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## Very low-grade metamorphism in external parts of the Central Alps: Illite crystallinity, coal rank and fluid inclusion data

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### ABSTRACT

Very low-grade regional Alpine metamorphism has been studied along three cross sections of the Helvetic Alps (Kien valley, Lake Lucerne-Reuss valley and Glarus Alps) and in the Pennine Falknis nappe. Illite crystallinity ( $IC$ ) and coal rank by means of the reflectivity ( $R_m$  and  $R_{max}$ ) were determined for 107 samples. In addition, the fluid inclusions from fissure quartz at 50 localities were studied and yielded information about the fluid composition during metamorphism and, in some cases, minimum temperatures and pressures of formation.

In general,  $IC$  and  $R_m$  increase from tectonically higher to lower units and from external to internal parts within the same tectonic unit. Deep diagenetic conditions occur in the Border chain, the northern part of the Wildhorn nappe, the Drusberg nappe and the northern part of the Axen nappe. Anchizone conditions are reached in the Gellihorn nappe and the middle to southern part of the Axen nappe while anchi- to epimetamorphic conditions are found in the Doldenhorn nappe and in the autochthonous cover of the Aar massif.

The fluid composition in fluid inclusion from fissure quartz crystals shows a general evolution related to metamorphic grade as determined by  $IC$  and  $R_m$  data. In the deep diagenetic zone, fluids with >1 mole-% of higher hydrocarbons were found with homogenization temperatures of less than 200 °C. The low- and medium-grade anchizone is dominated by methane-bearing fluids with minimum formation temperatures between 200° and 270 °C. In the higher-grade anchizone and the epizone, water-bearing fluids were encountered.

The following examples of inverted metamorphism have been found, i.e. cases where higher-grade units have been thrust on lower-grade units: a) The high-grade anchimetamorphic Pennine Niesen nappe is lying on Helvetic units belonging to the deep diagenetic zone. b) The southern part of the Axen nappe with medium to high anchizone grade is thrust on the North Helvetic Flysch zone with deep diagenetic to anchimetamorphic conditions. Post-metamorphic thrusting, therefore, is an important phenomenon in external parts of the Central Alps.

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Good but different correlations between  $IC$ ,  $R_m$  (or  $R_{max}$ ) and fluid inclusion data were found for the Kien valley and Lake Lucerne-Reuss valley sections. No generally valid relationship between these three parameters seem to exist.

Divergent relations between  $IC$ ,  $R_m$  and fluid inclusion data were found in the Pennine Falknis nappe. According to the  $IC$  data this nappe either belongs to the diagenetic or medium-grade anchimorphic zone. However, the  $R_m$  data indicate deep diagenetic to anchizonal conditions during a prekinematic stage with some additional syn- to postkinematic coalification under anchizonal conditions. Fluid inclusion data indicate medium-grade anchizonal conditions for the upper- and higher-grade anchizonal conditions for the lower part of the Falknis nappe.

## ZUSAMMENFASSUNG

Die schwache alpine Regionalmetamorphose wurde an drei Profilen durch das Helvetikum (Kiental, Urnersee-Reusstal, Glarner Alpen) und in der penninischen Falknis-Decke studiert. An 107 tonigmergeligen Proben wurden die Illit-Kristallinität ( $IK$ ) und der Inkohlungsgrad des organischen Materials mit Hilfe des mittleren bzw. maximalen Reflexionsvermögens ( $R_m$  bzw.  $R_{max}$ ) bestimmt. Zusätzlich wurden an Kluftquarzen von 50 Lokalitäten die Gas- und Flüssigkeitseinschlüsse studiert, um Angaben über die Zusammensetzung der fluiden Phase während der Metamorphose und, wenn möglich, minimale Bildungstemperaturen und -drücke zu ermitteln.

Im allgemeinen nehmen  $IK$  und  $R_m$  von tektonisch höheren zu tektonisch tieferen Einheiten zu, desgleichen von der Stirn zur «Wurzel» innerhalb derselben Decke. Die Randkette, der nördliche Teil der Wildhorn-Decke, die Drusberg-Decke sowie der Nordteil der Axen-Decke gehören noch dem Diagenesebereich an. In der Gellihorn-Decke und im mittleren und südlichen Teil der Axen-Decke wurden anchimetamorphe Bedingungen angetroffen, während die Doldenhorn-Decke und das Autochthon des Aar-Massivs in den Grenzbereich von Anchi- zu Epimetamorphose zu stellen sind.

Die Zusammensetzung der Gas- und Flüssigkeitseinschlüsse der Kluftquarze zeigt einen einfachen Zusammenhang mit dem Metamorphosegrad. Höhere Kohlenwasserstoffe ( $> 1$  Mol-%) sind charakteristisch für den starken Diagenesebereich mit maximalen Homogenisations-Temperaturen von  $200^\circ\text{C}$ . Die schwach- und mittelgradige Anchizone zeichnet sich durch methanhaltige Einschlüsse aus mit minimalen Bildungstemperaturen von 200 bis  $270^\circ\text{C}$ . Wasserhaltige Einschlüsse sind typisch für die höhergradige Anchizone und die Epizone.

In folgenden Fällen konnte eine invertierte Metamorphose nachgewiesen werden, indem tektonisch höhere Einheiten einen stärkeren Metamorphosegrad aufweisen: a) Die stark anchimetamorphe Niesen-Decke liegt auf helvetischen Einheiten, welche dem Diagenesebereich angehören. b) Der mittel- bis hochgradig anchimetamorphe Südteil der Axen-Decke ruht auf nordhelvetischem Flysch, welcher dem Grenzbereich Diagenese-Anchimetamorphose angehört. Postmetamorphe Überschiebungen spielen demnach in den Externbereichen der Zentralalpen eine bedeutende Rolle.

Für zwei Profile wurden gute, aber unterschiedliche Korrelationen zwischen  $IK$ ,  $R_m$  (bzw.  $R_{max}$ ) und der Zusammensetzung der Gas- und Flüssigkeitseinschlüsse ermittelt. Zwischen den drei Parametern scheint keine allgemeingültige Beziehung zu bestehen. Für die Falknis-Decke zeigen die drei Methoden voneinander abweichende Ergebnisse. Die  $IK$  weist auf Bedingungen der Diagenese oder der mittelgradigen Anchizone hin. Demgegenüber weist  $R_m$  auf Bedingungen im Grenzbereich Diagenese-Anchimetamorphose während eines präkinematischen Stadiums hin, gefolgt von einer zusätzlichen syn- bis postkinematischen Nach-Inkohlung. Die Untersuchungen an Gas- und Flüssigkeitseinschlüssen verweisen die höheren Anteile der Falknis-Decke in die mittelgradige, die tieferen Anteile hingegen in die höhergradige Anchizone.

## 1. Introduction

Conditions of incipient metamorphism may be assessed through a) the determination of mineral equilibria, b) the illite crystallinity, c) coal rank data and d) fluid inclusion studies. Basic volcanic rocks or graywackes are particularly sensitive to low temperature alterations (e.g. COOMBS 1954). Pelitic and marly rocks, on the other hand, show relatively few zone markers (ZEN & THOMPSON 1974) at these low temperatures, but the continuous increase of illite crystallinity (KÜBLER 1967a,

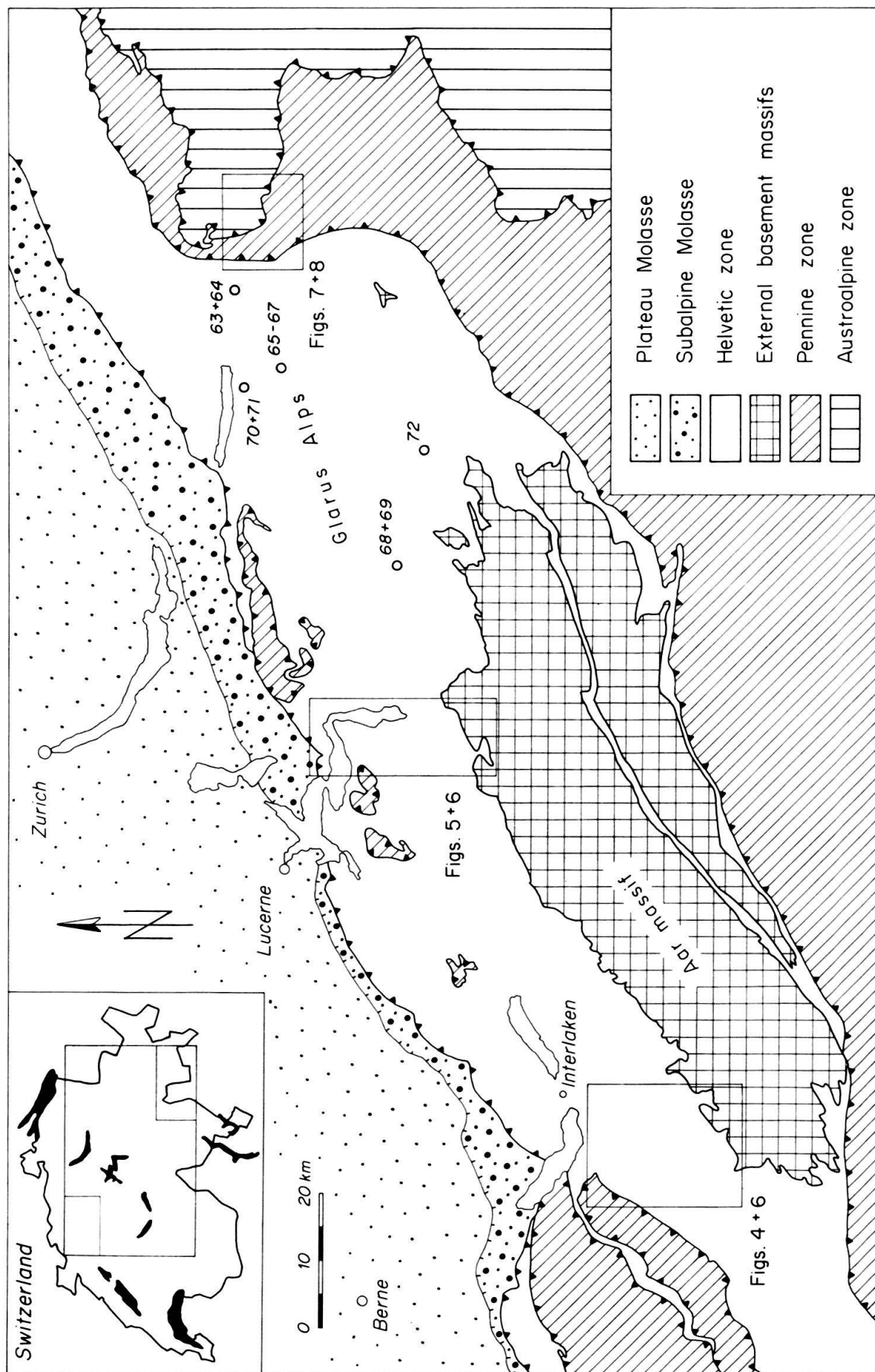


Fig. 1. Simplified geologic map of the Central Alps, Switzerland, showing studied areas (Fig. 4-8) and single sample localities of the Glarus Alps (63-72).



1968) may be used as an indication of metamorphic grade (e.g. DUNOYER 1970). Coal rank determinations by reflectivity measurements on carbonaceous material is a powerful tool in recognizing an increase in temperature (e.g. TEICHMÜLLER 1970; BOSTICK 1974, STACH et al. 1975). Fluid inclusion studies can provide important information on the composition of the fluid phase during metamorphism. In addition, this method may furnish temperature and pressure estimates independent of those derived from the mineral equilibria provided that the fluid inclusions were trapped in their host crystal near the peak of metamorphism (e.g. MULLIS 1979).

Recognition of very low-grade metamorphism in the external parts of the Central Alps - the Helvetic nappes, the Prealpes and related Klippen (see Fig. 1) - began in the fifties when NIGGLI et al. (1956) described stilpnomelane and riebeckite as products of Alpine metamorphism in sediments of the eastern Aar massif. In the following years research was aimed at the Taveyanne graywacke carrying assemblages with laumontite + corrensite, pumpellyite, prehnite, epidote and rarely pumpellyite + actinolite (MARTINI & VUAGNAT 1965, 1970; KÜBLER 1973; KÜBLER et al. 1974; COOMBS et al. 1976); pelitic and marly rocks were shown to contain, besides illite and chlorite, pyrophyllite, mixed-layer paragonite/muscovite, paragonite and rarely chloritoid (FREY 1970, 1978; FREY & WIELAND 1975; WIELAND 1976); and finally, glauconite-bearing formations yielded assemblages with stilpnomelane, K-feldspar and biotite (FREY et al. 1973).

Coal rank data based on earlier chemical analyses from the external parts of the Central Alps were summarized by FREY & NIGGLI (1971). Data based on reflectivity measurements have been published only recently (TEICHMÜLLER & TEICHMÜLLER 1978; KÜBLER et al. 1979, STALDER 1979), and further work is in progress (KISCH, in press).

