Variscan regional metamorphism in north Devon and west Somerset

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Metamorphism of the Devonian and Carboniferous strata in north Devon and west Somerset ranges from a high diagenetic to low greenschist facies level. The Devonian sequences range from anchizone to epizone, and the highest grade is developed typically in well cleaved slates; a low pressure facies series *is* indicated by b_0 data. The Carboniferous sequences range from diagenetic levels to anchimetamorphic and closely corelates with previously published vitrinite reflectance data for Carboniferous rocks in the west of the area. High geothermal gradients are again indicated. Oxygen isotope data suggest that the rocks approached isotopic equilibrium with the fluid phase during metamorphic recrystallization, although disequilibrium relationships now prevail.

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Introduction and methodology

The geology of the north Devon and west Somerset region is dominated by a clastic sedimentary sequence of Devonian and Carboniferous age, which has been variably deformed and altered in the Variscan orogeny (Matthews 1977). The Variscan regional metamorphism of the Devonian-Carboniferous succession in the southern part of the southwest peninsula has attracted much interest, where it has been shown to vary from a diagenetic to low greenschist facies level (Phillips 1928; Brazier et al. 1979; Robinson and Read 1981; Primmer 1985a, 1985b). However the metamorphic character of the north Devon/west Somerset region has received virtually no attention apart from a preliminary account (Kelm 1986), and an initial examination of vitrinite reflectance in some Carboniferous sediments (Cornford et al. 1987)

In this study the variation in metamorphic grade across the region has been determined by measurement of the illite crystallinity (IC) of some 250 samples of pelitic character. This technique is now well known in the examination of metamorphism at a sub-greenschist level and the peak width method of Kubler (1967) has been utilised in this study.

Clay material was prepared by ultrasonic disaggregation of small rock chips followed by centrifuge separation of appropriate grain sizes. The machine conditions for measurement of IC values were Ni filtered, Cu Kα radiation at a tube setting of 40kV and 40mA, using an automatic divergence slit, receiving slit 0.1mm, time constant of 2, graphite crystal monochromator at a scanning speed of $1/2°$ 2θ/minute. Measurement of the b_0 parameter (Sassi and Scolari 1974) as an indicator of metamorphic facies series has also been undertaken, and the 58-63° 2θ range was scanned at $1/4°$ 2θ /minute, using the quartz 211 peak as an internal standard. Use was made of a special mount (Robinson 1981) which enhanced the 060 peak, relative to the overlapping 331 peak, which causes error in the true determination of the 060 value (Frey et al. 1983; Robinson and Bevins 1986).

Stable isotope analyses of $^{18}O/^{16}O$ ratios of co-existing pairs of quartz and illite have been undertaken on slatey samples from a north-south traverse of the Devonian-Pilton Shale sequence of the Exmoor Forest. This work was in order to determine the absolute temperatures of this anchizone-greenschist transition, and to establish the oxygen isotope character of the metamorphic fluid.

The material for isotopic analysis was treated for removal of Fe, Mn and Ti oxides (Mehra and Jackson 1963), and organic matter by using 30% H_2O_2 solution. The mineralogy of each sample was then determined by XRD analysis; where chlorite was present the samples were further treated with 1M HCl at 80°C until no further chlorite was detected by XRD. Portions of the size fractions were subjected to a combined fusion and leaching process (Syers et al. 1968) to produce quartz separates

whose purity was checked by XRD analysis. Oxygen was liberated from the silicates by reaction with CIF3 following the technique of Clayton and Mayeda (1963) as modified by Borthwick and Harmon (1982), at a temperature of 600°C for quartz and 650°C for the phyllosilicates. CO_2 was produced by reaction of the oxygen with a hot, platinised graphite rod for isotopic analysis on a triple collector, fully automated VG903 mass spectrometer. Before isotopic analysis non-structural water was removed from the samples by heating samples at 200°C for 1.5 hours under vacuum and by brief prefluorination. All data are reported as ‰ relative to SMOW; the NBS28 quartz standard gives $\delta^{18}O=+9.6‰$ with a 1σ precision of ±0.1‰.

