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CARBON ISOTOPE RATIOS OF GRAPHITES IN THE BÜNDNERSCHIEFER SERIES OF THE TAUERN WINDOW AND THE KÖSZEG-RECHNITZ WINDOWS (AUSTRIA AND WESTERN HUNGARY): ORIGIN OF ORGANIC MATTER AND SEDIMENTARY FACIES CORRELATION

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Abstract: Carbon isotope ratios of graphites were studied in carbonate-free metasediments of the Tauern Window and the Köszeg-Rechnitz Windows of eastern Austria and western Hungary that belong to the Alpine Bündnerschiefer series. The stable carbon isotopic compositions of the graphites range from about -22 ‰ to -12 ‰ and show systematic regional distribution patterns. Variations in $\delta^{13}C$ are explained as mixing of a normal marine component (-22 ‰) and a detrital carbonaceous component (-12 ‰). The detrital component is similar to the carbonaceous matter of black pebbles of conglomerate bodies that occur in the same sedimentary sequence. Based on the $\delta^{13}C$ values, a distinction can be made between sediments deposited in the southermost part of the Penninic ocean basin and sediments deposited more to the north. The results are used for possible sedimentary facies correlations between rocks in the Alpine Penninic windows.

Key words: stable carbon isotopes, graphite, Alpine Bündnerschiefer, facies correlation.

Introduction

The Bündnerschiefer series is one of the most important units of the Alpine edifice and crops out in several windows in the Central and Eastern Alps. This series formed during the opening of the Penninic ocean basin and consists of variably metamorphosed ophiolitic magmatites and flysh-like sediments deposited in the basin (Janoschek & Matura 1980; Trümpy 1980). Hence, investigations on these rocks can provide valuable data for the formation and evolution of the basin and for the later metamorphic influences in the course of the Alpine orogeny.

In the Eastern Alps the Bündnerschiefer series crops out from below the Austroalpine nappes in three main areas: the Engadine Window, the Tauern Window and the Köszeg-Rechnitz Windows. Among these series, the rocks of the Tauern Window have been studied most extensively as not only the P-T conditions and age relationships of the metamorphic events have been determined (e.g. Höck 1980; Droop 1985; Cliff et al. 1985), but also the original sedimentary characteristics and sedimentary facies distributions have been described (see for example Janoschek & Matura 1980).

Although the other two areas mentioned above have not been studied in such details, the general characteristics of the rocks and the metamorphic conditions in the Köszeg-Rechnitz series have been described (e.g. Koller & Pahr 1980; Koller 1985). These investigations mainly dealt with the metamagmatites of the series, so the formation and evolution processes of the metasediments are not well determined.

As graphite-bearing metasediments are frequent in the Bündnerschiefer series and as the graphite not in contact with other carbon reservoirs (e.g. carbonates) more-or-less reflects the isotopic composition of the original organic material (see Discussion), such studies can provide valuable data for the comparison of the metasediments of the well known Tauern Window and the Köszeg-Rechnitz series.

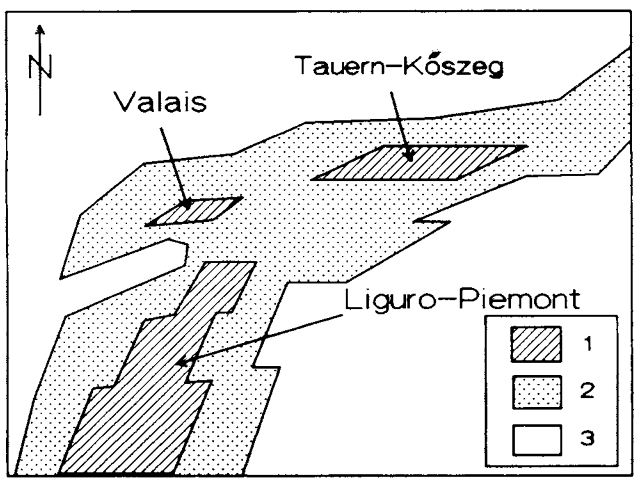

Fig. 1. Palaeogeographic sketch of the Penninic ocean basin with the main ophiolite-bearing complexes.
1 - oceanic crust; 2 - thinned continental crust; 3 - continental crust. After Lemoine & Trümpy (1987).