Fluid inclusion studies in the external zones of the Central Alps are still in their infancy. Homogenization temperatures up to 220 °C were obtained by STALDER & TOURAY (1970) on fissure quartz from many localities in the Helvetic nappes between Lake Lucerne and the Rhone valley. These temperatures are believed to represent minimum temperatures of metamorphism. In the parautochthonous flysch of the Val d'Iliez, the first generation of fissure quartz formed at minimum temperatures of around 250 °C and at a minimum pressure of about 1.5 kbar (MULLIS 1975, 1976). All these fluid inclusions from fissure quartz show high proportions of CH<sub>4</sub> and H<sub>2</sub>O and very low proportions of CO<sub>2</sub> and higher hydrocarbons (HHC).

A strong correlation between the fluid composition of fissure quartz and Alpine regional metamorphism, based on illite crystallinity data and mineral assemblages, has been established for the outer zones of the Central Alps (MULLIS 1979). In the diagenetic or nonmetamorphic zone, included fluids consist of HHC (>1 mole-%) + CH<sub>4</sub> + H<sub>2</sub>O + CO<sub>2</sub>, and this zone was called the HHC-zone (HHC-bearing fluid inclusions); in the low- and medium-grade anchizone, the fluid inclusions contain CH<sub>4</sub> + HHC (<1 mole-%) + H<sub>2</sub>O + CO<sub>2</sub> defining the CH<sub>4</sub>-zone (CH<sub>4</sub>-bearing fluid inclusions); in the higher-grade anchizone and the epizone only H<sub>2</sub>O + CO<sub>2</sub> were found defining the H<sub>2</sub>O-zone (H<sub>2</sub>O-bearing fluid inclusions). Other gas species not detectable with microthermometry are found in small quantities: Ar (J. Hunziker, pers. communication 1979) and H<sub>2</sub>S (DHAMMELINCOURT et al. 1979).

It is the purpose of the present paper

- a) to show how coal rank, illite crystallinity and fluid inclusion data can contribute to the understanding of very low-grade metamorphism in external parts of the Central Alps,
- b) to present new information on the relationship between coal rank, illite crystallinity and the composition of fluid inclusions and finally
- c) to discuss the interpretation of P-T data from fluid inclusion studies.

As study areas three cross sections through the Helvetic nappes and an additional one through the Pennine Falknis nappe were chosen (Fig. 1). For these areas detailed mineralogical data are already available or work is in progress as follows:

- a) Kien valley area: KÜNZI (1975) and thesis work in progress;
- b) Lake Lucerne - Reuss valley: A. Breitschmid, thesis work in progress;
- c) Glarus Alps: FREY (1969, 1970, 1978) and work in progress;
- d) Falknis nappe: GRUNER (1976) and B. Schwizer, thesis work in progress.

The following abbreviations will be used:

HHC = higher hydrocarbons

IC = coefficient of illite crystallinity

$R_m, R_{max}, R_{min}$  = mean, maximum, minimum reflectivity

## 2. Methods

### *Material studied*

Coal rank and illite crystallinity determinations were performed on the same hand specimen, except at locality No. 4, where two different specimens were used. The fissure quartzes for fluid inclusion studies had to be sampled in most cases from different outcrops. Within one fissure, always the earliest formed quartz generation was studied.

### *Illite crystallinity*

The so-called "illite crystallinity", that is the width of the first order illite basal reflexion at half height (KÜBLER 1967), was measured with an X-ray diffractometer. The X-ray mounts were prepared by sedimentation of the  $< 2 \mu$  fraction, and air-dried or glycolated. Glycolated mounts were prepared in a glycol steam bath at 70 °C overnight. The fraction  $< 2 \mu$  was obtained by sedimentation in Atterberg cylinders of samples finely ground in a tungsten-carbide Sieb mill for 30 sec. For the separation of different grain size fractions (Table 3) the mechanical grinding was avoided and rock chips were disintegrated in an ultrasonic bath. Illite crystallinity standards were kindly provided by Prof. B. Kübler. The nonmetamorphic (or diagenetic)/anchimetamorphic boundary and the anchimetamorphic/epimetamorphic boundary correspond to a crystallinity index of 7.5 and 4.0, respectively.

### *Coal rank determination*

Coal rank was determined through reflectivity measurements on finely dispersed organic matter, vitrinite if possible (for details see STACH et al. 1975, p. 263-273 and 303-307). Working conditions with a Leitz Orthoplan photometer microscope were as follows: oil-immersion, monochromatic polarized light (546 nm), magnification  $\times 500$ ,  $3 \mu$  area of measurement.

From each sample a polished, untreated sample cut perpendicular to the bedding and a particulate polished block (grain size 3 mm) were used. Average values of  $R_m, R_{max}$  and  $R_{min}$  were almost identical for both sections. Therefore, the arithmetic mean of each pair of single values was calculated. For samples 22, 23, 88-90 and 99, which were very poor in coaly material, the organic matter was concentrated by a chemical treatment with HCl and HF.

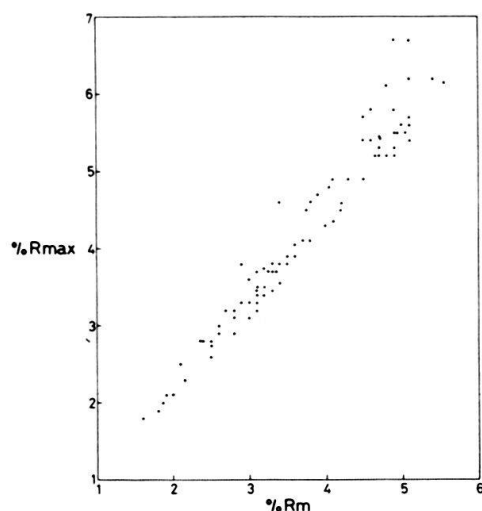


Fig. 2. Correlation of the mean reflectivity ( $R_m$ ) versus the maximum reflectivity ( $R_{max}$ ) of the samples studied (see text).

Due to the limited number of  $R_{max}$  determinations (in many cases the particle size was too small for measurements of  $R_{max}$  and  $R_{min}$ ) the  $R_m$  values are shown graphically in Figures 4–6. However, as shown in Figure 2, there exists a good linear relationship between  $R_m$  and  $R_{max}$  values up to values of 4.8%  $R_m$  and 5.4%  $R_{max}$ , respectively (see also TEICHMÜLLER & WEBER 1979).

Most samples were extremely poor in measurable coaly inclusions. For many limestones and marls earlier bituminous constituents (meta-bituminite or meta-exsudatinite) had to be included in the reflectivity measurements. Such a procedure is possible in the anthracitic stage where meta-bituminite and vitrinite reach very similar reflectivities. The small number of measuring points ( $n$ ) used for many samples might throw some doubt on the quality of our data. As a check on this, the  $R_m$  values of always two different samples from the same outcrop were plotted in Figure 3. This figure seems to indicate that reliable data can be obtained even with a small number of measuring points.

In a few cases fluorescence microscopy was used as a support of the reflectivity measurements.

#### Fluid inclusion studies

Fluid inclusions were studied by microthermometric methods, that is, temperatures of phase transitions were determined on a microscope equipped with a heating and cooling stage (type Chaix Meca). Quartz slabs, 0.5–0.8 mm thick and polished on both sides, were used.

The fluid composition was derived from the homogenization temperatures ( $T_{hom}$ ), which are as follows for the fluids encountered in this study:  $T_{hom} < -82.4^\circ\text{C}$  for methane with  $< 1$  mole-% of HHC;  $T_{hom} > -82.4^\circ\text{C}$  for methane with  $> 1$  mole-% of HHC;  $T_{hom} > 80^\circ\text{C}$  for water.

Minimum temperature and pressure conditions during the trapping event of methane-bearing fluids are determined by three steps: a) Methane-rich fluid inclusions (with a purity greater than 99 moles-%)

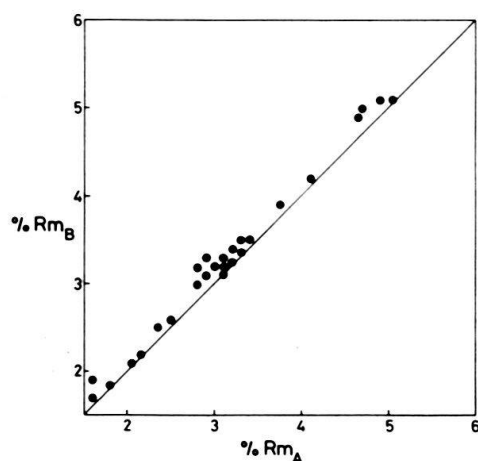


Fig. 3. Plot of the mean reflectivity ( $R_m$ ) of two different samples ( $A$  and  $B$ ) from the same locality (see text).

Table 1a: Coal rank and illite crystallinity data for section I: Kien valley.

No. original specimen	coordinates/elevation	tectonic/stratigraphic position	lithology	Rm (%)	Rmax (%)	Rmax-Rmin	n	IC glyc/air	remarks
1	BK 74/12	Ni Flysch	slate	5.1	5.7	1.3	14(4)	n.d./(6.8)	pa/mu
2	BK 75/124	Zc Schattwald-Zone	marl	2.1	2.5	0.5	5	10.7/19	ka
3	BK 75/95	Zc Wildflysch	limestone	2.8	2.9	0.2	14	8.5/12	
4	BK 75/33	Wi Brackwasser-Sch.	marly lst.	2.5	2.6	0.3	50	8.2/n.d.	
5	BK 75/23	Wi "Oxfordien"	marly lst.	3.8	4.1	0.6	13(7)	6.5/7.1	
6	BK 75/108	Wi "Oxfordien"	marly lst.	3.7	4.1	0.6	22	5.5/n.d.	
7	BK 75/77b	Wi Glockhaus-Serie	slate	3.4	4.6	2.0	27	5.4/n.d.	
8	BK 75/99	Wi Glockhaus-Serie	slate	4.3	4.9	1.1	25	(7.5/8.3)	pa/mu
9	BK 75/40	IF Dachschiefer	slate	2.0	2.1	0.3	11	7.7/9.3	
10	BK 75/18	IF Dachschiefer	slate	2.4	2.8	0.6	26(16)	6.3/6.7	
11	BK 75/43	IF Dachschiefer	slate	3.1	3.2	1.1	7(5)	6.9/7.5	
12	BK 75/90	?IF Wildflysch	slate	3.6	3.9	0.7	22	5.3/n.d.	
13	BK 75/146	IF Dachschiefer	slate	4.1	4.9	1.4	21	4.5/4.7	
14	BK 75/161	Ge Zementstein-Sch.	marly lst.	5.1	5.4	0.7	13(3)	6.1/6.3	
15	BK 73/8	Ge Zementstein-Sch.	marly lst.	5.55	6.15	1.2	27	n.d./4.1	
16	BK 75/86	Do Oehrlialk	marly lst.	4.8	5.2	0.7	12	3.8/n.d.	
17	BK 75/151	Do "Tertiary"	marly lst.	4.8	6.1	2.1	8	4.6/4.9	
18	BK 75/103a	Do Zementstein-Sch.	marly lst.	4.9	5.5	1.3	5	n.d./3.1	
19	BK 75/117	Do "Dogger"	slate	4.9	5.8	1.4	20	3.7/n.d.	
20	BK 75/155	Au "Tertiary"	marly lst.	5.4	6.2	1.4	4	3.5/3.5	
21	BK 75/111	Au "Dogger"	marly lst.	4.7	5.2	0.8	4	n.d./3.6	

Tables 1a-d: Abbreviations used

Tectonic position:	Au = Autochthonous	Ge = Gellihorn nappe	Ni = Niesen nappe
	Ax = Axen nappe	Ih = Intrahelvetic complex	NF = North Helvetic Flysch zone
	Bo = Border chain	IF = Intermedial Flysch zone	Sä = Säntis-Drusberg nappe
	Do = Doldenhorn nappe	Kl = Klippen nappe	Wi = Wildhorn nappe
	Dr = Drusberg nappe	Mü = Müritschen nappe	Zc = Zone des cols

n = number of measurements  
 glyc/air = glycolated/air-dried  
 ka = kaolinite, pa = paragonite, pa/mu = mixed-layer paragonite/muscovite, py = pyrophyllite

Table 1b: Coal rank and illite crystallinity data for section II: Lake Lucerne - Reuss valley.

