ±0.05 (34)	0.33 ±0.03 (106)

Note: Uncertainties given as one standard error on the mean value. Number in parentheses is the number of analyses used to compute the average and standard error.

*Total iron as Fe₂O₃.

†Includes H₂O⁺ and H₂O⁻.

‡Loss on ignition for XRF analyses.

**Approximate H₂O + CO₂, computed by averaging H₂O + CO₂ for wet chemical analyses with loss on ignition (LOI) from XRF analyses. LOI is not exactly equivalent to H₂O + CO₂ because: (1) C and sulfide S are also lost on ignition, adding to the LOI value, and (2) the sample takes on oxygen during ignition due to oxidation of Fe²⁺ to Fe³⁺, which in turn tends to make the LOI value less than the actual volatile content. However, in pelitic compositions these two effects will tend to cancel, and therefore LOI values are here taken to be roughly equivalent to H₂O + CO₂.

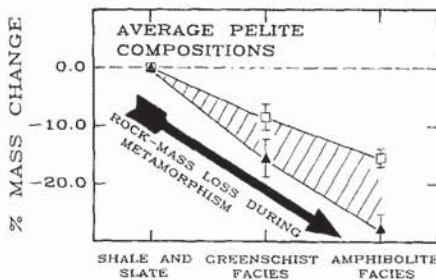


Figure 1. Change in rock mass as function of metamorphic grade relative to shale and slate for average pelite compositions of Table 2. Squares and triangles represent calculated T_{Al} and T_{Ti} values, respectively. Error bars given as one standard error on mean T_i . Note that regardless of which element is taken as reference species, T_i values all indicate substantial overall loss of rock mass during regional metamorphism. As discussed in text, bulk of lost mass is silica.

to the medium- and high-grade rocks, as discussed previously by Shaw (1956).

Consideration of the average compositional data of Table 2 also leads to the conclusion that metamorphism of pelites may involve significant mass loss, but the overall pattern is somewhat different from that indicated by the Littleton Formation. Here, the concentrations of the major elements Ti, Al, Fe, and Mg all increase with metamorphic intensity. Using each of these elements to calculate T_i yields mean $T_{Ti} = -28\% \pm 2.4\%$, $T_{Fe} = -21\% \pm 2.5\%$, $T_{Mg} = -17\% \pm 4.0\%$, and $T_{Al} = -16\% \pm 1.6\%$ for the mean amphibolite facies composition relative to shale and slate. Because Ti is likely to have the lowest solubility under metamorphic conditions (see O'Hara and Blackburn, 1989), the value of $T_{Ti} = -28\% \pm 2.4\%$ is here taken to be the "best" approximation for the total mass loss. However, regardless of which element is chosen as the geochemical reference frame, all the above T_i values are consistent with substantial overall mass loss during metamorphism (Fig. 1). As in the Littleton Formation, most of this mass is silica. Average τ_i^{Si} values range from $-33\% \pm 2.3\%$ to $-21\% \pm 1.7\%$ for the amphibolite facies, depending upon which reference element is used. However, unlike the Littleton, τ_i^{Ca} and τ_i^{Na} are negative. For example, $\tau_{Ti}^{Ca} = -23\% \pm 9.2\%$ and $\tau_{Ti}^{Na} = -16\% \pm 6.7\%$ when the amphibolite facies group and the shales and slates are compared. Although the uncertainties in the calculated mean τ_{Ti}^{Ca} and τ_{Ti}^{Na} are relatively large, it is probable that Ca and Na can be mobile during metamorphism. In addition, $\tau_{Ti}^K = -20\% \pm 4.0\%$ for the amphibolite facies, indicating that K is depleted in the higher grade rocks.

At present, it is uncertain why the geochemistry of the Littleton Formation appears to differ in detail from the average pelite compositional trends. It is certainly reasonable to expect that because no two metamor-

phic belts are identical in terms of protolith character and geologic evolution, differences in chemical behavior should occur. However, it is also true that the number of Littleton Formation samples considered was relatively small ($N = 18$), and this may not be a large enough sample to elucidate fully geochemical trends.

