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COAL RANK VERSUS ILLITE CRYSTALLINITY AND ESTIMATED P-T CONDITIONS: SOME PROBLEMS CONCERNING THE PINDOS, TRIPOLITZA AND PHYLLITE-QUARTZITE SERIES IN CRETE

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ABSTRACT

Different techniques such as illite crystallinity, coal rank and calculated multivariant equilibria have been applied to characterize siliciclastic rocks of the Pindos Series, the Tripolitza Series and the Phyllite-Quartzite Series of the nappe pile in Crete.

For the Pindos flysch and the Tripolitza flysch, it is possible to distinguish two diagenetic/metamorphic events. Detrital illitic material and resedimented organic particles, probably derived from more internal parts of the Hellenides, reflect an older pre-neohellenic anchimetamorphism. In contrast, reflections of "in situ" carbonized vitrinite correspond to the final neohellenic overprint of diagenetic to anchimetamorphic grade.

The Phyllite-Quartzite Series was affected by high-pressure/low-temperature metamorphism. This is evident from calculated mineral equilibria. However, the coal ranks of this unit are much to low with respect to the estimated temperature. Apparently, coalification is retarded because of rapid burial and uplift.

Surprisingly, the same coal rank characteristics have been observed in the Ravdoucha Beds. Hence, a high-pressure character of this unit can also be inferred. This is supported by finding of high-pressure diagnostic minerals and phengite geobarometry.

In conclusion, not only the Phyllite-Quartzite Series but also the Ravdoucha Beds represent supracrustal rocks buried to significant depth in contrast to the Tripolitza Limestone and overlying units.

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INTRODUCTION

Illite crystallinity and coal rank usually correlate well in rocks subjected to burial diagenesis (e.g. TEICHMÜLLER et al. 1979), anchimetamorphism (FREY 1987) and telemagmatic activity (e.g. BRAUCKMANN 1984). KÜRMANN & RICHTER (1989) proposed a generalized correlation of these two parameters which furthermore allows estimate of temperatures during diagenesis and low-grade metamorphism. Coal rank of 2.7% Rm and illite crystallinity of 222 Hb_{rel} mark the boundary between diagenesis and anchizone, which corresponds to about 200°C. The boundary between anchizone and epizone (5.5% Rm, 125 Hb_{rel}) corresponds to a temperature of about 300°C (LUDWIG 1972, BREITSCHMID 1982, KISCH 1983, FREY 1986).

We determined relevant data for the Pindos Series, Tripolitza Series and Phyllite-Quartzite Series of the nappe pile in Crete. However, temperature estimates based on illite crystallinity and coal rank subdivisions show serious deviations from correlations found elsewhere. As will be show below, these deviations are caused partly by detrital clay minerals, partly by retarded coalification. The latter point can give an indication for the baric type of metamorphic overprint.

GEOLOGICAL SETTING

The island of Crete is part of the external Hellenides. Late Alpine nappe structures (cf. Fig. 6) and subsequent horst and graben tectonics are the dominant geologic features. A recent review of the geology of Crete is presented by SEIDEL & WACHENDORF (1986).

The autochthonous (?) base of the Cretean nappe pile is formed by the Plattenkalk Series. Its stratigraphic age ranges from Permian to Oligocene (EPTING et al. 1972, FYTROLAKIS 1972). The Plattenkalk Series mainly consists of cherty marbles and some siliciclastics at the base (Sisses Beds), and the top (Kalavros Beds). The grade of metamorphism has been estimated on metabauxites occurring in the northern part of Central Crete. Mineral assemblages with magnesiocarpholite, chloritoid, pyrophyllite and diaspore suggest high-pressure/low-temperature metamorphism at about 350°C, 10 kbar (SEIDEL 1978 and 1983, THEYE et al. 1992). Metamorphism was accompanied by large-scale folding.

The Phyllite-Quartzite Series represents the lowermost unequivocally allochthonous nappe unit. It comprises metamorphic siliciclastics (partly in form of different types of gravity flows), limestones, evaporites and volcanics. Biostratigraphic records range from Upper Carboniferous to Upper Triassic (KRAHL et al. 1983, 1986). The age of metamorphism of the Phyllite-Quartzite Series is Miocene as [illegible] Diagnostic minerals such as Na-pyroxene, blue amphibole, lawsonite and aragonite give evidence of a high-pressure/low-temperature type of the metamorphism (SEIDEL 1978, THEYE 1988). Intensive deformation is ubiquitous, ranging from ductile (up to three generations) to semibrittle and brittle.

