# Illite crystallinity and coal rank associated with lowest-grade metamorphism of the Taveyanne greywacke in the Helvetic zone of the Swiss Alps

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# Illite crystallinity and coal rank associated with lowest-grade metamorphism of the Taveyanne greywacke in the Helvetic zone of the Swiss Alps

By HANAN J. KISCH[1])

## ABSTRACT

Illite crystallinities and vitrinite reflectances were determined in pelitic rocks associated with earlier-described zeolite-facies and prehnite-pumpellyite facies assemblages of the Taveyanne greywacke and other Lower Tertiary flysches of the Helvetic zone in order to compare the evolution of these parameters of incipient metamorphism.

Variation of the illite 10 Å peak width upon various chemical treatments is discussed; the anomalous peak broadening upon K-saturation in some samples is ascribed to the incorporation of previously resolved diffraction effects of illite/smectide mixed-layers into the main 10 Å peak.

Illite crystallinities associated with the laumontite-bearing greywacke occurrences (Diablerets, Taveyanne, Lower Kiental, and Balmen near Kandergrund) are "diagenetic", as are those not associated with diagnostic Ca-Al-silicate minerals from higher Helvetic nappes in the Kander valley. The mean vitrinite reflectances from the laumontite occurrences range from 0.85 to 1.3% $R_{max}$ (high-volatile *A* and medium-volatile bituminous ranks); some flysches from the Wildhorn nappe in the Kander valley are associated with similarly low reflectances, but others show higher mean reflectances of 1.6 to 2.3% $R_{max}$ (low-volatile bituminous and semi-anthracite ranks).

Pelitic rocks associated with laumontite-free, prehnite- and pumpellyite-bearing Taveyanne greywacke and with flysches without diagnostic Ca-Al-silicate minerals from the Kander valley eastwards towards the Linth valley, have anchimetamorphic (predominantly middle- to high-grade anchimetamorphic) illite crystallinites; associated vitrinite reflectances are largely 3.3 to 4.2% $R_{max}$ (medium- to high-rank anthracite).

Within the low-grade and the high-grade group of localities no detailed correlation between illite crystallinity and coal rank could be established.

Illite crystallinities from two localities, La Tièche (with pumpellyite; no laumontite) and Seedorf near Altdorf show a wide spread of values; they are tentatively assigned to the onset of the anchizone (2.1-2.9% $R_{max}$ at Seedorf).

The onset of anchimetamorphism as apparent from illite crystallinity seems to be approximately in the coal-rank range 2.3 to 3.3% $R_{max}$ (high-rank semi-anthracite to low-rank anthracite).

The pelites associated with Carboniferous anthracites of the Rhone valley show low-grade epizonal illite crystallinities; the real grade may be somewhat higher, in view of peak-broadening by the almost ubiquitous paragonite. The associated reflectances are not much higher than in the Lower Tertiary flysch between the Reuss and Linth valleys: as found earlier in Jämtland, reflectance does not seem to be a sensitive parameter of degree of metamorphism within the range high-grade anchizone to low-grade epizone, at least in terranes with medium- to high-pressure metamorphism.

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ZUSAMMENFASSUNG

Illit-Kristallinität und Vitrinit-Reflektivität wurden an pelitischen Gesteinen bestimmt, die mit früher beschriebenen Zeolit- und Prehnit-Pumpellyit-Faziesgesellschaften in der Taveyanne-Grauwacke und anderen Untertertiärflyschen der helvetischen Zone vorkommen, um die Entwicklung dieser Parameter der Anfangsmetamorphose zu vergleichen.

Durch chemische Behandlungen erzeugte Veränderungen in der Halbwertbreite des 10-Å-Diffraktionspeaks im Illit werden erörtert; die in Einzelfällen durch K-Sättigung erzeugte anormale Peakverbreitung wird dem Zusammenlaufen vorher resolvierter Diffraktionseffekte der Illit/Smektit-Mixed-Layers mit dem 10-Å-Hauptpeak zugeschrieben.

Die mit den Laumontit-haltigen Grauwacke-Vorkommen (Diablerets, Taveyanne, Nieder-Kiental und Balmen bei Kandergrund) vergesellschafteten Illit-Kristallinitäten sind «diagenetisch» sowie die nicht mit diagnostischen Ca-Al-Silikatmineralen vergesellschafteten Illit-Kristallinitäten von höheren helvetischen Decken im Kandertal. Die mittleren Vitrinit-Reflektivitäten aus den Laumontit-Fundstellen schwanken zwischen 0,85 und 1,3% $R_{max}$ (Gasflammkohle bis Fettkohle); einige Flysche der Wildhorndecke im Kandertal sind mit ähnlich niedrigen Reflektivitäten assoziiert, andere aber zeigen höhere mittlere Reflektivitäten von 1,6 bis 2,3% $R_{max}$ (Ess- und Magerkohle).

Pelitische Gesteine in Verbindung mit Laumontit-freier, Prehnit-Pumpellyit-haltiger Taveyanne-Grauwacke, und mit Flyschen ohne diagnostische Ca-Al-Silikatminerale vom Kandertal ostwärts nach dem Linthtal, weisen anchimetamorphe (vorwiegend mittel- bis hochgradig anchimetamorphe) Illit-Kristallinitäten auf; die Vitrinit-Reflektivitäten sind hier meist 3,3 bis 4,2% $R_{max}$ (mittel- bis hochgradige Anthrazite).

Innerhalb der niedriggradigen und hochgradigen Fundortsgruppen konnte keine detaillierte Korrelation zwischen Illit-Kristallinität und Inkohlungsgrad festgestellt werden.

Die Illit-Kristallinitäten von zwei Lokalitäten, La Tièche (mit Pumpellyit; kein Laumontit) und Seedorf bei Altdorf, zeigen breite Streuung; sie sind mit Vorbehalt dem Anfang der Anchizone eingeschaltet (2,1–2,9% $R_{max}$ bei Seedorf).

Der durch Illit-Kristallinität angegebene Anfang der Anchimetamorphose liegt ungefähr im Inkohlungsbereich 2,3 bis 3,3% $R_{max}$ (niedriggradigen Anthrazit).

Die mit den karbonischen Anthraziten des Rhonetals vorkommenden Pelite zeigen niedriggradig epizonale Illit-Kristallinitäten; der wirkliche Grad könnte höher sein im Hinblick auf die Peakverbreiterung durch den weitverbreiteten Paragonit. Die vergesellschafteten Reflektivitäten sind nicht viel höher als die im tertiären Flysch zwischen Reuss und Linthtal: wie vorher schon in Jämtland gezeigt, scheint Vitrinit-Reflektivität im Bereiche hochgradige Anchizone–niedriggradige Epizone kein empfindlicher Metamorphoseparameter zu sein, am wenigsten in Gebieten der Mittel- bis Hochdruckmetamorphose.

## Introduction

Attempts to arrive at a correlation between various parameters of burial diagenesis and incipient metamorphism have been made by this writer over a number of years (Kisch 1966, 1968, 1969, 1974, 1980). Initially relations were studied between zeolite facies and rank of bituminous coals in a number of areas (Kisch 1966). In a later stage data were collected – largely from published information – bearing on the association between illite-crystallinity and zeolite- and prehnite–pumpellyite facies assemblages on one hand, and coal rank on the other (Kisch 1974), and between zeolite- and prehnite–pumpellyite facies and changes in clay mineralogy (Kisch, in press). A field study was made of relations between illite crystallinity and coal rank in the Jämtland Supergroup of the eastern Caledonides in western Sweden (Kisch 1980). These correlations showed that the anchimetamorphic zone, as defined by a specific range of illite-crystallinity values, is by and large associated with anthracitic to low-grade meta-anthracitic coal ranks (ASTM classification).

No attempts have thus far been made at direct correlation of the three types of parameters of burial diagenesis and incipient regional metamorphism in a terrane where all three can be measured, i.e. in a terrane in which pelitic slates or shales with carbonaceous matter is associated with lithic-feldspathic or volcanic greywackes containing lowest-grade diagenetic to metamorphic mineral assemblages characteristic of the laumontite zone of the zeolite facies and the prehnite-pumpellyite-metagreywacke facies. Such a study has been undertaken by the writer in the Upper Eocene to Lower Oligocene Taveyanne greywacke-"Dachschiefer" (roofing slate) formation, the youngest flysch in the Lower Helvetic nappes of the Swiss Alps.

## Burial metamorphism in the Taveyanne greywacke formation

### *The Taveyanne greywacke formation*

The Taveyanne greywacke formation ("Grès de Taveyanne") and the associated shales or slates ("Dachschiefer") of Upper Eocene to Lower Oligocene age constitute the youngest clastic sedimentary rocks in the Lower Helvetic (North-Helvetic) nappes of Switzerland and the autochthonous sedimentary cover of the external central massifs of the French and Swiss Alps. The greywackes are characterized by a very high content of detritic plagioclase (now largely albitized), clinopyroxene, brown hornblende and volcanic rock fragments, with minor quartz, K-feldspar and biotite. Up to 90% of the detritus is of basaltic andesite derivation (VUAGNAT 1974, p. 419; KÜBLER et al. 1974, p. 462), and is supposed to have been derived from a cordillera or island arc which is not presently extant.

### *Zeolite and prehnite-pumpellyite facies metamorphism*

MARTINI & VUAGNAT (1965, 1970; see also MARTINI 1968, 1972; VUAGNAT 1974; KÜBLER et al. 1974) have described the wide-spread occurrence of secondary laumontite, prehnite and pumpellyite in the Taveyanne greywackes.

According to their secondary mineralogy, MARTINI & VUAGNAT (1965, 1970; see also MARTINI 1968) distinguish three main types in the volcanic greywackes:

1. Laumontite greywackes

These are characteristically mottled arenites ("Grès mouchetés"), containing laumontite-rich patches several millimeters in diameter. The laumontite replaces both plagioclase and the vitreous matrix of the andesites, and occurs as a cement. The plagioclase is largely pseudomorphosed by albite. KÜBLER (1973, see also KÜBLER et al. 1974, p. 465) has noted the common presence of the regular chlorite/swelling chlorite mixed-layer corrensite in the fine fraction of the laumontite greywackes.

2. Pumpellyite-prehnite greywackes

These are also commonly mottled. Two end-members can be distinguished:

a) Prehnite greywacke, in which coarse xenomorphic has a textural position similar to that of laumontite in the former type.

b) Pumpellyite greywacke, in which pumpellyite appears in small crystals in plagioclase and in the matrix.