Illite crystallinity

At diagenetic and anchizone levels of metamorphic transformation it is the character of the clay minerals and clay assemblages, in particular the illite/smectite (I/S) mixed-layers and illitic materials, that have been most useful in recognising prograde variation. These variations have been quantified by XRD methods of the measurement of the illite content in I/S, and the change in the peak-width of the 10Å peak which can be used to trace relative grade changes at diagenetic and anchizone levels respectively.

In many areas in which rocks show only diagenetic grade, the concentration of I/S in the pelitic units is often too low, especially at a high diagenetic level, to allow satisfactory identification of the appropriate diffraction peaks necessary to measure compositional variation in smectite contents. In these circumstances use is often made of the illite crystallinity technique, where the width of the 10Å peak of illitic material is believed to vary in response to loss of the final smectitic components (< 15%) in I/S (Srodon 1984). This is not a "normal" use of the technique which is considered to be a measure of the increasing size of the illite crystallites in the "c" direction as metamorphic grade increases (Merriman and Roberts 1985). As such the method has been applied most successfully to the definition of, and recognition of variation in grade within, the anchizone.

Many of the sequences in the area of study, particularly those of Carboniferous age, have not suffered major metamorphic change. XRD investigation of the pelitic units of these sequences has shown that I/S is not a major component (Kelm 1988). It has not been possible, therefore, to study the variation in composition of I/S in the diagenetic areas, and where present I/S had <15% smectite content. It was thus necessary to resort to the less satisfactory method of tracing variation in grade at diagenetic level by measuring variation in IC. In doing so a study has been made of the variation in IC of four different size fractions of clay material, rather than the single < 2μm fraction normally utilised.
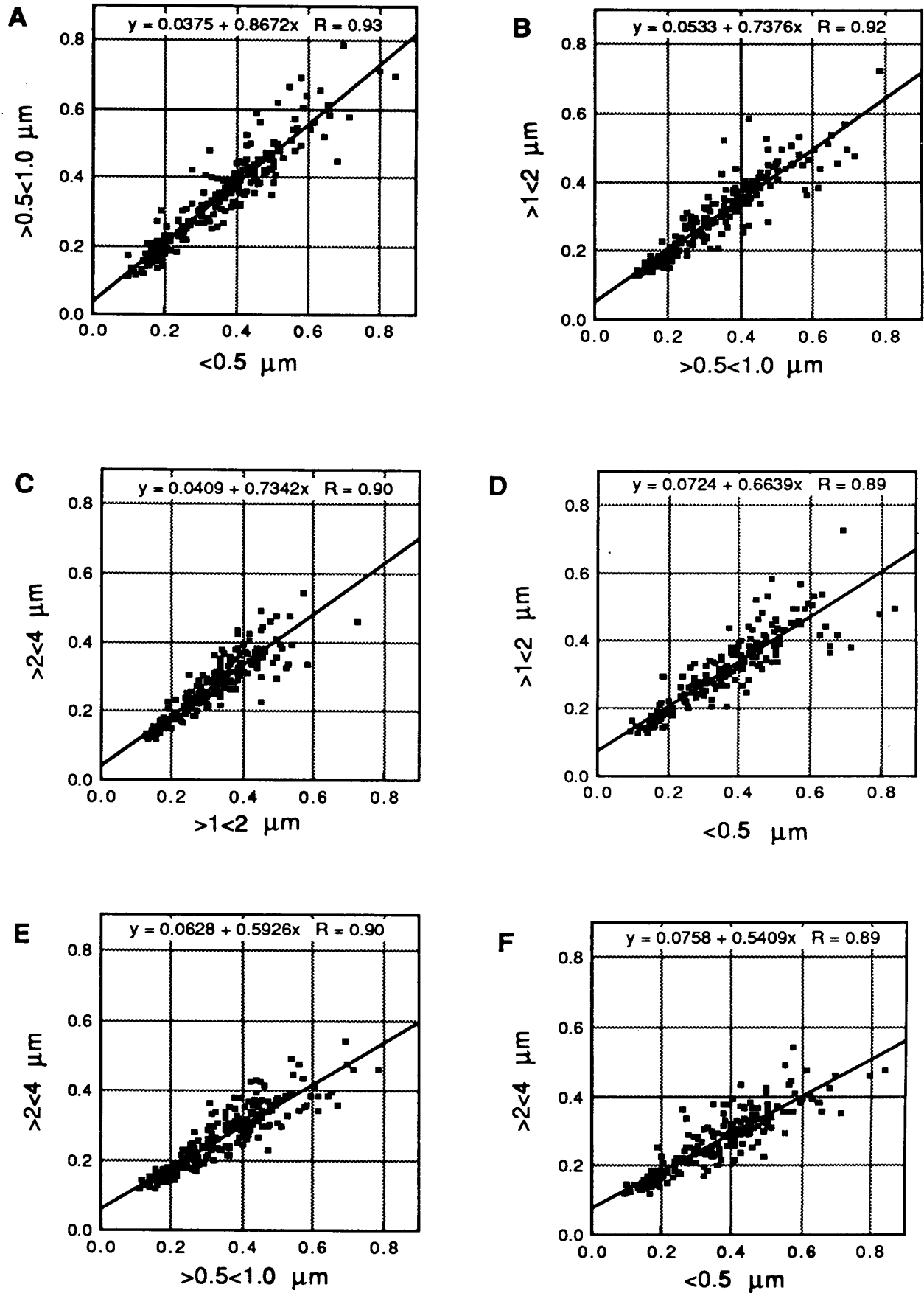
Figure 1. Bivariate data plots showing linear relationships between illite crystallinity values (in °2θ) of four different grain size fractions.

