

## Heavy minerals from sedimentary rocks of the Malcov Formation and their palaeogeographic implications for evolution of the Magura Basin (Western Carpathians, Slovakia) during the Late Eocene–Late Oligocene

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Detrital heavy minerals separated from the Malcov Fm. deposits (Magura Nappe) indicate their source rocks and areas. The heavy mineral assemblages predominantly consist of garnet, zircon, tourmaline, rutile and Cr-spinel. EPMA analyses reveal a few groups of garnets: zoned and unzoned Grs almandines, Prp-Sps almandines, unzoned Prp almandines, almandines, Sps almandines and rare zoned spessartine grains (~85 mol% Sps). The garnet composition indicates that gneisses, mica schists, amphibolites and granites were their main source rocks, but low-grade metapelites with Mn mineralisation probably contributed as well. The detrital dravitic tourmalines were mostly derived from paragneisses and mica schists. Cr-spinel indicates a volcanic source. Based on heavy mineral assemblages, coupled with palaeoflow analysis, we conclude that the Marmarosh Massif and Fore-Marmarosh Suture are the most probable source areas. Additionally, the Malcov sedimentary basin was supplied by material from the crystalline complexes of the Tisza Mega-Unit and Pieniny Klippen Belt (PKB). The bulk of the clastic deposits comprise classical turbidites. These lithofacies were deposited from either turbidity currents or from concentrated density flows. The palaeoflow record is varied and highlights the contribution of sedimentary material from several directions and/or diversion of gravity currents from the main flow direction (SE–NW). The marginal parts of the Malcov sub-basins were formed of deformed and uplifted older formations of surrounding units of the Magura Nappe and PKB (submerged ridges). Older (Late Cretaceous to Eocene) flysch sediments may have been redeposited from these ridges to neighbouring sub-basins in a transverse direction (NE–SW).

Key words: Western Carpathians, Magura Nappe, Malcov Formation, provenance, heavy minerals, mineral composition.

### INTRODUCTION

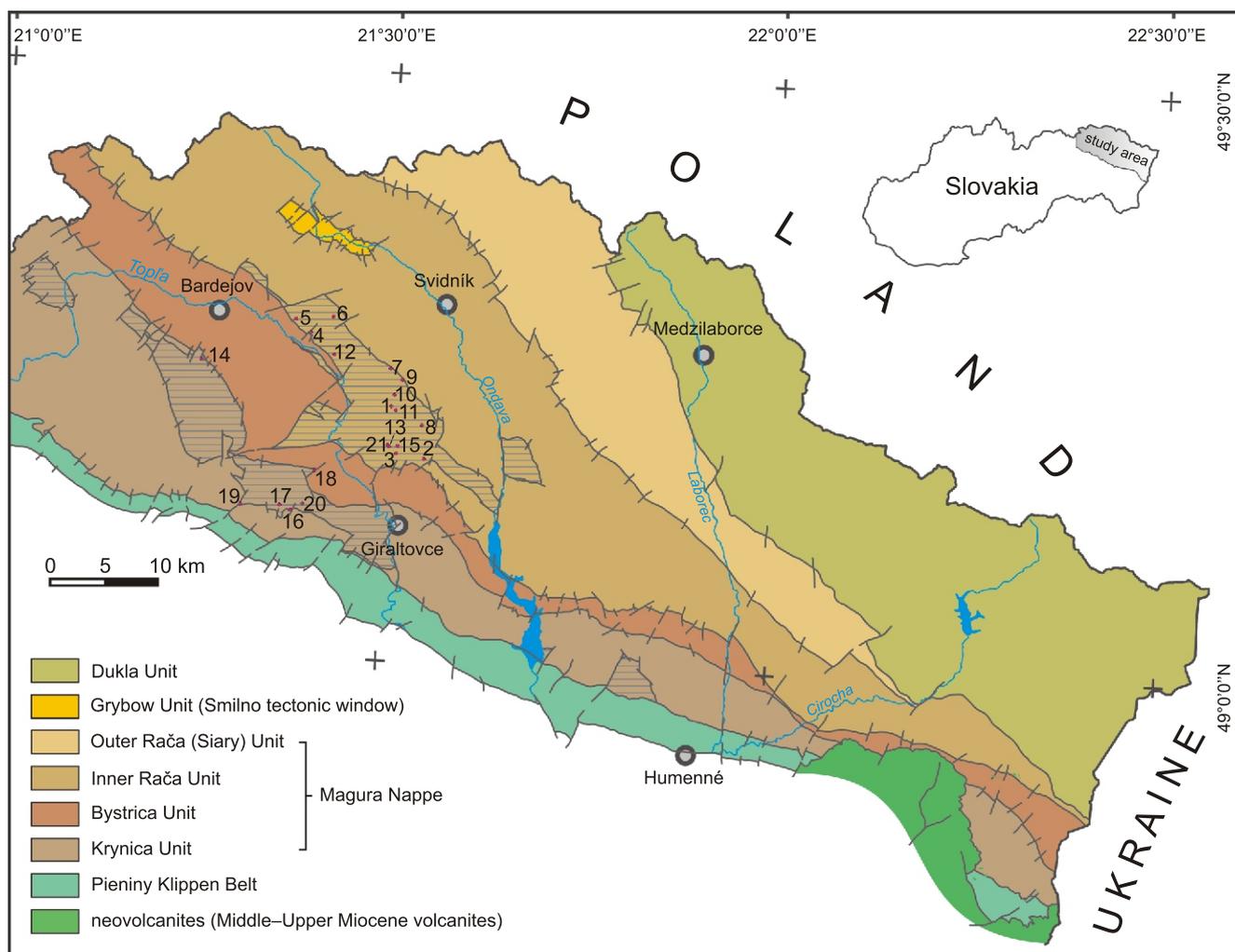
The analysis of heavy minerals is a widely used tool in provenance reconstruction of ancient and modern clastic sedimentary rocks (e.g., Morton, 1984, 1987; Morton and Hallsworth, 1994, 1999; Morton et al., 2005). Heavy minerals, such as garnet, tourmaline, zircon and Cr-spinel, are resistant to weathering, mechanical effects of transport and diagenesis. They usually resist intrastratal dissolution, as well. Their chemical composition is generally dependent on the parent rock composition, temperature and pressure conditions under which they originated (magmatic, postmagmatic/hydrothermal or metamorphic

processes) and therefore heavy minerals are good provenance indicators, and provenance data can then be used in palaeogeographic reconstructions.