3. Greywackes of the "type vert"

This type is not macroscopically mottled; it consists essentially of albite and chlorite, and is almost devoid of secondary Ca-Al-silicates. Compared to the previous types, this green type is poor in Ca and rich in Na, hence KÜBLER et al. (1974, p. 463) suggest the designation "spilitic facies". A somewhat aberrant green type ("type vert micromoucheté") contains fine-grained radial aggregates of prehnite replacing matrix.

The Taveyanne greywackes do not show any metamorphic schistosity.

The secondary nature of the albite is indicated by the local conservation of calcic plagioclase (e.g. in some impermeable calcareous concretions; MARTINI & VUAGNAT 1970, p. 56-57) and by the regional distribution of the various alteration types.

The degree of incipient metamorphism in the Taveyanne greywacke increases both perpendicular to the strike southeastwards (eastwards in the Frenche Alps) and in the strike of the Alpine arc northeastwards. According to MARTINI & VUAGNAT (1970, p. 55-57) the distribution of the laumontite and prehnite-pumpellyite types - the "type vert" occurs in both zones - is essentially as follows:

The laumontite greywackes characterize the western part of the occurrence of the Taveyanne formation: the Champsaur (south of the Pelvoux massif), the Morcles nappe of Haute-Savoie, part of the Diablerets nappe in the type region.

Transitions between the laumontite and prehnite-pumpellyite types - which facially belong to the zeolite facies rather than to the prehnite-pumpellyite facies - occur in Haute-Savoie (southern part of the Massif de Platé), in the frontal part of the Diablerets-Gellihorn nappe in the Diablerets area and in the Lower Kander and Kien valleys (Berner Oberland), and probably also at Merligen, north of Lake Thun.

Prehnite-pumpellyite greywacke without laumontite appears locally in Haute-Savoie (southern part of the Massif de Platé), but is fully developed only towards the northeast, in the more internal parts of the Helvetides of the Berner Oberland (Upper Kander and Kien valleys) and in eastern Switzerland.

East of the Reuss valley greywacke types with pumpellyite and epidote - representing a further advance in degree of incipient metamorphism - are common, but prehnite continues to occur (MARTINI 1968, p. 585; MARTINI & VUAGNAT 1965, Fig. 1).

Locally, e.g. east of Leuk (Valais), near the root zone of the Helvetic nappes south of the Kander valley, a schistose modification of the Taveyanne greywackes shows the even higher-grade metemorphic assemblage pumpellyite-actinolite (MARTINI & VUAGNAT 1970; COOMBS et al. 1976).

## Incipient-metamorphic phyllosilicate mineralogy in the Swiss Helvetides

### *a) Illite crystallinity*

FREY (1969, 1970), in a major study of phyllosilicate mineralogy and illite crystallinity in the Upper Triassic and the Lower Liassic of the Glarus Alps, has established a lowest-grade metamorphic zoning for the Helvetic zone of eastern Switzerland. The Upper Trias and Lower Liassic of the southern Glarus Alps including the Sernf, upper Linth and Urnerboden valleys (FREY 1970, Fig. 7) show illite crystallinities characteristic for the high-grade part of the anchizone (4.0 to 5.1 mm half-height peak width in the scale of KÜBLER, 1968). Diagnostic anchimetamorphic minerals such as pyrophyllite, paragonite/phengite (or paragonite/muscovite) mixed-layers, and minor rectorite (regular mica/montmorillonite mixed-layer) appear already in the lower-grade parts of the anchizone, in the northern Glarus Alps (ibid., p. 265–269 and Fig. 10); irregular illite/montmorillonite mixed-layers are absent.

West of the Reuss valley, KÜBLER (1970) noted the occurrence of deep "diagenesis" – but not of anchimetamorphism – in the Upper-Helvetic Wildhorn nappe at the Schynige Platte, north of the Aar massif, as well as further southwest in the frontal part of the Morcles nappe, and in the Ultra-Helvetic nappe of Tour-d'Anzeinde between the Aar and Aiguilles-Rouges massifs, in the canton Vaud.

### *b) Appearance of stilpnomelane in glauconitic lithologies*

In a subsequent study of the progressive lowest-grade metamorphism of glauconite-bearing beds (of Lower Cretaceous and Eocene age) in the Helvetic zone of eastern Switzerland, FREY et al. (1973) found partial replacement of glauconite by stilpnomelane within a zone approximately 3 km wide; the illite crystallinities in this zone are characteristic for the middle part of the anchizone. In samples associated with the high-grade part of the anchizone in the southern Glarus Alps, e.g. in the Linthal and Urnerboden areas, the replacement of glauconite by stilpnomelane was largely completed (op. cit., p. 199–201).

Similarly, DURNEY (1974), in a study of the Helvetic nappes in Valais, places the appearance of stilpnomelane west of the Morcles nappe in the Rhone valley in the middle of the anchizone, and correlates a stilpnomelane in the root zone of the Helvetic nappes – assigned to the middle of the stilpnomelane zone on the basis of fluid-inclusion temperature data – to the high-grade limit of the anchizone.

Further to the southwest stilpnomelane has been claimed to appear at even lower degrees of incipient metamorphism: the Taveyanne greywacke, now removed by erosion, that overlaid its abundant occurrence in Albian glauconitic sandstones ("Gault") of the Morcles nappe at the Massif de Platé (Haute-Savoie), is extrapolated from nearby occurrences to have been at the limit of the laumontite and prehnite-pumpellyite facies (MARTINI & VUAGNAT 1970, p. 58; KÜBLER et al. 1974, p. 467).

### *c) Relationship between secondary Ca-Al-silicate minerals in the "Grès de Taveyanne" and illite crystallinity: published data*

Relatively few precise correlations between the lowest-grade mineral facies in the "Grès de Taveyanne" and illite crystallinity of associated rocks can be extracted from the literature. The main reason is that the bulk of the data on illite crystallinity and phyllosilicate mineralogy in the Helvetic zone have been obtained on different stratigraphic units – largely from Upper Triassic and Lower Liassic of the Glarus Alps (FREY 1969, 1970) and in the case of the appearance of stilpnomelane from glauconitic beds in Middle Jurassic, Lower Cretaceous and Eocene (FREY 1970; FREY et al. 1973) – and in part even on different nappe units.

Moreover, as pointed out by KÜBLER et al. (1974, p. 466), the crystallinity of the illites from the greywackes themselves cannot be used as a parameter of diagenetic evolution, since the evolution of the rocks leads to destruction of the clastic micas.

Correlations thus have to be based on crystallinity of illites from the pelitic and marly shales and slates ("Dachschiefer") intercalated with the Taveyanne greywackes, although it appears that at least in a few cases the illites from nearby Mesozoic sedimentary rocks indicate similar degrees of burial-diagenetic evolution.

1. A few kilometers externally from the prehnite- and pumpellyite-bearing Taveyanne greywackes from the parautochthonous units south of the Schächen (canton Uri) and Linth valley (canton Glarus), middle- and high-grade anchizonal illite crystallinities and the occurrence of pyrophyllite and paragonite (FREY 1970, Fig. 9 and 10) have been established in the Upper Triassic and Lower Liassic of the tectonically overlying Axen nappe. This nappe according to MARTINI (1972, p. 263) is more schistose and more "metamorphosed" than the underlying parautochthon; nevertheless, a similar high-grade anchizonal illite crystallinity has been reported from "Dachschiefer" directly associated with the prehnite-pumpellyite-epidote assemblages from the Linth valley (FREY & NIGGLI 1971, p. 232).

2. KÜBLER (1970) has noted that the illite crystallinities in Jurassic and Cretaceous limestones both of the frontal part of the Helvetic Morcles nappe and of the overlying Ultrahelvetic nappe, only a few kilometers south of the abundant laumontite greywackes in the intervening Diablerets nappe, have not yet attained the anchizone.

KÜBLER et al. (1974, p. 465-466) report that illites from Eocene shales and Senonian limestones underlying laumontite-bearing Taveyanne greywackes in western Switzerland and France have a low crystallinity, indicating that the low-grade limit of the anchizone has not been reached. Similarly poor illite crystallinities are associated with laumontite-bearing Taveyanne greywackes in the Wageten chain, northernmost Glarus Alps (FREY & NIGGLI 1971, p. 232).

MARTINI (1972, Fig. 2) has shown that the marls and argillites associated with the laumontite greywackes in the Morcles nappe between Arve and Giffre (Haute-Savoie) still contain traces of expanding minerals, suggesting a similarly weak degree of burial-diagenetic alteration.

In contrast, the crystallinity of illites from some shales intercalated with pumpellyite-prehnite greywackes from central Switzerland (no details on localities are given) corresponds to the anchizone-"epizone" limit.

The illite crystallinities in the vicinity of the pumpellyite- and actinolite-bearing Taveyanne semi-schist occurrence northeast of Leuk (Valais) lie in the low-grade part of the "epizone" (KÜBLER et al. 1974, p. 466); the area lies well within the chloritoid zone for pelites as recently defined by FREY & WIELAND (1975).

KÜBLER et al. (1974, p. 466) note that the onset of anchimetamorphic illite crystallinities takes place "somewhere in the pumpellyite-prehnite zone", but regard this situation as anomalous: they would have expected laumontite greywackes showing extensive transformation of original mineralogy to be associated already with anchimetamorphic illite crystallinities (but admit that more work to establish this correlation is required). This expectation, in this writer's view, seems unwarranted in the light of the wide-spread association of laumontite-bearing assemblages with montmorillonite and montmorillonite/illite mixed-layers, e.g. in the classic Triassic sequence of the Taringatura and Hokonui Hills of Southland, New Zealand (COOMBS 1953; KISCH, in prep.), and the association of the onset of pumpellyite-prehnite assemblages in some areas with lower coal ranks than the onset of anchimetamorphism (KISCH 1974).

Rather, it is to be fully expected that anchimetamorphic illite crystallinities should make their appearance only at or beyond the degree of incipient metamorphism causing the disappearance of laumontite from volcanic greywackes and similar suitable host rocks.

## Vertical zoning and age of the incipient metamorphism in the Swiss Helvetides

The tectonic superimposition of somewhat schistose volcanics of the Glarus Verrucano with greenschist facies mineral assemblages - and of similar Verrucano of the Axen nappe - over parautochthonous Taveyanne greywacke in the prehnite-pumpellyite facies, led MARTINI & VUAGNAT (1970, p. 59) and MARTINI (1972, p. 263) to the conclusion that the metamorphism of the higher nappes took place prior to their final emplacement.