The four fractions analysed in this work are 1) < 0.5µm; 2) > 0.5 < 1.0µm; 3) >1 < 2µm; 4) >2 <4µm. A finest fraction such as <0.2µm has not been examined because of the peakbroadening effect on the 001 peak that results with such material (Kisch 1983). The bivariate relationships between the IC values of grain size pairs are shown in Fig. 1A-F. These plots demonstrate that there are simple linear relationships with high correlation coefficients between all the different size fractions (Fig. 1). The regression lines, however, show quite different slopes between the different plots indicating that significant differences in relative grade are indicated according to the size fraction selected. For example as seen from Fig. 1F, an IC value from the <0.5µm fraction of 0.4°2θ, which might be regarded as equivalent to the onset of the anchizone, is matched by an IC value from the >2 <4µm fraction of 0.29°2θ, and indicative of the mid-anchizone.

From examination of Fig. 1 it can be ascertained that the closest correspondence in IC values is between the <0.5 and > 0.5 < 1.0µm size fractions which show a relationship close to 1:1. This correspondence is such that the apparent difference in grade indicated between these two size fractions is in the order of the typical machine error (1σ) for reproducibility of the IC measurement (c. 0.02°2θ)> and in reality there is noexperimental difference between the values. The lowest correspondence occurs, as expected between the finest and coarsest fraction, namely the <0.5 and >2<4µm fractions (Fig. 1F). Here a value in the 0.5µm fraction, corresponding approximately to the onset of the anchizone, of 0.4°2θ, is equivalent to a mid-anchizone value of 0.29°2θ. A comparison (Fig. 1D) between the finest fraction (<0.5µm) and the normal size fraction used in IC studies (< 2µm) shows that the disparity between the data, at a mid-anchizone level of approximately 0.3°2θ, is some 0.03° and thus is close to the machine error. Thus there would be little difference in designation of metamorphic grade of an area between these two size fractions once grade reached the mid-anchizone level. In all the diagrams it is noticeable that the correspondence between the IC values is close for samples where values approach those of the onset of the epizone (c. 0.21°2θ).