Heavy mineral assemblages have been successfully used in sedimentary research of the External Western Carpathians. The use of heavy mineral associations for the interpretation of source areas in the Flysch Belt of the Western Carpathians was exploited by Leško et al. (1959), Starobová (1962), Ďurkovič (1965, 1966), Koráb and Ďurkovič (1966, 1973) and Fejdiová (1990); more decisive results were obtained by using the electron probe combined with optical methods (Otava et al., 1997, 1998; Salata and Oszczytko, 2000; Salata, 2002a, b, 2004, 2013, 2014a, b; Oszczytko and Salata, 2004, 2005; Grzebyk and Leszczyński, 2006; Bónová et al., 2009a, b, 2010a, b; Salata and Uchman, 2012, 2013).

In this article we deal with the Malcov Formation deposits from the Rača and Krynica tectono-lithofacies units of the Magura Nappe, providing brief lithological and petrographic characteristics, and the chemical compositions of selected heavy minerals in order to indicate the source rock, as well as

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**Fig. 1. Simplified and partly modified structural sketch map of the NE part of the Slovak Flysch Carpathians (according to Stráňik, 1965; Koráb, 1983; Nemčok, 1990; Žec et al., 2006; Kováčik et al., 2011; Geological map..., 2013) with sampling locations**

The numbers correspond to the marked samples in Appendix 1\*; horizontal pattern shows the occurrence of the Malcov Fm.

analysis of the depositional environment. This study uses heavy mineral analysis to better locate the sediment sources supplying the Malcov Basin. The results of petrographic-mineralogical research of these deposits, supplemented by palaeoflow analysis, are additional tools for the identification of source areas.

## GEOLOGICAL SETTING

This study analyses the provenance and sedimentology of the youngest fill (Potfaj et al., 2008; Kováčik et al., 2011) of the Magura Basin, which is represented by Malcov Formation, situated in the western part of the Nízke Beskydy Mts. outcropping in the northeastern part of Slovakia (Fig. 1).

The Malcov beds were distinguished from the Richvald “series” by Świdziński (1961). Later the term Malcov Formation was used (Leško and Samuel, 1968). The Malcov Fm. is widespread within NE Slovakia – Nízke Beskydy and the Čergov Mts. (cf. Nemčok, 1961, 1990; Nemčok and Koráb, 1963;

Stráňik, 1965; Leško and Samuel, 1968; Nemčok et al., 1990). Its occurrence is also known from the Polish part of the Flysch Carpathians, southwards from Nowy Sącz (Oszczypko, 1973), near Nowy Targ (Cieszkowski and Olszewska, 1986) and the village of Leluchów (Blaicher and Sikora, 1967). Potfaj (1983) and Potfaj et al. (1991) described the Malcov Fm. within the Oravská Magura Mts. The occurrence and continuation of the Malcov Fm. is shown on the Geological Map of the Western Carpathians and adjacent areas (Lexa et al., 2000).

According to recent work on the region (Kováčik et al., 2011, 2012), the Malcov Fm. is an integral part of the Krynica and Rača tectono-lithofacies units. Both units, together with the Bystrica Unit, form the Magura Nappe of the Flysch Belt adherent to the Outer Western Carpathians (e.g., Lexa et al., 2000). The lithological content of these units consists of the deep-sea, mostly siliciclastic deposits of Late Cretaceous to Oligocene age. In the south, the Magura Nappe is tectonically bounded by the Pieniny Klippen Belt, while in the north-east it is in tectonic contact with the Dukla Unit belonging to the Fore-Magura group of nappes. The Grybow Unit, as the innermost unit of the

\* Supplementary data associated with this article can be found, in the online version, at doi: 10.7306/gq.1285

Fore-Magura group of nappes, crops out in the Smilno tectonic window beneath the Inner Rača Unit (Fig. 1).

The Rača Unit represents the northernmost tectono-lithofacies unit of the Magura Nappe in the area investigated. On the basis of lithofacies differences in its northern and southern parts, two zones are distinguished – an external (Outer Rača Unit, equivalent to the Siary Unit in Poland) and an internal (Inner Rača Unit, Rača Unit s.s. in Poland) in terms of the current geological map of the region at 1:50,000 scale (Kováčik et al., 2011). In view of this division, the Malcov Fm. is a part of the Inner Rača Unit, where it is the main building block of the Brezovka and Olšava synclinoria (cf. Bóna et al. in Kováčik et al., 2012). The Inner Rača Unit is built of the following formations (Fig. 2): the Kurimka Fm. (*sensu* Samuel, 1990), the Beloveža Fm., the Zlín Fm. and the Malcov Fm. The Outer Rača Unit has a narrower stratigraphic range and consists of the Beloveža and Zlín formations (Kováčik et al., 2011, 2012).

The Bystrica Unit is overthrust on the Inner Rača Unit to the north. In the south, it is in tectonic contact with the Krynica Unit. The lithostratigraphy of the Bystrica Unit is similar to the Outer Rača Unit, the older Beloveža Fm. being overlain by the Zlín Fm.

The Krynica Unit is the southernmost tectono-lithofacies unit of the Magura Nappe in the area studied and consists of the Proč, Čergov, Strihovce and Malcov fms. The Proč Fm. is commonly regarded as a part of the PKB (e.g., Nemčok, 1990; Lexa et al., 2000). However, later research in the study area showed a facies transition (Jasenovce Member) between the Proč and Strihovce fms. and so both formations constitute an integral part of the Krynica Unit (Potfaj in Žec et al., 2006; Žec et al., 2011).