FREY et al. (1973, p. 213), on the other hand, argue that the gradual downward increase of the illite crystallinity through the Helvetic Bächistock-Axen-Mürtschen-Glarus nappe sequence in the Glärnisch, north of Linthal, indicates that during the incipient metamorphism the sequence of the Helvetic nappes (excluding the southern Helvetic nappes) was similar to the present and had been covered by a major sequence of higher tectonic units. They conclude that the peak of metamorphism - dated by K-Ar ages on minerals at 31-36 my - must have been shortly after the main Lower-Oligocene orogenic phase of the Helvetic Alps. Local younger K-Ar ages of 20-16 my from minerals on joints and young thrusts are considered to reflect a younger, early Miocene phase which thrusted the whole northern and middle Helvetic nappe complex northward over the Alpine margin.

In accordance with these conclusions are the findings of TEICHMÜLLER & TEICHMÜLLER (1978) of a downward coalification increase from high-volatile bituminous coal to anthracite with 2.8% $R_m$ through approximately 5000 m of Helvetic nappes in the well Maderhalm 1 in the Allgäu Alps (southern Germany), but with poor correlation with burial depth; further east, the Oligocene molasse underlying the nappe complex yielded reflectances of only 0.5 to 0.7% $R_m$. This confirms that the coalification in the Helvetic complex post-dates its internal structure, but pre-dates its emplacement upon the molasse.

## Experimental methods

### *a) Identification of hydrous Ca-Al-silicates in greywackes*

Pumpellyite and prehnite were identified in thin sections. The presence or absence of laumontite in the Taveyanne greywackes was confirmed by X-ray diffraction of both the silt-clay fraction and the 2.50-float fraction of these greywackes. The greywackes were ground to pass a 100 µm sieve and the powder dispersed in water. The clay-silt fraction was obtained by repeatedly decanting the suspension remaining after settling for about 2 minutes in 10 cm of water; the $D<2.50$ fraction of the remaining coarser (100 to about 30 μm) powder was obtained by float-sink separation in a tetrabromoethane-NN-dimethylformamide solution. Laumontite, if present, was strongly enriched in both these fractions due to its low specific gravity of 2.25; additional phases detected in the silt-clay and the 2.50-float fractions were chlorite, corrensite as well as albite, quartz and minor calcite and K-feldspar ($D=2.5$).

### *b) Illite crystallinity*

Approximately 5 g of sample were ground in a hammer mill to pass a 0.2 mm strain sieve. The powders were then treated with 6% $H_2O_2$ solution and with 0.1 HCl to remove organic matter and carbonates, respectively, and dispersed in sedimentation cylinders. The −20 μm, −6 μm and −2 μm size fractions were pipetted off after the appropriate settling periods and sedimented on glass slides; in later stages of the project the "−2+6 μm" and −2 μm size fractions were prepared (for details see Kisch 1980).

Two portions of the −2 μm fraction were treated with respectively 0.5 N KCl solution and 1 N $MgCl_2$ solution (cf. Dunoyer de Segonzac 1969) in order to saturate the interlayered smectite and vermiculite layers, and also sedimented on glass slides.

The half-height width of the illite 10 Å diffraction peaks were measured on diffractograms run on a Philips X-ray diffractometer using Ni-filtered $CuK_\alpha$ radiation. Settings were as follows: slits 1° - 0.2 mm - 1°; time constant 2 at scale $1\times10^3$ or $2\times10^3$ (usually the former), or rarely time constant 4 at scale $4\times10^2$; goniometer 0.5° $2\theta$/min and chart 20 mm/min, so that 1° $2\theta$ equals 40 mm on the chart. The peak widths are expressed in degrees $\Delta2\theta$ rather than in millimeters.

Under these conditions the narrowest 10 Å peak widths measured on well-crystallized muscovite are approximately 0.11° $\Delta2\theta$ (4.5 mm). The values corresponding to the low- and high-grade limit of Kübler's (1967, 1968) anchizone - as established by measurement of polished-slate inter-laboratory standards in both Professor Kübler's and the writer's laboratory - are respectively 0.38 and 0.21° $\Delta2\theta$. For further comparisons with the operating conditions and limiting values for the anchizone as used by Kübler (1967, 1968), Dunoyer de Segonzac (1969), Frey (1969) and other workers the reader is referred to Kisch (1980, particularly Table 1).

### *c) Vitrinite reflectances*

The vitrinite-reflectance measurements were carried out on polished grain mounts of grains up to 5 mm in diameter in cold-setting polyester resin.

The measurements of reflectance were made on a reflected-light Zeiss photomicroscope equipped with photomultiplier and stabilized power supply in plane-polarized monochromatic green light (525 nm), using an EPI 0.40/0.85 Pol oil-immersion objective. This procedure allows reflectance measurement on vitrinite grains down to some ten microns in diameter.

For further details on the sample-preparation and measurement techniques see Kötter (1960), de Vries et al. (1968) and Stach et al. (1975, p. 240-243 and 263-274). For a recent $R_{max}$ versus volatile-matter curse for the low-volatile bituminous, semi-anthracite and anthracite rank range see Wolf (1972, Fig. 2).

## Material studied

"Dachschiefer" associated with Taveyanne greywackes were collected from a number of localities. These include - from southwest to northeast - Taveyanne[2]), Diablerets, Balmen (middle Kandertal) and Lower Kiental, from which Martini & Vuagnat (1965) have reported laumontite, and La Tièche (near "Varnerkumme" of Martini & Vuagnat 1965, Fig. 1), Reichenbachfall near Meiringen and Trübsee

[2]) The samples collected at Taveyanne are possibly in part of flysch of the Ultrahelvetic Plaine Morte nappe of the "Zone des Cols".

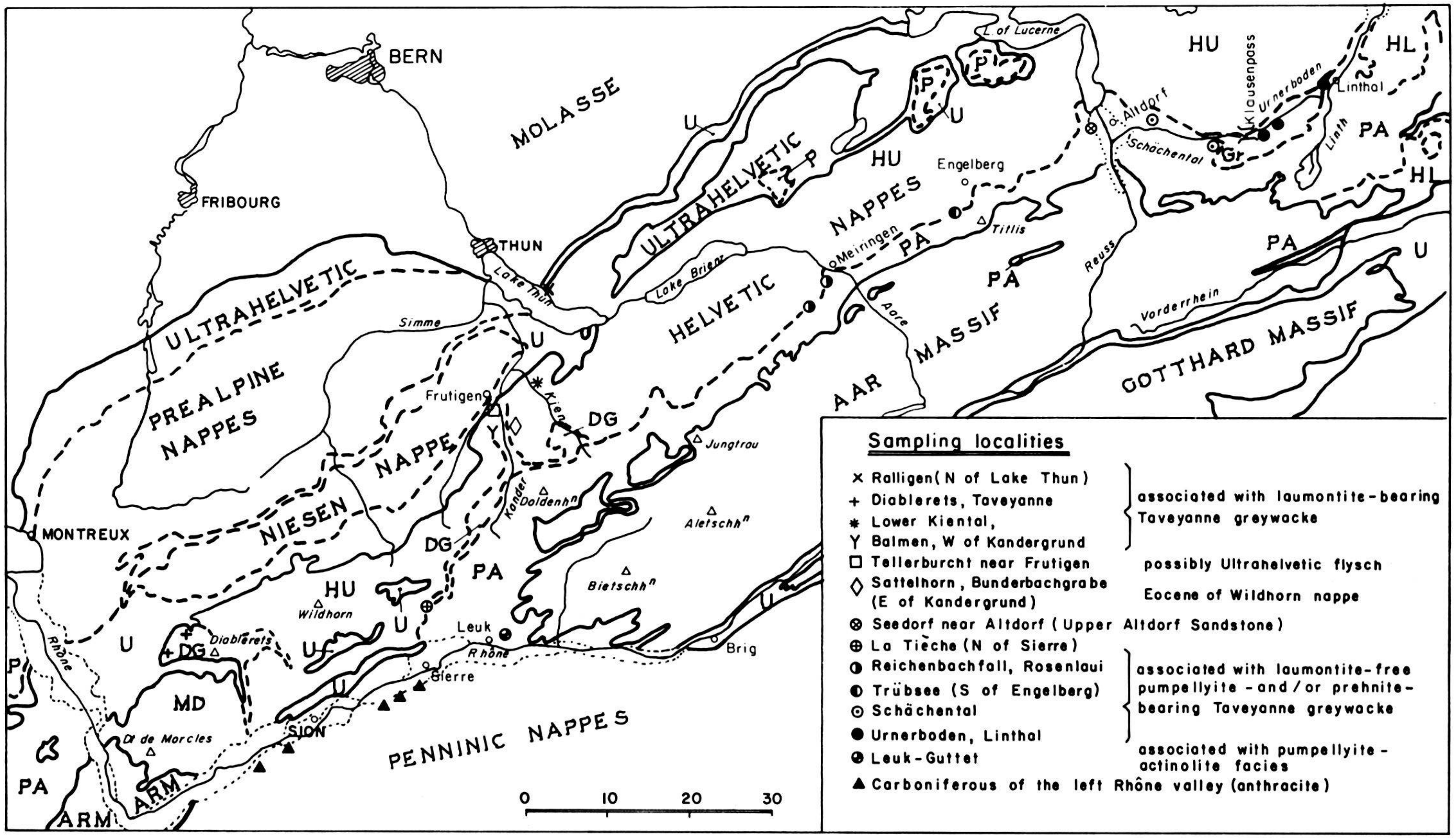


Fig. 1. Simplified tectonic map of the external Alpine zones of central Switzerland, showing the localities sampled. The limits between the Prealpine and Ultrahelvetic nappes, and between the Helvetic nappes, are shown as broken lines. The boundaries of major valley fills are stippled.

Abbreviations of tectonic units: *P* = Prealpine nappes (exclusive of Niesen nappe). *U* = Ultrahelvetic nappes (including the Garvera-Urseren-Furka zone and the Mesozoic cover of the Gotthard massif). *HU* = Upper Helvetic nappes: Wildhorn, Axen, Säntis-Drusberg nappes. *HL* = Lower Helvetic nappes mainly: Glarus, Mürtschen, Diablerets-Gellihorn (DG) nappes; Griesstock (Gr) and other subhelvetic nappes. *PA* = Parautochthonous and autochthonous Helvetic (including the Morcles-Doldenhorn nappe, MD). *ARM* = Aiguilles-Rouges massif.

near Engelberg from which only prehnite and pumpellyite were reported, and from various localities in the Schächen and Urnerboden valleys and near Linthal; in the latter areas no precise localities are indicated by MARTINI & VUAGNAT (1965), but these authors as well as FREY (1970, Fig. 10) report the occurrence of pumpellyite, prehnite and epidote in the absence of laumontite.