We mention here that although the degree of well developed graphite was not reached in some samples (for example in the pebbles of conglomerates discussed later), the majority of the samples fall in the range of well developed graphite established by Landis (1971) (see Demény 1990), therefore we use the word "graphite" throughout this paper.

As a contribution to the knowledge of the sedimentary characteristics of the Alpine Bündnerschiefer series, this paper deals with the carbon isotopic compositions of graphites of carbonate-free rocks and raises the possibility of a facies correlation between Bündnerschiefer units of the Eastern Alps.

Geological background

The history of the Alpine Penninic rocks to which the studied series belong can be devided into three phases:

1 - opening of the Penninic ocean and sedimentation
2 - subduction and related HP/LT metamorphism
3 - Tertiary metamorphism and tectonism.

Several paleogeographic pictures have been proposed for phase 1. Fig. 1 shows a reconstruction of the Penninic oceanic basin during sedimentation in Jurassic to Lower Cretaceous times. The sediments were rich in clastic material that derived

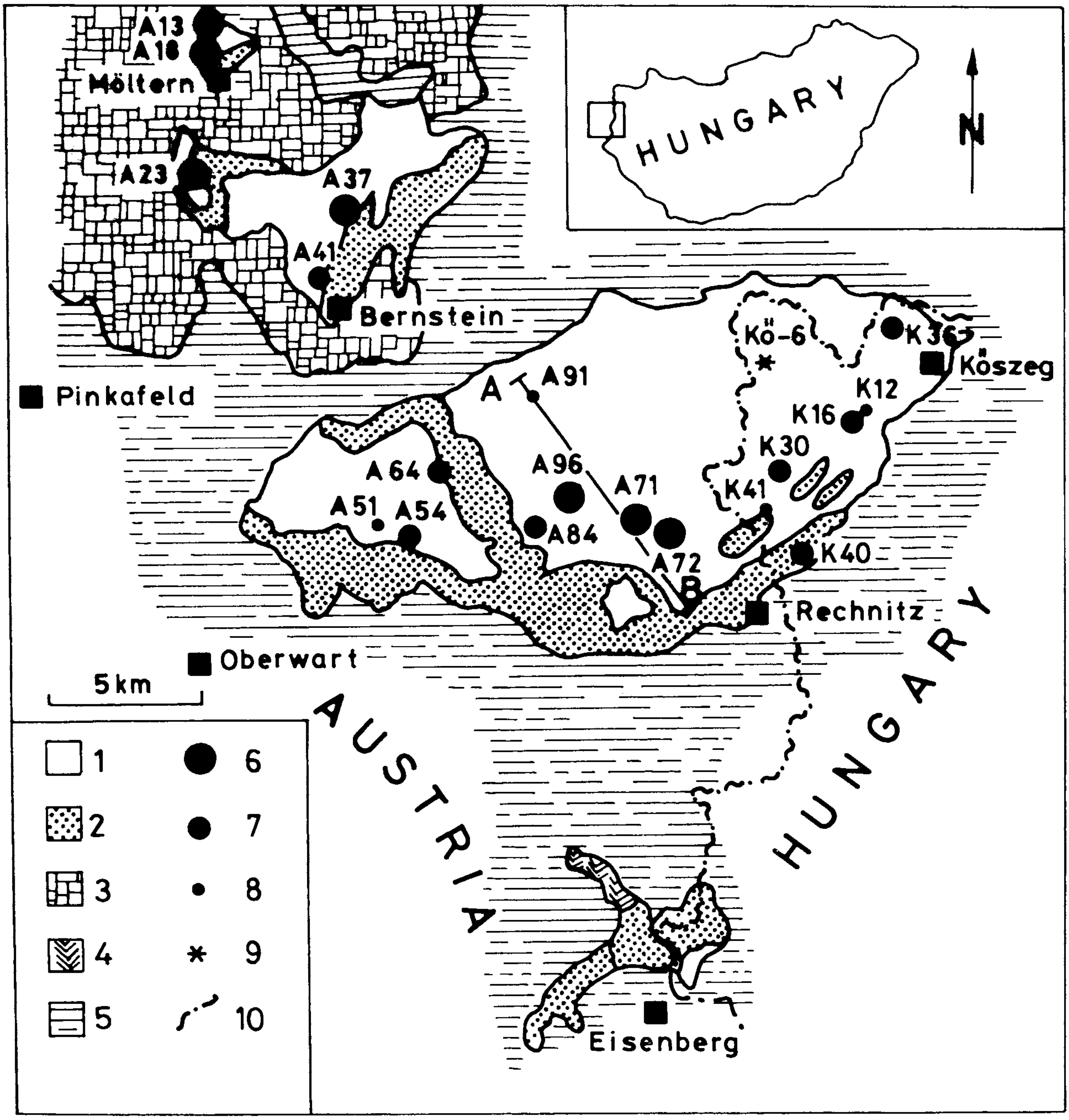

Fig. 2. Distribution of the Penninic windows in the eastern end of the Alps (after Koller 1985) and sample localities with the stable carbon isotopic compositions of graphites.
1 - Penninic metasediments; 2 - Penninic metamagmatites; 3 - lower Austroalpine nappes; 4 - upper Austroalpine nappes; 5 - Tertiary sediments; 6 - $\delta^{13}C$ < -20‰; 7 - $\delta^{13}C$ = -17 to -20‰; 8 - $\delta^{13}C$ >-17 ‰; 9 - bore-hole; 10 - boundary.

from erosion of rocks that were exposed south-east of the through (Tollmann 1980). Subduction phase 2 during the Lower - Upper Cretaceous caused HP/LT metamorphism, deformation and early nappe formation. Tertiary metamorphism and tectonism phase 3 came in two subphases, 40 - 35 My and about 20 My ago, respectively. The first subphase produced the well known metamorphic zoning in the Lepontine Alps (Hurford 1986) and in the Tauern Window (Cliff et al. 1985); the second subphase caused doming and uplifting.

The Köszeg-Rechnitz series is the easternmost occurrence of the Alpine Bündnerschiefer unit and crops out in four small windows from below the lower Austroalpine nappes. From these windows we are going to deal with the metasedimentary rocks of the Möltern Window, the Bernstein Window and the Köszeg-Rechnitz Window (Fig. 2), the latter being the largest and best known one. Földváry et al. (1948) and Bandat (1928, 1932) made the first geological maps, while Koller & Pahr (1980) and Koller (1985) discussed the petrology of the area. The newest maps were made by Pahr (1984) and Ferencz et al. (1988).

