

## Oxygen isotope study of the highly fractionated Podlesí granite system, Krušné hory Mts., Czech Republic

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**Abstract.** The extremely fractionated, Li-, P- and F-rich, and Sn-, W-, Nb-, and Ta-bearing minor granite stock cropping out near Podlesí east of Potůčky, western Krušné hory/Erzgebirge Mts., was studied using oxygen isotope geochemistry of quartz and other silicate minerals. A total of 39 monomineral analyses were taken from 13 rock samples.  $\delta^{18}\text{O}$  values were obtained from quartz (9.2 to 11.6‰), feldspar (7.1 to 10.6‰), and mica of protolithionite and zinnwaldite composition (5.8 to 8.6‰, SMOW). The  $\delta^{18}\text{O}$  relationships among minerals indicate the influence of hydrothermal processes in sub-solidus conditions, which especially affected the feldspars. The oxygen isotope systematics of these rocks results from the activity of high- $\delta^{18}\text{O}$  fluids, while no major involvement of low- $\delta^{18}\text{O}$  fluids was evident. Whole-rock  $\delta^{18}\text{O}$  data, estimated based on mineral  $\delta^{18}\text{O}$  data and modal composition, range between 9.5 and 10.5‰ SMOW, which demonstrate the limited effect of hydrothermal activity and point to a crustal origin of the Podlesí granite.

**Key words:** Krušné hory/Erzgebirge Mts., granite, oxygen isotopes, isotope thermometry

### Introduction

The Podlesí granite system, located in the western part of the Krušné hory (Erzgebirge) Mts. east of Potůčky, Czech Republic, is a well-known example of an extremely fractionated, phosphorus- and fluorine-rich rare-metal granite (Breiter et al. 1997, 2005; Breiter 1998, 2001, 2002). The tongue-shaped granitic body, cropping out in an area of about 0.1 km<sup>2</sup>, represents the youngest intrusion of the multistage, late-Variscan, peraluminous, tin-bearing Eibenstock-Nejdek pluton. The Podlesí granite intruded Ordovician phyllites and biotite granite of a previous intrusive phase. Kostitsyn and Breiter (2001) obtained a Rb-Sr isochron age of 315 ± 2 Ma from a set of 8 rock samples from Podlesí.

The internal structure of this stock is well documented from several boreholes that reach maximum depth of 350 m. According to Breiter (2002), the Podlesí granite body consists mainly of albite-protolithionite-topaz granite in several sub-facies (Li-mica protolithionite, in the sense of Weiss et al. 1993). The upper facies constituting the uppermost 30 to 40 m of the stock is fine-grained and porphyritic, while the lower facies of the main part of the body is medium grained and non-porphyritic. In the uppermost 100 m, the granite stock is intruded by several flat-lying dykes of albite-zinnwaldite-topaz granite (zinnwaldite is a transitional member between polyolithionite and siderophyllite). The upper contact of a major dyke cropping out in an old quarry contains unidirectional solidification textures (UST) with quartz-alkali feldspar laminae alternating with comb quartz, comb orthoclase, and/or layers of oriented, fan-like, zinnwaldite aggregates. A marginal pegmatite zone (Stockscheider) is locally present at the upper

contact of the Podlesí stock. Breiter et al. (1997) and Breiter (2002) interpret this granite body as originating from a relatively shallow intrusion of volatile-rich melt resulting from the pronounced fractionation of the Eibenstock-Nejdek granite pluton. Cooling and crystallization of the upper part of the Podlesí body enabled the opening of brittle structures, into which residual melts extremely enriched in F, P, Li, and water intruded, thus forming a set of flat dykes. Crystallization in the dykes was related to the repeated opening of the system, undercooling, and the escape of fluid phases.

The chemical data of the rock-forming minerals show distinct differences between the stock granite and the dyke granite, reflecting the above mentioned two crystallization stages during their magmatic evolution (Breiter et al. 1997). The entire magmatic system at Podlesí shows only minor effects of post-magmatic processes, particularly greisenization, and no extensive meteoric-hydrothermal alteration (no extensive alteration of feldspars or micas, see Breiter et al. 1997, 2005, Breiter 2002, for detailed petrologic characteristics of the Podlesí stock). The system is therefore suitable for the study of  $\delta^{18}\text{O}$  systematics of the individual magmatic rock types. The preliminary results of the oxygen isotope study of the Podlesí granites were published by Žák et al. (2001). The position of the studied samples within a simplified geological cross section of the Podlesí granite is shown in Fig. 1.