In addition, samples were collected from a number of Eocene flysches not associated with Taveyanne greywacke: from the Wildhorn nappe at Bunderbachgrabe and at the abandoned coal mine at Sattelhorn, from possibly Ultrahelvetic at Tellerburcht near Frutigen in the middle Kander valley, and from the uppermost autochthonous flysch, the Upper Altdorf Sandstone (which stratigraphically overlies the Taveyanne greywacke), at Seedorf near Altdorf in the Reuss valley (cf. BRÜCKNER 1967).

## Effects of the presence of illite/smectite mixed-layers and paragonite on the illite peak widths

The illite crystallinities of the K- and Mg-saturated $-2$ µm fractions of all Tertiary and Carboniferous clastic rocks measured are listed in Table 1; also listed are the $I_{002(5\,Å)}/I_{001(10\,Å)}$ intensity ratios for these fractions. For some samples the peak width of the glycolated $-2$ µm fraction (without previous cation saturation), measured to assess the extent of the effects of montmorillonitic mixed-layers on the peak widths, is also given.

### *Effects of K-, Mg- and ethylene-glycol saturation upon the peak widths*

In many of the less crystalline samples (both saturated peak widths narrower than about 0.30° $\Delta 2\theta$), there is a marked difference between $B_{K\text{-sat}}$ and $B_{Mg\text{-sat}}$, the latter value generally being the larger one (see Fig. 2 and 3).

The Mg-saturated samples also commonly show an increase in the $I_{002\,(5\,Å)}/I_{001\,(10\,Å)}$ ratio, sometimes by up to 100%; comparison with peaks of phases unaffected by cation saturation shows this increase to be due to the attenuation of 001 (10 Å), as well as an extension of the low-angle "tail" of the 10 Å diffraction peak to lower diffraction angles upon Mg-saturation.

This indicates that these changes are due to the reduced contribution of mixed-layers with an Mg-expandable component to the height of the 10 Å peak.

Whether broadening, no change, or narrowing of the half-height width of the 10 Å peak will result, depends on the extent of detachment of the Mg-expanded mixed-layer "tail" from the main 10 Å peak compared with K-saturation. If the "tail" remains above half height, without appreciable resolution from the 10 Å peak, broadening will occur. If, on the other hand, the expansion results in lowering of the "tail" below half height, or its increased resolution from the main 10 Å peak, sharpening at half height may take place. The latter effect - rare upon Mg-saturation as compared with K-saturation - is almost ubiquitous upon EG-saturation (see Table 1), which expands the smectite component of the mixed-layers more than does Mg-saturation (to 17 Å and 14–15.5 Å, respectively).

Glycolation of the weakly crystalline samples resulted in an increased symmetry of the 10 Å peak profiles. Similar effects found earlier during a study of similar

Table 1: *Illite crystallinities of the K-saturated, Mg-saturated and glycolated – 2 μm size fractions of the samples studied.*

| Locality | Sample nr. | Lithology | $B_{001}$ (°Δ2θ) 1) K-sat | $B_{001}$ (°Δ2θ) 1) Mg-sat | $I_{002}/I_{001}$ 2) K-sat | $I_{002}/I_{001}$ 2) Mg-sat | $B_{001}$ (°Δ2θ) EG 1) | Parag; pyroph |
|---|---|---|---|---|---|---|---|---|
| Shales etc. associated with laumonitite-bearing Taveyanne greywacke (mainly Diablerets-Gellihorn nappe) | | | | | | | | |
| Diablerets | 75-66B | shale | .56-.62 | .70-.70 | .21* | .37 | | |
| | 66C | marl | .53-.56 | .52-.53 | .28* | .33 | .37+ | |
| | 68 | marl | .95+ | 1.2-1.3+ | .15** | .43* | .46 | |
| | 69B | marly shale | .55-.58 | .54-.62 | .21* | .40 | | |
| | 69C | slightly calcar. shale | .42-.43+ | .42-.48+ | .25* | .34* | | |
| | 70A | carb.-free shale | .46-.48 | .42-.46 | .16* | .16 | | |
| Taveyanne (poss. Ultrahelvetic in part) | 75-72C | shale | .34-.36 | .36-.39 | .28 | .30 | | |
| | 72D | fine micaceous ss | .38-.36 | .32-.31 | .27 | .29 | | |
| | 73 | marl | .46-.45 | .49-.54 | .23 | .42* | | |
| Ralligen near Merligen | 75-82A | micaceous ss | .54+ | .40-.43+ | .22* | .37 | | |
| | 82C | micaceous ss | .27-.30 | .40 | .25* | .43* | | |
| Kiental | 75-79A | slightly calcar. shale | .55-.56 | .67-.68 | .28 | .30 | | |
| | 79B | slightly calcar. shale | .60-.64 | .70-.75 | .17* | .35 | | |
| | 80A | arenite in carb. grains | .46-.48++ | .56-.58+ | .34** | .38* | | |
| | 74-3B | marly shale | .37-.38+ | .53-.55 | .26** | .28** | | |
| Balmen, W of Kandergrund | 75-89A | marl | .42-.45 | .38-.45 | .22 | .25 | .37-.41+ | |
| | 90A | fine gr. black | .50++ | .42++ | ** | ** | .38-.50++ | |
| | 90B | marl | .53-.56 | .46-.50 | .22* | .27 | | |
| "Diagenetic" to low grade anchizonal illite crystallinities associated with flysches without diagnostic Ca-Al Silicates | | | | | | | | |
| (a) Kandertal (upper Helvetic and Ultrahelvetic) | | | | | | | | |
| Tellerburcht | 75-83A | slightly calcar. shale | .78-.79 | .94-.96 | .19 | .33 | | |
| | 84A | marl | .68-.69 | .77-.78 | .17 | .46 | | |
| Sattelhorn (Wildhorn nappe) | 75-85C | marl | .52 | .66-.72 | .38 | .46 | .41-.55++ | |
| | 85D | marl | .55+ | .50-.54+ | .20* | .24* | .37-.44++ | |
| | 85H | marl or slaty | .55++ | .35++ | ** | ** | .40-.50++ | |
| Bunderbachgrabe (Wildhorn nappe) | 75-86A | marl | .42-.50++ | .42-.45 | .26* | .29 | | |
| | 76C | slate | .45-.49 | .39-.44 | n.d. | .35 | | |
| | 74B | marl | .41-.43 | .44-.45 | .29 | .35 | | |
| (b) Seedorf nr Altdorf (autochthonous Upper Altdorf Sandstone) | | | | | | | | |
| Grube Gasperini | 75-40A | coarse siltst.+ shale | .54-.55 | .53 | - .17* | .28 | .42-.43 | |
| | 40B | coarse siltst.+ shale | .53-.60 | .58-.64+ | .23* | .23* | .38-.43 | Parag. |
| | 40C | coarse siltst. | .34-.38+ | .34-.36+ | .26** | .31* | | |
| | 40D | coarse siltst. | .30-.32+ | .32-.34 | .26* | .26 | | |
| | 40E | coarse siltst. + shale (fold hinge) | .22-.29 | .25-.26 | .26 | .29 | | |

Table 1 (continued)

| Locality | Sample nr. | Lithology | $B_{001}$ (°Δ2θ) 1) K-sat | Mg-sat | $I_{002}/I_{001}$ 2) K-sat | Mg-sat | $B_{001}$ (°Δ2θ) EG 1) | Parag; pyroph |
|---|---|---|---|---|---|---|---|---|
| Shales etc. associated with laumontite-free Taveyanne greywacke and other anchizonal illite crystallinites associated with flysches without diagnostic Ca-Al-silicates | | | | | | | | |
| (a) West of Reuss Valley | | | | | | | | |
| La Tièche | 75-44A | silty clay shale | .38-.42 | .40-.43 | .17** | .24** | .35-.38$^{+}$ | |
| | 44B | | .33-.37 | .41-.43 | .20 | .25 | | |
| | 46A(new) | marl | .28-.30 | .43-.47 | .22 | .25 | .35-.36 | |
| | 46B | silty clay shale | .48-.49 | .42-.45 | .19* | .26* | .34-.38 | |
| | 47A(new) | marl | .28-.29 | .32-.33 | .27 | .33 | | |
| | 47B | sandy ls | .21-.24 | .24-.25 | .29 | .31 | .22-.23 | |
| | 47B(new) | | .24-.25 | .26 | .33 | .33 | | |
| | 48 | ls or marl | .23 | .23 | .30 | .36 | .21-.22 | |
| | 49B | schistose ls | - | - | - | - | | |
| Leuk-Guttet | 75-51A | green shale | .18$^{+}$ | .13-.14$^{+}$ | .16* | .21 | | |
| | 51C | slate | .20 | .20-.22 | .39 | .31 | | |
| Rosenlaui | 75-11B | slightly calcar. shale | .25-.26 | .23-.20 | .27 | .29 | .21-.22 | |
| | 12A | marl | .24-.25 | .26-.26 | .27 | .28 | .23-.24 | |
| | 12C | slightly calcar. shale | .24-.25 | .22-.24 | .28 | .32 | | |
| Reichenbachfall nr Meiringen (Parautochthonous) | 75-9C | shale | - | - | - | - | | |
| | 9D | black marl | .28 | .26-.27 | .22 | .28 | | |
| | 10C | slate | .28 | .27-.30 | .27 | .33 | | |
| idem (Aalenien of Wildhorn nappe) | 75-13A | black shale | .36 | .38-.40 | .23 | .39 | | |
| | 13B | black shale | .31 | .39-.40 | .26 | .34 | .26-.29 | |
| Trübsee nr. Engelberg | 74-23A | slaty marl or calcar. slate | .27-.28 | .29 | .34 | .38 | | |
| | 23B | slate | .27 | .28 | .35 | .35 | | |
| | 24B | ss | .20$^{++}$ | .17$^{++}$ | ~.1** | < .1** | | |
| | 25 | ss | .18$^{++}$ | .18-.22$^{++}$ | ~.3** | ** | | |
| | 27A | mudstone | .28-.30 | .27-.28 | .27 | .30 | | |
| | 27B | slate | .26 | .22 | .30 | .27 | | |
| | 28A | silty marl | .23-.24 | .21-.22 | .31 | .33 | | |
| | 28A | silty marl | .22 | .22-.24 | .34 | .39 | | |
| (b) East of Reuss Valley (Schächental-Urnerboden-Linthal) | | | | | | | | |
| Schroten (Witerschwanden) (autochthonous lower Altdorf sandstone) | 75-39A | grey ls or calc.ss | .28-.29 | .27-.27 | .31 | .36 | | Parag. |
| | 39B | slate | .25-.28 | .35-.36 | .34 | .29 | | |
| Rübi (S side of Vorderschächen) | 75-38C | interb. marl slate | .24-.25 | .30-.32 | .31 | .31 | | |
| Oberalp-Äsch (Parautochthonous) | 75-29B | carb.-poor shale | .24-.24 | .23-.24 | .29 | .35 | | |
| | 29C | carb.-poor shale | .30-.31 | .26-.27 | .30 | .39 | .25-.26 | |
| | 33B | silty shale | .26-.26 | .28-.28 | .19* | .30 | .23-.27$^{+}$ | |
| | 34B | carb.-poor shale + marl | .23-.22 | .25-.26 | .29 | .32 | | |
| Klausenpass (Upper Triassic, Axen nappe) | 75-24B | | .36-.35 | .34-.35 | .23 | .40 | | {Pyroph; parag. |
| Chlus ("Flyschfenster") (Autochthonous) | 75-22B | slate + silty ls | .26-.26 | .20-.20 | .30 | .34 | | |
| | 22C | silty ls | .17-.17 | .16-.18 | .33 | .33 | | |
| | 23C | slaty silty ls | .26-.26 | .20-.20 | .29 | .35 | | |