The stratigraphic range of the Malcov Fm. is Late Eocene to Late Oligocene. The thickness of the strata in the Krynica Unit is considerably greater (approximately 1500–2000 m) than in the Rača Unit (about 800–1200 m) and its deposition began earlier – in the older part of the Late Eocene (nannoplankton zone NP18), while in the Rača Unit this occurred at the Eocene–Oligocene boundary (zone NP21). The deposition of the formation lasted at least until the Late Oligocene (zone NP24, Kováčik et al., 2012; Fig. 2). The Late Oligocene age of the Malcov Fm. was also confirmed in the Polish part of the Magura Nappe (nannoplankton zones NP24 and NP25, Oszczypko-Clowes, 2001).

The Malcov Fm. of the Rača Unit grades from older strata of the Zlín Fm. (with interbedding of both formations' lithofacies in the northern part of the Inner Rača Unit in the Early Oligocene), whereas in the Krynica Unit, it is developed in the overlapping of the Strihovce Fm. – in the region studied as a part of Raslavice and Richvald synclinoria. The lithofacies infills of the Malcov Fm. are similar in both units. Its dominant and main facies is represented by grey calcareous claystones and siltstones with interbeds of quartzose-carbonate sandstones (flysch facies). This facies has a classic flysch character. There are a number of other, subordinate lithostratigraphic units (or facies) of relatively large stratigraphic significance. At the bottom of the formation, the Globigerina Marls are present (Leluchów Marl Member in the Polish part of the Magura Nappe, cf. Birkenmajer and Oszczypko, 1989), together with variegated claystones, laminated Tylawa limestones and the oldest part of the Menilite Member. This member (hard dark brown quartzose “menilite” shales, pelocarbonates, calcareous claystones and sandstones; Smreczek Shale Member in the Polish part of the Magura Nappe, cf. Birkenmajer and Oszczypko, 1989) is present at two younger stratigraphic levels. Compared with the Menilite Mb. of outer units of the Flysch Belt (Grybów or Dukla units), the absence of black cherts – menilites – is characteristic of their lithology. A coarse-grained, sandstone-conglomerate facies is locally also present – it forms one part of the Malcov

Fm. of the Inner Rača Unit and about four levels are developed in the Krynica Unit.

The dominant Malcov Fm. palaeoflow direction from SE to NW was documented by Koráb et al. (1962), Ďurkovič (1966), Leško and Samuel (1968), Nemčok and Ďurkovič (1989), Oszczypko (2006) and Oszczypko and Oszczypko-Clowes (2009).

In terms of provenance, acid igneous rocks and metamorphic rocks coupled with carbonates are regarded as source rocks for the Malcov deposits (Koráb et al., 1962; Nemčok and Ďurkovič, 1989). The carbonates were derived from the Pieniny Klippen Belt (Koráb et al., 1962; Ďurkovič, 1966; Nemčok and Ďurkovič, 1989; Olszewska and Oszczypko, 2010), rocks of crystalline basement origin were supplied by “the source area situated in the South” (Koráb et al., 1962) or by “multipoint source areas” (Oszczypko and Oszczypko-Clowes, 2009).

## MATERIAL AND ANALYTICAL METHODS

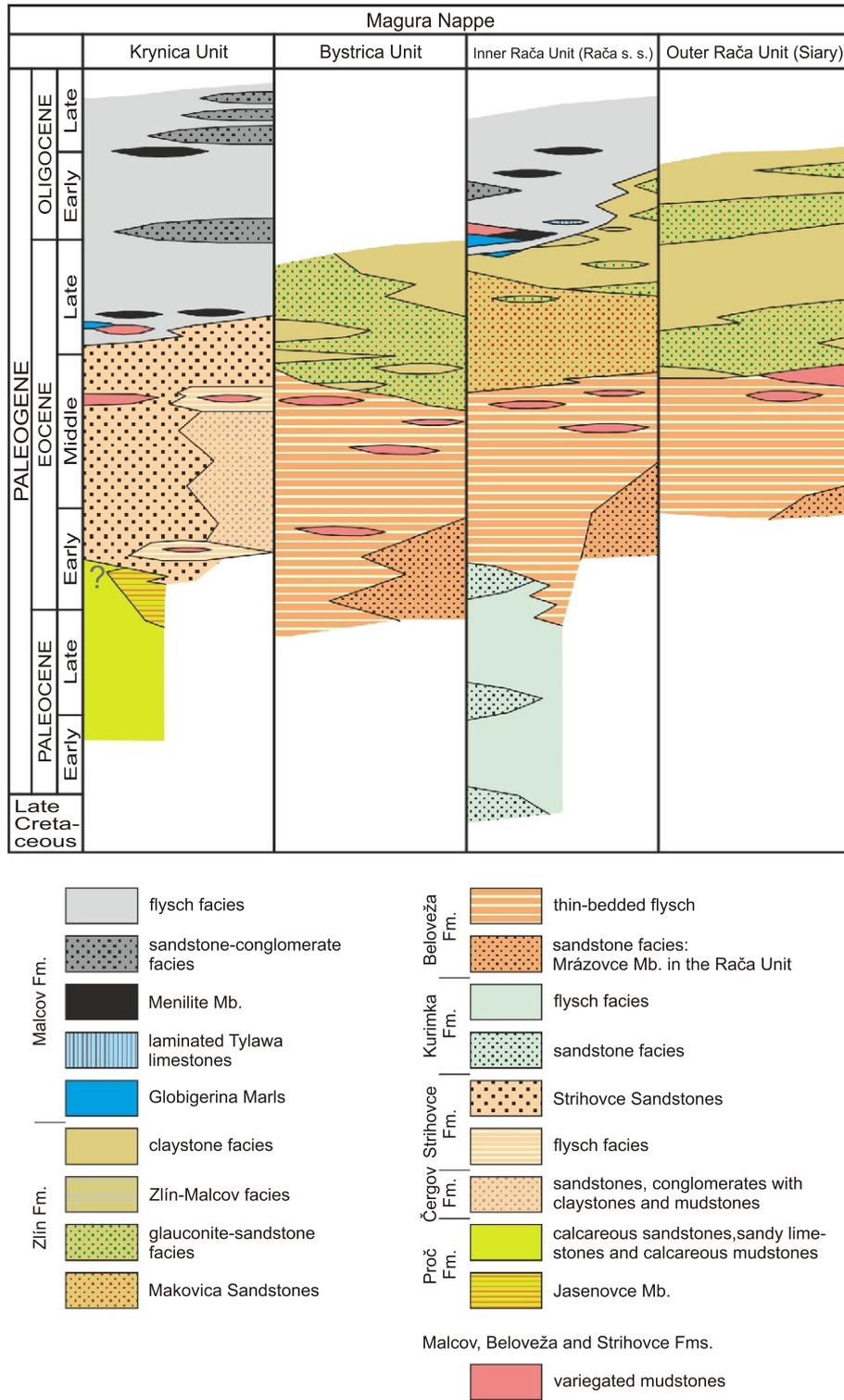
Sedimentological data were acquired through investigation of the successions and by the geological mapping. The sedimentary successions were investigated bed by bed. Lithofacies were specified on the basis of grain size, bed thickness and geometry, sedimentary structures and textures, as well as composition (*sensu* Pickering et al., 1986). Depositional environments were interpreted using lithofacies associations. Data on the direction and orientation of sedimentary structures of the lower bed surfaces are also important to the provenance analysis. Palaeoflow direction indicators were corrected with respect to the tilted bed surfaces, which were restored into a horizontal position.