No. original specimen	coordinates/elevation	tectonic/stratigraphic position	Lithology	Rm (%)	Rmax (%)	Rmin (%)	n	IC glyc/air
22 S 26	680.960/199.630/1330	K1 Zoophycos-Sch.	marly lst.	1.8	1.9	0.2	15(7)	7.8/9.2
23 S 27	680.970/199.690/1340	K1 Zoophycos-Sch.	marl	1.85	2.0	0.2	29(16)	9.0/10.9
24 S 22	681.330/199.290/1170	F Flysch	marl	1.0			24	12.5/25
25 S 24	680.870/199.100/1340	F Flysch	marl	0.9			27	10.0/27.5
26 V 3	684.950/207.450/1420	F Flysch	marl	1.6			8	8.3/10.5
27 V 4	684.950/207.450/1420	F Flysch	marl	1.7			7	8.0/11.2
28 V 5	685.220/207.210/1650	Bo Drusberg-Sch.	marl	1.9	2.1	0.4	4	10.5/18.4
29 V 6	685.220/207.210/1650	Bo Drusberg-Sch.	marl	1.6	1.8	0.3	4	10.9/21.7
30 V 12	685.000/206.750/1390	Bo "Berriasien-Va-langinienmergel"	marl	1.6	1.8		16	13.9/25.5
31 U 2	689.260/203.450/480	Dr Kieselkalk	marly lst.	2.3			22	9.0/16
32 S 15	682.470/200.920/850	Dr "Gault"	marly lst.	1.4			10	9.3/9.6
33 S 37	684.930/199.880/1840	Dr "Gault"	marly lst.	1.6			11	13.7/13.0
34 U 4	689.375/202.590/440	Dr Kieselkalk	marly lst.	2.0	2.1	0.2	15(3)	11.0/25
35 U 6	689.650/201.390/450	Dr Kieselkalk	marly lst.	2.05			9	11.3/18
36 U 7	689.650/201.390/450	Dr Kieselkalk	marly lst.	2.1			10	10.1/16
37 U 9	689.665/201.350/450	Dr Kieselkalk	calc.slate	2.0			7	9.5/15
38 U 11	689.990/199.815/470	Ax Drusberg-Sch.	marly lst.	2.15	3.3	0.3	16(6)	12.0/16
39 U 12	689.990/199.815/470	Ax Drusberg-Sch.	marly lst.	2.2	2.85	0.4	17(2)	10.3/19
40 U 16	687.740/196.585/600	Ax Drusberg-Sch.	calc.slate	2.8			6	7.1/8.3
41 U 18	687.725/196.490/560	Ax Drusberg-Sch.	calc.slate	3.0	3.3	0.4	5(3)	7.2/7.7
42 I 34	683.910/193.320/1710	Ax Drusberg-Sch.	calc.slate	2.9	3.3	0.5	7	8.8/8.8
43 A 47	687.250/191.470/1550	Ax Zementstein-Sch.	limestone	4.1	4.35	0.5	7	7.2/6.9
44 A 48	687.250/191.450/1550	Ax Zementstein-Sch.	limestone	4.2	4.5	0.5	15	6.6/7.4
45 I 55	682.240/190.340/2100	Ax "Aalénien"	slate	4.5	5.7	2.1	23	6.4/6.5
46 H 25	681.500/188.770/2880	Ax "Aalénien"	marly lst.	4.05	4.8	1.0	17	5.7/5.6
47 A 23	685.000/188.580/2260	Ax "Aalénien"	slate	4.6	5.4	1.5	17	6.3/6.7
48 A 15	684.360/188.010/2390	Ax Sexmor-Serie	marly lst.	4.9	5.3	0.7	22	6.0/6.1
49 A 6	681.460/187.560/2180	Ax "Cretaceous"	marly lst.	4.6	5.8	1.9	10	4.7/4.7
50 U 23	687.890/195.310/440	NF Dachschiefer	slate	2.6	3.0	0.65	21	8.8/10.8
51 U 24	687.890/195.310/440	NF Dachschiefer	slate	2.5	2.8	0.5	35(15)	8.5/9.8
52 A 51	687.880/192.600/1280	NF Dachschiefer	slate	3.0	3.3	0.8	22	9.8/11.9
53 A 52	687.890/192.600/1280	NF Dachschiefer	calc.slate	3.2	3.5	0.6	10	6.8/8.1
54 U 27	689.165/192.350/520	NF Dachschiefer	calc.slate	3.75	4.5	2.5	13	7.1/8.8
55 U 28	689.165/192.350/520	NF Dachschiefer	calc.slate	3.9	4.7	1.7	20	6.7/6.9
56 A 19	684.570/187.900/2230	NF Dachschiefer	marly lst.	3.8	4.6	1.2	10	6.8/7.0

Table 1b: *Continued*

No. original specimen	coordinates/elevation	tectonic/stratigraphic position	lithology	Rm (%)	Rmax-Rmin (%)	n	IC glyc/air
57 A 10	681.530/187.440/2140	NF Dachschiefer	calc.slate	3.1	3.7	1.1 14	7.8/8.5
58 MF 1792	692.400/187.800/550	Au "Aalénien"	slate+py	5.1	6.2	2.55 15	5.1/5.3
59 A 35	688.380/186.080/1560	Au "Aalénien"	calc.slate	5.1	6.7	2.5 10	3.7/3.7
60 A 38	688.420/186.120/1590	Au "Aalénien"	marly lst.	4.8	6.1	2.5 10	4.7/5.2
61 H 16	686.160/181.890/3050	Au "Aalénien"	slate	4.7	5.3	1.1 25	4.6/4.6
62 H 21	682.050/181.250/2900	Au "Aalénien"	calc.slate	4.5	5.4	1.35 24	4.6/4.8

Table 1c: *Coal rank and illite crystallinity data for section III: Glarus Alps.*

No. original specimen	coordinates/elevation	tectonic/stratigraphic position	lithology	Rm (%)	Rmax-Rmin (%)	n	IC glyc/air	remarks
63 MF 1243	752.530/219.550/1160	Sä Drusberg-Sch.	marly shale	3.1	3.5	17	-	ka, py
64 MF 1244	752.530/219.550/1160	Sä Drusberg-Sch.	marly shale	3.2	3.5	10(4)	6.9/7.7	
65 MF 644	740.850/210.500/1960	Ax lower "Liassic"	slate	4.9	6.7	3.15 28	-	pa, pa/mu
66 MF 649	740.850/210.500/1960	Ax lower "Liassic"	calc. slate	5.1	5.6	0.8 19	n.d./(6.4)	pa, pa/mu
67 MF 651	740.850/210.500/1960	Ax lower "Liassic"	calc. slate	5.1	5.4	0.6 18	n.d./(5.6)	pa, pa/mu
68 MF 548	711.500/194.600/1440	Ax lower "Liassic"	calc. slate	5.05	5.5	0.6 26	4.6/4.9	
69 MF 555	711.500/194.600/1440	Ax middle "Liassic"	calc. slate	5.1	5.6	0.8 50	3.9/4.2	
70 MF 582	738.675/217.250/1035	Mu lower "Liassic"	slate	4.65	5.2	1.1 26	n.d./(6.9)	pa, pa/mu
71 MF 585	738.675/217.250/1035	Mu lower "Liassic"	calc. slate	4.9	5.2	0.5 36	n.d./(6.4)	pa, pa/mu
72 MF 738	727.540/190.180/2610	Ih lower "Liassic"	slate	4.5	4.9	1.0 17	4.4/4.1	pa, pa/mu



Table 1d: Coal rank and illite crystallinity data for section IV: Falknis nappe.

No. original specimen	coordinates/elevation	stratigraphic position	lithology	Rm (%)	Rmax	Rmax-Rmin	n	IC glyc/air
73	GR 75/3	762.050/212.660/2100	? "Liassic/Dogger"	4.0	4.3	0.6	30(15)	(10.3/9.1)
74	All 76/206	757.620/213.770/550	Falknisbreccien-Serie	4.95	5.5	0.9	57(27)	5.5/5.6
75	All 76/210	758.370/213.050/700	Falknisbreccien-Serie	5.0	5.6	1.2	19(9)	5.0/5.2
76	All 76/211	758.370/213.050/700	Falknisbreccien-Serie	4.7	5.45	1.4	32(16)	5.3/5.8
77	All 76/230	762.700/212.160/2050	Falknisbreccien-Serie	4.2	4.5	0.9	10	5.3/5.4
78	All 76/217	762.680/211.570/2200	Falknisbreccien-Serie	3.3	3.8	1.45	5	6.1/6.4
79	All 76/201	761.900/213.800/2060	Falknisbreccien-Serie	3.1	3.3	0.5	12	5.5/5.5
80	All 76/203	761.900/213.800/2060	Falknisbreccien-Serie	3.1	3.3	0.5	14	5.4/5.6
81	All 76/221	762.100/213.050/2220	Falknisbreccien-Serie	3.6	4.05	0.8	7(5)	5.3/5.8
82	All 76/235	764.780/212.690/1900	Falknisbreccien-Serie	3.3	3.7	0.8	24	5.5/6.2
83	All 76/236	764.780/212.690/1900	Falknisbreccien-Serie	3.5	3.9	0.7	15	6.2/6.6
84	TGr 21	766.630/212.970/2000	Falknisbreccien-Serie	3.3	3.7	0.6	21(11)	6.1/6.1
85	TGr 23	766.630/212.970/2000	Falknisbreccien-Serie	3.35	3.7	0.6	15(8)	5.6/5.0
86	All 73/109	759.250/217.800/660	Neokomflysch-Serie	3.2	3.5	0.5	14(8)	10.0/15
87	All 73/110	759.250/217.800/660	Neokomflysch-Serie	3.1	3.4	0.4	6	10.8/18
88	BS 78/110	759.380/216.865/1015	Neokomflysch-Serie	3.3	3.45	0.25	25(7)	10.2/8.9
89	BS 78/112	759.310/216.800/965	Neokomflysch-Serie	3.2	3.4	0.2	14(4)	11.2/11.1
90	BS 78/113	759.310/216.800/965	Neokomflysch-Serie	3.25	3.7	0.5	11(3)	9.6/9.4
91	BS 76/019	760.700/217.980/1340	Neokomflysch-Serie	2.5	2.75	0.4	6	8.2/12.4
92	BS 76/027	760.700/217.980/1340	Neokomflysch-Serie	2.35	2.8	0.7	31	8.2/11.2
93	All 73/113	760.280/215.750/1250	Neokomflysch-Serie	3.5	3.8	0.6	17(8)	9.0/9.5
94	All 73/114	760.280/215.750/1250	Neokomflysch-Serie	3.4	3.55	0.5	5	9.2/10.0
95	GR/MF 73	761.185/212.540/2200	Neokomflysch-Serie	4.2	4.6	0.6	25(17)	8.2/8.2
96	BS 77/064	763.830/213.770/2260	Neokomflysch-Serie	2.7	3.2	0.9	32	10.2/9.9
97	BS 76/006	765.050/213.310/2020	Neokomflysch-Serie	3.4	3.8	0.95	10	7.9/8.3
98	BS 76/091	765.050/213.310/2020	Neokomflysch-Serie	3.2	3.75	0.9	21	9.3/10.0
99	BS 78/111	759.470/216.560/1040	Tristelschichten	3.0	3.1	0.25	36(10)	10.8/9.3
100	BS 76/050	760.720/217.990/1380	Tristelschichten	2.6	2.9	0.4	19	8.2/12.8
101	BS 76/101	760.310/214.230/2005	Tristelschichten	3.3	3.7	0.5	16	9.6/12.4
102	BS 76/128	760.310/214.230/2005	Tristelschichten	3.1	3.45	0.8	50	7.9/13.1
103	BS 76/130	760.310/214.230/2005	Tristelschichten	2.9	3.8	1.4	48	12.8/17.9
104	BS 77/076	763.770/214.020/2320	Tristelschichten	3.0	3.6	0.9	21	10.4/9.6
105	BS 77/087	763.770/214.020/2320	Tristelschichten	2.8	3.1	0.75	22	13.9/13.0
106	BS 77/100	763.770/214.020/2320	Tristelschichten	3.2	3.4	0.6	30	8.5/8.2
107	BS 38/3	766.990/213.540/2530	Tristelschichten	2.8	3.2	0.6	18	10.7/12.1

Table 2a: Fluid inclusion data for section I: Kien valley.

No. original specimen	locality	tectonic/stratigraphic position	quartz type	n incl. xx	fluid comp.	T <sub>hom.</sub> (°C)	CH <sub>4</sub> (g/cm <sup>3</sup> )	P <sub>CH<sub>4</sub></sub> (bar)
201 Mu 122	Allebach	Ni Flysch	F	18	H <sub>2</sub> O	104/101-107	-	-
202 Mu 17	Stigelbach	Ni Flysch	F	37	H <sub>2</sub> O	106/101-109	-	-
203 Mu 121	Tschentenalp	Ni Flysch	P	10	H <sub>2</sub> O	97/ 91-100	-	-
204 Mu 18	Schmitte	Ni Flysch	P	20	H <sub>2</sub> O	91/ 86-98	-	-
205 Mu 19	Sackgrabe	Ni Flysch	P	15	H <sub>2</sub> O	96/ 95-97	-	-
206 Mu 185	Reichenbach	Ni Flysch	P	15	H <sub>2</sub> O	113/109-115	-	-
207 Mu 1	Därliigen	Wi Hohgant-Sandstein	S	55	CH <sub>4</sub>	199/193-207	0.282	1260
208 Mu 15	Wätterlatte	Wi Hohgant-Sandstein	P	27	HHC	193/187-201	-	-
209 Mu 3	Rengglipass	Wi "Berriasien-Valangi-nienmergel"	FP	2	CH <sub>4</sub>	223/216-228	0.332/0.327-0.334	2170
210 Mu 4	Spiggen	Wi Glockhaus-Serie	F	69	CH <sub>4</sub>	261/258-266	0.306/0.299-0.312	1890
211 Mu 2	Saustal	Wi "Berr.-Val.mergel"	F	37	CH <sub>4</sub>	244/236-249	0.286/0.277-0.295	1490
212 Mu 5	Steinenberg	Wi "Sinémurien-sandst."	F	73	CH <sub>4</sub>	265/258-272	0.286/0.232-0.318	1360
213 Mu 7	Finstertal	Wi "Berr.-Val.mergel"	PS	2	CH <sub>4</sub>	248/241-253	0.312/0.306-0.318	1920
214 Mu 9	Bundstock NW	Wi lower "Liassic"	F	122	CH <sub>4</sub>	262/256-271	0.272/0.248-0.286	1400
215 Mu 195	Rafliwald	IF Taveyenne sandst.	P	29	HHC	195/187-201	-	-
216 Mu 20	Loosplatte	IF Taveyenne sandst.	FP	30	CH <sub>4</sub>	238/235-241	> 2 mole-%	-
217 Mu 6	Mitholz	IF Wildflysch	F	110	CH <sub>4</sub>	241/237-247	0.309/0.302-0.315	1830
218 Mu 14	Oeschinensee	IF Taveyenne sandst.	P	14	H <sub>2</sub> O+CO <sub>2</sub> ?	224/214-229	-	-
219 Mu 8	Bundstock E	IF Taveyenne sandst.	P	23	CH <sub>4</sub>	263/261-265	0.267/0.261-0.272	1340
220 Mu 11	Wermtfluh	IF Taveyenne sandst.	F	64	CH <sub>4</sub>	263/258-268	0.295/0.240-0.299	1710
221 Mu 10	Bundstock S	IF Taveyenne sandst.	P	54	CH <sub>4</sub>	267/253-276	0.302/0.295-0.318	1850
222 Mu 12	Wermtfluh S	IF Taveyenne sandst.	F	45	CH <sub>4</sub>	257/252-264	> 2 mole-%	-
223 Mu 13	Gamchi	Do "Priabonien-sandst."	F	42	H <sub>2</sub> O	163/152-176	-	-

Tables 2a-c: Abbreviations used

Quartz type: F = "Fadenquarz", P = prismatic, S = sceptre habit  
 xx/incl = number of crystals/inclusions

Table 2b: Fluid inclusion data for section II: Lake Lucerne - Reuss valley.

