

# Very low-grade metamorphism of the Alps - an introduction

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## **Very low-grade metamorphism of the Alps – an introduction**

by *Martin Frey*<sup>1</sup>

### **Abstract**

Until recently, studies of very low-grade metamorphism in the Alps have been based largely based on illite “crystallinity” and index mineral data and, to a lesser extent, on coal rank and fluid inclusion data. These four methods are briefly outlined and a correlation is presented in Fig. 7. The distribution of mineral assemblages in metabasites and metagraywackes as well as the location of reaction-isograds and mineral zone boundaries with respect to anchimetamorphic areas are shown on a large-scale map of the Alps (Plate 1). In the Eastern Alps, very low-grade metamorphism is predominantly of Eo-Alpine age, while in the Central and Western Alps, it is predominantly of Meso-Alpine and Neo-Alpine age. Finally, some suggestions for future work are given.

*Keywords:* low grade metamorphism, isograds, illite crystallinity, coal rank, fluid inclusion data, metagreywackes.

### **1. Introduction**

Very low-grade metamorphism in the sense of WINKLER (1979) ranges from diagenesis to greenschist facies, an approximate temperature range of from 150–200 to 350–400 °C. In an orogenic belt, regional metamorphism is followed by uplift and erosion, yielding a clockwise pressure (P) – temperature (T) path (ENGLAND and RICHARDSON, 1977; ENGLAND and THOMPSON, 1984). Along this path there are three possible ways of passing through the P-T field of very low-grade metamorphism (Fig. 1): (A) during prograde metamorphism. In most cases, however, transformations at higher temperature will obliterate the imprint of very low-grade metamorphism; (B) during retrograde metamorphism. Such “late” overprints are important in unravelling a P-T path but will not be considered further here. (C) Maximum P and T are reached within the realm of

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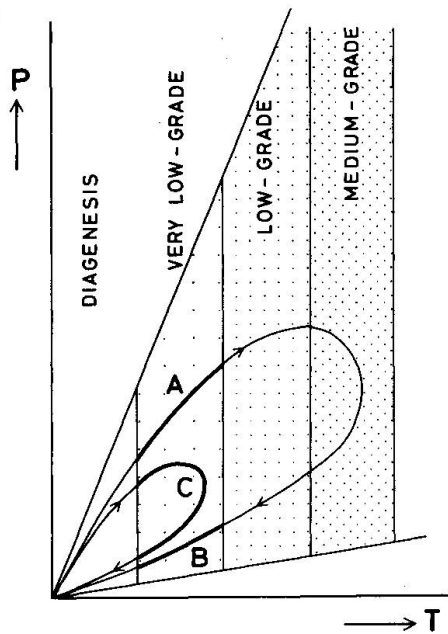


Fig. 1 Hypothetical pressure-temperature-time paths for two regionally metamorphosed rocks bodies. Transects through very low-grade metamorphism are shown in heavy lines: A, during prograde metamorphism; B, during retrograde metamorphism; C, during culmination of metamorphism. See text for discussion.

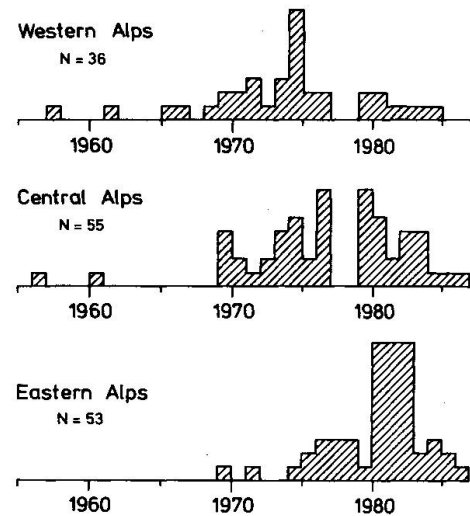


Fig. 2 Papers dealing with very low-grade metamorphism of the Alps grouped by year of publication.

very low-grade metamorphism. This paper deals exclusively with this final possibility.

In the Western and Central Alps, the study of very low-grade metamorphic rocks started some 25 years ago. However, research in this field in the Eastern Alps was initiated somewhat later (Fig. 2). While for this paper some 140 articles were consulted, only a few illustrative examples will be mentioned.