Table 1 (continued)

| Locality | Sample nr. | Lithology | $B_{001}$ (°Δ2θ) 1) K-sat | $B_{001}$ (°Δ2θ) 1) Mg-sat | $I_{002}/I_{001}$ 2) K-sat | $I_{002}/I_{001}$ 2) Mg-sat | $B_{001}$ (°Δ2θ) EG 1) | Parag; pyroph |
|---|---|---|---|---|---|---|---|---|
| Waldhüttli (Griesstock nappe) | 75-21B | silty slaty ls | .36-.36 | .35-.35 | .31 | .44 | .34-.35 | |
| | 21C | slaty marl + ls | .21-.21 | .24-.25 | .35 | .36 | | |
| Linthal | 75-16 | clay shale | .23-.23 | .25-.25 | .33 | .38 | | |
| | 19C | silty marly shale | .24-.24 | .23-.24 | .31 | .32 | | |
| | 20B | silty marl | .18-.18 | .18-.19 | .32 | .35 | | |
| | 20C | silty marl | .23-.23 | .23-.23 | .33 | .29 | | |
| Shales associated with Carboniferous anthracites in the left Rhône Valley, SW of Sierre and Sion | | | | | | | | |
| Chalais | 75-54C | sericite phyllite | .15-.16 | .14-.15 | .29 | .28 | | Parag. |
| Rêchy | 55C | sericite phyllite | .17-.18 | .17-.18 | .27 | .27 | | Parag. |
| | 55E | slate | .19-.20 | .20 | .27 | .26 | | Parag. |
| | 56A | black slate | .16 | .16 | .28 | .27 | | Parag. |
| Grône-Rêchy | 58E | carbonaceous slate | .15 | .16-.17 | .29 | .26 | | Parag. |
| | 58F | slate | .18-.19 | .16-.18 | .24 | .25 | | Parag. |
| | 58G | sericite phyllite | .17-.19 | .15-.15 | .32 | .30 | | Parag. |
| Aproz | 63B | quartzitic black slate | .20 | .19-.20 | .28 | .30 | | Parag. |
| | 63C | black sericite phyllite | .17-.16 | .16 | .30 | .30 | | Parag. |
| Haute Nendaz | 59C | phyllite | .15-.16 | .15 | .28 | .26 | | |
| | 61A | weathered carbonaceous slate | .19-.19 | .18-.17 | .34 | .33 | | |

Footnotes

1) In the $B_{001}$ column inaccurate determinations of $B_{001}$ due to low intensity of the illite 10 Å peak are indicated by + and ++ for I < 150 c.p.s. and I < 100 c.p.s. respectively.

2) Particularly unaccurate determinations of the $I_{000}/I_{001}$ ratio due to low signal/noise of the illite 5 Å peak are indicated by asterisks, * and ** indicating signal/noise ratios of 5 to 2 1/2 and less than 2 1/2 respectively.

samples from Jämtland, Sweden (Kisch 1980), were ascribed by this writer to the superposition of an unresolved, slightly higher-angle $(001)_{01\,\text{Å}}/(002)_{17\,\text{Å}}$ peak of random illite/smectite mixed-layer with less than 30% expandable layers upon the 10 Å peak of illite (cf. Heller-Kallai & Kalman 1972; Reynolds & Hower 1970, p. 34 and Fig. 9).

This effect is at variance with that described for "open illites" of type $I_M$ as defined by Lucas (1963): $I_M$-type "open illite" - with presence of interlayers considered by Lucas to behave similar to montmorillonite - is characterized by increase in the *asymmetry* of the background to the *low-angle* side of the 10 Å diffraction peak upon glycolation.

### *Applicability of "illite crystallinity" in the presence of mixed-layers*

These variable effects of K- and Mg-saturation on the 10 Å peak widths of illites - particularly in the "diagenetic" zone and the low-grade part of the anchizone - raise the question in how far these peak widths may be used as parameters of burial diagenesis and incipient metamorphism.

Strictly speaking, one should compare only peak widths in rocks with sufficient supply of K to achieve maximum illitization of such mixed-layers allowed under the lowest-grade metamorphic conditions attained; availability of such K might be assessed from the presence of unstable detrital K-minerals such as K-feldspar or biotite (cf. KISCH, in press). Porosities and permeabilities should be similarly high, to allow circulation of the K-bearing solutions.

However, since restriction of measurement of illite crystallinity to such rocks would greatly restrict the applicability of the illite-crystallinity method to clay-rich rocks at large, as a compromise we merely restrict ourselves to exclusion of rock

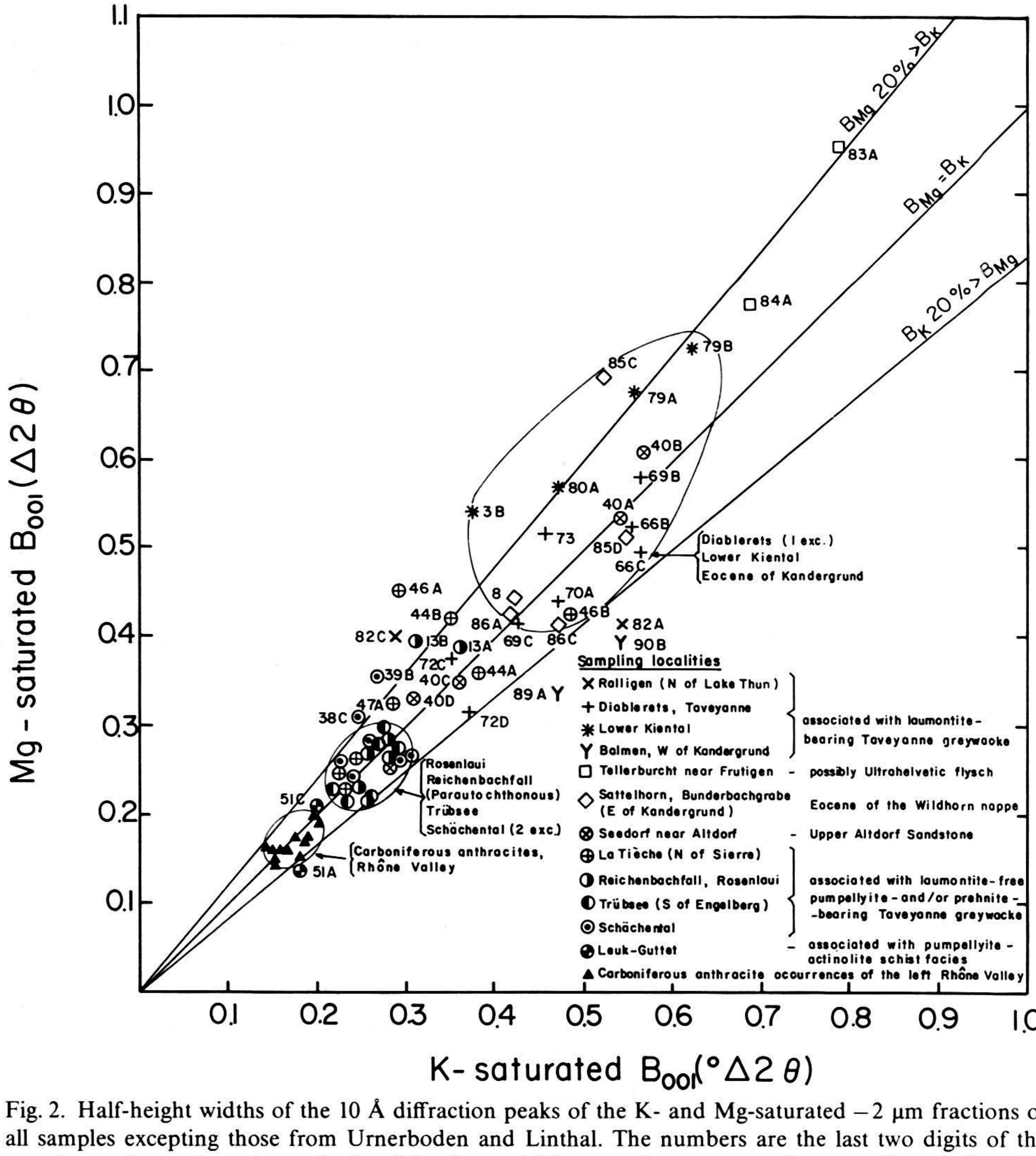


Fig. 2. Half-height widths of the 10 Å diffraction peaks of the K- and Mg-saturated $-2$ µm fractions of all samples excepting those from Urnerboden and Linthal. The numbers are the last two digits of the sample numbers, plus a letter for localities from which more than one sample was collected. Samples with 10 Å reflections of less than 100 cps intensity in both the saturated fractions have been omitted.

types such as highly calcareous shales and marls, which have been shown empirically to give anomalous peak widths (probably due to their low porosity), and of sandstones (in which the micas may be predominantly clastic).