No. original specimen	locality	tectonic/stratigraphic position	quartz type	n incl. xx	fluid comp.	T <sub>hom.</sub> (°C)	S <sub>CH<sub>4</sub>3</sub> (g/cm <sup>3</sup> )	P <sub>CH<sub>4</sub></sub> (bar)
224 Mu 27	Zingel	Wi Kieselkalk	S	3 45	HHC	118/112-132	-	-
225 Mu 28	Unterschönenbuch	Dr Seewerkalk	S	2 27	HHC	165/158-175	-	-
226 Mu 29	Sisikon N	Dr "Valanginien"	S	2 22	HHC	174/152-201	-	-
227 Mu 37	Sisikon S	Ax Drusberg-Sch.	S	1 10	HHC	?	-	-
228 Mu 31	Isleten	Ax Drusberg-Sch.	P	2 45	CH <sub>4</sub>	210/204-219	0.306/0.286-0.332	1630
229 Mu 230.1	Grosstal-Isental 1	Ax "Valanginien"	S	2 36	CH <sub>4</sub>	230/228-231	0.346/0.339-0.351	2560
230 Mu 230.2	Grosstal-Isental 2	Ax "Valanginien"	S	2 43	CH <sub>4</sub>	234/233-235	0.286/0.286-0.295	1460
231 Mu 238.2	Brunnistock 2	Ax "Dogger"	P	2 33	H <sub>2</sub> O	181/176-187	-	-
232 Mu 238.1	Brunnistock 1	Ax "Dogger"	P	2 14	H <sub>2</sub> O	199/193-204	-	-
233 Mu 34	Seedorf	NF Dachschiefer	FS	4 71	CH <sub>4</sub>	259/256-261	0.282	1510
234 Mu 35	Attinghausen	NF Dachschiefer	FS	6 243	CH <sub>4</sub>	262/258-264	0.302/0.299-0.306	1820
235 Mu 236	Grat-Attinghausen	NF Dachschiefer	FS	4 73	CH <sub>4</sub>	264/259-268	0.334/0.327-0.344	2490
236 Mu 237	Angistock	NF Dachschiefer	PS	2 38	CH <sub>4</sub>	255/251-259	0.267/0.254-0.272	1310
237 Mu 239	Waldnacht	NF Dachschiefer	P	2 33	CH <sub>4</sub>	250/246-252	0.302/0.295-0.315	1770

Table 2c: Fluid inclusion data for section IV: Falknis nappe.

No. original specimen	locality	stratigraphic position	quartz type	n incl. xx	fluid comp.	T <sub>hom.</sub> (°C)	S <sub>CH<sub>4</sub>3</sub> (g/cm <sup>3</sup> )	P <sub>CH<sub>4</sub></sub> (bar)
238 Mu 207.2	Triesen 2	Neokomflysch-Serie	F	1 14	H <sub>2</sub> O	142/140-143	-	-
239 Mu 207.1	Triesen 1	Neokomflysch-Serie	P	2 10	H <sub>2</sub> O	163/149-177	-	-
240 Mu 256	Münz-Triesen	Neokomflysch-Serie	P	1 8	H <sub>2</sub> O	174/171-177	-	-
241 Mu 206	Ans-Fläsch	Falknisbreccien-Ser.	P	2 23	H <sub>2</sub> O	102/ 95-108	-	-
242 Mu 205	Dürwald-Fläsch	Falknisbreccien-Ser.	P	2 24	H <sub>2</sub> O	118/116-122	-	-
243 Mu 199	Fläschertal	Falknisbreccien-Ser.	P	2 21	H <sub>2</sub> O	85/ 82-87	-	-
244 Mu 255	Lavenatal	Neokomflysch-Serie	F	3 28	H <sub>2</sub> O-CH <sub>4</sub>	222/214-226	> 1 mole-% CH <sub>4</sub>	-
245 Mu 200	Maienfelder Alp	Falknisbreccien-Ser.	F	1 26	H <sub>2</sub> O-CH <sub>4</sub>	200	> 2 mole-% CH <sub>4</sub>	-
246 Mu 229	Barthümeljoch SE	Falknisbreccien-Ser.	P	1 13	H <sub>2</sub> O-CH <sub>4</sub>	196/195-197	> 2 mole-% CH <sub>4</sub>	-
247 Mu 202	Ijes Fürggli	Tristelschichten	F	2 70	CH <sub>4</sub>	227/222-232	0.337/0.329-0.342	2310
248 Mu 203	Ijes Fürggli SW	Neokomflysch-Ser.	S	2 88	CH <sub>4</sub>	238/232-242	0.388/0.385-0.390	4060
249 Mu 204	Barthümeljoch SW	Neokomflysch-Ser.	F	3 147	CH <sub>4</sub>	226/223-228	0.367/0.367-0.369	3120
250 Mu 201	Ijes-Maienfelder Alp	Neokomflysch-Ser.	S	2 70	CH <sub>4</sub>	249/246-252	0.388/0.387-0.390	4170

allow the measurement of the density of  $\text{CH}_4$ . b) Water-rich fluid inclusions more or less saturated with  $\text{CH}_4$  during homogenization yield homogenization temperatures above  $180^\circ\text{C}$  which can be interpreted as minimum temperatures of formation. Water-rich fluid inclusions strongly undersaturated with hydrocarbons or  $\text{CO}_2$  during their homogenization generally show low homogenization temperatures ( $T_{\text{hom}} < 200^\circ\text{C}$ ) which are meaningless for geothermometric purposes. c) Provided that the density of methane and the minimum temperature of formation from fluid inclusions of the same or the next following quartz generation are known, the minimum pressure of formation can be determined graphically after the PVT-data of ZAGORUCHENKO & ZHURAVLEV (1970).

### 3. Illite crystallinity, coal rank and fluid inclusion studies in sections I-IV

The geologic setting of the external parts of the Central Alps, an especially well studied area, will not be repeated here. For general reviews the reader is referred to TRÜMPY (1960) and BERNOULLI et al. (1964).

Results are given in Tables 1-2 and presented in Figures 4-8. In order to facilitate a visual comparison of  $IC$  and  $R_m$  data in the figures, the size of the enclosing circle indicates the relative size of the parameter (inverse in the case of  $IC$ , since low values indicate high crystallinity). At each locality the  $IC$  value of a single sample (or the mean value of two or three samples) is given in order to allow direct comparison with  $R_m$  data of the same specimen. We are aware of the fact the  $IC$  data should be treated on a statistical basis. Therefore, our conclusions are based on much more  $IC$  data than presented here, e.g. 300  $IC$  values each for sections I and II. Single  $IC$  values which do not conform to the general pattern will be discussed below.

#### *Section I: Kien valley (Fig. 4 and 6a)*

This cross section comprises the Pennine Niesen nappe in the northwest and a complete succession through the Helvetic zone until the autochthonous of the Aar massif in the southeast. The Intermedial Flysch zone (KÜNZI, in prep.) corresponds to the upper part of the Gellihorn nappe s.l. as used up to now.

#### *Illite crystallinity*

The Pennine Niesen nappe belongs to the deeper anchizone. Note that the  $IC$  value of sample No. 1 would actually be better than 6.8, since the presence of mixed-layer paragonite/muscovite causes an enlargement of the  $10 \text{ \AA}$  illite reflexion. The flysch from the "Zone des Cols" (No. 2 and 3) and the frontal part of the Wildhorn nappe (No. 4) belong to the diagenetic zone. This seems to indicate that the slight metamorphism of the Niesen nappe is older than the thrusting of this Pennine nappe onto the Helvetic zone. The southern part of the Wildhorn nappe (No. 5-7), on the other hand, has reached the anchizone. Samples 4-7 show a regular increase in  $IC$  from northwest to southeast within this same nappe. The next deeper tectonic unit, the Intermedial Flysch zone, reaches in its frontal part the diagenesis/anchizone boundary (No. 9-11) but towards the southeast the deeper anchizone (No. 12 and 13). The Gellihorn nappe s.str. yielded  $IC$  values of the middle to deep anchizone (No. 14 and 15). With the exception of sample 17 all other samples from the parautochthonous Doldenhorn nappe (No. 16, 18 and 19) and the

SE

NW

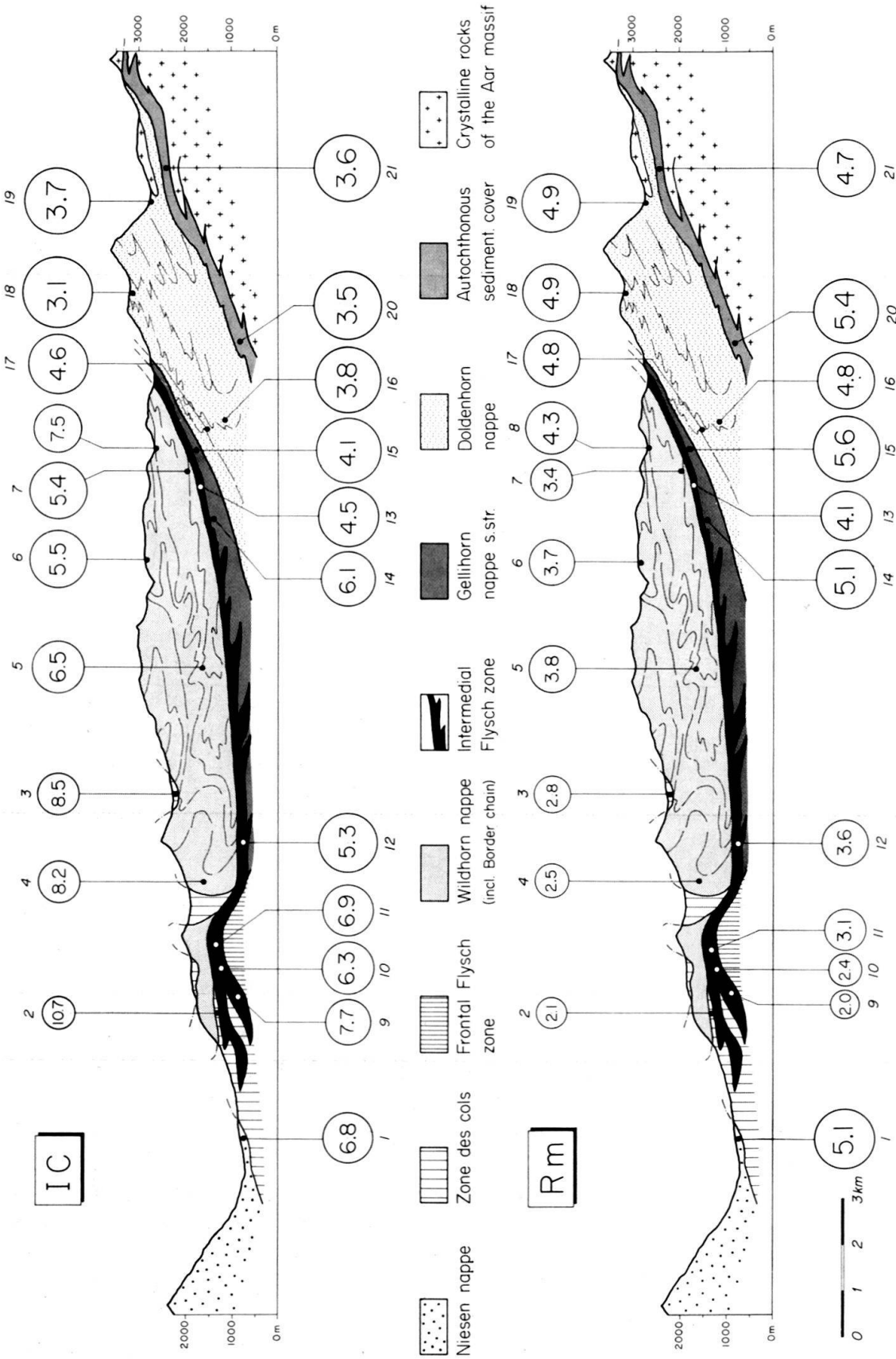


Fig. 4. Distribution of illite crystallinity (IC) and coal rank (R<sub>m</sub>) in a simplified cross section of the Kien valley, Bernese Oberland (section I). Sample localities (1–21) correspond to those of Table 1a.

autochthonous sedimentary cover of the Aar massif (No. 20 and 21) indicate epimetamorphic conditions.