The rocks can be divided into metamagmatites and metasediments (Fig. 2). The metamagmatites consist of serpentinites, metagabbros and greenschists. According to Kubovics (1983) and Koller (1985) these rocks represent ophiolites and can be correlated with the Glockner Nappe of the Tauern Window.

The metasedimentary rocks are phyllites with variable amounts of quartz, micas, carbonates, chlorite and albite. All the samples studied contain graphite (with graphite contents from 0.1 to 4%). In this study only carbonate-free rocks were selected excepted one sample, as the carbonate-graphite isotope exchange could affect the original isotopic composition of the carbonaceous matter. This exceptional sample - black carbonate pebbles containing carbonaceous material - was collected from a conglomerate body and will be discussed in details later. Accessory minerals such as tourmaline, zircon, rutile, apatite and opaque minerals are also frequent (Demény 1988).

Conglomerate bodies with black-grey dolomite and limestone pebbles and minor amount of metamorphic debris occur sporadically. These conglomerates indicate sedimentation close to a coast where unmetamorphosed carbonate rocks together with some granites and metamorphic rocks were exposed (Demény 1988). The origin of these conglomerate bodies has long been debated as they occur in fine-grained sedimentary rocks of deep-marine character (Koller pers. communication 1989). On the other hand, the Penninic ocean basin was bordered by transform zones with steep slopes (Weissert & Bernoulli 1985) which could have resulted in mixing of shallow-water sediments into deep-water ones.

The age of the sediments has been established by Schönlaub (1973) who found Middle Cretaceous sponge spiculae in some calcschists. Mostler & Pahr (1981) made a significant contribution to the studies on the conglomerate problem describing Triassic fossiles in the carbonate pebbles.

As for the further evolution of the series, three metamorphic events have been recorded in the rocks of the Köszeg-Rechnitz series: oceanic hydrothermal activity, subduction-related HP/LT metamorphism and Barrovian-type young Alpine metamorphism.