Samples from the sandstone-conglomerate, flysch and menilite facies of Rača and Krynica units were evaluated by petrographic (modal composition) and petrofacies methods. For the Rača Unit, we have analysed 15 samples from flysch facies, 2 samples from menilite facies and 3 from sandstone-conglomerate facies. For the Krynica Unit, 4 samples were taken from flysch, 1 sample from menilite and 7 samples from sandstone-conglomerate facies.

Heavy mineral associations and the chemical composition of selected minerals were investigated for 4 clastic sedimentary samples: 2 samples from the Rača Unit (Štefurov and Okružle localities) and 2 from the Krynica Unit (Kľušov and Koprivnica localities).

Samples weighing 5–10 kg were collected for the preparation of heavy mineral concentrates. Preparation of the samples was carried out in the laboratories of the Department of Applied Technology of Raw Minerals (State Geological Institute of Dionýz Štúr – Regional centre Košice, Slovak Republic). Heavy mineral concentrates were obtained using the standard methods, from the 0.025 to 0.063 mm size fraction and by final separation in heavy liquid (tribromomethane with  $D = 2.89 \text{ g/cm}^3$ ). Concentrates were qualitatively and quantitatively evaluated with a focus on the translucent heavy minerals. Totals of 350 to 400 grains were optically evaluated.

Garnet, tourmaline and Cr-spinel were analysed in polished thin sections using an electron microanalyzer CAMECA SX 100 (State Geological Institute of Dionýz Štúr, Bratislava, Slovak Republic) with the WDS method at accelerating voltages of 15 kV, beam current of 20 nA and electron beam diameter of 5  $\mu\text{m}$ . To measure concentrations of various elements the following natural and synthetic standards were used: fluorapatite (PK), orthoclase (SiK),  $\text{TiO}_2$  (TiK),  $\text{Al}_2\text{O}_3$  (AlK), Cr (CrK), fayalite (FeK), rhodonite (MnK), forsterite (MgK), wollastonite (CaK),  $\text{SrTiO}_3$  (SrK), albite (NaK), LiF (FK) and NaCl (ClK). Chemical data of detrital garnets were taken



**Fig. 2. Lithostratigraphy of the Magura Nappe in the Nízke Beskydy Mts. and adjacent areas (compiled after Kováčik et al., 2012; Potfaj in Žec et al., 1997; Žec et al., 2011 and Nemčok et al., 1990)**

from the BZK-322a and BZK-208 samples (Rača Unit) and from the BZKo-765 and MF-1 samples (Krynica Unit). Crystallochemical formula of garnet was normalized to 12 oxygens and conversion of iron valence ( $\text{Fe}^{3+}$  and  $\text{Fe}^{2+}$ ) according to ideal stoichiometry. Analysed points for tourmaline (BZK-322a, BZK-208 and MF-1 samples taken from the Rača and Krynica units, respectively) were located in the centre, on the core-rim and on the rim of the grains. Tourmaline structural formula was calculated on the basis of 31 oxygens,  $(\text{OH}+\text{F}) = 4$  a.p.f.u.,  $\text{B} = 3$  a.p.f.u. Chemical data of Cr-spinel grains were obtained from BZK-322a (Rača Unit) and BZKo-765 (Krynica Unit) samples. Analyses of spinel were calculated on the basis of 3 cations.  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$  in spinel were allocated according to the ideal stoichiometry. Cathodoluminescence was used for observation of the zircon zoning. It was carried out in the same instrument at accelerating voltage of 8 kV and a beam current of  $1 \cdot 10^{-3}$  nA.

## RESULTS

### SEDIMENTOLOGY

The bulk of the flysch facies has the character of classic turbidites (lithofacies D2.1, C2.1, C2.2 and C2.3, *sensu* Pickering et al., 1986). Thin to thick-bedded sandstone-mudstone (C2.1 to C2.3) or siltstone-mudstone (D2.1) couplets generally show normal grading with partial or complete Bouma sequences (Ta–Te, *sensu* Bouma, 1962). The most common sole structures are flute, prod and groove casts. These lithofacies were deposited from turbidity currents or concentrated density flows (*sensu* Mulder and Alexander, 2001) and shaped as flat laminar bodies formed especially in the middle and distal parts of submarine lobes and in the spaces between them.

Locally, thick to very thick beds of greywacke quartz-carboniferous sandstone, overlain by very thick layers of grey calcareous silty claystone (lithofacies C2.4, *sensu* Pickering et al., 1986), are present. The sandstone-mudstone couplets commonly reach up to 5 m in thickness. Sandstone beds have a typical composite lamination with alternation of ripple-cross, sinusoidal and parallel lamination in a vertical direction. The internal structures may show different flow directions. This lithofacies originated from large-volume concentrated gravity currents, which were reflected or deflected from the flow direction on the edges of sub-basins.