Variations in IC values with selection of varying grain sizes has been noted previously (Weber 1972; Kelm 1986); the usual explanation is that at diagenetic levels the presence of relict detrital grains in the coarser size fractions affects the IC value giving an anomalous result. The increasing scatter in Kubler values observed as the crystallinity decreases (Fig. I) offers support to the variable presence of relict detrital material with higher crystallinity values.

In considering the application of the IC technique to the interpretation of metamorphic grade in the region, it is apparent that below approximately mid-anchizone levels there will be a dependency upon the choice of size fraction. The very close correspondence seen between the two finest fractions (Fig. 1A) suggests that the significant difference in observed grade will lie between size fractions above and below the 1µm limit. This feature is clearly seen in Fig. 2 which shows the distribution of the Kubler data relative to the diagenetic, anchizone and epizone levels taken here at °2θvalues of > 0.40°, 0.40-0.21° and < 0.21° respectively for the < 2µm (Fig. 2A) and < 0.5µm (Fig. 2B) size fractions. It is noticeable that in both diagrams the proportions of epizone values are almost identical, supporting the earlier conclusion that correspondence between all size fractions is achieved in the epizone. The very noticeable feature in Fig. 2A and 2B is the marked difference in the proportions of the anchizone and diagenetic samples, with the 0.5µm fraction having more than doubled in size relative to the < 2µm fraction. Metamorphic maps of the area studied are shown in Fig. 3A and 3B based on the <0.5µm size fraction and the <2µm fraction respectively.

The map based on the <0.5µm fraction (Fig. 3A) shows that there is a general concordance between grade and stratigraphic position.
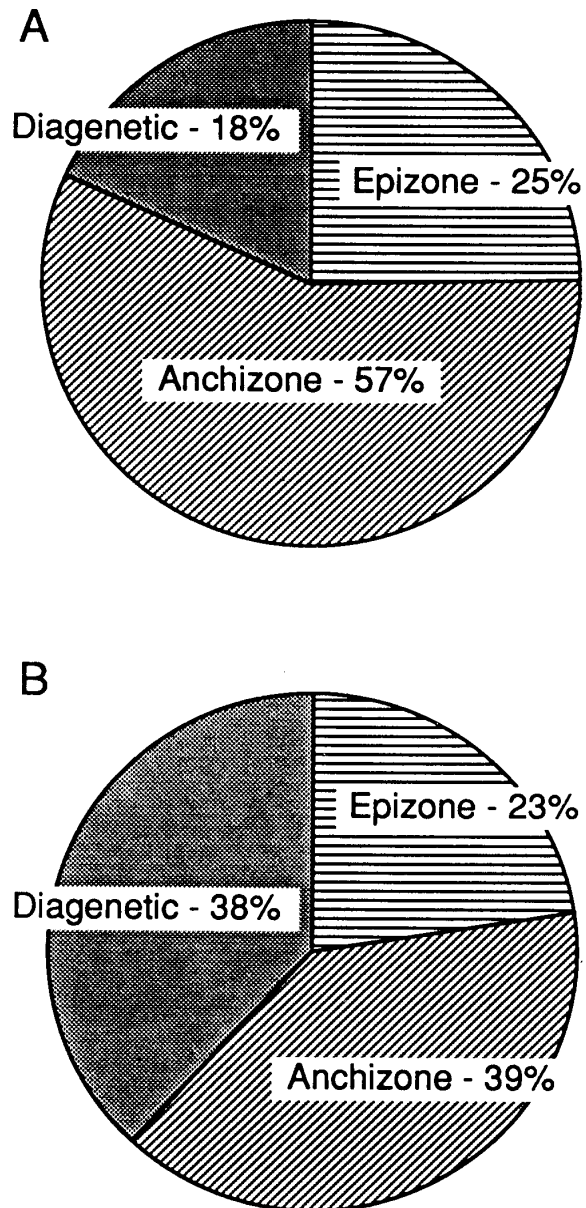

Figure 2. Pie diagrams showing relative proportions of measured illite crystallinity values in terms of diagenetic level (>0.40°2θ), anchizone (0.40-0.21°2θ) and epizone (<0.21°2θ) for the <2µm (A) and <0.5µm (B) size fractions.