The nappe overlying the Phyllite-Quartzite Series is the diagenetic to anchimetamorphic Tripolitza Series. It is composed of the Tripolitza limestone ranging from Upper Triassic to Eocene and flysch sediments of Eocene to Lower Oligocene age (WACHENDORF et al. 1980). Upper Triassic siliciclastics, the Ravdoucha Beds (SANNEMANN & SEIDEL, 1976), occur at the base of this series and are regarded as the underlying sedimentary stratum of the Tripolitza limestone (KOPP & OTT 1977). Deformation of the Ravdoucha Beds was ductile (up to two generations) and finally brittle.

The third allochthonous unit of the Cretean nappe pile is the Pindos Series consisting of Halobien beds (U. Triassic), cherty limestones (Jurassic-Cretaceous), a first flysch sequence of Cenomanian age and a second flysch sequence of Lower Tertiary age (SEIDEL 1968, 1971). The Pindos Series is non-metamorphic. Only one generation of ductile deformation was developed until brittle deformation took place.

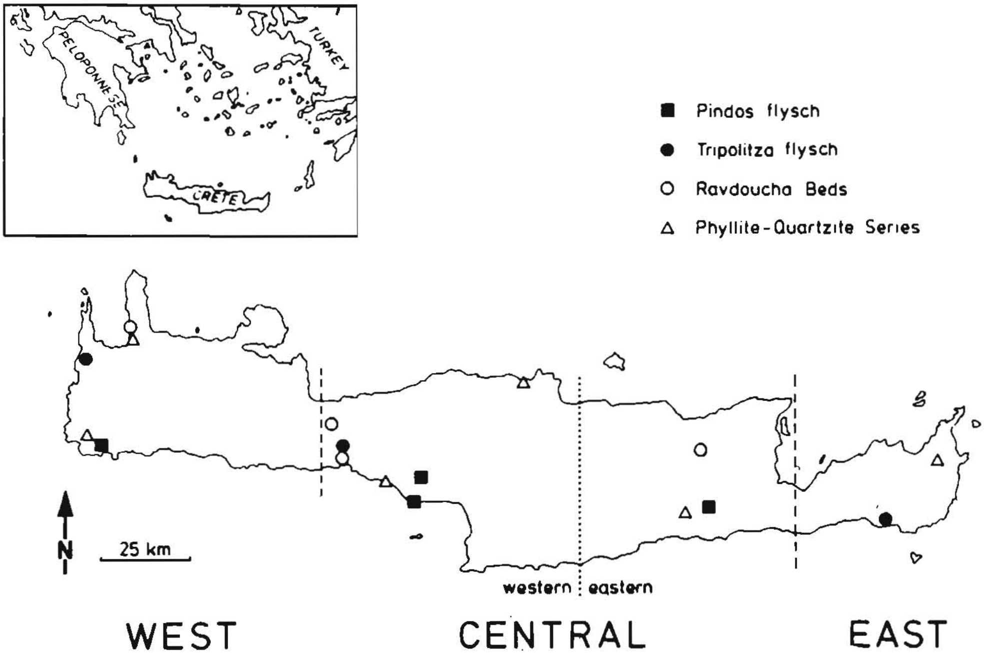

Figure 1: Sketch map of Crete with sample locations (cf. Table 1).

The Uppermost Unit is a composite nappe comprising ophiolites as well as slices of various metamorphic, igneous and sedimentary rocks.
We focussed our examinations on the siliciclastic parts of the Pindos, Tripolitza and Phyllite-Quartzite Series (flysch and pelites/metapelites) because of the abundance of organic particles and of phyllosilicates as well. The sample localities cover western, central and eastern Crete so that the significance of the data can be evaluated on a regional scale (Fig. 1).