### Sampling approach and methods

Two methods are available for preparing samples of granitic rocks for oxygen isotope studies. Either whole-rock

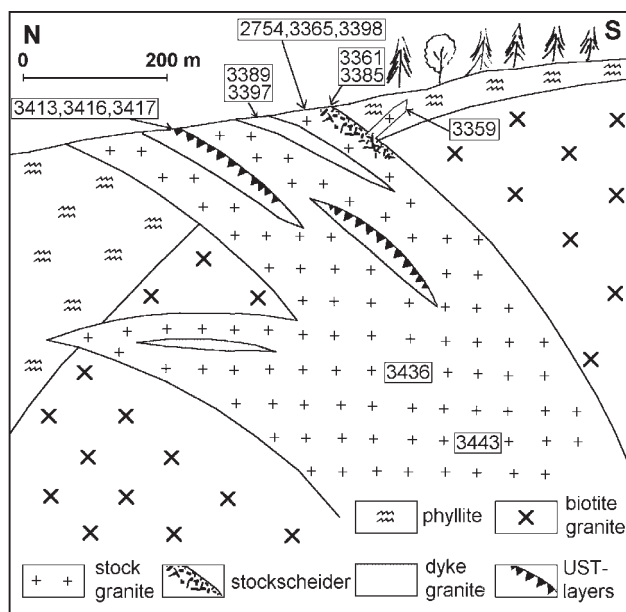


Figure 1. Simplified geological cross section of the Podlesí granite with location of the studied samples.

$\delta^{18}\text{O}$  values can be obtained from homogenized rock samples, or individual rock-forming minerals can be separated, and the  $\delta^{18}\text{O}$  values determined for each one independently. Data obtained using the first approach represent an average  $\delta^{18}\text{O}$  value for the whole sample, including all post-crystallization alterations of the original isotopic relationships. The second approach theoretically enables the calculation of  $[n \cdot (n - 1)]/2$  isotopic temperatures for  $n$  rock-forming minerals, and the evaluation of the processes of isotope exchange and the degree of equilibrium achieved between mineral phases and fluids. In reality the number of suitable pairs is usually lower, since oxygen isotope fractionation in some mineral pairs is too small relative to the analytical uncertainty. When  $\delta^{18}\text{O}$  data of individual minerals are available, the oxygen isotope systematics of the rock-forming minerals can be evaluated in  $\delta$  vs.  $\delta$  plots, which are the fundamental coordinate systems for stable isotope data (Gregory and Criss 1986). Another advantage of the second approach is the possibility of analysing only the most suitable mineral grains that do not exhibit evident signs of alteration. Furthermore, when using the second approach, whole-rock  $\delta^{18}\text{O}$  values can be estimated from the  $\delta^{18}\text{O}$  data of individual minerals if the modal composition of the rock is known.

For the study of the Podlesí granite monomineral samples (~10 mg each) were obtained by hand picking mineral grains from rock samples that had been crushed and sieved into 0.063–0.3 mm fractions. Grains of feldspars and micas showing signs of alteration (those that were very fine-grained or without cleavage) or mineral intergrowths were discarded. A total of 39 monomineral separates were obtained from 13 rock samples.

Oxygen for the isotopic analysis was liberated by a fluorination technique (using  $\text{BrF}_5$ ) similar to the method of Clayton and Mayeda (1963). After the cryogenic cleaning of oxygen from the reaction products and the removal of

the excess fluorination agent, the purified oxygen was converted into  $\text{CO}_2$  on a heated graphite rod. Measurements were done on a Finnigan MAT 251 mass spectrometer. The overall analytical uncertainty of the  $\delta^{18}\text{O}$  values, tested by repeated analyses of the NBS-28 standard, was  $\pm 0.2\%$ .

## Results

The oxygen isotope data, sample locations, and petrographic characteristics are given in Table 1.

The  $\delta^{18}\text{O}$  values for the analysed minerals are relatively high when compared to the general range of granitic rocks, and the order of enrichment of  $^{18}\text{O}$

quartz > feldspar > protolithionite/zinnwaldite is preserved in most cases. Apparent disequilibrium conditions with a disturbed enrichment order were recorded only in one case (with feldspar having higher  $\delta^{18}\text{O}$  than quartz).