The Upper Carboniferous sequences are of diagenetic to anchizone levels of alteration, while the Devonian sequences are anchizone to epizone. Within the Upper Carboniferous, however, the oldest strata in the Bideford Formation, to the southwest of Westward Ho!, have yielded the lowest crystallinity values of near 0.80°2θ. A very clear and gradual increase in grade is observed in the transition from the Upper Carboniferous through the Pilton Shales into the Devonian of the north Devon region (Fig. 3A). Kelm (1986) showed a detailed cross section with a steady increase in crystallinity from > 0.4 to 0.20°2θ in a south-north traverse along the western coastal region.

Where the metamorphic map is based on the < 2µm fraction (Fig. 3B) the same general features are seen but the main difference is in the greater extent of the anchizone areas and correspondingly smaller areas of diagenetic level.
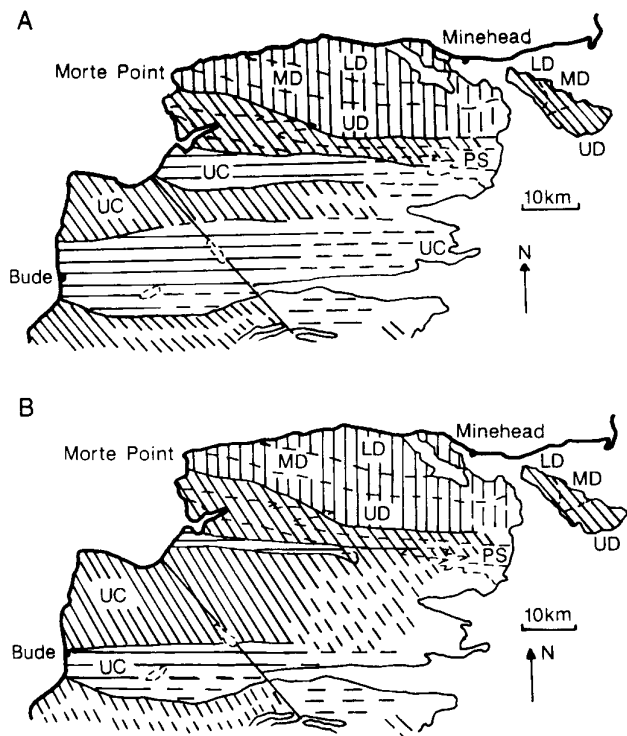
A