## 2. Four successful approaches

The following four methods have been successful in the investigation of very low-temperature metamorphism in the Alps: Illite "crystallinity", index minerals, coal rank, and fluid inclusion studies. The different variables of each method will be summarized, but for a detailed discussion the reader is referred to a forthcoming textbook (FREY, 1987b).

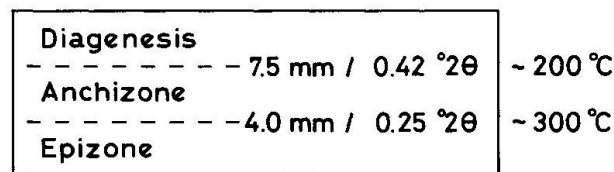
It has turned out that a combination of the various methods yields the most fruitful results, although usually only one or two methods were applied in any given study (Table 1). There exist only two studies where all of the four methods were combined (BARLIER et al., 1974; BREITSCHMID, 1982).

Tab. 1 Correlations based on the number of methods employed for a given study. IC = illite "crystallinity", IM = index minerals, R = coal rank determination by reflectivity measurements, FI = fluid inclusion studies.

<i>One method used</i>		<i>Two methods used</i>	
IM	30	IM + IC	52
IC	8	IM + FI	1
R	6	IC + FI	1
FI	4		
<i>Three methods used</i>		<i>Four methods used</i>	
IC + R + FI	1	IM + IC + R + FI	2
IM + IC + R	5	BARLIER et al. (1974)	
IM + IC + FI	2	BREITSCHMID (1982)	

2.1. ILLITE "CRYSTALLINITY"

The determination of illite "crystallinity", i.e. the peak width at half height of the first illite basal reflection on X-ray diffractograms (KÜBLER, 1967), has become most popular in the Alps. This is because the method involves little analytical expense and because illite-bearing sediments are widespread. With the aid of illite "crystallinity" data the following three zones can be distinguished with increasing metamorphic grade: diagenetic (or unmetamorphosed) zone, anchizone, epizone. Note that the "anchizone" is recognized only on the basis of illite "crystallinity" data (KISCH, 1983, p. 309). In the Alps, the anchizone corresponds approximately to the temperature range between 200 and 300°C (Fig. 3) as derived from fluid inclusion studies (see below). The term "epizone" will be used in this paper only in connection with illite "crystallinity" data. In earlier Alpine studies, epizone and greenschist facies were regarded as synonymous. However, as discussed in section 3, this usage needs some revision.



Major controlling variables:

- temperature
- lithology (porosity, pore solution composition)
- mineralogy (interfering basal reflections)
- illite chemistry
- experimental conditions (diffractometer, sample preparation)

Minor controlling variables:

- stress
- time

Fig. 3 Illite "crystallinity": division of metamorphic grade and controlling variables.

Illite “crystallinity” data have facilitated the mapping of an almost continuous zone of anchimetamorphic grade all the way from the Mediterranean Sea to Vienna (see Plate 1). In a few areas, it was possible to map lines of equal “crystallinity” or isocrysts (MERRIMAN and ROBERTS, 1985), as for example in the Northern Calcareous Alps near Innsbruck (KRUMM, 1984). In other areas, discontinuities in illite “crystallinity” were detected at nappe boundaries, with the higher tectonic unit showing higher metamorphic grade. This phenomenon was referred to as “transported metamorphism” (e.g. FREY et al., 1980a; BREITSCHMID, 1982), but “inverse metamorphic zonation” seems to be a more appropriate designation.

## 2.2. INDEX MINERALS

Very low-grade index minerals encountered in four different lithologies are given in Table 2. Investigations in the Taveyanne graywacke of Eocene-Oligocene age have produced a regular mineral zonation in the Western and Central Alps (MARTINI and VUAGNAT, 1965; MARTINI, 1968; MARTINI and VUAGNAT, 1970; SAWATZKI and VUAGNAT, 1971; MARTINI, 1972; KÜBLER et al., 1974; SAWATZKI, 1975; STALDER, 1979; KISCH, 1980; LIPPMANN and ROTHFUSS, 1980; BUSSY and EPARD, 1984), see Fig. 4. The facies series encountered is typical for an intermediate pressure regime (compare with LIU et al., 1987). In the Western Alps metabasites have yielded useful index minerals, including lawsonite and glaucophane, which are indicative of a high pressure—low temperature re-