Only sporadic remnants exist from the mineral assemblages of oceanic hydrothermal effect (barroisite, Mg-hornblende, Cr-andradite, chromite), while rocks related to the HP/LT metamorphism (blueschists, crossitites), as well as mineral relicts (glaucophane, lawsonite, pumpellyite) in greenschists and metagabbros are preserved at some localities in spite of the extensive effect of the young Alpine metamorphism (Lelkes-Felváry 1982; Kubovics 1983; Koller 1985). Koller (1985) determined temperature and pressure ranges for these phases with T=750 °C for the oceanic hydrothermal influence and T=330 - 370 °C, P=6 - 8 kbar for the HP/LT metamorphism. The young Alpine event is the major phase preserved in the metasediments; the conditions are 350 - 430 °C, increasing southwards, and a maximum pressure of 3 kbar (Koller 1985). This major influence resulted in the graphitization of the original organic matter of the sediments in variable degree depending not only on the temperature effect, but also on the nature of metamorphic fluids and tectonic movements (Demény 1990). Cooling and uplifting after the young Alpine event took place 15.1 - 18.5 My ago as defined with zircon fission track studies by Demény & Dunkl (1991).

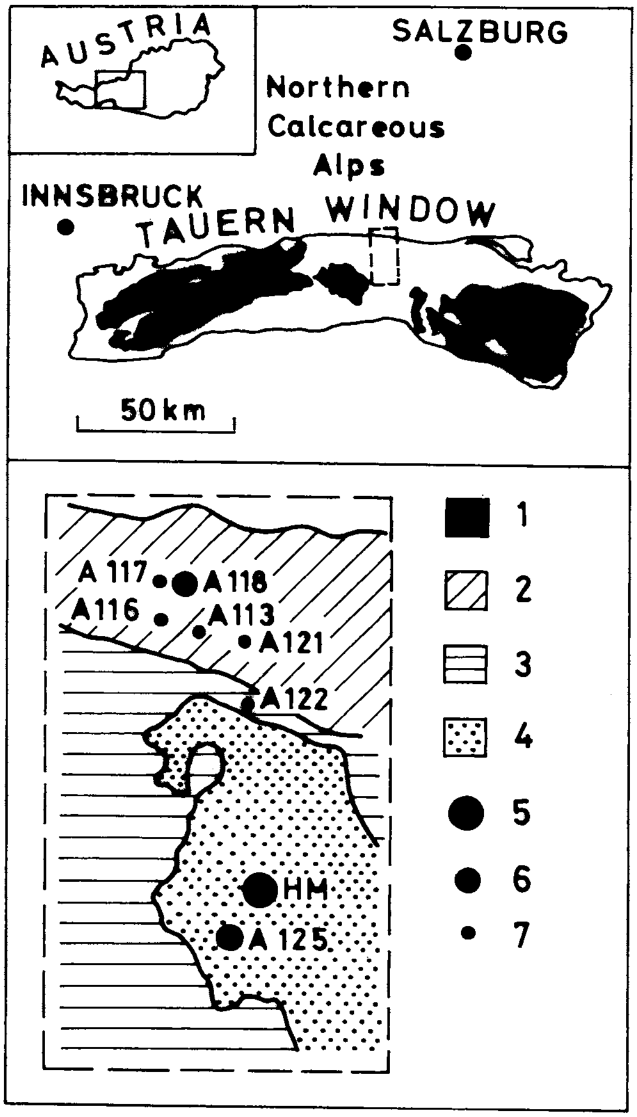

Fig. 3. Geological sketch map of the Tauern Window (after Janoschek & Matura 1980; Droop 1985) with sample localities and stable carbon isotopic compositions of graphites.
1 - Central Gneiss; 2 - Fusch facies; 3 - Glockner facies; 4 - Brennkogel facies; 5 - $\delta^{13}C < -20‰$; 6 - $\delta^{13}C = -17$ to $-20‰$; 7 - $\delta^{13}C > -17‰$ (values to PDB).

The geology of the Tauern Window is reviewed by Janoschek & Matura (1980) and Tollmann (1980). The Tauern Window consists of three main units: the Central Gneiss, the Paleozoic cover schists and the Bündnerschiefer series (Fig. 3). The latter consists of metasediments (mainly phyllites and schists) and metamagmatites of ophiolite origin. In this study we will only deal with the metasediments. Four sedimentary facies belts have been defined in the Bündnerschiefer: the Hochstegen, the Brennkogel, the Glockner and the Fusch facies. At the time of sedimentation, the Hochstegen facies formed in the northern part of the Penninic ocean, whereas the Fusch facies formed along the southern coast. Later tectonic movements turned the facies distribution over, so that the Fusch facies is now in the north. The Fusch facies contains breccias and conglomerate bodies that indicate a strong terrestrial influence. The metamorphic history is similar to that of the Köszeg-Rechnitz series. The Tertiary Barrovian-type metamorphic conditions were somewhat higher than in the Köszeg-Rechnitz windows: from 450 °C (Fusch facies) to 500 °C (Brennkogel facies) and 4 - 5 kbar.