B

Figure 3. Metamorphic maps of north Devon and west Somerset based on illite crystallinity values. (A) Based on <0.5μm clay fraction values; (B) based on <2μm fraction values. Horizontal ornamentation - diagenetic zone; diagonal ornamentation - anchizone; vertical ornamentation - epizone. LD, MD, UD - Lower, Middle and Upper Devonian respectively; PS - Pilton Shale; LC, UC - Lower and Upper Carboniferous respectively.
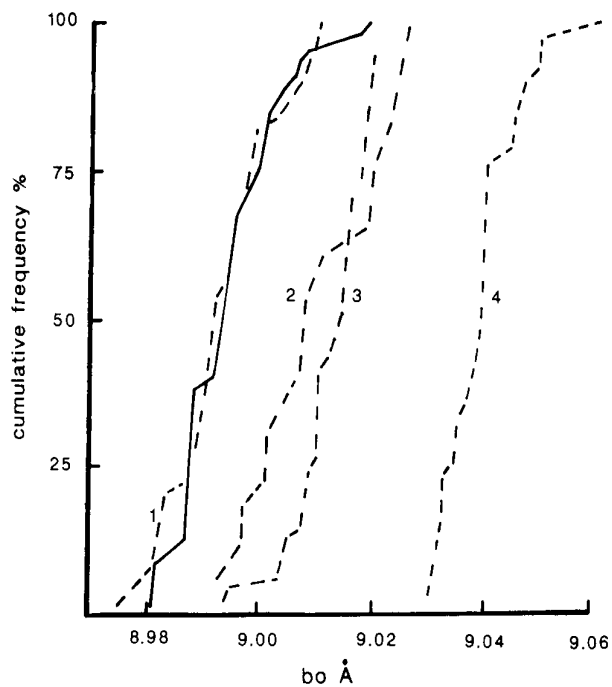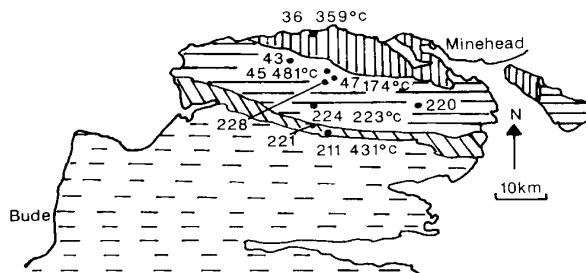

Figure 5. Map of Devonian and Pilton Shale sequences, showing location of samples used in the stable isotope analysis and the sample numbers and calculated temperatures. Vertical and horizontal ornamentation - Lower and Mid Devonian respectively; diagonal ornamentation - Pilton Shale; dashed ornamentation - Upper Carboniferous.

In detail there are discrepancies between the grade and stratigraphic level, for example the lowest crystallinity values in the Bude area do not correspond to the youngest stratigraphic level (Westphalian C) near Duckpool. The deformation style in this area is of open chevron folds and the cleavage is spaced at cm to dm intervals. To the north grade increases towards the high strain zone of Hartland (Hancock 1973), before decreasing to the area of Westphalian outcrop of the Bideford Formation. In the Devonian, the epizone level of metamorphism is associated with a narrowing of the cleavage spacing, and there is the noticeable difference with the anchizone level of metamorphism recorded in the Devonian of the Quantock Hills to the east compared with the main Devonian outcrop to the west (Fig. 3).

b_0 parameter

Fifty-two Devonian pelitic samples have been analysed for the b_0 parameter and the results are shown in Fig. 4. Here the cumulative frequency curve of the samples shows a very close correspondence to the curve representative of the low-pressure Bosost type metamorphism (Sassi and Scolari 1974). This shows that the metamorphic character of the Variscan metamorphism in this region has been of a low pressure facies series associated with high geothermal gradients. This confirms the provisional findings of Kelm (1986) based on a smaller data set.

Stable isotope results

Oxygen isotope analysis has been undertaken on separated quartz and illitic material from nine pelitic samples of Devonian age, which were all of epizone metamorphic grade, apart from

Figure 4. Cumulative frequency curve of b_0 data. Solid line is curve for the Devonian samples; other curves are from Sassi and Scolari (1974), 1 = Bosost; 2 = North New Hampshire; 3 = Ryoke; 4 = Otago.
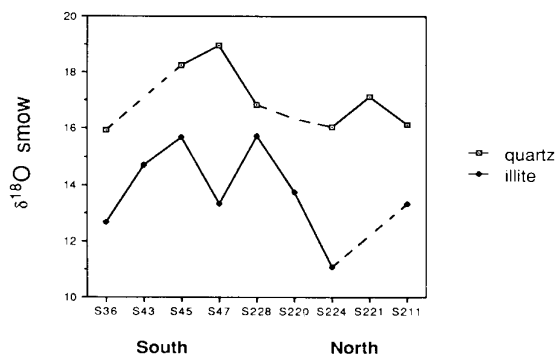

Figure 6. $\delta^{18}O$ values of quartz and illite in north to south traverse across Exmoor Forest. Sample locations shown in Fig. 5. Dashed portions indicate that no analysis is available for the intervening sample (see Table 1).