Tab. 2 Very low-grade index minerals from the Alps

### *Metabasites*

Prehnite  
Pumpellyite  
Epidote  
Lawsonite  
Actinolite

### *Metaclastites*

Kaolinite  
Pyrophyllite  
Paragonite  
Mg-Fe-carpholite  
Chloritoid

### *Metagraywackes*

Heulandite  
Laumontite  
Prehnite  
Pumpellyite  
Epidote  
Actinolite  
Corrensite

### *Fe-rich metalimestones*

Glaucosite  
Stilpnomelane  
Biotite

Presence of Index Mineral =  $f(T, P, X_{\text{rock}}, X_{\text{fluid}}, \dots)$

gime. In comparison, metabasites and metagraywackes are extremely rare in the anchimetamorphic zone of the Eastern Alps (see Plate 1). Two comments are offered regarding pumpellyite-bearing mineral assemblages mentioned in the Plate caption. First, it is not always evident from the literature with which phases pumpellyite coexists. Secondly, these assemblages may not belong to the same Alpine regional metamorphic event and some may even have originated during ocean-floor metamorphism.

FACIES MINERAL	ZEOLITE		PREHNITE - PUMPELLYITE	PUMPELLYITE - ACTINOLITE	GREENSCHIST
	GRAYWACKES				
Heulandite	—————				
Laumontite	—————				
Prehnite		---	—————		
Pumpellyite		---	—————	—————	
Epidote			—————		—————
Actinolite				-----	—————
SHALES, SLATES					
Smectite	—————				
Illite/smectite	—————				
Illite					
Corrensite					
Chlorite					
	DIAGENESIS		ANCHIZONE	EPIZONE	

Fig. 4 Distribution of secondary minerals in the Taveyanne graywacke of the Western and Central Alps. See text for sources of data.

A few reaction-isograds have been mapped in metaclastites and Fe-rich metalimestones. The reaction-isograd kaolinite + quartz = pyrophyllite + H<sub>2</sub>O was mapped in the high-grade anchizone of the Eastern Alps (SCHRAMM, 1978) and in the low-grade anchizone of the Central Alps (FREY, 1987a; see Plate 1). The difference in metamorphic grade is ascribed to the effect of different fluid compositions. In the first example, the presence of hematite points to a water-rich fluid while in the second example organic material produced a methane-rich fluid with a water activity much less than unity (FREY, 1978). The appearance of chloritoid in metaclastites is governed by the reaction: pyrophyllite + chlorite = chloritoid + quartz + H<sub>2</sub>O, and a corresponding reaction-isograd has been mapped at the beginning of the epizone of the Eastern and Central Alps (SCHRAMM, 1978; FREY and WIELAND, 1975). It is interesting to note that chloritoid appears at considerably higher grade in the polymetamorphic basement of the Ötztal nappe. In this case early Alpine chloritoid formed by a hydration-reaction at the expense of pre-Alpine staurolite (PURTSCHELLER, 1969).

Similar observations have also been made in the Western Alps: in "wet" Mesozoic metasediments, chloritoid appears at lower metamorphic grade than in the "dry" pre-Alpine basement (J. DESMONS, pers. comm. 1986).

The following two reaction-isograds have been established in Fe-rich limestones of the Helvetic nappes: glauconite  $\pm$  chlorite + quartz = stilpnomelane + K-feldspar + H<sub>2</sub>O + O<sub>2</sub> near the beginning of the anchizone and stilpnomelane + chlorite + K-feldspar = biotite + H<sub>2</sub>O near the anchizone—epizone boundary (FREY et al., 1973; BREITSCHMID, 1982; FREY, 1987c), see Plate 1.

### 2.3. COAL RANK

In the Alps, the determination of coal rank through reflectivity measurements on phytoclasts has proved to be a reliable method of determining very low-grade metamorphic conditions (e.g. BARLIER et al., 1974; KÜBLER et al., 1979; STALDER, 1979 in the Western Alps; KÜBLER et al., 1979; STALDER, 1979; FREY et al., 1980a; KISCH, 1980; DORTMANN, 1982; GROSHONG et al., 1984 in the Central Alps; EGGERT et al., 1976; TEICHMÜLLER and TEICHMÜLLER, 1975, 1978; HUFNAGEL et al., 1981 in the Eastern Alps). In the diagenetic realm and at the beginning of very low-grade metamorphism, coal rank from sub-bituminous coal to anthracite (Fig. 5) is a more reliable method than illite "crystallinity" measurements.