METHODS

The illite crystallinity was determined on ethyleneglycolated sedimentation samples after WEBER (1972): fraction <2 µm; density of clay layer 1.56 mg/cm^2; 4.26 Å - quartz peak as external standard with 3.2 mm half height widths; Philips PW 1050/25 diffractometer, Cu Kα_1 radiation, 40 kV, 35 mA, AMR-monochromator.
Coal rank was determined after TING & LO (1978) on a Leitz microscope-photometer MPV2.
Metamorphic pressure and temperature conditions of the Phyllite-Quartzite Series were estimated on the basis of calculated mineral equilibria involving minerals such as Mg-carpholite, Mg-chloritoid, sudoite, clinochlore, pyrophyllite, and quartz. Calculations were done applying the Ge0-Calc software (BROWN et al. 1988) and thermodynamic data of BERMAN (1988) and VIDAL et al. (1992). Because of the limited availability of thermodynamic data, the calculations were restricted to chemical system MgO-Al_2O_3-SiO_2-H_2O. Ideal activity models were used to consider actual mineral composition, as determined by electron microprobe. Representative mineral analyses used for these calculations are presented in THEYE et al. (1992).
For the sample locations, see Table 1.

Table 1: Sample locations

	Western	Central		Eastern
		western	eastern	
Pindos flysch	Paleochora	Kap Melissa SE Asiderotas	S Lasithi Sikologos	
Tripolitza flysch	Platanos	Sellia		Pilalimata
Ravdoucha Beds	Ravdoucha	Sellia Asi Gonia	Kera	
Phyllite-Quartzite Series	Nopigia Ag. Theodori	Kerames N Fodele	Vianos	Rousa Eklisia

RESULTS

Flysch sediments

The illite crystallinity of the Pindos and of the Tripolitza flysch ranges between 215 and 242 Hb_{rel}, suggesting conditions close to the diagenesis/anchizone boundary (Fig. 2, Table 2a and 2b). The coal ranks, however, show considerable scattering and partly significant deviations from the correlation curve of KÜRMANN & RICHTER (1989).

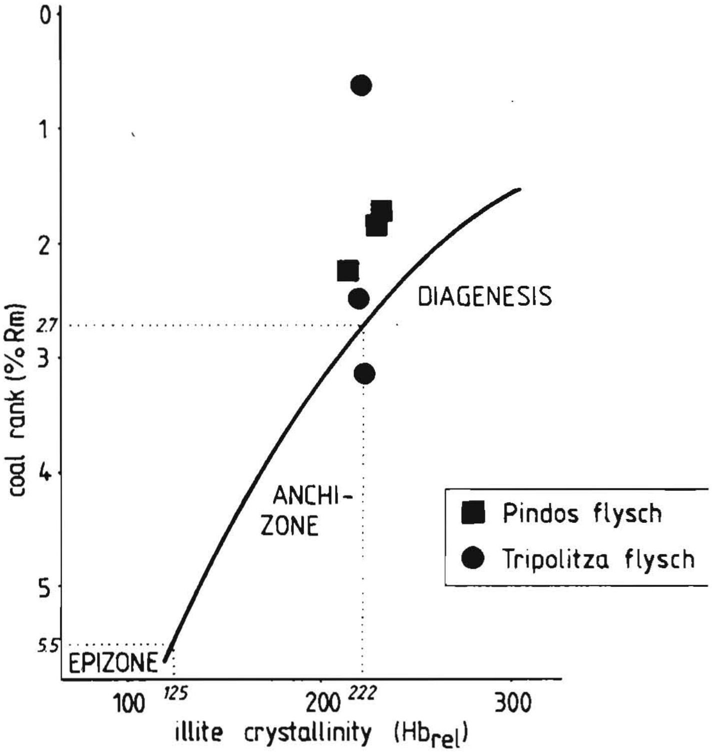

Figure 2: Illite crystallinity vs. coal rank (only "in situ" carbonized particles) diagram for the Pindos flysch and Tripolitza flysch. Correlation curve after KÜRMANN & RICHTER (1989).