### *Coal rank*

The  $R_m$  distribution with respect to tectonic units is similar to that of the *IC* data. One sample from the Pennine Niesen nappe (No. 1) shows a much higher coal rank than the six samples from the adjacent and underlying tectonic units (No. 2–4, 9–11), that is 5.1% versus 2.0–3.1%  $R_m$ . As with the *IC* data, this indicates that the metamorphism of the Niesen nappe occurred before the overthrusting onto the Helvetic zone. Within the Wildhorn nappe,  $R_m$  generally increases from northwest (No. 4,  $R_m = 2.5\%$ ) to southeast (No. 8,  $R_m = 4.3\%$ ). The same trend can also be observed in the Intermedial Flysch zone where  $R_m$  increases from 2.0% (No. 9) to 4.1% (No. 13). The two samples from the Gellihorn nappe (No. 14 and 15) show slightly higher  $R_m$  values than the tectonic units above and below. More data would be needed to evaluate their significance. The six samples from the Doldenhorn nappe (No. 16–19) and the autochthonous cover of the Aar massif (No. 20–21) have high and relatively uniform  $R_m$  values ranging from 4.7 to 5.4%.

### *Fluid inclusions*

The distribution of the fluid composition (Fig. 6a) shows a general dependence on metamorphic grade as derived by the *IC* and  $R_m$  data. The Pennine Niesen nappe contains exclusively  $H_2O$ -bearing fluids (No. 201–206), typical for metamorphic conditions of the higher-grade anchizone and epizone (MULLIS 1979). By contrast, the underlying Helvetic zone shows in its northwestern part a much less evolved fluid in fissure quartz, that is HHC (>1 mole-%) at localities 208 and 215 and  $CH_4$ -bearing fluids at localities 206 and 207. This proves in our opinion a pre-overthrust evolution of the fluids from the Niesen nappe. The Wildhorn nappe (without the Border chain) and the southern part of the Intermedial Flysch zone are dominated by  $CH_4$ -bearing fluids.  $H_2O$ -bearing fluids were encountered in sample No. 218 of the Intermedial Flysch zone and sample No. 223 of the Doldenhorn nappe and indicate metamorphic conditions near the anchi-/epizone boundary.

Minimum formation temperatures in the Wildhorn nappe and the Intermedial Flysch zone show an overall increase from about 190–200 °C in the northwest to about 260–270 °C in the southeast. Homogenization temperatures from  $H_2O$ -bearing fluids have no geothermometric meaning (see p. 185).

Minimum fluid pressures range from 1260 to 2170 bar with no evident relation to tectonic overburden (compare e.g. samples 212 and 213). Some of these pressures may therefore be interpreted as fluid over- or underpressures, cf. MULLIS (1979). A fluid overpressure could be generated by cracking HHC during a temperature increase, whereby the generated fluid could not or only partly escape until it was trapped in fissure quartz. Such possible fluid overpressures are often found in thick piles of slates (MULLIS 1979). Fluid underpressures can result from the opening and enlargement of Alpine fissures (MULLIS 1976).



The main conclusions can be summarized as follows:

- a) The high-grade anchizonal metamorphism of the Pennine Niesen nappe is obviously older than the overthrusting of this nappe onto the Helvetic zone.
- b) Some tectonic units (Wildhorn nappe, Intermedial Flysch zone) show a general increase in  $IC$ ,  $R_m$  and fluid evolution from northwest to southeast. However, no vertical variations within the same tectonic unit could be detected, which may be due to insufficient sampling.
- c) In the Helvetic zone of the Kien valley section there exists an overall increase in  $IC$ ,  $R_m$  and fluid evolution from higher to lower tectonic units.

#### *Section II: Lake Lucerne – Reuss valley (Fig. 5 and 6b)*

This is a complete cross section through the Helvetic zone from the Subalpine Flysch in the north to the autochthonous of the Aar massif in the south. Some Pennine Klippen occur in the northern part of the section.

#### *Illite crystallinity*

The Pennine Klippen and the Drusberg nappe belong to the diagenetic zone. Note that two samples from the Pennine Klippen (No. 22 and 23) have a slightly better  $IC$  than the underlying flysch (No. 24 and 25). The possible significance of these data will be discussed together with the coal rank determinations. Some of the samples from the northern part of the Axen nappe (No. 38 and 39, 42) still indicate conditions of the diagenetic zone whereas the southern part of this nappe has clearly reached the anchizone.  $IC$  values from the North-Helvetic Flysch zone (No. 50–57) are in the transition range from diagenesis to the anchizone. The two flysch samples No. 56 and 57 show a less advanced  $IC$  than the overlying samples No. 47–49 from the Axen nappe. This inversion in the  $IC$  data could be an indication for a pre-thrusting anchizonal metamorphism of the Axen nappe. 3 km to the north, however, no such inversion could be observed (compare samples No. 42–45 versus No. 50–55). Samples from the autochthonous sedimentary cover of the Aar massif show  $IC$  values of the deep anchizone (No. 58, 60–62) or already epizonal conditions (No. 59).

#### *Coal rank*

The coal rank data at the base of the Pennine Klippen are of special interest. For two samples of the Pennine Klippen (No. 22 and 23)  $R_m$  values of 1.8% were measured while the underlying flysch shows only 0.9%  $R_m$  (No. 24 and 25) and one sample from the higher part of the Drusberg nappe 1.4%  $R_m$  (No. 32). Both  $IC$  and  $R_m$  data point to the possibility that the Pennine Klippen reached their diagenetic stage before arriving at their present tectonic position. This conclusion is supported by fluorescence investigations. Both samples from the Pennine Klippen did not show any liptinite fluorescence, while sample No. 25 from the underlying flysch contains vitrinite and bituminite with redbrown fluorescence colours and also sample No. 32 from the upper Drusberg nappe still shows some liptinite fluorescence.

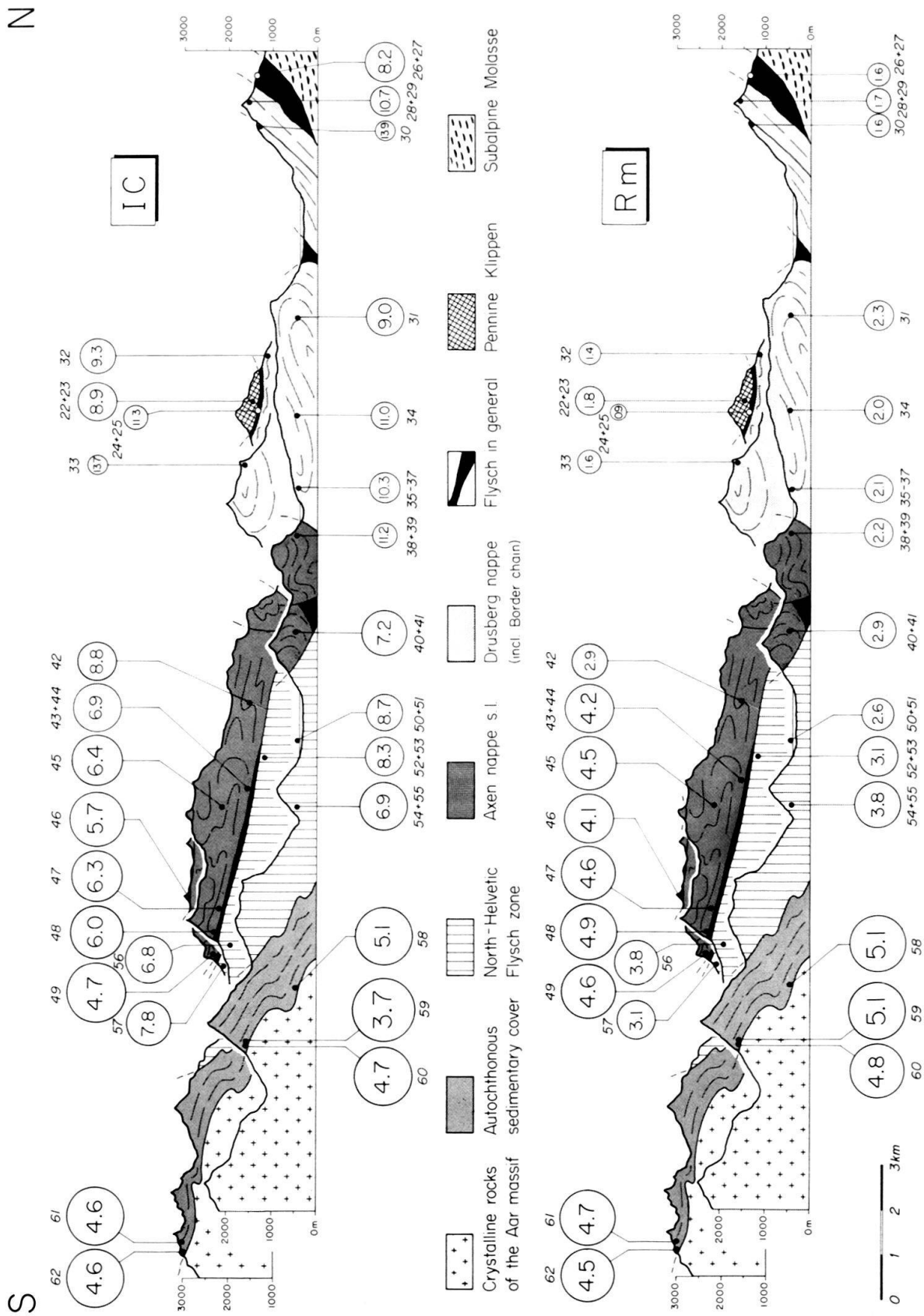


Fig. 5. Distribution of illite crystallinity (IC) and coal rank (R<sub>m</sub>) in a simplified cross section of Lake Lucerne - Reuss valley (section II). Sample localities (22-62) correspond to those of Table 1b. Note that north and south are reversed in this section as compared with Figure 4.

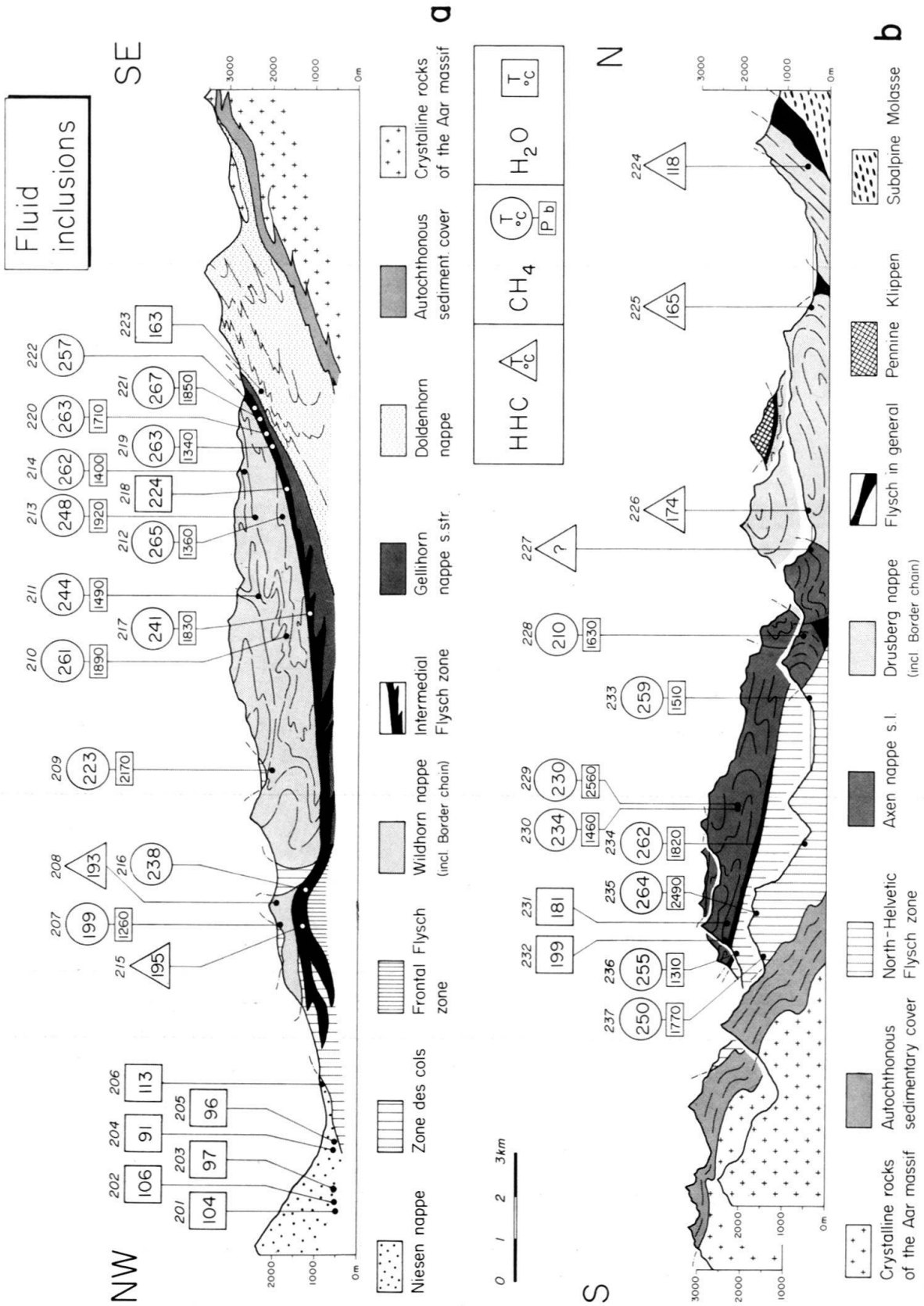


Fig. 6. Fluid inclusion data from fissure quartz of a section I and b section II (cf. Figures 4 and 5). Sample localities (201–237) correspond to those of Tables 2a and 2b.