Several conditions must be fulfilled for the application of oxygen isotope thermometers for estimating the magmatic crystallization temperatures of a mineral pair. Firstly, an exchange of oxygen isotopes must have occurred between the two mineral phases at some stage during their common history (usually via a fluid phase), leading to isotopic equilibrium. Secondly, the isotopic equilibrium between the phases must be frozen in order to preserve the isotopic signal. Finally, the isotopic composition of the minerals cannot have been changed by later processes. It is quite clear that these conditions were not fulfilled in most of the mineral pairs studied here, since the calculated temperature ranges are too wide to reflect real magmatic crystallization temperatures.

Temperatures calculated from the quartz-feldspar pairs (after Bottinga and Javoy 1973, 1975) range widely from below 300 °C (i.e. pairs with  $\Delta^{18}\text{O}$  quartz-feldspar of ~3‰) to unrealistically high temperatures (i.e. pairs with  $\Delta^{18}\text{O}$  quartz-feldspar below 1‰, see Table 1, Fig. 2). The quartz-feldspar pairs clearly do not reflect real crystallization temperatures in most cases. A quartz-feldspar pair from the greisenised granite shows lower  $\delta^{18}\text{O}$  values for both quartz and feldspar, with a  $\Delta^{18}\text{O}$  quartz-feldspar of 2.1‰, corresponding to a temperature of ~400 °C. The feldspar phenocrysts (typical grain size X.0 cm) have in both analyzed cases lower  $\delta^{18}\text{O}$  values than the fine-grained matrix feldspar (typical grain size 0.X to X.0 mm) of the same sample.

Temperature calculations based on quartz-mica pairs (which are generally more precise because of the larger oxygen isotope fractionation between them) are problematic because the oxygen isotope fractionation factors for Li-micas (zinnwaldite, protolithionite) are not precisely known. Since the micas studied here belong to the biotite group, the use of a quartz-biotite fractionation (Bottinga and Javoy 1973, 1975, Zheng 1993b) is the first possible approach. The observed  $\Delta^{18}\text{O}$  quartz-mica values differ from less than 1‰ to more than 4‰ (see Fig. 3). These values also correspond to wide temperature range that does not reflect real crystallization temperatures.

Table 1. Sample location, petrographic characteristics, and O isotope data ( $\delta^{18}\text{O}$ , ‰ SMOW). Temperatures corresponding to the difference in the  $\delta^{18}\text{O}$  of individual mineral pairs are calculated for the quartz-feldspar pairs using the equations of Zheng (1993a), and Bottinga and Javoy (1975), and for the quartz-mica pairs using the equations of Zheng (1993b), and Bottinga and Javoy (1975).