The sandstone-conglomerate facies consists of several 10 m thick bed successions within the flysch facies of the Malcov Fm. We have observed several lithofacies (*sensu* Pickering et al., 1986): lithofacies A2.4 – a very thick to thick-bedded, clast-supported conglomerate is commonly normally graded to stratified pebbly sandstone, lithofacies A2.8 – a normally graded pebbly sandstone usually overlain by stratified (ripple-cross, sinusoidal and parallel lamination) granule sandstone, lithofacies B1.1 – thick to medium-bedded massive sandstones, lithofacies B1.2 – thin-bedded massive sandstones and lithofacies C2.1 – thick-bedded sandstone-mudstone couplets with well-developed Bouma intervals, mostly Tac(d)e, Ta(d)e, Tbc(d)e. The lithofacies distinguished originated mostly from concentrated density flows in channels or in transition zones between channels and lobes. Deposition from turbidity currents was of minor importance in this case.

The most common coarser-grained clastic lithofacies associated with Menilite-type black shales are thin to medium-bedded very fine-grained sandstones and siltstones (Tcd, Td) nor-

mally graded to thin to thick-bedded grey calcareous mudstones (lithofacies C2.2, C2.3 and D2.1 *sensu* Pickering et al., 1986). The deposition of these lithofacies was from turbidity currents.

Flute and groove casts are the most important sedimentary structures for the palaeoflow analysis (Fig. 3B). They are usually present on the bases of the sandstone layers belonging mainly to lithofacies class C. The palaeoflow record is very varied and highlights the contribution of sedimentary material from several directions and/or diversion of gravity currents from the main flow direction. Besides the dominant flow direction from SE to NW (longitudinal direction, Fig. 3), transverse input of clastics is also of great significance. We have recorded three other important palaeoflow directions (Fig. 3A): (1) from S and SW to N and NE (mainly in the Krynica Unit), (2) from E and NE to W and SW (mostly in the Rača Unit), (3) from NW to SE (locally observed in both units).

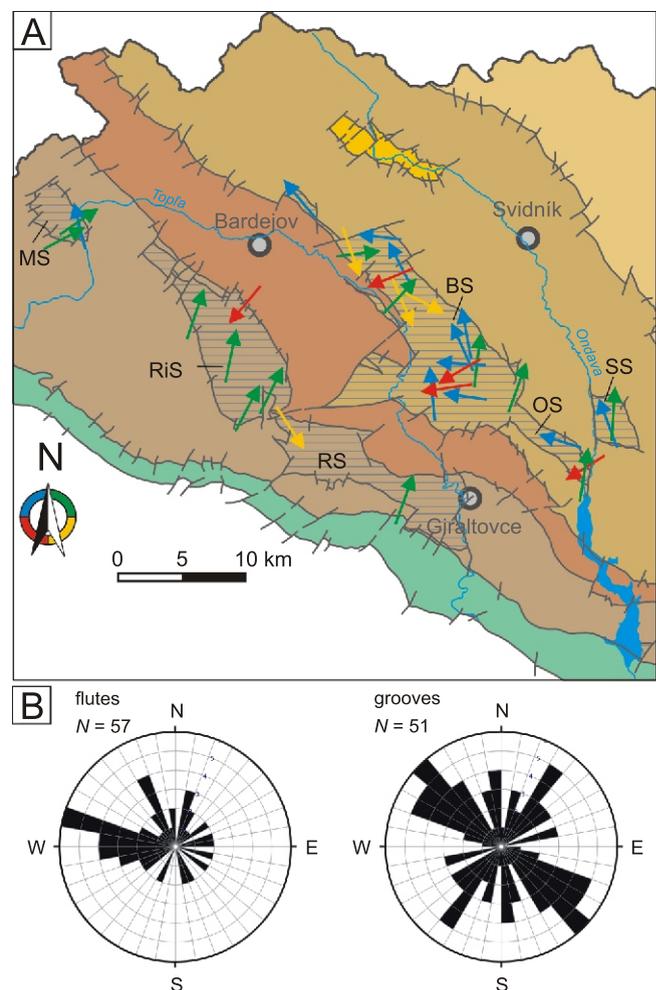


Fig. 3A – geological map showing the main palaeocurrent trends in the Malcov Fm. deposits (compiled after Koráb et al., 1962; Stránik, 1965; Nemčok and Ďurkovič, 1989 and our data); B – rose diagrams with palaeocurrent trends in the Brezovka and Oľšava synclinoria

BS – Brezovka synclinorium, OS – Oľšava synclinorium, SS – Sitník syncline, MS – Malcov syncline, RiS – Richvald synclinorium, RS – Raslavice synclinorium; horizontal pattern shows the occurrence of Malcov Fm.; other explanations as in Figure 1

## PETROGRAPHY (AND DETRITAL MODES)

The results of detrital mode investigations of the Malcov Fm. deposits are given in [Appendix 1](#).

For the **Rača Unit**, the *flysch facies* is represented by sublitharenites and rarely by lithic greywackes ([Fig. 4; Pettijohn et al., 1972](#)). They are medium- to coarse-grained, poorly to better sorted, angular to subangular. Monocrystalline quartz dominates over polycrystalline quartz ( $Q_m/Q_p = 2.31\text{--}6.40$ ) in samples. Grains tend to be slightly rounded, but angular clasts are also present. Sericitised plagioclase is found more often than K-feldspar, the ratio  $Plg/Kfs$  varying from 1.09 to 1.44 (rarely 3), sporadically vice-versa ( $Plg/Kfs = 0.86$ ). Clastogenic micas are represented by chloritised, baueritised biotite and muscovite. Muscovite prevails over biotite. The accessory mineral assemblage comprises detrital (rounded and euhedral) zircon, detrital and acicular tourmaline, rutile, spinel, glauconite and framboidal pyrite (especially near the fossils). Lithic fragments are moderately common: limestones (micritic and sparitic), sandstones, phyllites, mica schists, gneisses, granitoids, volcanic rocks (also volcanic glass). The binding material is formed by sparitic cement (somewhere up to 19%) and matrix.

Unlike the *flysch facies*, the *sandstone-conglomerate facies* contains sublitharenites and lithic greywackes ([Fig. 4; Pettijohn et al., 1972](#)) with slightly dominant polycrystalline quartz ( $Q_m/Q_p = 0.61\text{--}0.92$ ). Clastogenic plagioclase and K-feldspar occur almost in identical amounts ( $Plg/Kfs = 1\text{--}1.17$ ). The matrix, which is formed from fine quartz aggregates and clay minerals (up to 20%), prevails over sparitic cement. As well as the lithic fragments noted above, basic rocks are also present.