A more detailed examination of organic particles reveals that scattering of coal ranks is regular with a more or less bimodal frequency distribution (Fig. 3). This observation suggests the presence of "resedimented" organic particles (2-3 % Rm) as well as "in situ" carbonized particles (0.6-3.2 % Rm). The distinction of two vitrinite generations is possible especially for the Pindos flysch of central end eastern Crete and for the Tripolitza flysch of eastern Crete because in these cases vitrinite reflectance of "in situ" carbonized particles is clearly lower than those of the "resedimented" particles (Table 2a and 2b). The

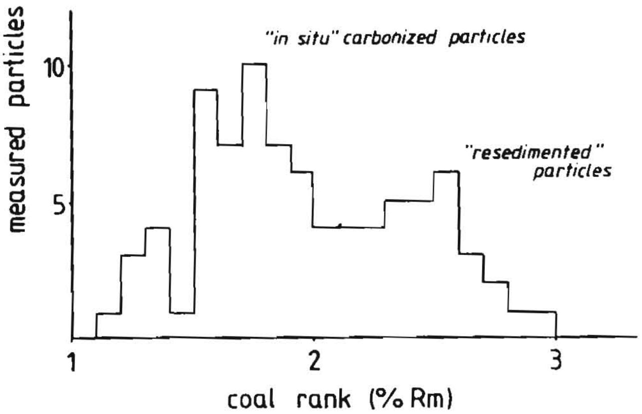

Figure 3: Typical bimodal distribution of organic particle reflectance in the Pindos flysch. 6 specimens from Central Crete.

"resedimented" particles could be derived from source rocks that underwent an older anchimetamorphism, possibly in more internal terrains of the Hellenides. Therefore, only the "in situ" carbonized particles reflect the final diagenesis/anchimetamorphism that affected the flysch series as wholes. In line with the reasoning above, the illite crystallinity is also assumed to be governed by detrital illite/muscovite that can be related to the "resedimented" organic particles. Accordingly, measured illite crystallinity can not be attributed to the final overprint. Some examples of specimens on which it was possible to determine both the reflectance of "in situ" carbonized particles and illite crystallinity are given in Table 2c.

The same interpretation was given by KISCH (1981) for the Pindos flysch and for the westhellenic flysch in the Greek mainland. Very poor vitrinite reflectance ("Glanzbraunkohle", 0.44-0.65% Rm) does not correspond to "anchizonal peak width of illite" (0.26-0.35°2θ). KISCH (l.c.) also attributed this observation to the presence of detrital clay minerals.

In summary, only "in situ" carbonized particles give reliable information on the thermal overprint in the Pindos and Tripolitza flysch. Accordingly, the Pindos flysch shows only a diagenetic imprint, without any significant regional variation. In contrast, the Tripolitza flysch underwent slightly higher temperatures corresponding to diagenetic to anchimetamorphic grade. Regional variation of coal ranks of "in situ" carbonized particles in the Tripolitza flysch suggests increasing temperatures from eastern (diagenetic) to western Crete (anchizonal). An estimate of pressure was not possible.

Table 2a: Coal rank and illite crystallinity data of the Pindos flysch in Crete. n refers to measured samples; for half height width of white K-mica (Hb_{rel}) the minimum, average and maximum value are given.

	coal rank (%) "in situ"				coal rank (%) resedimented	illite crystallinity (Hb_{rel})	
	n	Rmin	Rm	Rmax	Rm	n	
western Crete	5	1.9	2.2	2.5		6	191-215-263
central Crete	6	1.6	1.7	2.0	2-3	9	227-241-252
eastern Crete	4	1.6	1.8	2.0	2-3	5	210-242-285

Table 2b: Coal rank and illite crystallinity data of the Tripolitza flysch in Crete.

	coal rank (%) "in situ"				coal rank (%) resedimented	illite crystallinity (Hb_{rel})	
	n	Rmin	Rm	Rmax	Rm	n	
western Crete	4	2.9	3.2	3.6		9	193-222-254
central Crete	4	2.4	2.6	2.8		6	186-218-243
eastern Crete	3	-	0.6	-	2-3	7	193-220-234

Table 2c: Examples of specimens with measured coal rank of "in situ" carbonized particles as well as illite crystallinity.