The fact that in virtually all cases where wide differences existed between the peak widths of the K- and Mg-saturated fractions, the glycolated fraction showed a narrower 10 Å peak than either, might suggest adoption of the glycolated peak width as a more dependable parameter of illite crystallinity; this approach has not been adopted in the current paper, but its merits will be discussed in a further study.

*Peak-broadening due to the presence of paragonite and/or pyrophyllite*

A wide range of crystallinities at one locality can be caused by the broadening of peaks in one or more of the samples through presence of another mica or mica-like

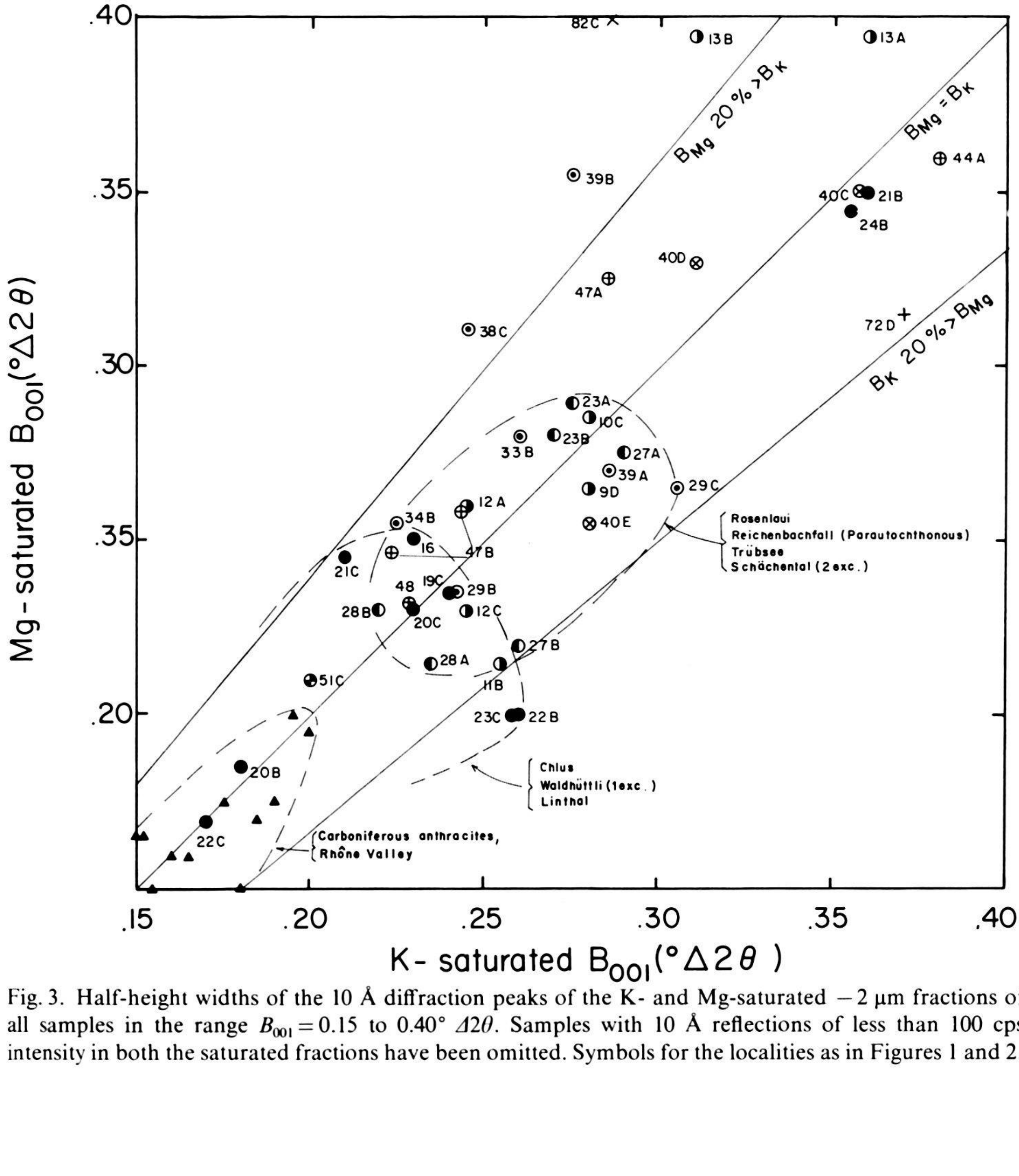


Fig. 3. Half-height widths of the 10 Å diffraction peaks of the K- and Mg-saturated $-2$ µm fractions of all samples in the range $B_{001} = 0.15$ to 0.40° $\Delta 2\theta$. Samples with 10 Å reflections of less than 100 cps intensity in both the saturated fractions have been omitted. Symbols for the localities as in Figures 1 and 2.

mineral, such as pyrophyllite (THOREZ 1976, p. 16), paragonite or the paragonite/phengite mixed-layers described by FREY (1969, 1970, 1978).

Peak broadening due to the presence of paragonite and/or paragonite/phengite mixed-layer may be assumed if these minerals can be identified from their higher-order diffraction peaks at about 4.83 Å and respectively at 3.21 and 3.25 Å, which are usually resolved from the corresponding illite or muscovite peaks; an anomalous peak width due to presence of paragonite should be unaffected by cation saturations or glycolation.

A rather extreme case in point is the peak width of 0.35° $\Delta 2\theta$ measured in a pyrophyllite-rich sample from the Triassic of the Klausenpass (cf. FREY 1970, 1978), which is obviously due to presence of such minor paragonite/muscovite (phengite) mixed-layers, the 001 diffraction peak of which is not resolved from that of muscovite; moreover, the strong pyrophyllite diffraction peak at 9.2 Å is only partially resolved from the illite 10 Å peak.

The 10 Å peak widths of the paragonite-bearing slates from the Carboniferous of the Rhone valley are also likely to be broader than they would be in the absence of paragonite.

## Lowest-grade metamorphic mineralogy: regional distribution of the results

### *Taveyanne greywackes*

As the diagnostic minerals occurring in each area are only schematically represented on a sketch map by MARTINI & VUAGNAT (1965) and no precise localities are given, the writer studied the Taveyanne greywackes at the sampling localities for diagnostic minerals such as laumontite and pumpellyite.

Abundant laumontite was found in all Taveyanne greywackes from Diablerets, the Lower Kiental and Balmen (west of Ausser Kandergrund), and in one of the possibly Ultrahelvetic greywackes from Taveyanne itself. The samples from these localities almost invariably also contained the chlorite-swelling chlorite mixed-layer *corrensite*, identified by its characteristic low-angle peak at about 28–29 Å, which shifted to about 30–31 Å upon glycolation of the sample; the stronger second-order reflection at 14 Å also moved to a somewhat higher d-value upon glycolation. In most of these samples small amounts of bluish green pleochroic pumpellyite were detected optically.

Pumpellyite, in some cases with prehnite, but no laumontite was found in the Taveyanne greywackes of La Tièche (near "Varnerkumme" of MARTINI & VUAGNAT 1965), the parautochthonous of Reichenbachfall near Meiringen, Trübsee near Engelberg and in the Schächen and Urnerboden valleys, in agreement with the findings of MARTINI & VUAGNAT (1965) and FREY (1970). Neither laumontite nor pumpellyite was found in the non-andesitic sandstones at Tellerburcht near Frutigen in the Kandertal and in some of the samples from Taveyanne (both probably Ultrahelvetic) and in the flysch of the Gasperini quarry at Seedorf near Altdorf, presumably due to the absence of suitable primary clastic constituents such as calcic plagioclase and volcanic fragments.

### *Scatter of the illite-crystallinity values*

In most localities, particularly those with high-grade anchizonal or "epizonal" illite crystallinities (all saturated peak widths narrower than about 0.30° $\Delta 2\theta$) the range of peak widths measured in the samples from any one locality is reasonably narrow, enabling unequivocal determination of the degree of incipient metamorphism.

However, at some of the medium-grade localities, a wider range of peak widths covering a range from high-grade "diagenetic" to high-grade anchimetamorphic was encountered, that may be ascribed to differences in either host-rock lithology or tectonic position.

### *Effects of lithology on illite crystallinity*

Outstanding examples of localities showing a wide range of illite 10 Å peak widths are La Tièche and the Seedorf quarry. The mean saturated peak widths at these two localities range respectively from 0.23 to 0.45° $\Delta 2\theta$ (8 samples) and from 0.27 to 0.59° $\Delta 2\theta$ (5 samples).

The comparatively wide 10 Å peaks of the shales and marls associated with Taveyanne greywacke in the northern part of the La Tièche section indicate a low degree of incipient metamorphism, close to the onset of the anchizone. The marked differences in peak widths between the K- and the Mg-saturated fractions (e.g. Z75-44B, 46A and 46B) reflect presence of smectitic mixed-layers, as confirmed by the peak sharpening by up to 0.07° $\Delta 2\theta$ upon glycolation of the unsaturated fractions. The relatively wide peak widths of 0.34 to 0.38° $\Delta 2\theta$ in the glycolated −2 µm fractions exclude higher than low-grade anchizonal degrees of incipient metamorphism.

In contrast, the marls, and particularly the limestones of the southern part of the La Tièche section (Z75-47A, 47B and 48) show mean saturated peak widths of 0.23 to 0.31° $\Delta 2\theta$, indicating high-grade anchimetamorphism. These high illite crystallinities - which are accompanied by much higher $I_{002}/I_{001}$ ratios - in the latter samples may reflect the highly calcareous matrix of the host rocks; however, it is also conceivable that these samples come from a different, lower tectonic unit, viz. from the parautochthonous, which underlies the Diablerets-Gellihorn nappe containing the Taveyanne greywackes.

For the purpose of correlation with the metamorphic mineral assemblages in the Taveyanne greywackes of the La Tièche locality, the lower illite crystallinities of the associated shales and marls have been adopted.

The wide spread of illite-crystallinity values of samples from the Gasperini quarry at Seedorf near Altdorf seems to be related to the fact that they are all from coarse siltstones, in part interlaminated with shale; the peak broadening in one of the samples (Z75-40B) may be due to the presence of paragonitic mica.