Analytical techniques

Samples were treated with 35 % HCl to remove traces of carbonates and washed until reaching neutral pH. Depending on the graphite content, 50-150 mg of sample was burnt at 900 °C in 0.2 atm oxygen. The burning products were repeatedly circulated through CuO at 900 °C for complete oxidation. H_2O and CO_2 were condensed in melting acetone and liquid nitrogen cooled traps, respectively. The isotope ratios of CO_2 were measured with a VG SIRA-24 mass spectrometer. Traces of SO_2, which could interfere with the isotope measurement, were checked-for in brackground scans. It appears, however, that SO_2 is quantitatively trapped as H_2SO_4 in the oxidation line. The isotope results are expressed as $\delta^{13}C$ relative to PDB. The combustion method was calibrated on NBS21 graphite (standard now exhausted) and on NBS22 oil. $\delta^{13}C$ values of -28.2 and -29.7 ‰ respectively are obtained on these reference samples. The overall reproducibility was better than 0.15 ‰.

Table 1: $\delta^{13}C$ values of graphites of the Köszeg-Rechnitz series, the Tauern Window and the lower Austroalpine Wechsel series. All data are expressed in ‰ and relative to PDB (Pee Dee Belemnite).

sample	$\delta^{13}C$	sample	$\delta^{13}C$	sample	$\delta^{13}C$
		Közeg-Rechnitz series			
A13	-21.5	A18	-22.3	A23	-21.7
A37	-21.2	A41	-18.9	A51	-16.5
A54	-17.5	A64	-19.8	A71	-21.2
A72	-20.4	A84	-18.6	A91	-14.8
A96	-21.5	K12	-14.2	K16	-18.3
K30	-17.9	K36	-18.1	K40	-18.1
K41	-12.3	Kö-6/1	-20.1	Kö-6/2	-19.3
		Tauern Window			
A113	-14.0	A116	-13.9	A117	-16.0
A118	-18.1	A121	-14.9	A125	-17.5
HM	-21.0				
		Wechsel series			
Fr-403	-29.4	Fr-404	-30.2	Fr-407	-30.7
Fr-423	-30.0	Fr-2048	-28.8	B9	-30.1
B	-33.6				

Results

The analytical results on graphites from the Köszeg-Rechnitz Windows and the Tauern Window are listed in Table 1. Plotting the data on a map, one can see systematic regional distribution patterns (Figs. 2 and 3). Graphites in the Möltern Window and in the northern part of the Bernstein Window have d13C values close to -22 ‰, whereas in the southern part of the Bernstein Window we find values around -19 ‰. Graphites in the Köszeg-Rechnitz Window show a concentric distribution pattern with values around -20 ‰ in the central part, and -18 down to -12.3 ‰ at the margins. Two samples collected from a bore-hole (Kö-6/1, 2) at depths of 118 and 157 m gave -20.1 and -19.3 ‰, respectively.

Samples collected from the Tauern Window also show a regional distribution pattern (Fig. 3). Graphites from the Fusch facies (samples A113-A121) vary between -13.9 and -18.1 ‰, while those from the Brennkogel facies gave -17.5 and -21.0 ‰.

Discussion

The origin of isotopic compositions

The regional distribution patterns cover a continuous $\delta^{13}C$ range from about -22 to -12 ‰ and suggest mixing of two types of organic matter. Values of -22 ‰ indicate normal marine organic matter (e.g. Newman et al. 1973; Schultz & Calder 1976; Fontugne & Duplessy 1986). The isotopic composition of the other end-member (graphite of -12 ‰) is less common, and more difficult to explain.

Generally, variations in $\delta^{13}C$ may be related to several factors, the most important ones are discussed below. The effects of changes in the climate and the sedimentary environment are excluded, as they are not relevant for the observed differences in $\delta^{13}C$ (Andersen & Arthur 1983; F. Koller, personal communication, 1989).