Within the Drusberg nappe a slight increase in  $R_m$  values from 1.6% in the north to 2.0–2.3% in the south can be observed. Some vertical increase in the  $R_m$  values is indicated by samples 32 and 33 (1.4–1.6%  $R_m$ ) versus samples 31, 34–37 (2.0–2.3%  $R_m$ ).

Proceeding from the Drusberg nappe to the Axen nappe the  $R_m$  values show a continuous increase (samples 34–42). This seems to indicate that the coalification is younger than the tectonic juxtaposition of the Drusberg with the Axen nappe. The rapid increase in  $R_m$  values within the Axen nappe over a short distance of only about 5 km is remarkable. Samples No. 40–42 show  $R_m$  values of 2.9% while the more southerly located samples 43–49 have 4.1–4.9%  $R_m$ . The strong decrease of the degree of coalification towards the north may be explained by a corresponding rapid decrease of the former overburden with Pennine and Austroalpine nappes.

The North-Helvetice Flysch zone shows a clearly lower coal rank than the overlying Axen nappe, compare samples 50–56 (2.5–3.9%  $R_m$ ) versus samples 43–49 (4.1–4.9%  $R_m$ ). This means that the coalification of the Axen nappe (and probably also the Drusberg nappe, see above) occurred before the overthrusting onto the North-Helvetice Flysch zone.

High  $R_m$  values were found in the autochthonous sedimentary cover of the Aar massif (samples No. 58–62). Note the slight decrease in  $R_m$  values from 5.1% in the north to 4.5% in the south.

#### *Fluid inclusions*

In accordance with the  $IC$  and  $R_m$  data the fluid composition shows an evolution from external to internal tectonic position. In the Drusberg nappe (samples 224–226) and the frontal part of the Axen nappe (No. 227) fluids from fissure quartz contain >1 mole-% HHC, while the major part of the Axen nappe and the underlying North-Helvetice Flysch zone are dominated by  $CH_4$ -bearing fluids. In two samples from the southern part of the Axen nappe (No. 231 and 232)  $H_2O$ -bearing fluids were encountered, indicating slightly higher metamorphic conditions than in the flysch below, in agreement with the conclusion reached by the  $IC$  and  $R_m$  data.

Minimum formation temperatures increase from about 120 °C near the Alpine border (No. 224) to about 250–260 °C in the North-Helvetice Flysch zone (No. 233–237). The unexpected low temperatures of 230 °C (No. 229) and 234 °C (No. 230) for two samples of the overlying Axen nappe may be caused by one or both of the following reasons. First, the aqueous fluid may not have been saturated with  $CH_4$  during the homogenization of the fluid inclusion. Second, relatively late sceptre quartz were used for samples 229 and 230 while earlier quartz crystals (“Fadenquarz”, prismatic and early sceptre quartz) were used from the North-Helvetice Flysch zone. As shown by MULLIS (1975, 1976) there may occur a considerable temperature decrease of up to 70 °C from early to late quartz generations within the same Alpine fissure.

Minimum fluid pressures range from 1310 to 2560 bar. As for the Kien valley section there exists no evident relation to tectonic overburden (compare e.g. samples 234 and 235) and the pressure values may show a large variation of >1 kbar with two neighbouring outcrops (compare samples 229 and 230). Fluid over- and

underpressures are believed to be a reasonable explanation for this phenomenon (see p. 187).

The main conclusions can be summarized as follows:

- a) There exists some indication that the diagenesis of the Pennine Klippen happened before the present tectonic position was reached. However, more data are needed to substantiate this conclusion.
- b) The Helvetic Drusberg and Axen nappes show a general increase in  $IC$ ,  $R_m$  and fluid composition evolution from north to south with an especially strong rise of the degree of coalification within the Axen nappe. The  $R_m$  data point also to a vertical increase of the coal rank within the Drusberg nappe.
- c) The inversion in  $IC$  and  $R_m$  data between the North-Helvetic Flysch zone and the overlying Axen nappe as well as the fluid composition data are interpreted as a pre-thrusting anchizonal metamorphism of the Axen nappe.

### *Section III: Glarus Alps*

Due to the limited number of samples the results are not presented in a cross section. Sample locations are indicated in Figure 1.

#### *Illite crystallinity*

The presence of pyrophyllite, paragonite and mixed-layer paragonite/muscovite in most of the samples made an  $IC$  determination impossible or yielded too high values. The few undisturbed  $IC$  values presented here as well as already published (FREY 1970; BRIEGEL 1972) and unpublished values lead to the following results.

The Säntis-Drusberg nappe belongs to the diagenetic zone but has reached in some places the diagenesis/anchizone boundary (samples No. 63 and 64). Samples studied from the tectonically lower Axen nappe (No. 65-69) and Mürtschen nappe (No. 70-71) belong to the anchizone. Sample No. 72 from the Infrahelvetic complex (MILNES & PFIFFNER 1977) has reached the anchizone/epizone boundary.

#### *Coal rank*

The two samples from the Säntis-Drusberg nappe (No. 63 and 64) have much lower  $R_m$  values of 3.1-3.2% than the remaining samples No. 65-72, which show relatively uniform  $R_m$  values ranging from 4.5 to 5.1%. Note that there exists no difference in coal rank from Axen nappe samples south of Lake Walen (No. 65-67) and the Klausenpass area (No. 68 and 69), respectively.

### *Section IV: Falknis nappe (Fig. 7 and 8)*

This section was chosen in order to study the relationships between  $IC$ , coal rank and fluid inclusions in more detail within one single nappe. The stratigraphic sequence of the Falknis nappe ranges from Triassic to Lower Eocene (TRÜMPY 1916; ALLEMANN 1957; GRUNER 1979) and the main Alpine folding and thrusting occurred presumably in Upper Eocene to late Lower Oligocene times. The nappe



NW

SE

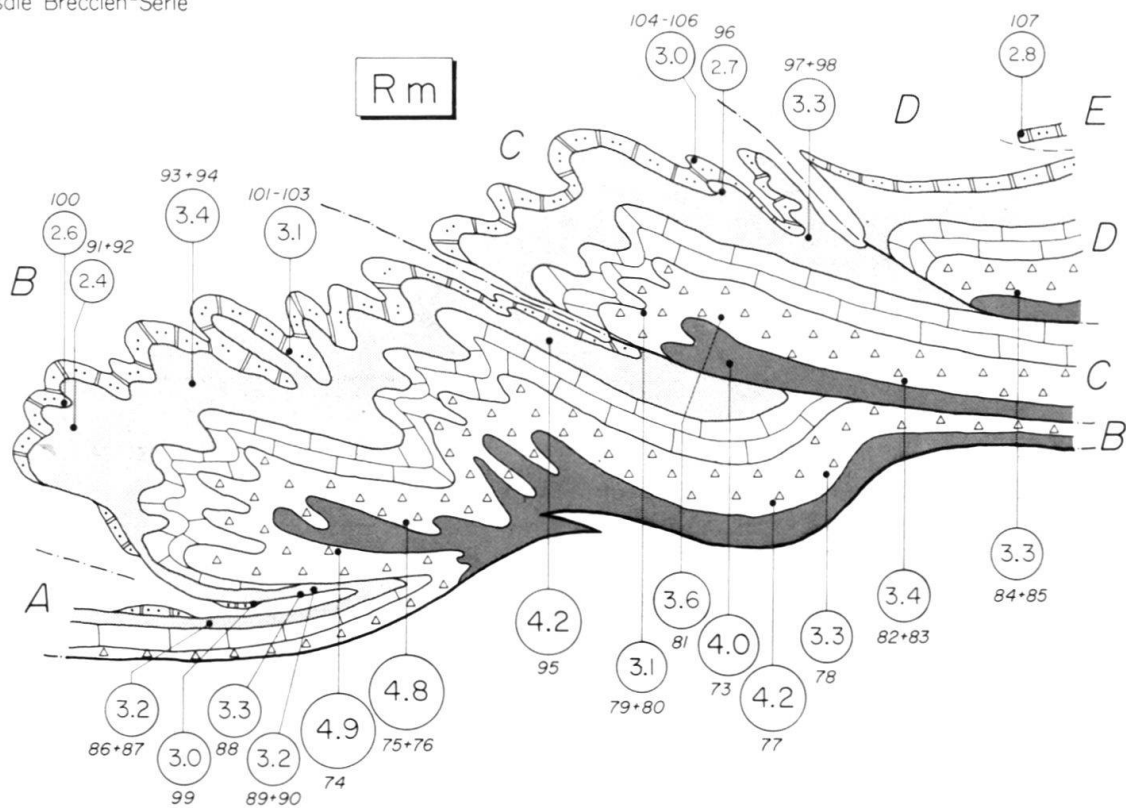
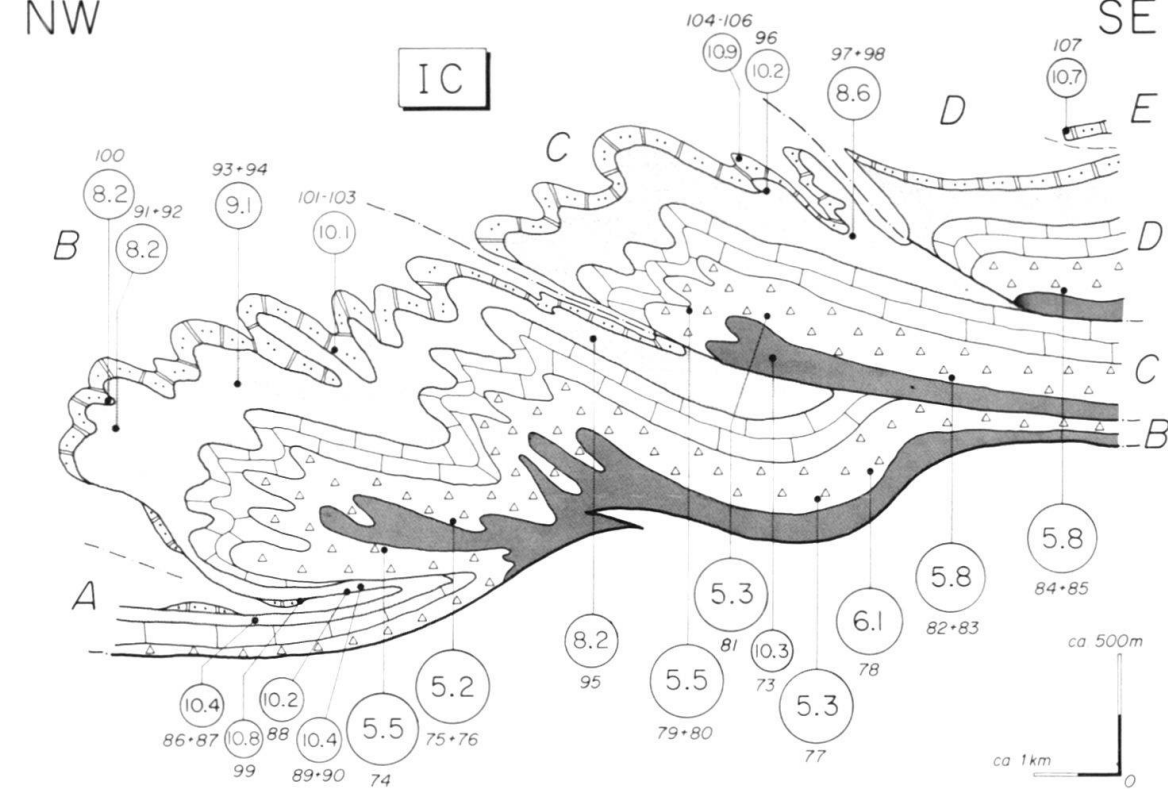


Fig. 7. Distribution of illite crystallinity ( $IC$ ) and coal rank ( $R_m$ ) in a simplified cross section of the Falknis nappe (section IV). Sample localities (63-107) correspond to those of Table 1d. The designation of the schuppen is as follows: A = Basalschuppe; B = Gleckhorn-Schuppe; C = Falknis-Schuppe; D = Tschingel-Schuppe; E = Obere Tschingel-Schuppe (German nomenclature).