Table 1. Oxygen isotope data.

Sample	quartz(‰)	illite(‰)	Δ quartz-illite (‰)	°C	grade
36	15.95+	12.67	3.28	359	epizone
43	-	14.68	-	-	"
45	18.26	15.68	2.58	481	"
47	18.97+	13.31	5.66	174	"
228	16.83	15.71	1.12	1792	
220	-	13.75	-	-	"
224	16.05	11.30	4.75	223	"
224*	-	10.84	5.21	(196)	"
221	17.15	-	-	-	"
211	16.14	13.32	2.82	4.31	anchizone

+ quartz samples with < 2μm size, all other quartz separates were <1μm

* repeat analysis on illite

all illite material was of a <0.5μm size fraction

sample 211 of anchizone grade (Fig. 5). The quartz $\delta^{18}O$ values range from 15.95-18.97‰, which is similar to the range recorded in other anchizone-greenschist pelitic samples (Weaver *et al.* 1984; Primmer 1985a, 1985b). The absence of low values (< 10‰) suggests that detrital material of a crystalline origin has not been a major component in the separated quartz fractions.

The illite separates show a range of 10.84-15.71‰ which is also similar to the ranges seen in other anchizone to greenschist transitions (Weaver *et al.* 1984; Primmer 1985a, 1985b). No trend in $\delta^{18}O$ values is apparent in the south to north sequence of samples (Fig. 6) representing the anchizone to greenschist gradation. In all the samples quartz values are always more positive than the illite material (Fig. 6) suggesting that wholescale recrystallization of both phases has occurred in these samples so that an approach to equilibrium has developed.

Isotopic temperatures have been calculated using the mineral pair quartz-illite and temperatures are given in Table 1 and Fig. 5. The quartz-illite temperatures were calculated from the fractionation data of Eslinger and Savin (1973) which gives the equation:

$$1000 \ln\alpha_{\text{quartz-illite}} = 0.95 \, (10^6 \, T^{-2}) + 0.88$$

As can be seen from Table 1, apart from a totally unrealistic value of 1792°C, a wide range of temperatures from 174-481°C is apparent. The variability in these temperatures shows that isotopic equilibrium if attained has not been preserved in the samples. Temperature estimates for the anchizone/epizone boundary vary from an approximate level of 300-350°C to the value of 360°C suggested from isotope geothermometry by Weaver *et al.* (1984). The two values of 431°C (211) and 481°C (45) thus appear too high for epizone slates in the absence of any higher grade phases such as biotite or chloritoid. Sample 36 from the Lower Devonian in the north of the traverse (Fig. 5) would appear to be the only realistic temperature for this epizone area of metamorphism. Samples 244 and 47 with temperatures of 223°C and 174°C are too low and would appear to be more compatible with a low anchizone/high diagenetic level of metamorphism. Sample 224 is from the entrance of an abandoned Cu-Fe mine works at Heasley Mill and it is possible that the mineralisation process which was later than the cleavage may have lead to the re-adjustment of isotopic values. Sample 211 from the Pilton Shales is of anchizone grade and clearly the high temperature is incompatible with the anchizone character. This anamolous result may be related to an ^{18}O determination on an unseparable mixture of illite and paragonite in this sample.