1 - Effect of metamorphism. Barker & Friedman (1969), McKirdy & Powell (1974) and Hoefs & Frey (1976) observed shifts towards higher $\delta^{13}C$ with increasing metamorphism of organic matter and attributed this to the liberation of large amounts of isotopically light CH_4. On the other hand, no such effects have been found in other studies (e.g. Gavelin 1957; Hahn-Weinheimer & Hirner 1981; Douthitt 1982).

Data reported in earlier XRD studies of the graphites in the Köszeg-Rechnitz window (Demény 1990) show no correlation with degrees of the main Alpine metamorphism, nor with the $\delta^{13}C$ values.

Beside these observations, it is apparent that both $\delta^{13}C$ end-members with -20 and -12 ‰ exist in the Köszeg-Rechnitz Window at about the same metamorphic temperature, therefore the temperature effect could not cause the observed $\delta^{13}C$ shift.

Isotope exchange with carbonates and metamorphic fluids can also modify the isotopic composition of graphite. Our samples, however, were collected from carbonate-free rocks that makes the influence of carbonates unlikely. As for the effect of fluids, the observed isotope shift would require huge amount of pervasive fluid with high CO_2 content. On the contrary, studies on the nature of fluids of the Alpine metamorphism established that these fluids were rather channelized than pervasive (Hoernes & Friedrichsen 1978, 1980; Friedrichsen & Morteani 1979).

2 - Detrital material. Two possible sources of detrital material are land-derived plant material and erosion of preexisting rocks. Land-derived plant material normally has more negative $\delta^{13}C$ values than marine organic matter (Newman et al. 1973; Schultz and Calder 1976; Fontugne and Duplessy 1986) and therefore be ruled-out. Admixing of detrital carbonaceous material made available by erosion of preexisting rocks is an attractive hypothesis for rocks that show a strong terrestrial influence (such as the Fusch facies). Since the isotopic composition of the admixed component is rather uncommon ($\delta^{13}C$ = -12 ‰), stable isotopes form a powerful tool to correlate the detrital graphite with its source rock.

We have studied two possible source rocks of detrital carbonaceous matter. The first one is the lower Austroalpine Wechsel unit in which graphitic schists are abundant. At the time of sedimentation, this unit was situated near the southern coast of the Penninic ocean (Tollmann 1980). Graphites in 6 out of 7 samples however, gave a $\delta^{13}C$ of -29.9 ± 1 ‰ and one sample showed a value of -33.6 ‰ (Tab. 1). As the graphite of this unit is perfectly ordered (Szabó 1988) it was probably formed under high grade metamorphic conditions. Well ordered graphite is rather insensitive to low-temperature overprinting and therefore we can expect the graphite to have remained unchanged during the Alpine metamorphism. From these considerations we can conclude that our data range can not be explained by denudation of the graphite-bearing Wechsel schists, as these graphites could not cause the observed isotope shifts in positive direction.

Another possible source rock is found in the conglomerate bodies that occur not only in the Tauern Window, but also in the Kőszeg-Rechnitz series. The conglomerates contain black-grey dolomite and limestone pebbles and minor amounts of metamorphic debris. The carbonate pebbles are only slightly recrystallized preserving the original sedimentary structures and fossils (Mostler & Pahr 1981). We have conducted stepped heating experiments in order to compare the carbonaceous matter of the pebbles with the general graphite of the studied Bündnerschiefer rocks. The main combustion temperatures of the graphites range between 500 and 700 °C. Most of the carbonaceous matter of the pebbles gave the same combustion temperature, but a significant fraction of the material - about 10 % - burned below 400 °C indicating that this fraction was not graphitized. It can be interpreted as unmatured organic matter formed during the graphitization process and trapped in the pebbles due to the low permeability. This low permeability excludes the effective isotope exchange between the carbonate and the carbonaceous matter which process would have operated in a fluid-rich environment.

The bulk carbonaceous matter gave a $\delta^{13}C$ of -12.7 ‰, similar to the highest values in our data range. As discussed above, the unusually high $\delta^{13}C$ of the carbonaceous matter in the conglomerates is probably not a result of calcite-graphite isotope exchange, regarding the weak recrystallization of the pebbles.

Crawford & Valley (1990) have made similar conclusions studying graphite- and carbonate-bearing rocks of amphibolite and granulite facies. They found that no significant carbonate-graphite carbon isotope exchange took place in fine-grained gneisses even in the amphibolite facies. Therefore it is no surprise that the carbonaceous matter in our pebbles retained its original isotopic composition in spite of the presence of carbonate. Instead, we think that the isotopic composition is characteristic of the precursor organic matter.