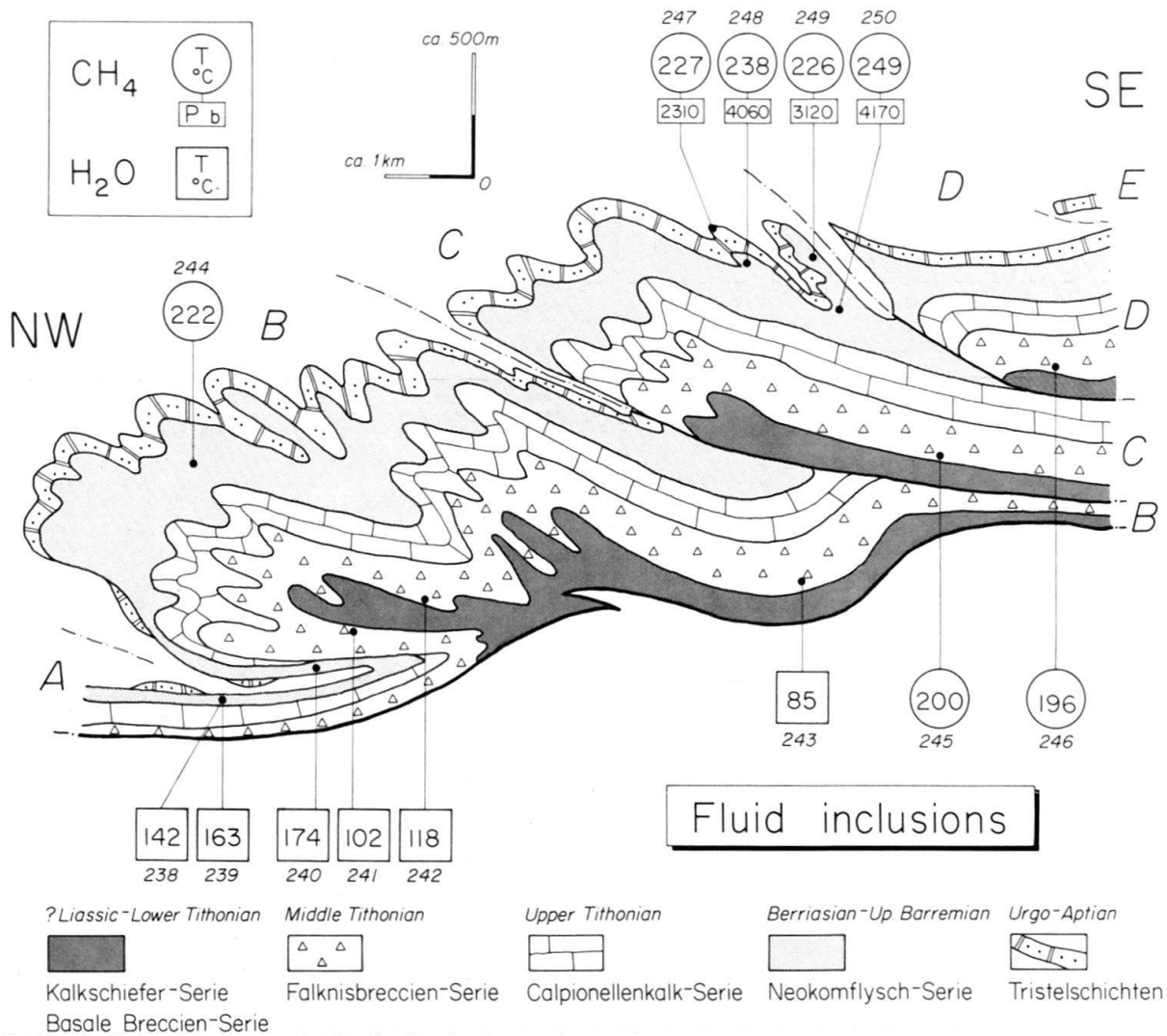


Fig. 8. Fluid inclusion data from fissure quartz of the Falknis nappe. Sample localities (238–250) correspond to those of Table 2c.

consists of five schuppen, named here A to E from bottom to top. Note, that there exists in Figures 6 and 7 a threefold vertical exaggeration.

*Illite crystallinity*

At a first glance, the IC distribution seems to be rather erratic. However, there exists a strong relationship between IC and lithology. The IC of the Upper Jurassic Falknisbreccien-Serie<sup>5)</sup> (5.0–6.0) indicates medium anchizonal conditions while in the Lower Cretaceous Neokomflysch-Serie (7.9–10.8) as well as in the Lower Cretaceous Tristelschichten (7.9–13.9) only diagenetic conditions are recorded. It is well-known that the IC of the diagenetic zone is influenced by the lithology, but it is generally assumed that this influence would diminish in the anchizone (see e.g. KÜBLER 1968). The question arises, therefore, which formation is recording the “true” IC. It might be suggested that the Falknisbreccien-Serie contains a too high

<sup>5)</sup> We retain here the German nomenclature for the various formations, as they appear in the literature.

Table 3: *Illite crystallinity values of different grain sizes for four samples from the Falknis nappe.*

No.	Original sample	< 2 $\mu$ *	2-6 $\mu$	6-20 $\mu$	20-63 $\mu$
78	All 76/217	5.8/6.0	4.7/5.0	4.7/5.1	4.5/5.0
84	TGR 21	n.d.	4.0/4.5	4.3/4.6	5.2/6.0
91	BS 76019	6.5/11.3	5.5/9.4	4.0/6.1	7.4/11.4
92	BS 76027	8.7/15.5	5.2/14.2	4.9/8.3	5.8/8.6

*IC*: The first value refers to the glycolated state, the second to the air-dried state.

No. 78 and 84 = Falknisbreccien-Serie, No. 91 and 92 = Neokomflysch-Serie.

\* Note that the *IC* values of the < 2  $\mu$  fraction are different from those of Table 1d probably due to different sample preparation (see p. 177).

*IC* since this formation shows the largest clastic influence and the *IC* would have been enhanced by an inherited *IC* from a high-grade source area. In order to test this assumption the *IC* was determined on several grain size fractions of four samples (Table 3). In one sample from the Falknisbreccien-Serie (No. 78) the 2-6  $\mu$  fraction showed actually a higher *IC* (lower coefficient) than the < 2  $\mu$  fraction, but the same trend was also observed in the Neokomflysch-Serie (No. 91 and 92). However, this trend is reversed in the coarsest fraction (No. 84, 91 and 92) but is not understood at present. On the other hand, the *IC* of the Neokomflysch-Serie and the Tristelschichten might have been lowered due to the presence of another sheet silicate, which causes a broadening of the 10 Å peak, as e.g. pyrophyllite, paragonite or mixed-layer paragonite/muscovite. However, no such minerals could be identified. A too low *IC* might be assumed for the carbonate-rich Tristelschichten, since an insufficient supply of potassium in limestones with low porosity may inhibit the process of illitization. However, this argument would not hold true for the Neokomflysch-Serie.

Summarizing the above arguments it must be admitted that the *IC* data are contradictory and either indicate medium-grade anchizonal or diagenetic conditions for the Falknis nappe.

### Coal rank

The  $R_m$  values of the organic particles indicate the anthracite and meta-anthracite coal rank (German coal classification, see STACH et al. 1975, p. 42). The coal rank is relatively low in the Tristelschichten and the Neokomflysch-Serie (2.6-3.4%  $R_m$  with the exception of sample No. 95 with 4.2%  $R_m$ ) and is barely higher in the Falknisbreccien-Serie of schuppen C and D (3.1-3.6%  $R_m$ ). Such a distribution of similar  $R_m$  values seems to indicate a *pre-schuppen coalification* where differences in the stratigraphic overburden were never greater than about 600 m. A higher meta-anthracite rank was reached in some parts of schuppe B (No. 74-77 and 95 with 4.2-5.0%  $R_m$ ) and at the base of schuppe C (No. 73, 4.0%  $R_m$ ). Since the stratigraphic thickness was not larger here than in schuppen C and D, the higher coal rank may be due to a *syn-schuppen coalification* in addition to the pre-schuppen coalification. This later coalification may have been either caused by the deeper burial after the schuppen tectonics or by shear stress. One may argue that the  $R_m$  inversion between

schuppen *A* and *B* does not support the view that tectonic thickening was of any importance to contribute to the post-schuppen coalification. However, thrusting of the whole Falknis nappe (schuppen *B–E*) onto schuppe *A* may have been a late effect. On the other hand, in the strongly sheared central part of the Falknis nappe tectonic movements may have enhanced the coalification. The meta-anthracite coal rank represents a preliminary stage of the graphite formation. At this stage shear stress can promote the orientation and the extension of graphite-like crystallites, thus increasing the reflectance.

#### *Fluid inclusions*

The distribution of the fluid composition shows a simple pattern with CH<sub>4</sub>-bearing fluids in the upper and the middle part of the Falknis nappe (No. 244–250) and H<sub>2</sub>O-bearing fluids in the lower part of schuppe *B* (No. 241–243) and schuppe *A* (238 and 239). This distribution pattern argues for a post-schuppen generation of the fluid inclusions.

Minimum formation temperatures in the upper part of schuppe *C* range between 226° and 249 °C. The lower temperatures of 196–222 °C in the middle part of the nappe are probably due to a nonsaturated aqueous solution with respect to CH<sub>4</sub> during the homogenization of the fluid inclusions. The low homogenization temperatures of the H<sub>2</sub>O-rich fluids cannot be used for geothermometric purposes (see p. 185).

Minimum fluid pressures of 2310 to 4170 bar were obtained for four localities from the upper part of schuppe *C*. The high-pressure values and the large variation of fluid pressure within a small distance may be interpreted as fluid overpressures as discussed on page 187.

The above results obtained from *IC*, coal rank and fluid inclusion data may be summarized as follows:

- a) The determination of metamorphic grade by the different methods led to contradictory results for the Falknis nappe. According to the *IC* data either conditions of the diagenetic zone or the medium-grade anchizone were reached.
- b) The coalification was in part prekinematic with respect to the schuppen formation and occurred under conditions of deep diagenesis, but an additional syn- to postkinematic coalification continued in some places under anchizonal conditions.
- c) The distribution of fluid composition and the homogenization temperatures of schuppe *C* indicate medium-grade anchizonal conditions for the upper part and higher-grade anchizonal conditions for the lower part of the Falknis nappe.

#### **4. Correlation of illite crystallinity and coal rank**

##### *Previous work*

Several attempts have been made to correlate *IC* and coal rank data (KISCH 1971, 1974, 1975; FREY & NIGGLI 1971; WOLF 1975; TEICHMÜLLER & WEBER 1979, TEICHMÜLLER & TEICHMÜLLER 1979).

According to KISCH (1971) "the anchimetamorphic zone is found to be associated with anthracite (<8% volatile matter, corresponding to >2.5%  $R_m$ ) and low meta-anthracite (>0.7% hydrogen, corresponding to <2% volatile matter and >5%  $R_m$ ) rank". (Note, that Kisch refers to the North American coal rank classification.)

FREY & NIGGLI (1971) tried to correlate  $IC$  and coal rank indirectly on the basis of the coal rank associated with the deadline of oil (e.g. BARTENSTEIN & TEICHMÜLLER 1974, Table 1) and the definition for the beginning of the anchizone, which was supposed to be equivalent to the same deadline of oil (KÜBLER 1967, p. 111). From these data it was concluded that the beginning of the anchizone would fall within the "Fettkohlen-Stadium" ( $\approx 1.3\%$   $R_m$ ). Later studies have shown, however, that the beginning of the anchizone corresponds to the deadline of dry gas and not to that of oil (B. Kübler, personal communication 1976). For this reason the proposed correlation of FREY & NIGGLI (1971) becomes untenable.

From a study of telemagmatically metamorphosed and burial metamorphic rocks of the eastern Rhenish Schiefergebirge (West Germany) WOLF (1975) concluded that there exists no simple relation between  $IC$  and coal rank and that "each region is characterized by its own relationships".

In an extensive study based on 420 samples TEICHMÜLLER & WEBER (1979) found that  $IC$  and coal rank can not be correlated directly in all cases since  $IC$  and coal rank are influenced by different factors to different extents. Although both  $IC$  and coalification seem to depend mainly on temperature, the time relationship between temperature increase and deformation is important. In the absence of a synkinematic illite recrystallization, for example, the coalification will be enhanced relative to the  $IC$ . Because of the wide scattering of  $IC$  values at the boundary diagenesis/anchizone it was proposed to define the boundary between diagenesis and anchimetamorphism with a coal rank of approximately 3.5%  $R_m$  or 4%  $R_{max}$ , the anchi-/epimetamorphic boundary with the highest  $IC$  value ( $Hb_{rel.} = 120$ ), corresponding to values of 5–10%  $R_{max}$ . This means that the anchizone would correspond to the meta-anthracite coal rank (German coal classification).