The persistence of abundant clastic biotite in all samples measured from this quarry suggests a low degree of incipient metamorphism. Moreover, some of the samples show peak narrowing upon glycolation, indicating presence of smectitic mixed-layers. The locality has thus tentatively been assigned to the low-grade part of the anchizone.

The Trübsee locality above Engelberg provides a good example of the dependence between illite crystallinity and lithology: narrow and extremely weak peaks of less than 0.22° $\Delta 2\theta$ are restricted to greywackes whereas the carbonate-poor slates and marls show mean saturated peak widths of 0.22 to 0.29° $\Delta 2\theta$, which are considered to reflect the high-anchizonal grade of incipient metamorphism in this locality.

### *Regional variation of the illite crystallinity*

The regional variation of the illite crystallinity may be assessed using plots of the peak widths for the K-saturated against the Mg-saturated fractions (Fig. 2 and 3), and an ESQUEVIN diagram showing the relationship between $I_{002(5\,Å)}$ and $B$ for the K-saturated fractions (Fig. 4).

Uniformly "diagenetic" peak widths are associated with the laumontite-bearing Taveyanne greywackes at Diablerets, Lower Kiental and Balmen (west of Kandergrund), and in three other localities in the Kander valley that are not associated with Taveyanne greywacke, Tellerburcht (near Frutigen) and Sattelhorn and Bunderbachgrabe (east of Kandergrund); some of the values found at Taveyanne overlap the onset of the anchizone (~0.38° $\Delta 2\theta$).

The locality at Seedorf (with its wide spread in peak-width values), the northern part of the La Tièche section, and the Wildhorn nappe at Reichenbachfall are considered to be close to the onset of the anchizone (see preceding discussion).

The Lower-Tertiary flysches at all the other localities sampled show unequivocally anchimetamorphic illite crystallinities. The samples from Rosenlaui, the parautochthonous of the Reichenbachfall, Trübsee and the Schächen valley show a particularly well-defined group on all plots; the samples from Chlus and Waldhüttli (both in the Urnerboden valley) and Linth valley show a partly overlapping group on the $B_{Mg\text{-sat}}$ vs. $B_{K\text{-sat}}$ plots (Fig. 2 and 3).

Saturated peak widths between 0.15 and 0.20° $\Delta 2\theta$ - characterizing KÜBLER's "epizone" - predominate in the slates from the anthracite mines in the Carboniferous of the left Rhone valley southwest of Sierre and Sion, Valais.

## Coal rank and its relation with lowest-grade metamorphic mineralogy

### *Vitrinite reflectance*

Vitrinite reflectance was measured on samples from the majority of the localities sampled. In many of the samples no vitrinite reflectance could be measured, either due to the extremely small grain size of the vitrinite, or the predominance of inertinite macerals. Results of the vitrinite-reflectance measurements are listed in Table 2.

### *Correlation of coal rank with zeolite facies in the Taveyanne greywacke*

The coal ranks measured at the laumontite-bearing localities Diablerets, Taveyanne and Lower Kiental range from 0.85 to 1.3% $R_{max}$ (i.e. high-volatile $A$ and medium-volatile bituminous rank).

The higher reflectances of 1.65 to 1.7% $R_{max}$ (i.e. low-volatile bituminous rank) measured at Balmen, west of Kandergrund, are from loose material formerly mined from the Wildhorn nappe tectonically overlying the Gellihorn nappe (which contains the laumontite greywackes), and thus cannot directly be correlated with the zeolite occurrences.

In all the localities with Taveyanne greywacke without laumontite - including all the Eocene flysches east of the Kien valley with the exception of the Seedorf quarry,

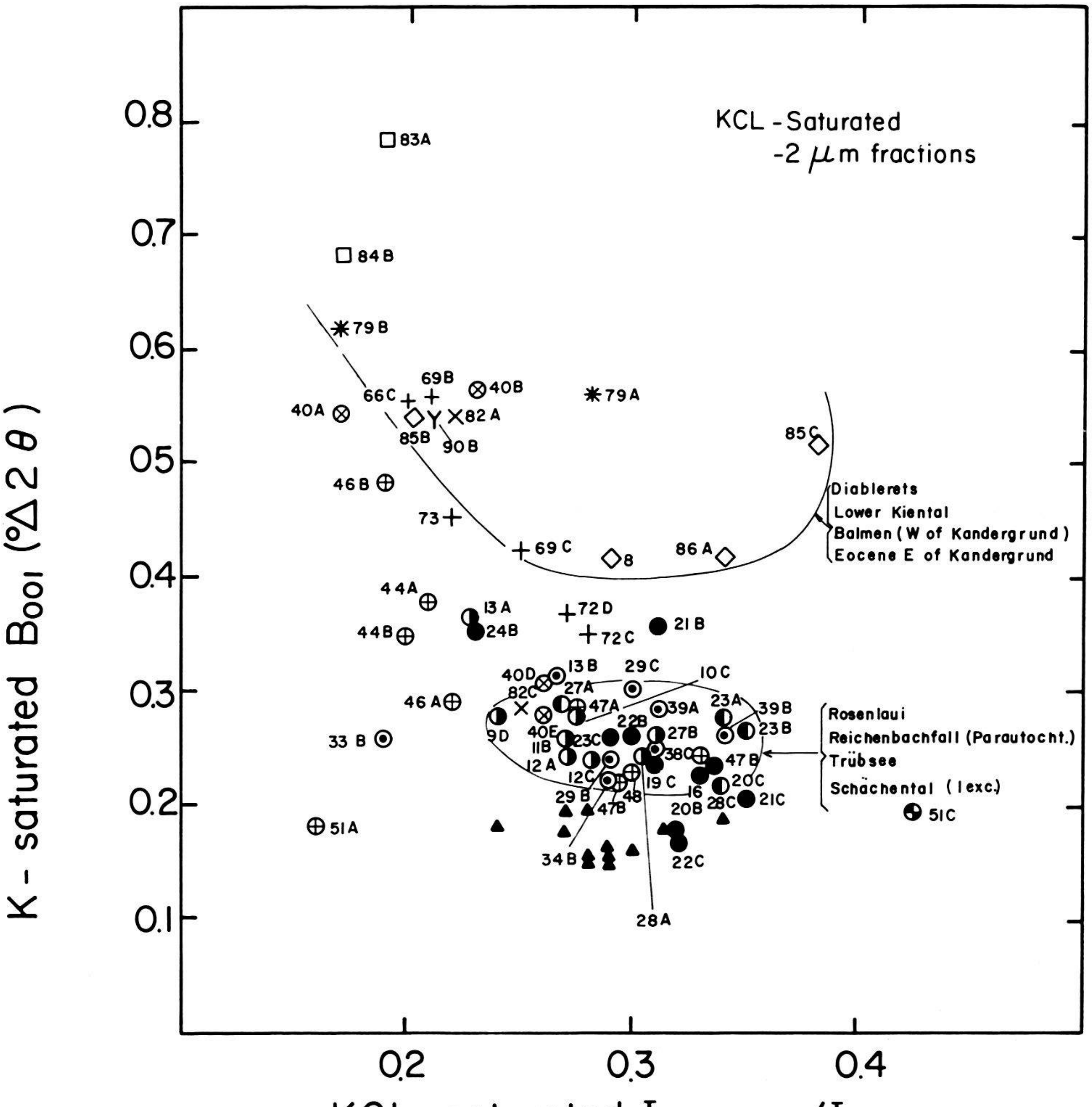


Fig. 4. ESQUEVIN diagram showing the relationship between the width of the illite 10 Å peak at half height, and the ratio of the intensities of the illite (002) and (001) diffraction peaks for the KCl-saturated −2 μm fractions. The numbers are the last two figures of the sample numbers (and the letter for localities from which more than one sample was collected).
Samples with 10 Å reflections of less than 100 cps intensity, and/or with 5 Å reflections with signal/noise ratios of less than 2.5 have been omitted.

Table 2: *Vitrinite reflectance of coaly matter from predominantly Lower Tertiary clastic rocks in the Helvetic zone of the Swiss Alps and some Carboniferous rocks in the Rhone valley, Valais.*

| Locality | Sample nr | $R_{max}$ (oil) % range (nr of measurements) | $R_{max}$ (oil) % mean | |
|---|---|---|---|---|
| Associated with laumontite-bearing Taveyanne greywackes (mainly Diablerets-Gellihorn nappe) | | | | |
| Diablerets | Z75-66A | 0.75-1.00 (4) | 0.90 | |
| | Z75-68 | 0.91-1.21 (4) | 1.03 | |
| | Z75-69B | 1.23 (1) | 1.23 | |
| | Z75-70A | 1.20-1.32 (7) | 1.27 | |
| Taveyanne (poss. Ultrahelvetic in part) | Z75-73 | 1.25-1.26 | 1.25 | |
| Kiental | Z75-79A | 0.79-0.90 (7) | 0.85 | |
| | Z75-79B | 1.19-1.21 (2) | 1.20 | |
| Balmen, W of Kandergrund | Z75-87E | 1.51-1.70 (7) | 1.65 | loose; ? Wildhorn |
| | Z75-89B | 1.54-1.70 (6) | 1.68 | |
| Associated with flysches without diagnostic Ca-Al-silicates ("diagenetic" to low-grade anchizonal illite crystallinities) | | | | |
| Tellerburcht (poss. Ultrahelvetic) | Z75-83A | 0.97-1.18 (8) | 1.07 | |
| Sattelhorn (Wildhorn nappe) | Z75-85A | 2.13-2.34 (17) | 2.29 | |
| | Z75-85B | 1.86-2.14 (10) | 1.97 | |
| Bunderbachgrabe (Wildhorn nappe) | Z75-86A | 0.87-1.09 (25) | 0.96 | |
| | Z75-86B | 0.83-0.97 (3) | 0.88 | |
| Seedorf nr Altdorf (autochthonous) | Z75-40A | 2.10 (2) | 2.10 | |
| | Z75-40C | 2. 8-3. 1 (16) | 2.91 | |
| | Z75-40E | 2. 6 (1) | 2.60 | |
| Associated with laumontite-free Taveyanne greywackes, and with flysches without diagnostic Ca-Al-silicates (anchizonal illite crystallinities) | | | | |
| La Tièche | Z75-44A | 3. 3 (1) | 3. 3 | |
| | Z75-47A | 3. 8-4. 1 (2) | 3.95 | |
| | Z75-49A | 3. 8 (1) | 3. 8 | |
| | Z75-50A | 3. 2-3.55 | | |
| Rosenlaui | Z75-11B | 3. 1 (1) | 3. 9 | |
| Reichenbachfall (Parautochthonous) | Z75-9B | 3. 7-4. 1 (7) | 3.89 | |
| | Z75-10C | 3.75-3. 9 (3) | 3.80 | |
| (Aalenien of Wildhorn nappe) | Z75-13A | 2. 6-2. 8 (4) | 2.67 | |
| | Z75-13B | appr. 3.3 (1) | | |
| Oberalp-Äsch (Parautochthonous) | Z75-33A | 3. 4-3. 6 (5) | 3.47 | |
| | Z75-33B | 3.25-3. 4 (6) | 3.34 | |
| Waldhüttli (Griesstock nappe) | Z75-21B | 3. 5 (1) | 3. 5 | |
| Carboniferous anthracites of the left Rhône Valley, Valais | | | | |
| Réchy | Z75-55A | 3. 7-4. 6 (50) | 4.38 | |
| Grône-Réchy | Z75-58B | 4. 1-4. 4 (12) | 4.25 | |
| Haute Nendaz | Z75-59A | 2.15-3. 2 (13) | 2.65 | |
| | Z75-60A | 1.84-2.33 (7) | 1.97 | |

as well as the La Tièche locality - anthracitic reflectances between 3.3 and 4.2% $R_{max}$ were measured, corresponding to 5 to 4% V.M.