According to the present knowledge, organic matter of seagrasses (Parker 1964; Frey et al. 1977), salt marshes (Delaune 1986) and algal mats (Behrens & Frishman 1971) can provide similar values. It is therefore interesting to note that we have found algal mat-like structures in these pebbles (Fig. 4). These results indicate that the source rocks of the conglomerates may have provided part of the carbonaceous matter presently found in our rocks, through denudation and shedding into the ocean basin.

For such an explanation of the data, large scale continental weathering is a necessary condition. Such condition is supported by the work of Simoneit (1986) who suggested a rather humid climate for the Atlantic-Penninic areas and found evidence for a strong terrestric influence with detrital carbonaceous matter in Atlantic rocks of the same age.

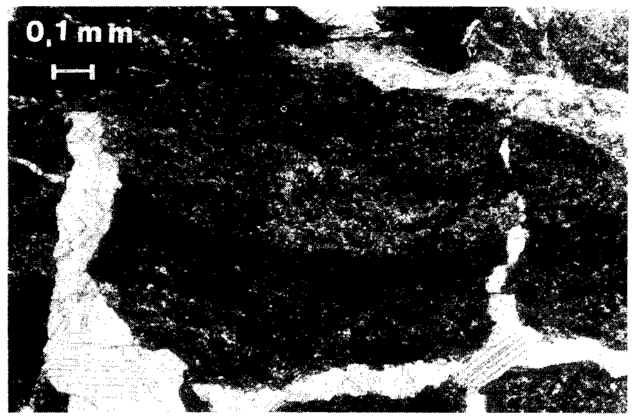

Fig. 4. Algal mat-like structures in a black carbonate pebble of the conglomerate described in the text. Thin section. 1 nicol.

Facies correlation

A number of studies showed that the carbon isotopic composition of graphite in carbonate-free rocks does not normally change with metamorphism (see references above). Hence, we can use variations in the isotopic composition of graphites as a tool for sedimentary facies correlation. This is especially true, if the graphites have carbon isotopic compositions that strongly deviates from the usual values around -25 ‰. Graphites in the present sample set seem therefore particularly suitable for this purpose.

The data in Tab. 1 and Figs. 2 and 3 show that graphites in both the Tauern Window and the Kőszeg-Rechnitz Windows cover a range from about -22 up to -14 ‰. In the Tauern Window, the unusally high $\delta^{13}C$ values occur in the Fusch facies, whereas the Brennkogel facies shows the normal low $\delta^{13}C$ values. This distribution is opposite to what could be expected from the metamorphic conditions, as the least metamorphosed material showed the most positive compositions. According to Janoschek & Matura (1980) and Tollmann (1980), the Fusch facies was deposited in the southernmost part of the Penninic ocean and the Brennkogel facies more to the north. Thus, the heavy carbon must have come from erosion of rocks exposed along the southern coast of the Penninic ocean. Later tectonic movements reversed the positions of the Fusch facies and the Brennkogel facies.

In the Kőszeg-Rechnitz Windows, high $\delta^{13}C$ values similar to those of the Fusch facies are found along the margins of the Kőszeg-Rechnitz Window (Fig. 2). The central part of the Kőszeg-Rechnitz Window, as well as the Bernstein Window and the Möltern Window, have normal marine $\delta^{13}C$ values such as found in the Brennkogel facies.

Two samples from a bore-hole near Kőszeg (Kő-6/1, 2 at 118 m and 157 m, respectively) give a clue to a better understanding. These bore-hole samples have values around -20 ‰, similarly to graphites in the central part of the Kőszeg-Rechnitz Window, while surface samples at the same locality have higher values around -18 ‰.

Although more data would be needed to determine the $\delta^{13}C$ distributions of the graphites in the Bündnerschiefer facies belts, the observed differences point to the complex structure of the Kőszeg-Rechnitz window and to the correlation possibilities between the Tauern window and the Kőszeg-Rechnitz series.