| No.  | Sample location, petrography, and description  | $\delta^{18}\text{O}$<br>quartz          | $\delta^{18}\text{O}$<br>feldspar       | $\delta^{18}\text{O}$<br>mica | Temperature (°C)<br>quartz-feldspar | Temperature (°C)<br>quartz-mica |
|------|--|--|---|-------------------------------|-------------------------------------|---------------------------------|
| 2754 | Outcrop south of the quarry, fine-grained albite-protolithionite granite with abundant topaz, represents upper part of the stock, not influenced neither by contact, nor dyke rocks, nor greisenization.   | 11.2                                     | 9.2                                     | 8.4                           | 390/420                             | 760/770                         |
| 3359 | Dyke granite, minor apophysis penetrating into phyllite, sampled 20–30 cm thick steep dyke of aplitic appearance with minor stockscheider along the upper contact, albite-protolithionite granite with topaz. The rock contains up to 2 cm large phenocrysts of feldspar.                  | 9.8                                      | 10.6<br>(matrix)<br>9.7<br>(phenocryst) | 6.4                           |                                     | 650/690                         |
| 3361 | Typical stockscheider, smaller blocks in outcrop talus in the uppermost (southern) part of the outcrop of the granite stock. Fine-grained matrix is represented by albite-protolithionite granite with topaz, this matrix hosts large orthoclase and quartz crystals of the stockscheider. | 10.9<br>(matrix)<br>11.0<br>(pegmatitic) | 7.9<br>(matrix)<br>8.0<br>(pegmatitic)  | 7.1                           | 230/300                             | 590/640                         |
| 3365 | Free block in talus in the southern part of the stock. Dark-colored quartz-mica greisen formed by metasomatic replacement of stock-granite along steeply dipping joints. The mica corresponds to protolithionite (to Li-biotite).  | 11.6                                     |   | 8.3                           |                                     | 670/700                         |
| 3385 | Block close to southern contact of the stock, stock-granite sampled directly below the stockscheider, fine-grained albite-protolithionite granite with topaz, rarely are present bipyramidal quartz phenocrysts.   | 11.4                                     | 10.5                                    | 8.5                           | 810/770                             | 740/750                         |
| 3389 | Block close to southern contact of the stock, fine-grained white quartz-rich greisen with topaz and wolframite, formed by metasomatic replacement of dyke-granite.   | 9.7                                      |   | 6.0                           |                                     | 610/650                         |
| 3397 | Dyke-granite, outcrop in the southern part of the stock, uppermost found dyke of the dyke-granite, about 60 cm thick, fine-grained albite-zinnwaldite-topaz granite.   | 11.4                                     | 8.7                                     | 5.8                           | 270/330                             | 410/500                         |
| 3398 | Small block close to south-east contact of the stock, dark-colored quartz-mica greisen formed metasomatically from the stock-granite, contains relics of non-altered granite. Sampled dark-grey greisen.   | 9.2                                      | 7.1                                     | 8.3                           | 370/410                             | 1430/1300                       |
| 3413 | The quarry, main dyke of the dyke-granite, sampled lowermost part of the dyke with normal granitic texture, fine-grained albite-zinnwaldite-topaz granite.   | 10.9                                     | 10.5                                    | 8.6                           | 1300/1280                           | 870/860                         |
| 3416 | The quarry, main dyke of the dyke-granite, sampled upper part of the dyke with laminar texture, fine-grained albite-zinnwaldite-topaz granite.   | 11.0                                     |   | 8.2                           |                                     | 760/770                         |
| 3417 | The quarry, main dyke of the dyke-granite, sampled upper part of the dyke with UST texture, fine-grained albite-zinnwaldite-topaz granite with large oriented orthoclase crystals enclosing numerous small albite crystals, and with smaller oriented quartz crystals.                     | 11.1                                     | 10.1<br>(matrix)<br>9.2<br>(comb Kfs)   | 7.6                           | 750/710                             | 640/680                         |
| 3436 | PTP-1 drilling, depth 200 m, medium-grained albite-protolithionite granite with topaz.   | 11.3                                     | 8.4                                     | 6.8                           | 240/310                             | 510/580                         |
| 3443 | PTP-1 drilling, depth 290 m, medium-grained albite-protolithionite granite with topaz, this sample represents the deepest section of the stock reached by sampling.  | 10.6                                     |   | 6.3                           |                                     | 530/600                         |

## Discussion

### Isotopic relationships between rock-forming minerals

The observed  $\delta^{18}\text{O}$  data for quartz and other silicate minerals are the result of the combined effects of magmatic evolution and post-magmatic hydrothermal events. In the  $\delta$ – $\delta$  diagrams (Figs 2, 3), plotted with the lower exchange rate mineral (quartz) on the horizontal axis and the higher ex-

change rate mineral (feldspar, mica) on the vertical axis, the data from the studied rocks show vertical trends. Such trends are usually caused by open-system hydrothermal alteration (Gregory et al. 1989). This conclusion is consistent with the results of a petrologic study (e.g., Breiter et al. 2005) that indicates the repeated opening of the system for fluid escape even during the magmatic crystallization. Closed systems, with isotope systematics controlled only by isotopic exchange between minerals at variable tempe-