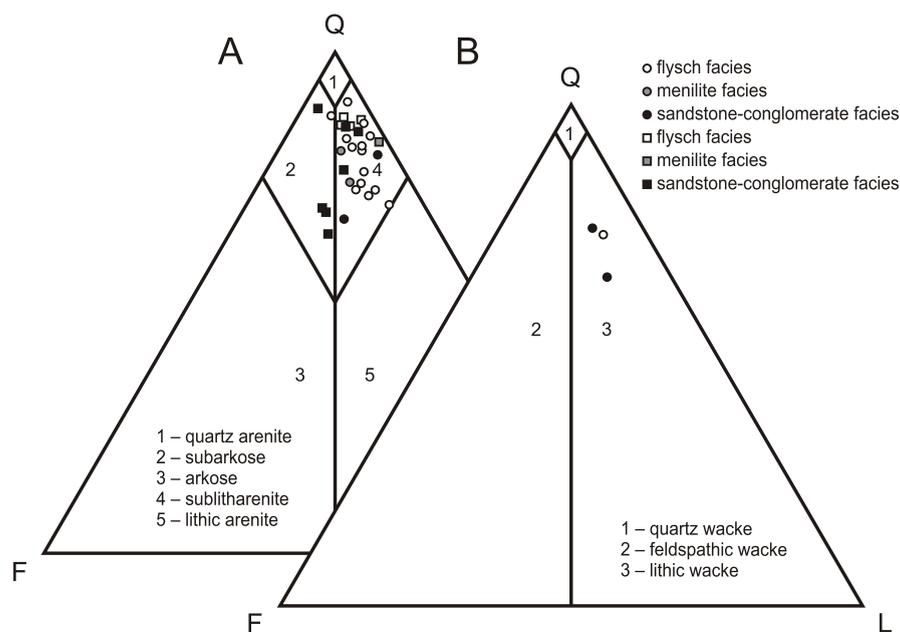
The *menilite facies* is represented by fine- to medium-grained arenites (sublitharenites *sensu* [Pettijohn et al., 1972; Fig. 4A](#)). The predominant monocrystalline quartz shows corrosion signs. Feldspar fragments are altered to sericite and clay minerals. Plagioclase slightly predominates over K-feldspar, clastogenic muscovite exceeds biotite. In places, biotite flakes are associated with chlorite. Zircon, tourmaline, rutile,

glauconite, spinel and framboidal pyrite occur as accessory minerals. Lithic fragments are represented by limestones (sparitic and micritic), pelites, phyllites, mica schists, gneisses, granites, silicic rock fragments and volcanic glass.

In the **Krynica Unit**, clastic deposits of the *flysch facies* are represented by sublitharenites (*sensu* [Pettijohn et al., 1972; Fig. 4A](#)). They consist of quartz and feldspar with minor biotite, muscovite and accessories. Among quartz, monocrystalline quartz predominates over polycrystalline quartz ( $Q_m/Q_p$  ratio varies in the interval from 3.7 to 8.14). Plagioclase exceeds potassium feldspar ( $Plg/Kfs = 1.7\text{--}3.5$ ). Microcline and perthite are rare. Clastogenic chloritised and baueritised biotite usually dominates over muscovite. Resistant heavy minerals such as zircon, detrital and acicular tourmaline, garnet, rutile, glauconite and pyrite are also present in the arenites. Limestones, metamorphic rocks represented by mica schists, gneisses and phyllites, and sandstones, silicic rock fragments, granites, volcanics and basics are also present within this rock framework. Two types of binding material are encountered, which are calcite cement and matrix.

The *sandstone-conglomerate facies* contains sublitharenites and subarkoses ([Fig. 4A; Pettijohn et al., 1972](#)) with dominant monocrystalline quartz ( $Q_m/Q_p = 3.4\text{--}10.7$ ). Plagioclase slightly exceeds potassium feldspar ( $Plg/Kfs = 1.3\text{--}1.9$ ), only one sample showing an opposite trend ( $Plg/Kfs = 0.67$ ). Clastogenic micas are represented by chloritised biotite and muscovite. Biotite prevails over muscovite, sporadically vice-versa. The accessory mineral assemblage comprises of detrital zircon, tourmaline, garnet, rutile, glauconite and framboidal pyrite. Lithic fragments are represented by limestones, sandstones, phyllites, mica schists, gneisses, granitoids, and volcanic and basic rocks. The binding material is formed by sparitic cement (somewhere up to 21%) and matrix (usually up to 6%), too.

Fine-grained sublitharenites with dominant monocrystalline quartz and abundant silicic rock fragments are typical for the *menilite facies*. Feldspar content is negligible. Clastogenic muscovite prevails over biotite, chlorite is scattered. Their acces-



**Fig. 4. Q-F-L classification of sandstones containing (A) less than 15% matrix and (B) more than 15% matrix after [Pettijohn et al. \(1972\)](#)**

Circle – Inner Rača Unit, square – Krynica Unit, Q – monocrystalline and polycrystalline quartz, F – plagioclases and K-feldspars, L – lithic debris

sory mineral assemblage consists of zircon, rutile, tourmaline, glauconite, spinel and framboidal pyrite. Lithic fragments show the same character as those ones in the Rača Unit.

Discrimination of sandstone provenance is according to the Dickinson's (1985) scheme and detrital modes were recalculated to 100% as the sum Qt, Qm, F, L and Lt. The Qt-F-L triangular plot emphasizes maturity, where as the Qm-Fm-Lt plot emphasizes primary deposition from source rocks. Dickinson's (1985) schemes suggest that the Malcov Fm. sediments were derived from a recycled orogen (Fig. 5A), mainly from a quartzose one (less transitional recycled orogen; Fig. 5B).