		coal rank (% Rm) "in situ"	illite crystallinity (Hb_{rel})
Pindos flysch			
central Crete:	Mf11	2.00	242
	Mf12	1.60	252
	Mf14	1.80	231
	Mf15	1.75	227
	Mf16	1.70	243
	Mf17	1.80	240
eastern Crete:	Ef1	1.60	260
	Ef2	1.75	229
	Ef3	2.00	227
	Ef5	1.95	210
Tripolitza flysch			
eastern Crete:	Ch9	0.65	227
	Ch10	0.55	218
	Ch13	0.75	193

Phyllite-Quartzite Series and Ravdoucha Beds

Pressure-temperature conditions of the high-pressure metamorphic Phyllite-Quartzite Series can be estimated by calculating relevant mineral equilibrium curves based on the actual mineral compositions (Fig. 4). In eastern Crete, mineral compositions of metapelites with the parageneses quartz-pyrophyllite-chlorite-carpholite and quartz-pyrophyllite-chlorite-sudoite can be used to calculate the equilibria

(1) sudoite + quartz = chlorite + pyrophyllite + H_2O

(2) chlorite + pyrophyllite + H_2O = carpholite + quartz.

The resulting intersection of these curves define P-T conditions of about 300°C and 8 kbar. Estimated uncertainties are in the range of ± 40°C, ± 3 kbar. In central Crete, chloritoid-carpholite-bearing schists can be related to the simple reaction

(3) carpholite = chloritoid + quartz + H_2O

which indicates a temperature of about 350°C. A minimum pressure of about 8 kbar can be derived from the lack of pyrophyllite + chlorite and the presence of carpholite according to reaction (2).
Chloritoid-carpholite schists also occur in western Crete, but with more Mg-rich mineral compositions. Corresponding temperatures are about 400°C. The pressure of metamorphism can be constrained by the presence of aragonite and, as in central Crete, by the lack of pyrophyllite + chlorite to a minimum of about 10 kbar.

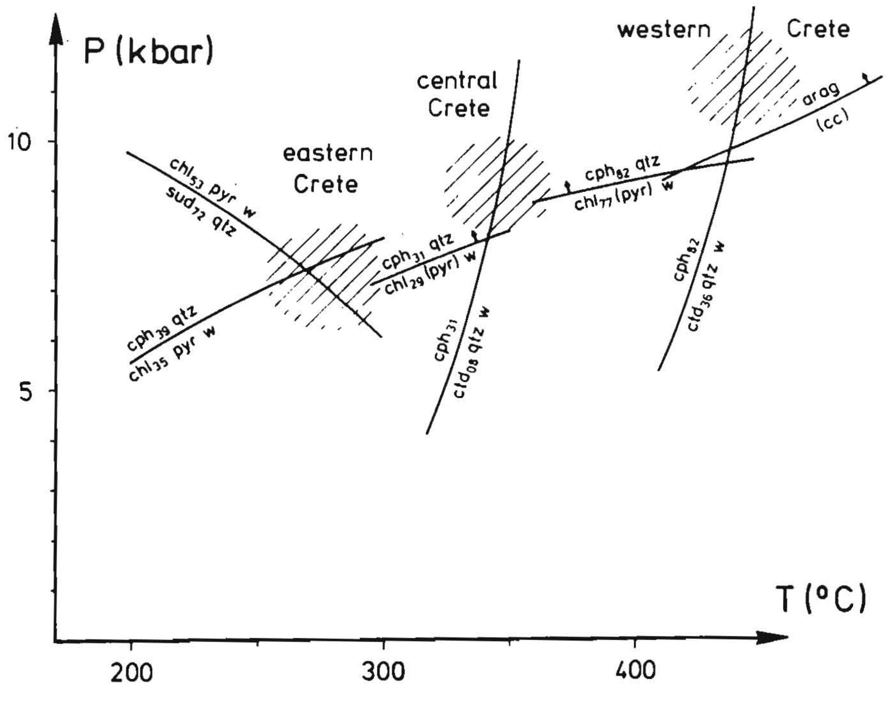

Figure 4: Calculated multivariant equilibria and resulting pressure-temperature conditions for the Phyllite-Quartzite Series of Crete. Abbreviations: arag=aragonite, cc=calcite, chl=chlorite, cph=carpholite, ctd=chloritoid, qtz=quartz, pyr=pyrophyllite, sud=sudoite, w=H_2O. Mineral names with figures refer to measured 100 Mg/(Mg+Fe^{2+}). Mineral names in brackets are absent in the measured specimens. Accordingly, these curves define minimum conditions. Reaction curve calcite = aragonite after JOHANNES & PUHAN (1971).