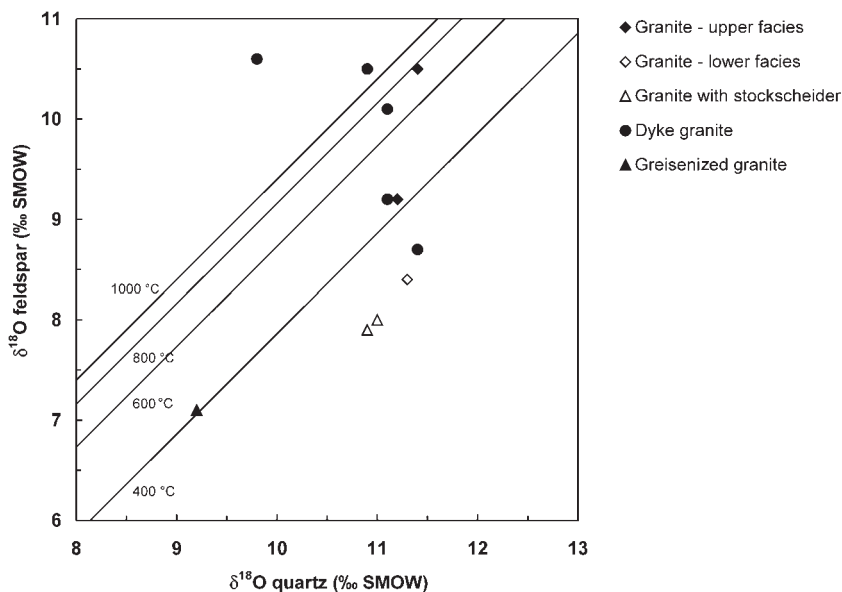


Fig. 2. Oxygen isotope data of quartz-feldspar pairs. Isotherms are based on the formula of Bottinga and Javoy (1975).

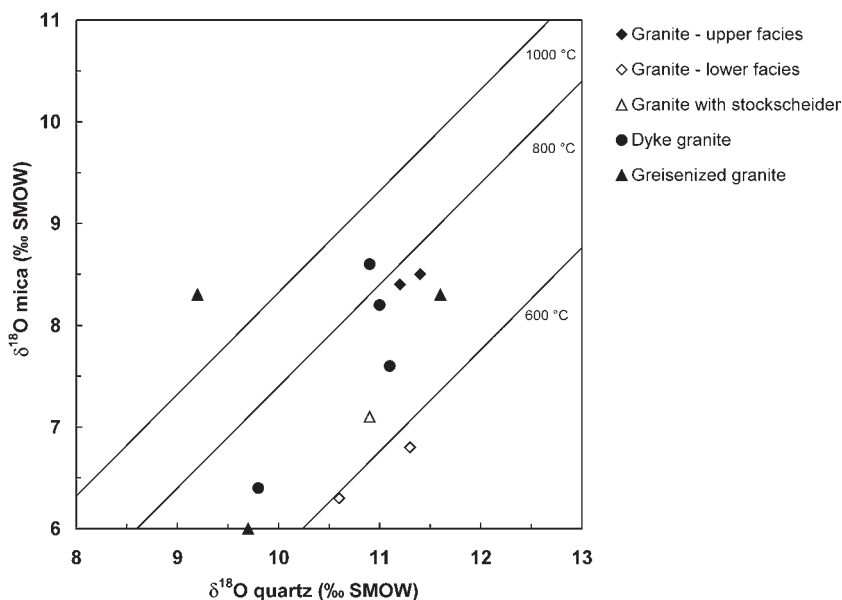


Fig. 3. Oxygen isotope data of quartz-mica pairs. Isotherms are based on the formula of Bottinga and Javoy (1975).

ratures, display negative slopes in  $\delta$ - $\delta$  space (Gregory and Criss 1986). The feldspar data are the most disturbed. Of the rock-forming minerals common in granitic rocks, the feldspars are usually the most sensitive to later isotope exchange. Nevertheless, no significant involvement of external, low- $\delta^{18}\text{O}$  meteoric waters was evident. Considering the quick cooling rate of the small Podlesí stock, the direct sub-solidus oxygen isotope exchange between minerals was probably very limited. The observed wide range of  $\Delta_{\text{quartz-mica}}$  and  $\Delta_{\text{quartz-feldspar}}$  values, and the shifted  $\delta^{18}\text{O}$  data (especially of feldspars) are either the result of isotope exchange between minerals and high- $\delta^{18}\text{O}$  magmatic fluids at sub-magmatic temperatures in a system opened to fluid phases (pairs with small  $\Delta^{18}\text{O}$  quartz-feldspar), or of infil-

tration of a small quantity of external fluids with slightly lower  $\delta^{18}\text{O}$  (pairs with large  $\Delta^{18}\text{O}$  quartz-feldspar). These fluids would have had slightly lower  $\delta^{18}\text{O}$  values than the magmatic fluid (*sensu stricto*) of the Podlesí granite itself. The influence of such a different fluid type is readily apparent for the quartz-feldspar pair from the greisen. Nevertheless, these processes did not significantly change the chemical composition of the rock-forming minerals (e.g., the magmatic phosphorus distribution in feldspars was not significantly altered in most of the samples, see Breiter et al. 2005).