Just a few interesting results will be mentioned here. DORTMANN (1982) determined an increase in coal rank with depth within the 2 km thick Niesen nappe from 3% to 5% maximum reflectivity. In the Vorderriss 1 borehole through the Northern Calcareous Alps (see Plate 1, No 12, for location) the intermediate Lechtal nappe I showed mean reflectivity (R<sub>m</sub>) values of 1.5–2.0 while the Lechtal nappe II above and the Allgäu nappe below showed considerably lower coal ranks with 0.4–1.1% R<sub>m</sub> (HUFNAGEL et al., 1981). Obviously, the Lechtal nappe I was exposed to higher temperatures before it was tectonically emplaced between the other two nappes.

Besides coal rank determination, a few other methods to determine the maturation of organic material have been applied in the Alps. GAUPP and BATTEN (1985) established a gradient in metamorphic grade based on colour changes of palynomorphs in Cretaceous pelitic rocks in the western part of the Eastern Alps. Conodont colour changes (e.g. EPSTEIN et al., 1977), on the other hand, has seen very little application in the Alps, but is of potential use in Triassic and Paleozoic limestones of the Eastern Alps.

RANK OF COAL		VITRINITE REFLECTANCE
Germany	USA	
Fettkohle	medium volatile bituminous	1.5 % Rm
Esskohle	low volatile bituminous	
Magerkohle	semi-anthracite	1.9 % Rm
Anthrazit	anthracite	2.5 % Rm 2.8 % Rmax
Meta-Anthrazit		4.0 % Rmax
Semigraphit	meta-anthracite	6.0 % Rmax
Graphit	graphite	< 2 % Rmin

Major controlling variables:

- temperature

Minor controlling variables:

- time
- stress
- lithology

Fig. 5 Coal rank: division of metamorphic grade and controlling variables.

HHC - Zone	~ 200 °C
CH <sub>4</sub> - Zone	
H <sub>2</sub> O - Zone	

Major controlling variables:

- temperature
- time

Minor controlling variables:

- lithostatic pressure
- lithology

Fig. 6 Fluid inclusions: division of metamorphic grade and controlling variables.

## 2.4. FLUID INCLUSIONS

Until a few years ago fluid inclusion studies concentrated on large quartz crystals from late Alpine clefts. Recently, MULLIS (1979) developed a method that leads to a three-fold zonation of very low-grade regional metamorphic areas using fluid inclusions from tiny fissure quartz. Three fluid zones were defined, namely (i) a zone of higher hydrocarbons covering the diagenetic or non-metamorphic range; (ii) a methane zone covering the low- and medium-grade anchizone as defined by illite "crystallinity" data; and (iii) a water zone beginning in the high-grade anchizone and extending into the epizone (Fig. 6). The precise boundary values used to define the fluid composition of the three mentioned fluid zones are tabulated in FREY et al. (1980b). It is believed that the early inclusions from the nonmetamorphic zone and the anchizone have a fluid composition approximately related to peak metamorphic conditions. Based on homogenization temperatures, the lower and upper boundaries of the methane zone were determined to be about 200° at 1.2 kbar and 270°C at 1.7 kbar, re-



spectively (MULLIS, 1979; see Fig. 6). This method, which has been applied so far only in a few subsequent studies (MULLIS in FREY et al., 1980a, 1980b; MULLIS in NIEDERMAYR et al., 1984) seems to be very promising.

### 3. Correlation of methods

As mentioned earlier, very few studies have applied all of the four methods discussed above. Illite "crystallinity" data, mineral assemblages and fluid zones yield a homogeneous picture. Even so, there is still some uncertainty as to how coal rank correlates with the other three methods. Fig. 7 shows the presently available scheme as worked out in the Alps (subject to slight modification in the future). This correlation supersedes an earlier attempt (FREY and NIGGLI, 1971; see also FREY et al., 1980a, p. 197 for a correction) and is in general agreement with the correlation proposed by KISCH (1983). According to data from the Western and Central Alps (Plate 1), the majority of occurrences of pumpellyite  $\pm$  prehnite + chlorite + albite  $\pm$  quartz are located in the anchizone, whereas all occurrences with pumpellyite + actinolite (or glaucophane) + chlorite  $\pm$  albite + quartz are located in the epizone as defined by illite "crystallinity" data.