IMPLICATIONS FOR VOLUMETRIC STRAIN

Clearly, mass loss of the magnitude indicated by the pelite compositional trends will result in substantial decreases in rock volume. These strains can be estimated by considering chemical systematics in conjunction with likely changes in rock density attending regional metamorphism. Owing to mass loss and the formation of dense minerals, including garnet and kyanite, it can be shown that rock density increases of at least 10% can be attained during the metamorphism of pelites to high grade.

As discussed by Brimhall et al. (1988), the percent volume strain calculated on the basis of a reference species i , here denoted ϵ_i , is given by

$$\epsilon_i = \left[\left(\frac{C_i^o}{C_i^f} \right) \left(\frac{\rho^o}{\rho^f} \right) - 1 \right] \times 100, \quad (3)$$

where ρ^o and ρ^f are the initial and final rock densities, respectively. Using a conservative estimate of 5% density increase from protolith pelite to amphibolite facies schist, we now employ equation 3 with the pelite compositional data.

Depending upon which reference element is used, volumetric strains are in the range of -16% to -20% for the medium- and high-grade Littleton Formation relative to the low-grade rocks (Table 1), and between -20% and -31% for the average amphibolite facies pelite relative to the shales and slates (Table 2). The calculated strains provide strong evidence that

the regional metamorphism of pelites is not an isovolumetric process, owing to density increases, devolatilization, and, most important, loss of major rock-forming elements. Although the strain estimates provide no direct information about the nature of shape change, it is reasonable to expect that a substantial amount of the volume loss occurred parallel to penetrative cleavage fabrics (see Wright and Platt, 1982).

DISCUSSION

The conclusion that progressive metamorphism of pelites is generally accompanied by mass loss clearly rests on the assumption that a suitable chemical reference frame has been chosen. In this study, the simplest interpretation of the increase in concentration of low-solubility elements such as Ti with metamorphic grade is that these elements are being enriched in a residual fashion, owing to the loss of more soluble species such as Si. Regardless of how the reference frame is chosen, the data in Tables 1 and 2 strongly suggest that changes in major element concentrations do occur during regional metamorphism.

If we accept the likely hypothesis that Ti and other low-solubility elements do in fact provide a geochemical reference frame, then a critical question arises: where is all the mass that is lost from the pelites going? The foregoing calculations provide strong evidence that the bulk of this lost mass is silica. The most obvious candidates for a silica sink are quartz veins, ubiquitous features of pelitic sequences worldwide. However, whether quartz veins can account for the total mass of transported silica is currently unknown. If both Al and Ti behave in a perfectly immobile fashion, then T_i values calculated with either element should be the same. However, for both the Littleton Formation and the average pelite compositions, the amount of mass loss predicted by Ti systematics is greater than that predicted by Al (Fig. 1). In view of this result, it is tempting to speculate that Al may in fact be mobile to some degree in metamorphic fluids. In conjunction with alkalis, this Al could contribute to the feldspar commonly present in the veins of high-grade metamorphic rocks (see Vidale, 1974). Evidence for Al mobility has been presented by Tracy et al. (1983) for metacarbonate systems and by Kerrick (1990) for more aluminous compositions.

In summary, it seems clear that, far from being an isochemical process, the regional metamorphism of pelites probably involves significant metasomatism such that the average pelite may lose as much as 30% of its mass and volume during progressive metamorphism to high grade. Thus, many fundamental questions now must be addressed. For example, what are the roles of advection and diffusion in hydrochemical fractionation? What are the length and time scales of mass transport? Is convection a significant agent of mass redistribution in the deep crust? The answers to these and other questions must await future petrologic, mass-balance, isotopic, kinetic, and modeling studies aimed at illuminating the chemical dynamics of progressive regional metamorphism.

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