#### HEAVY MINERALS

**Heavy mineral assemblages.** The percentages of the heavy mineral assemblage components are given in Appendix 2. Garnet, rutile and zircon belong to the dominant group. Apart from these minerals, tourmaline, apatite, pyroxene, Cr-spinel, and traces of glauconite, staurolite, kyanite, amphibole, zoisite and chlorite were also found. Garnet dominates (up to 51 vol.%) over zircon and rutile in both the Rača and Krynica units. The proportion of opaque minerals – pyrite and hematite (not given in Appendix 2), which are not important for provenance, significantly increases in the flysch facies (largely in the Rača Unit).

**Heavy mineral geochemistry.** The investigation of chemical composition and of the internal structure is focused on garnet, tourmaline, Cr-spinel and zircon, which are traditionally considered as moderately to highly stable detrital minerals indicative of provenance. The representative chemical compositions of detrital garnets, tourmalines and chromian spinels are shown in Appendix 3.

**Detrital garnet** in the Malcov deposits often appears as slightly irregular-shaped fragments, sometimes with signs of corrosion on grain surfaces without retention of the original shape. Isometric grains occur occasionally, euhedral forms being rare (Fig. 6A). They are usually pink, sporadically colourless or or-

ange. Inclusions of rutile, zircon, monazite, xenotime, quartz, apatite, calcite and ilmenite are commonly found in the garnets.

In the Rača Unit, from a compositional aspect, detrital garnets show a high variability. Grossular-almandine, pyrope-spessartine almandine, pyrope-almandine, almandine, spessartine-almandine and spessartine were recognized. Grossular-almandines are either (1) zonal, whereby the grossular (and partly spessartine) component ( $And_1Prp_5Sps_8GrS_{31}Alm_{55}$ ) slightly decreases towards grain peripheries with corresponding increase of the almandine molecule ( $Adr_0Sps_4Prp_7GrS_{26}Alm_{63}$ ), or (2) unzoned with a high almandine component ( $Adr_0Sps_{1-4}Prp_{4-8}GrS_{13-17}Alm_{75-77}$ ). Another type of garnet is zonal pyrope-spessartine almandine ( $Adr_{0-2}GrS_{2-12}Prp_9Sps_{11-17}Alm_{62-75}$ ) with increase of grossular component towards the rim ( $Adr_0Prp_{5-9}Sps_{6-9}GrS_{19-26}Alm_{63}$ ). Additional types of garnet are: unzoned pyrope-almandine with variable contents of the spessartine and grossular molecule ( $Adr_0GrS_{1-13}Sps_{1-9}Prp_{16-21}Alm_{66-76}$ ), almandine ( $Adr_0GrS_5Sps_8Prp_8Alm_{79}$ ) and spessartine-almandine ( $Adr_1GrS_7Prp_{14}Sps_{20}Alm_{58}$ ). A zonal spessartine garnet was also found – the spessartine component in grain centre reaches 85.5 mol% ( $Adr_1Prp_1GrS_6Alm_7Sps_{86}$ ), while it rapidly drops at the rim at the expense of the almandine component (35.1 mol% Sps;  $Adr_1Prp_3GrS_{14}Sps_{35}Alm_{46}$ ).

In the Krynica Unit, the chemical composition of detrital garnets is likewise heterogeneous. There are (1) almost unzoned pyrope-spessartine-almandines ( $Adr_0GrS_{1-3}Prp_{3-10}Sps_{17-25}Alm_{66-71}$ ), (2) garnets with increasing grossular (up to 27 mol%) and pyrope (up to 33 mol%) components at the expense of almandine (45–51 mol%) ( $Adr_1Sps_1Prp_{20-33}GrS_{18-27}Alm_{45-51}$ ; the high-temperature character of these garnets is indicated by a lack of zoning), (3) pyrope-almandine, in which grossular and spessartine contents are variable ( $Adr_0Sps_{2-13}GrS_{3-10}Prp_{13-16}Alm_{69-72}$ ), (4) grossular-almandine ( $Adr_1Sps_2Prp_8GrS_{14}Alm_{75}$ ) and (5) zonal almandine-spessartine (54 mol% Sps in the centre of the grain;  $Adr_0Prp_1GrS_{17}Alm_{29}Sps_{54}$ ), which is enriched in grossular and almandine components at the expense of spessartine in the rim ( $Adr_0Prp_3Sps_{12}GrS_{22}Alm_{63}$ ).

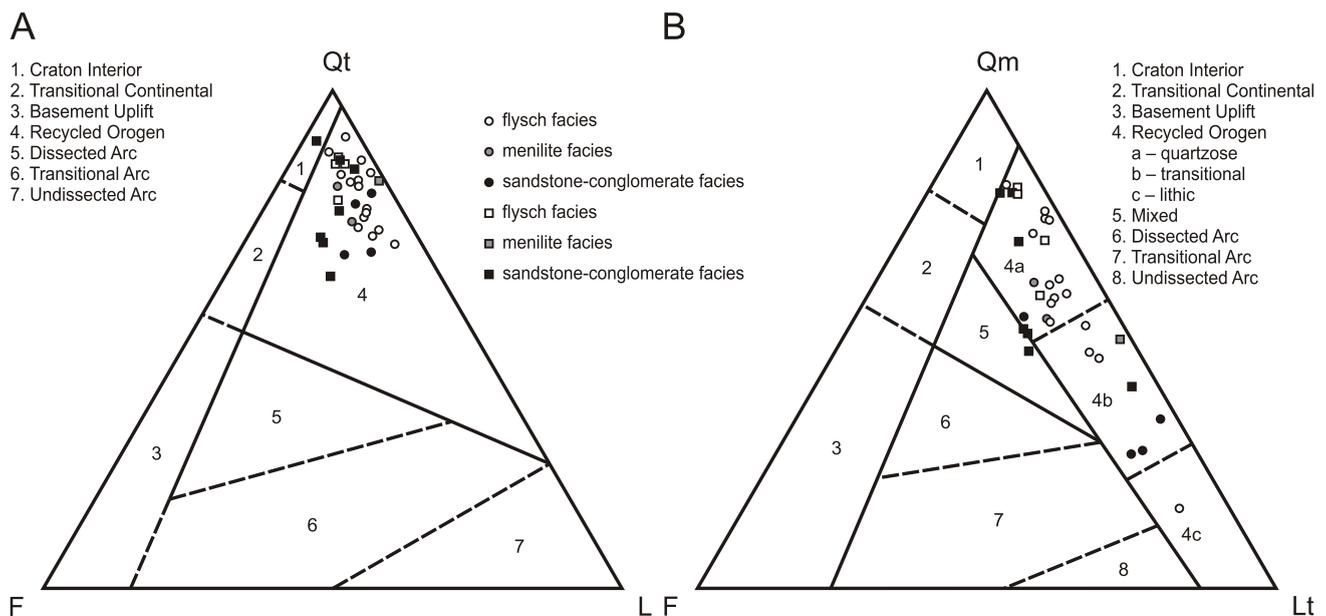


Fig. 5. Qt-F-L (A) and Qm-F-Lt (B) tectonic discrimination diagrams of sandstone source areas after Dickinson (1985)