### *Results and discussion*

The relationships between  $IC-R_m$  and  $IC-R_{max}$  of the four studied areas are shown in Figures 9 and 10, respectively, as well as calculated linear correlation equations with their corresponding correlation coefficients. Samples represented by open symbols were omitted from the correlation because in these samples the  $IC$  is apparently low due to the presence of mixed-layer muscovite/paragonite. Similar negative correlations with relatively high correlation coefficients ( $r = -0.84$  to  $-0.89$ ) were found for the Kien valley and the Lake Lucerne–Reuss valley areas. The broad bands formed by the data points may be due to the different factors affecting  $IC$  and coal rank. In this respect the influence of lithology, of weathering and of differential shear stress should be especially mentioned.

In sections I and II lithologies varied from slates to limestones (Tables 1a and 1b). It is well-known that the  $IC$  is dependent on the lithology and weathering, mainly in the diagenetic zone and to a lesser extent also at the beginning of the

anchizone (see e.g. KÜBLER 1968; TEICHMÜLLER & WEBER 1979). On the other hand it is generally assumed that the progress of coalification is not influenced by the chemical environment. The data presented by BOSTICK & FOSTER (1975) refer to very low rank coals and cannot be generalized, especially not for anthracites, as were studied in this paper.

Differential shear stress was very pronounced in these Alpine areas. According to KÜBLER (1967b) no correlation between schistosity zones and *IC* can be observed. FREY et al. (1973, p. 212) reported lower *IC* values (= higher crystallinity) in some samples from shear zones and FLEHMIG & LANGHEINRICH (1974) observed in an open fold that the *IC* increases with increasing tectonic strain. On the other hand, GRUNER (1976) found no such relationship in strongly folded slates of the Falknis nappe. Our data from different lithologies give no conclusive indication whether the *IC* was affected by differential tectonic movements.

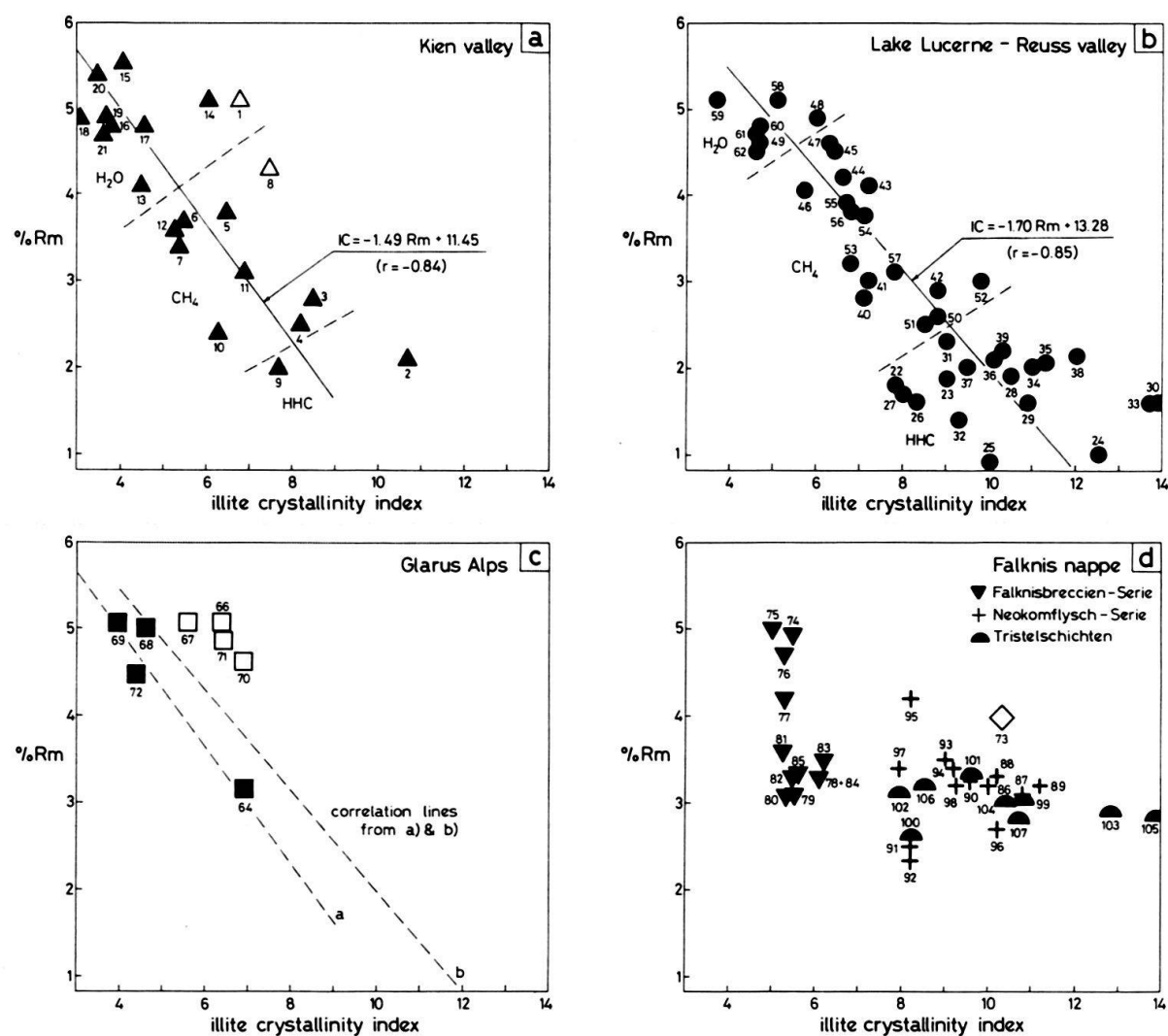


Fig. 9. Correlation of illite crystallinity (*IC*) versus the mean reflectivity ( $R_m$ ) of the four studied areas. Samples are numbered according to Tables 1a-d. Open symbols refer to samples whose *IC* is apparently lowered by the presence of other sheet silicates besides illite. Areas of different fluid composition (HHC,  $CH_4$ ,  $H_2O$ ) with respect to the *IC* and  $R_m$  data are schematically indicated in *a* and *b*. For discussion see text.



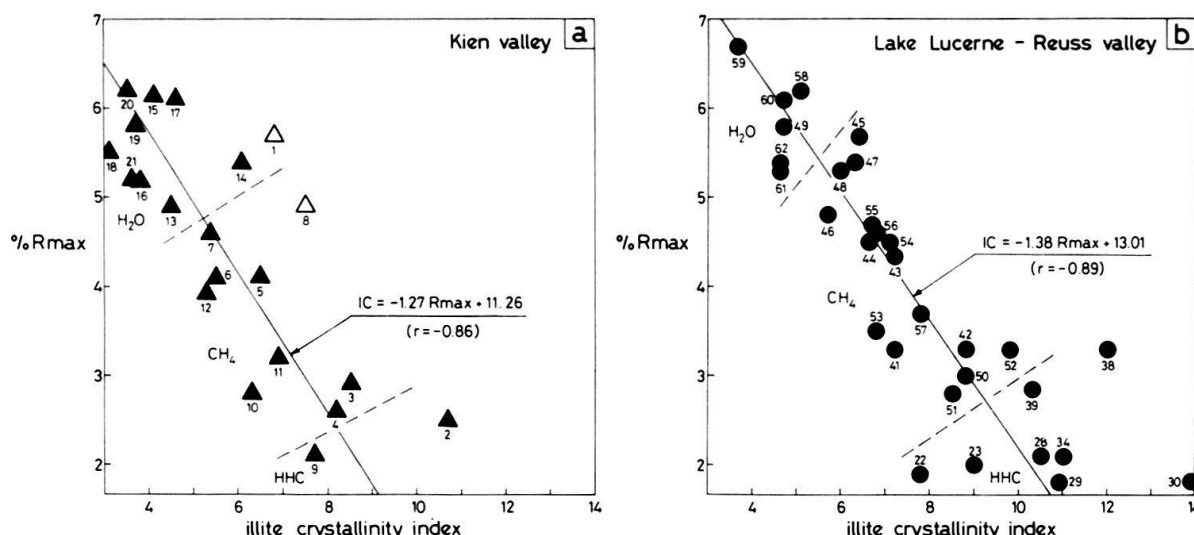


Fig. 10. Correlation of illite crystallinity ( $IC$ ) versus the maximum reflectivity ( $R_{max}$ ) of two out of the four studied areas. Symbols as in Figure 9. For discussion see text.

Approximately at the beginning of the anthracite stage ( $R_m = 2.5\%$ ) shear stress is able to increase  $R_{max}$  and reflectance anisotropy (see e.g. STACH et al. 1975, Fig. 120; TEICHMÜLLER & TEICHMÜLLER 1979; TEICHMÜLLER & WEBER 1979). In the Kien valley section samples 1, 5, 8, 14, 15 and 17 come from localities with particular high shear stress. From Figures 9a and 10a it can be seen that all these data points are actually situated above the correlation curves.

From section III (Glarus Alps) only four data points with undisturbed  $IC$  values (see p.192) are available (Fig.9c, filled symbols). The distribution of these four points is not in contradiction to those of sections I and II.

From the correlation curves of Figures 9a, b and 10a, b we calculated the coal rank limits for the anchizone of sections I and II (see Table 4a). The  $R_m$  limit for the

Table 4: Correlation of illite crystallinity and coal rank data.

a) Reflectivities of organolites for the lower ( $IC = 7.5$ ) and upper ( $IC = 4.0$ ) limits of the anchizone.

	Beginning of anchizone		End of anchizone	
	% $R_m$	% $R_{max}$	% $R_m$	% $R_{max}$
Section I	2.65	3.0	5.0	5.7
Section II	3.4	4.0	5.5	6.5
TEICHMÜLLER & WEBER (1979)	3.5	4.0	-	5-10
KÜBLER et al. (1979)	2.6-2.8	-	4.0	-
KISCH (1974)	~2.25	-	~4.0	-

b) Illite crystallinity values for the lower and upper limits of the anchizone as defined by coal rank data (TEICHMÜLLER & WEBER 1979).

	Beginning of anchizone	End of anchizone
	( $R_{max} = 4\%$ , $R_m = 3.5\%$ )	( $R_{max} = 5-10\%$ , $R_m > 5\%$ )
	$IC$	$IC$
Section I	6.3	4.8
Section II	7.4	5.7



beginning of the anchizone of section I is consistent with the value given by KÜBLER et al. (1979) but appreciably lower than the value proposed by TEICHMÜLLER & WEBER (1979). The  $R_{\max}$  limit for the end of the anchizone of this section is in agreement with the observations of TEICHMÜLLER & WEBER (1979). The reflectivity limits of section II agree well with the limits proposed by TEICHMÜLLER & WEBER (1979) but are much higher than those given by KÜBLER et al. (1979).

If coal rank data are used as a reference frame for the anchizone according to TEICHMÜLLER & WEBER (1979), then the  $IC$  limits for the beginning and the end of the anchizone of sections I and II can be calculated (Table 4b). This table shows that for section I – in contrast to section II – the boundary diagenesis/anchimetamorphism is characterized by relatively low  $IC$  values (= high crystallinity).

The  $IC$ -coal rank distribution diagrams of the Falknis nappe (Fig. 9d) are totally different from those discussed above. The correlations seem to be different for the three stratigraphic formations although slates were used in most cases (Table 1d). The Falknisbreccien-Serie shows a large variation in reflectivity at almost constant  $IC$  values while the Tristelschichten reveal the opposite relation. The Neokomflysch-Serie, on the other hand, shows an irregular  $IC$ -reflectivity distribution. Some possible explanations have already been discussed on pages 194–195.

### 5. Correlation of fluid inclusion data with illite crystallinity and coal rank

The fluid composition of sections I and II shows a relatively good correlation with the  $IC$  and  $R_m$  values. The transition from the HHC-zone to the  $CH_4$ -zone occurs at  $IC$  values ranging between 7 and 9 and  $R_m$  values of 2.5–3%, that is coincident with or just below the onset of the anchizone. This transition is located at minimum temperatures and pressures of 200 °C and 1200 bar, respectively.

The transition from the  $CH_4$ -zone to the  $H_2O$ -zone occurs at  $IC$  values of 4.5–5 and  $R_m$  values of 4.5–5%, that is in the higher-grade part of the anchizone. Minimum temperatures and pressures are 270 °C and 1700 bar, respectively. These correlations are schematically shown in Figures 9 and 10.

The continuous change of the fluid composition from north to south in sections I and II indicates that the fluid evolution is mainly dependent on the temperature increase during progressive Alpine metamorphism.

### 6. Conclusions

- a) The combined application of  $IC$ , coal rank and fluid inclusion data has turned out to be a successful approach to study the diagenesis and anchimetamorphism in external parts of the Central Alps.
- b) Metamorphic grade increases generally from tectonically higher to lower units and from external (in the north) to internal (in the south) parts of the same tectonic unit. However, there exist several inversions where obviously higher-grade metamorphic units were thrust onto lower-grade ones. Post-metamorphic thrusting seems to be a common feature in the external parts of the Alps.

- c) Although in some areas a good correlation between *IC*, reflectivity of coaly material and fluid inclusion data was found, there exists no generally valid relation between the three parameters. The relationship obviously depends on the level of diagenesis and metamorphism and on the tectonic history, especially upon the time relationship between temperature increase and tectonics.

### Acknowledgments

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