Table 3a: Coal rank data and estimated pressure-temperature conditions of the Ravdoucha Beds in Crete.

	T (°C)	P (kbar)	n	coal rank (%) Rmin	Rm	Rmax
western Crete	300	>3-4	8	2.4	3.2	4.0
central Crete						
(west. part)	200-300		2	2.1	2.3	2.6
(east. part)	200-300		1	1.7	2.1	2.6
eastern Crete	-	-	-	-	-	-

Table 3b: Coal rank data and estimated pressure-temperature conditions of the Phyllite-Quartzite Series in Crete.

	T (°C)	P (kbar)	coal rank (%) n	Rm
western Crete	400 40	>10	5	2.5-4.0
central Crete				
(west. part)	350 40	> 8		
(east. part)	"	"		
eastern Crete	300 40	8 3		

Measured coal ranks of carbonized particles in metapelites and aragonite marbles from western Crete are rather poor (2.5-4.0 % Rm) and in complete disagreement with temperatures estimated from mineral equilibria (Fig. 5 and Table 3b; no data are available for eastern and central Crete). This effect can be explained by retarded coalification because of the relatively high pressure/temperature ratio of the metamorphic conditions resulting from rapid burial and uplift (preservation of aragonite!).

Another example confirming this interpretation is the Jurassic/Cretaceous "Bündner-Schiefer" in the Engadine window of the Alps. Metamorphism took place under high-pressure/low-temperature conditions (300-350°C, 6-7 kbar) as evident from illite crystallinity, vitrinite reflectance, occurrence of lawsonite, phengite geobarometry and fluid inclusions study (STÖCKHERT et al. 1990). However, our new measurements show that coal ranks are much lower than expected (1.7-2.9 % Rm; Fig. 5). Hence, coal ranks of the "Bündner-Schiefer" are also not suitable to estimate temperatures but to indicate the baric type of overprint. Similar observations on other high-pressure metamorphic rocks have been made by DIESSEL et al. (1978), GOFFE & VILLEY (1984), DOMINE (1989), and have also been discussed in BARKER (1989).

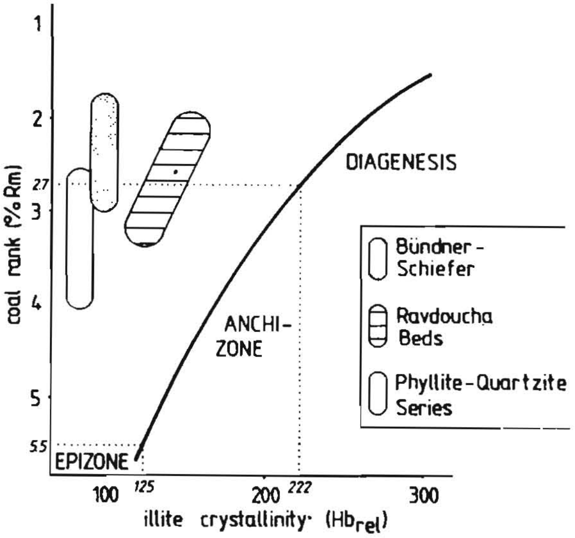

Figure 5: Illite crystallinity vs. coal rank diagram for the Phyllite-Quartzite Series, Ravdoucha Beds and "Bündner-Schiefer". White K-micas of the Phyllite-Quartzite Series and "Bündner-Schiefer" are well crystallized muscovite/phengite. Correlation curve after KÜRMANN & RICHTER (1989).