Looking at the collected data set - $\delta^{13}C$ values in the Tauern facies belts, areal distributions in the Kőszeg-Rechnitz window and the $\delta^{13}C$ differences within the Kő-6 bore-hole -, the interpretation of the $\delta^{13}C$ variability in the easternmost Penninic windows is that rocks of the southern facies (probably related to the Fusch facies) with isotopically "heavier" graphite were thrusted upon the northern facies belt characterized by isotopically "lighter" graphite. Later uplift resulted in exposure of the lower tectonic unit (northern facies belt) in the central part of the Kőszeg-Rechnitz Window and in the Bernstein and Möltern Windows. The upper tectonic unit (southern facies belt) was mostly eroded away, but is preserved along the margins of the Kőszeg-Rechnitz Window. The situation is illustrated by Fig. 5 showing a generalized cross-section through the Kőszeg-Rechnitz Window.

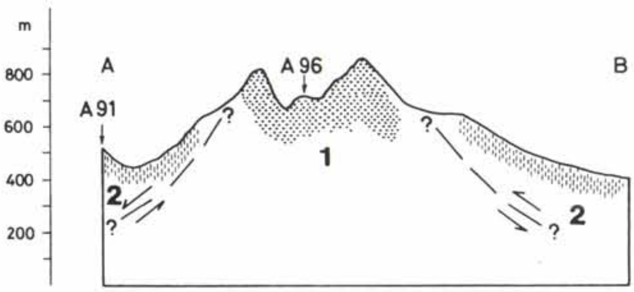

Fig. 5. Cross section through the Kőszeg-Rechnitz Window. The localities of points "A" and "B" are shown in Fig. 2.
1 - $\delta^{13}C$ < -20 ‰; 2 - $\delta^{13}C$ > -20 ‰ (values to PDB).

Fig. 6 shows a tentative reconstruction of the Penninic ocean basin with two sedimentary facies belts, conglomerate bodies, direction of sediment transport and isotope trends. Although this picture is based only on studies of the Kőszeg-Rechnitz Windows and the Tauern Window, the good correlation of the data from different areas could give a hint at the possibility of sedimentary facies correlation throughout the whole Alpine Bündnerschiefer series. It is also questionable whether this facies distribution ends in the series of the Kőszeg-Rechnitz Windows, or there is a continuation toward east (e.g. the Carpathians). This proposal calls for further investigations on graphite-bearing rocks of other Bündnerschiefer units of the Eastern and Central Alps and on Carpathian series of the same age.

Conclusions

1 - $\delta^{13}C$ variations of graphites in the Penninic Windows in Austria and western Hungary reflect mixing of normal marine organic matter with isotopically heavy detrital carbonaceous material.

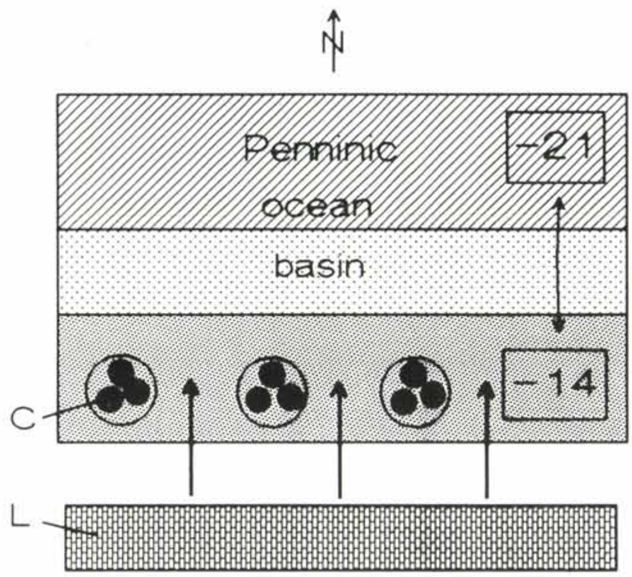

Fig. 6. Schematic picture of the Penninic ocean basin in the Tauern-Köszeg system with the isotope ranges of the two main facies belts (values are in ‰, relative to PDB), the conglomerate bodies ("C") and the position of their source terranes ("L", limestones). The arrows indicate the direction of the sediment transport. Not to scale.

2 - The detrital carbonaceous matter is characterized by $\delta^{13}C$ values of about -12 ‰ and probably originated from previously unmetamorphosed carbonate rocks containing algal mat structures.

3 - On the basis of isotopic compositions of graphites, two main sedimentary facies belts have been determined in the Kőszeg-Rechnitz Penninic Windows that might be correlated with the Brennkogel and the Fusch facies of the Tauern Window. From these results a uniform sedimentary facies distribution is proposed.

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