### The origin of high $\delta^{18}\text{O}$ magmas

Based on material-balance calculations, Taylor and Sheppard (1986) concluded that during magma differentiation the  $\delta^{18}\text{O}$  of the melt usually increases slightly (bulk cumulates are usually slightly lower in  $^{18}\text{O}$  than the residual silicate melt). The calculations of Zhao and Zheng (2003) verified the following sequence of  $^{18}\text{O}$  enrichment: felsic rocks > intermediate rocks > mafic rocks > ultramafic rocks. Nevertheless, the bulk  $\delta^{18}\text{O}$  value of a melt does not usually change by more than 0.2 to 0.8‰ during magmatic differentiation, as evidenced by data from rock suites. Based on the increment method model calculation, Zhao and Zheng (2003) concluded that for common magmatic rocks there is negligible oxygen isotope fractionation between the melt and the rock of the same composition.

The estimated  $\delta^{18}\text{O}$  whole rock values of our Podlesí granite samples (based on the  $\delta^{18}\text{O}$  data of individual minerals and the modal composition of rock samples, Table 1) range between +9.5 and +10.5‰ (SMOW). Harris et al. (1997) used the  $\delta^{18}\text{O}$  data of quartz for distinguishing between S-type and I-type (or A-type) granites, as this mineral is generally insensitive to later alterations. The observed quartz  $\delta^{18}\text{O}$  values from the Podlesí granite range between 9.7 and 11.6‰ (excluding the most altered sample 3398, see Table 1), which is within the range of S-type granites.

Dolejš and Štemprok (2001) found different  $\delta^{18}\text{O}$  whole rock data for the low-P, Li-mica granites of the Cínovec stock (5.2 to 8.5‰ SMOW) and the high-P, Li-mica granites of the Krásno area (10.4 to 10.6‰ SMOW). The Podlesí granite is clearly more similar to those of the Krásno area. Both of these granites are highly fractionated and represent similar genetic types from the

perspective of granite chemistry. Conversely, the highly fractionated granites from the Cínovec/Zinnwald area are A-type granites (Breiter et al. 1999, Förster et al. 1999). Arlt et al. (1995) and Irber et al. (1997) studied the oxygen isotope compositions of the Fichtelgebirge granites (Germany), located west of our study area. They found practically identical  $\delta^{18}\text{O}$  whole-rock values between +10 and +11‰ SMOW for the granites of both the Older and the Younger Intrusive Complexes.

High- $\delta^{18}\text{O}$  magmas are usually interpreted as having a crustal origin (Sheppard 1986, Taylor and Sheppard 1986). A crustal origin for the Podlesí granite melts is further supported by their high initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ratio of  $\sim 0.710$  (Kostitsyn and Breiter 2001). Based on the available data, the Podlesí granites were derived primarily from reworked continental crust.

## Conclusions

1. The granitic rocks of the highly fractionated Podlesí granite system show high  $\delta^{18}\text{O}$  whole rock values ( $10.0 \pm 0.5\%$  SMOW) common for peraluminous granites elsewhere in the European Variscides, supporting their crustal origin (see Sheppard 1986, for a review).
2. Magmatic hydrothermal processes at sub-solidus conditions modified the original oxygen isotope relationships among individual minerals in most of the rock samples. These processes especially influenced the feldspars, while quartz-protolithionite/zinnwaldite pairs have been less modified.
3. Submagmatic are a result of the activity of high- $\delta^{18}\text{O}$  fluids. No pervasive involvement of external low- $\delta^{18}\text{O}$  fluids (meteoric waters) was evident.

**Acknowledgements.** This study was financed through project VaV/3/630/00 "Complex geochemical research of the interaction and migration of organic and inorganic chemicals in rock environments" financed by the Ministry of Environment of the Czech Republic, and by Research Programme No. J13/98: 113100005 of the Czech Ministry of Education. The final modifications of the paper by K. Žák have been supported by the internal project "Evolution of the Bohemian Massif before and after its consolidation" of the Institute of Geology, Academy of Sciences of the Czech Republic, within Research Program AVOZ 30130516. The comments of K. Malý (Jihlava) and an anonymous reviewer significantly improved the manuscript and are greatly acknowledged.

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