Surprisingly, a similar behaviour has also been found in the Ravdoucha Beds: illite crystallinity indicates anchi-/epimetamorphic conditions (128-155 Hb_{rel}), but coal rank is of diagenetic to anchimetamorphic grade (2.1-3.2 % Rm) (Fig. 5, Table 3a). According to the reasoning above, the Ravdoucha Beds should also have experienced high-pressure conditions. This is supported by phengite geobarometry on metapelites of Ravdoucha Beds in western and central Crete suggesting a pressure of at least 3-4 kbar (method after MASSONNE & SCHREYER 1987). Furthermore, the high-pressure mineral ferrocarpholite has been found in Ravdoucha Beds near Asi Gonia (location: see KOPP & OTT 1977). The main rock type of this outcrop is a chlorite-pyrophyllite schist with abundant carpholite segregations. This mineral association indicates a metamorphic grade similar to the Phyllite-Quartzite Series of eastern Crete , i.e. pressure of at least 6 kbar and about 300 °C. In contrast, the Phyllite-Quartzite Series of the same location contains chloritoid but no carpholite and no chlorite + pyrophyllite suggesting higher temperature. High-pressure records are also known in the Peloponnese. From the Tyros Beds (KTENAS 1924) which occupy the same tectonic position as the Ravdoucha Beds in Crete, the high-pressure mineral lawsonite has been described as well as Si-rich phengite (PE-PIPER 1983, BALTATZIS & KATAGAS 1984). In summary, our results suggest high-pressure metamorphism of at least parts of the Ravdoucha Beds, based on retarded coalification of organic matter, phengite geobarometry and diagnostic mineral assemblages.

CONCLUSION

Combining different methods such as illite crystallinity, coal rank, phengite geobarometry and calculation of mineral equilibria from thermodynamic data, diagenetic/metamorphic conditions can be reconstructed to an extend which cannot be achieved by any single method alone.

Thus, in the Pindos flysch and Tripolitza flysch two generations of diagenesis/anchimetamorphism can be distinguished. "Resedimented" organic particles and detrital clay minerals reflect an older pre-neohellenic thermal overprint probably of internal parts of the Hellenides. Only "in situ" carbonized particles can be related to the final neohellenic diagenesis.

The high-pressure character of the Ravdoucha Beds has important geologic consequences (cf. Fig. 6). Obviously, most of the rock sequences sandwiched between the Plattenkalk Series and the Tripolitza limestones represent supracrustal rocks buried to significant depth decreasing from the base (up to 50 km in the Phyllite-Quartzite Series) to the top (about 15 km (?) in the Ravdoucha Beds). Hence, the thrust boundary separating these two rock sequences is equivalent to a low-angle normal fault omitting a considerable crustal section (cf. AVIGAD & GARFUNKEL 1991).

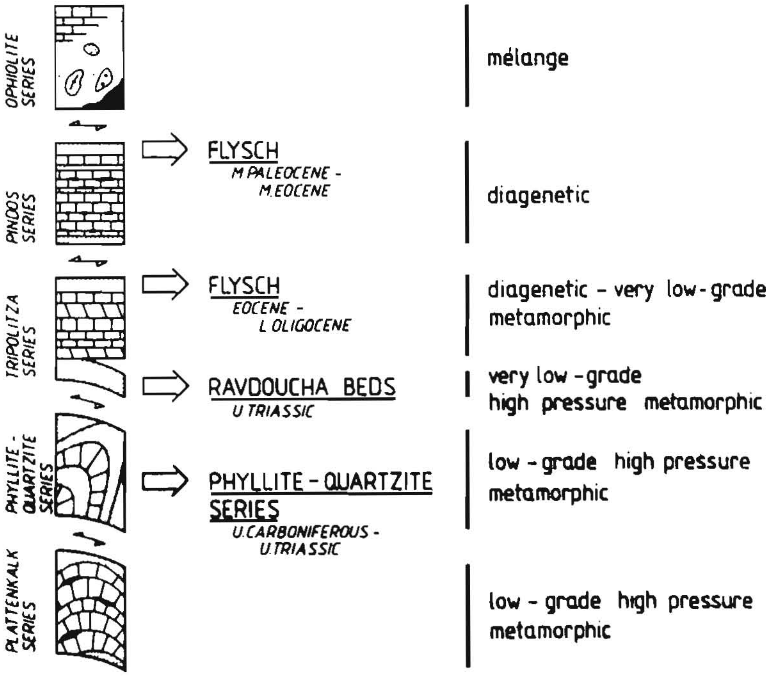

Figure 6: Sketch of the nappe pile in Crete and types of diagenesis metamorphism. Investigated rock sequences are indicated by arrows.

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