Discussion

In the Devonian strata, including the Pilton shale the increase in illite crystallinity is accompanied by an increasingly closely spaced slatey cleavage. There is no definable variation in grade either in IC values or in mineralogy (Kelm 1988) across the stratigraphic divisions of the Devonian, apart from the anchizone level of the Middle and Upper Devonian rocks of the Quantock Hills (Fig. 3). The Carboniferous succession is of a lower grade than the Devonian, but in detail there is discordance with the stratigraphic level. The Carboniferous Bideford Formation, near Westward Ho!, is the least tectonically disturbed and has the lowest crystallinity values. Upper Carboniferous rocks around Hartland Point, where there is local overturning, have an anchizonal grade.

Cornford *et al.* (1987) have also identified the Bideford Formation in the area of Westward Ho! as having a lower thermal maturation level, based on vitrinite reflectance data, compared to the Crackington and Bude Formation to the south. They postulated that the two areas had suffered different burial histories and suggested that the area of low maturation was tectonically distinct forming the so-called "Westward Ho! nappe". The area designated as the Westward Ho! nappe by Cornford *et al.* (1987) closely matches the area of diagenetic grade identified here in the coastal areas of Bideford Formation outcrop (Fig. 3). A review by Kisch (1987) of vitrinite reflectance values associated with the limit between the diagenetic and anchimetamorphic zones shows that they are in the range 2.3-3.1 %This matches closely with the present interpretation of a high diagenetic level in the Bideford Formation and the reflectance range of 1.5-2.5% recorded by Cornford *et al.* (1987). The estimate by Cornford *et al.* (1987) of a c. 5km depth of burial and temperatures in the order of 220°C for this formation closely relates to the clay data indicative of a high diagenetic level, where temperatures were probably in the range 150-200°C. This suggests a relatively high thermal gradient and agrees with the estimate of Cornford *et al.* (1987) of a geothermal gradient of some 40°C km^{-1}.

Cornford *et al.* (1987) have shown that the Bideford and Crackington Formations underwent thermal maturation related to depth of burial which thus occurred prior to folding and uplift. As the estimates of mineralogical maturity and temperature closely correlate with those of the vitrinite data it would appear therefore that the mineralogical maturation must also be an effect due to burial.

The Devonian sequences, from b_0 data, and the Carboniferous sequences, from vitrinite data, show evidence of a high geothermal gradient being responsible for the metamorphic alteration. The mechanism by which this high thermal gradient developed and affected both the Devonian and Carboniferous successions remains problematic. Robinson (1987) and Robinson and Bevins (1989) have, however, postulated that low grades of metamorphism may develop in extensional settings, where a high heat flow is generated in response to lithospheric thinning. While this model could well be applicable to the Carboniferous, the lack of metamorphic variation in the Devonian and the association with a well developed cleavage points to other factors, such as microtectonic or stress/strain, being relevant in the control of metamorphic development.

The Devonian rocks of the region have undergone major recrystallization as shown by the strong alignment of the phases into the closely spaced cleavage. This recrystallization appears to have generated an approach to isotopic equilibrium between minerals and fluid, as samples have $\delta^{18}O$ quartz values consistently higher than co-existing illite. However the wide range in calculated isotopic temperatures shows that any equilibrium relationships must have been reset, presumably subsequent to the peak of metamorphism. In similar low grade slatey rocks from North Wales there is much evidence of post metamorphic alteration of isotopic relationships as a result of fluid/rock interaction subsequent to the peak of metamorphism (Bottrell, pers comm. and Robinson, unpublished data). There the fluid/rock interaction has been such that lower isotopic temperatures have been generated suggesting interaction with a fluid having an isotopic

composition lighter than during the metamorphism. In the rocks studied here there are temperatures that are obviously both higher and lower than can be realistically expected. This implies that if fluid/rock interaction was subsequent to the metamorphic peak, there has been more than one episode of fluid/rock interaction and that fluids of variable isotopic compositions, possibly related to the mineralisation in the area, have been involved. The data set here is rather limited as a result of variation in mineralogy and tectonic fabric of the analysed samples, and confirmation of these interpretations should ideally be sought by further analysis to produce a larger and more uniform data.

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