ILLITE 'CRYSTALLINITY'	METAMORPHIC FACIES	FLUID ZONE	COAL RANK (USA)
DIAGENESIS	ZEOLITE	HHC	BITUM. COAL SEMI-ANTHRACITE
ANCHIZONE	PREHNITE- PUMPELLYITE	CH <sub>4</sub>	ANTHRACITE
	PUMPELLYITE- ACTINOLITE	H <sub>2</sub> O	META-ANTHRACITE
EPIZONE	GREENSCHIST		GRAPHITE

Fig. 7 Correlation between different indicators of very low-grade metamorphism based on data from the Alps.

### 4. Geothermometry and geobarometry

The derivation of temperature-pressure conditions in very low-grade metamorphism is a difficult undertaking. The analyses of mineral assemblages in

very low-grade metabasites and metagraywackes provides tight constraints on P-T conditions (LIU et al., 1987), but relevant mineral chemical data are very scarce in the Western and Central Alps (e.g. COOMBS et al., 1976). Some reaction-isograds in metaclastites and iron-rich metalimestones have been located in the Central and Eastern Alps as mentioned earlier, but only the univariant reaction kaolinite + quartz = pyrophyllite + H<sub>2</sub>O has been located in P-T space. Unfortunately, this dehydration reaction cannot be used for temperature estimates as long as the activity of water remains unknown. The calcite-dolomite geothermometry is limited at temperatures below 300°C because of low degree of solid-solution. Vitrinite reflectance data have yielded reasonable temperature estimates (e.g. STALDER, 1979) and the same may become true by application of the conodont colour method in the Eastern Alps. In this author's opinion, the most reliable temperature and pressure estimates can presently be extracted from fluid inclusion data (see section 2.4. for references). The b<sub>0</sub> geobarometer proposed originally by SASSI and SCOLARI (1974) for greenschist facies metapelites has been extended to anchizonal metaclastites of the Central Alps by PADAN et al. (1982) and needs further testing.

### 5. Radiometric dating

Radiometric dating of very low-grade metamorphic rocks is fraught with many difficulties (for a review see HUNZIKER, 1987). Notwithstanding these reservations, radiometric dating in the Alps has yielded important but sometimes conflicting results. Most commonly, the K-Ar and the Rb-Sr methods have been applied (Western Alps: BONHOMME et al., 1980; Central Alps: FRANK and STETTLER, 1979; HUNZIKER et al., 1986; Eastern Alps: JUNG, 1980; KRALIK et al., 1981; KRALIK, 1982, 1983a, b; HAMMERSCHMIDT, 1982), and in a few cases the <sup>39</sup>Ar/<sup>40</sup>Ar method was used (FRANK and STETTLER, 1979; HAMMERSCHMIDT, 1982; HUNZIKER et al., 1986). These studies indicate that most very low-grade metaclastites of the Eastern Alps were metamorphosed during the Eo-Alpine orogenic phase in late Cretaceous time. In the Central and Western Alps, on the other hand, the Meso-Alpine and Neo-Alpine phases of middle and late Tertiary age, respectively, seem to be predominant.

### 6. Suggestions for future work

The study of very low-grade metamorphism during the last two decades has enhanced the knowledge of metamorphism in the external regions of the Alps to a considerable extent. Our knowledge of the boundary between very low-grade and low-grade metamorphism is fairly complete, but much less is known

about the lower temperature realm encompassing advanced diagenesis to the beginning of very low-grade metamorphism. What is particularly needed, besides careful X-ray diffraction studies, are (i) microstructural and chemical information on the fine grained sheet silicates through the application of electron microscopy (SEM, TEM, HRTEM); (ii) more detailed studies of the organic matter, not only reflectivity studies on phytoclasts, but also using organic chemical methods and applying the method of conodont colours (Eastern Alps). (iii) Fluid inclusion studies on quartz from open fissures initiated by J. Mullis should be extended to the Western and Eastern Alps. In addition, such studies may also be applied to vein and rock forming quartz with a well defined structural history. (iv) Fission track dating with its low closure temperatures has much potential for dating the very low-grade metamorphism and subsequent uplift history but has, to the author's best knowledge, not yet been applied to Alpine areas covered in this review.

#### Acknowledgments

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