These results show that the laumontite occurrences are associated with appreciably lower coal ranks than the laumontite-free pumpellyite- and/or prehnite-bearing greywackes; however, the absence of determinations of semi-anthracitic ranks in association with Taveyanne greywackes leaves a rather unsatisfactorily wide wange of uncertainty regarding the coal rank associated with the prograde disappearance of laumontite.

*Correlations of coal rank with illite crystallinity*

Reflectances associated with unequivocally "diagenetic" illite crystallinities (Diablerets, Taveyanne, Lower Kiental, Balmen, Tellerburcht and Bunderbachgrabe) of $B > 0.39° \Delta 2\theta$ range up to 2.3% $R_{max}$ (semi-anthracite).

However, within this lowest-grade group of localities, there is some lack of correlation between vitrinite-reflectance and illite-crystallinity values: the lowest reflectances, 0.85 to 1.3% $R_{max}$ are found at Diablerets, Lower Kiental and Tellerburcht, but also at Taveyanne and Bunderbachgrabe, which show some illite crystallinities approaching the onset of the anchizone.

Slightly lower coal rank of 0.79% $R_m$ has been reported by TEICHMÜLLER & TEICHMÜLLER (1978) from the Eocene of the Wildhorn nappe of the Gemmen-Alphorn, some 6 km northwest of Merligen (north of Lake Thun).

Some other localities from the Wildhorn nappe in the Kandergrund area show similar "diagenetic" illite crystallinities, but appreciably higher reflectances, e.g. 1.9 to 2.3% $R_{max}$ at Sattelhorn; the reflectances of 1.6 to 1.7% $R_{max}$ at Balmen probably also come from the Wildhorn nappe, which tectonically overlies the laumontite greywackes and the associated "diagenetic" illite crystallinities.

For the time being the author has no satisfactory explanation for the comparatively advanced coal rank with respect to illite crystallinity in several occurrences from higher tectonic units the above from the Wildhorn nappe and also in the possibly Ultrahelvetic of Tellerburcht, with its very low illite crystallinities.

In the localities that have been tentatively assigned to the onset of the anchizone the reflectances are generally higher: from 2.1 to 2.9% $R_{max}$ (semi-anthracite to low-rank anthracite) at Seedorf, 2.6 to 3.3% $R_{max}$ in the Aalenien of the Wildhorn nappe, and 3.3% $R_{max}$ in the northern part of the La Tièche section.

All localities with unequivocally anchizonal illite crystallinities (Rosenlaui; Reichenbachfall/parautochthonous; Trübsee; Oberalp-Äsch; Waldhüttli) have coal ranks of $R_{max}$ 3.3 to 3.9% $R_{max}$, i.e. high-anthracitic ranks. The onset of anchimetamorphism as apparent from illite crystallinity thus appears to be approximately in the range 2.3 to 3.3% $R_{max}$, i.e. high-rank semi-anthracite and low-rank anthracite (ASTM classification).

The total number of vitrinite grains measured at most localities, less than 5, is insufficient to distinguish between higher-rank and lower-rank localities within the "anthracitic" group. Of the few localities in which more than 10 dependable measurements could be made, Oberalp-Äsch (Schächental), with 3.3 and 3.5% $R_{max}$ (or about 5% V.M.), seems to be slightly lower-rank than the parautochthonous at Reichenbachfall (3.8 and 3.9% $R_{max}$ or about 4.5% V.M.) and a similar value at

Rosenlaui (3.9% $R_{max}$); these differences are not matched by parallel differences in illite crystallinity, which ranges 0.23–0.31° $\Delta 2\theta$ (K-sat and Mg-sat) in both cases.

A problem in matching coal rank with wide-ranging illite crystallinities is posed at La Tièche; the high-rank anthracitic reflectances found (3.3, 3.8 and 3.9% $R_{max}$ or about 5 to 4.5% V.M.) are at variance with the relatively low illite crystallinities of the silty shales earlier interpreted to represent the degree of incipient metamorphism, and tally better with the 0.22–0.29° $\Delta 2\theta$ (K-sat) or 0.23–0.33° $\Delta 2\theta$ (Mg-sat) of the limestones and marls; the latter interpretation is also more consistent with the absence of laumontite from the Taveyanne greywackes. Closer investigation of this locality seems called for.

The anthracitic coal ranks in the Carboniferous zone left of the Rhone between Sierre and Sion, Valais, were confirmed by mean reflectance measurements of 3.7 to 4.6% $R_{max}$ at Réchy and Grône. KÜNDIG & DE QUERVAIN (1953, Table 1) quote $CO_2$-free volatile-matter yields of 4.3 to 8.0% (d.a.f.) for coals with 20 to 40% ash (dry basis) from this area.

Lower reflectances of 1.8 to 3.2% $R_{max}$ were measured on two samples from Haute-Nendaz, southwest of Sion, where the $CO_2$-free volatile-matter yields quoted by KÜNDIG & DE QUERVAIN (ibid.) are higher, 8.0 to 11.4% (d.a.f.) for coals with 11 to 27% ash (dry basis), i.e. semi-anthracite. The fact that the lower coal ranks from Haute-Nendaz are not matched by broader illite 10 Å peaks may well be related to the slates of this locality being the only ones from the Carboniferous zone to lack paragonite. The presence of paragonite in all slates from the remaining Carboniferous anthracite localities may indicate their higher degree of metamorphism, and cause the broadening of the apparent widths of the 10 Å diffraction peaks.

TEICHMÜLLER & TEICHMÜLLER (1978) report a rather higher average reflectance of 6.83% $R_{max}$ for Carboniferous meta-anthracites within the Helvetic massifs, which were not sampled for the present study.

### *Comparison with other areas*

The coal rank associated with the onset of the anchizone in the Helvetides shows good correspondence with that found in the Cambro-Silurian of the Jämtland Group in western central Sweden (KISCH 1980) in which the onset of anchizonal illite crystallinities is associated with reflectances around 2.7% $R_{max}$, i.e. close to the semi-anthracite to anthracite boundary, whereas all "diagenetic" illite crystallinities were associated with reflectances of less than 2.9% $R_{max}$.

KISCH (1974) suggested on the basis of the scattered evidence available at the time, that the onset of the anchizone was by and large associated with coals of anthracitic rank, in several cases middle- or high-rank anthracite (less than 6% V.M.) in some cases low-rank anthracite.

The areas in which low-rank anthracite already was inferred to appear in the advanced "diagenetic" zone included the Jurassic "Terres Noires" of the subalpine belt of the French Alps, and the Upper-Triassic "Quartenschiefer" of the Helvetic zone, Glarus Alps.

The association of highest-grade "diagenetic" to lowest-grade anchizonal illite crystallinities in the Lower Cretaceous of British Columbia (FOSCOLOS & KODAMA

1974; FOSCOLOS et al. 1976) with vitrinite reflectances of about 2.53% $R_0$, i.e. highest-rank semi-anthracite, agrees with the latter relationship.

However, other terrances show association of the onset of anchizonal illite crystallinities with higher, high-rank anthracitic coal ranks; such relationships are generally associated with heating by deep plutons, or high geothermal gradients generally (WOLF 1975). However, they are also found in the Mesozoic of the French Alps (CHATEAUNEUF et al. 1973; ROBERT 1971; BARLIER 1974; BARLIER et al. 1974), which is rather surprising in view of the present results from the Swiss Alps.

If further data confirm the association of the onset of the anchizone with higher coal ranks in Mesozoic than in Lower-Tertiary rocks, this difference might conceivably reflect either the influence of greater depth of burial of the Mesozoic before folding, or possibly the influence of earlier ("Eo-Alpine") folding phases. However, much additional data and refinement will be required until such speculations can be substantiated.

The association of the epizonal illite crystallinities in the Carboniferous of the Rhone valley with anthracitic reflectances not much higher than those from the anchizonal localities in the flysch between the Reuss and Linth valley, and at La Tièche, is similar to the relationship found earlier in the higher-grade zones in the Cambro-Silurian of Jämtland (KISCH 1980). In the highest-grade zone *D* the vitrinite reflectances are largely similar to those in the lower-grade zones *C* and *B*, with lower illite crystallinities. This might indicate that reflectance is not an accurate parameter of degree of metamorphism within the range high-grade anchizone to low-grade "epizone", at least in terranes with medium to high pressure type regional metamorphism. DTA studies currently in progress indicate that the temperature of the exothermic effect upon heating may provide a more sensitive distinction between the coal ranks in these zones of incipient metamorphism.

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Since the text of the present paper was concluded, the following papers dealing with illite crystallinity and coal rank data on the external zones of the Swiss Alps and Haute Savoie have appeared:

FREY, M., TEICHMÜLLER, M., TEICHMÜLLER, R., MULLIS, J., KÜNZI, B., BREITSCHMID, A., GRUNER, U., & SCHWIZER, B. (1980): *Very low-grade metamorphism in external parts of the Central Alps: illite crystallinity, coal rank and fluid inclusion data.* - Eclogae geol. Helv. *73/1*, 173-203.

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