Qt – monocrySTALLINE and polycrySTALLINE quartz, Qm – monocrySTALLINE quartz, Lt – lithic debris and polycrySTALLINE quartz; other explanations as in Figure 4

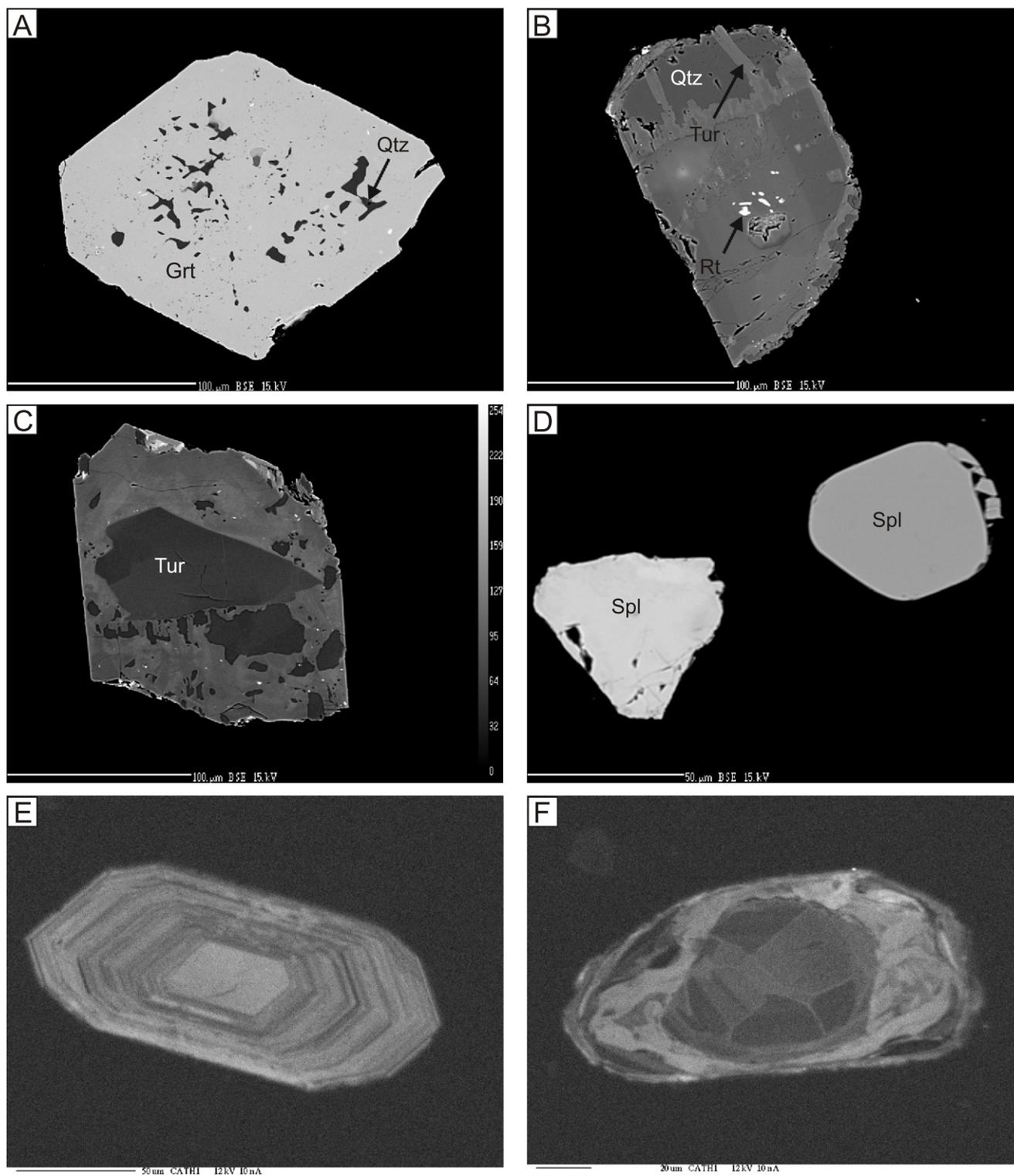


Fig. 6. Back-scattered electron (BSE) images of (A) garnet, (B, C) tourmaline, (D) Cr-spinels and (E, F) cathodoluminescence (CL) images of zircons from deposits of the Malcov Fm.

**Detrital tourmaline** occurs as short and abruptly terminated prismatic grains. Euhedral acicular grains appear rarely. Rounded and sub-rounded tourmalines are scarcer, usually of brown to dark brown colour. They are transparent to translucent with a glassy lustre. Some grains are inclusion-rich (Fig. 6B), with quartz, ilmenite and chlorite (Rača Unit). Zircon, rutile and quartz are enclosed in tourmalines from the Krynica Unit. For both units, tourmaline shows identical chemical composition.

Geochemistry of the tourmalines revealed that the dominant cation occupying the X-site position in all grains investigated – chemically homogeneous and zoned – is Na, which varies in the range of 0.5 to 0.9 a.p.f.u. Ca amount does not exceed of 0.2 a.p.f.u, while K content is in the range of 0.0 to 0.01 a.p.f.u. X-site vacancy is <0.4 a.p.f.u. Each of these may be classified as alkali tourmaline. Based on the dominant divalent cations in the Y-site position, which are Fe and Mg, the tourmalines studied belong to dravitic tourmaline (Henry et al., 2011). Molar Fe/(Fe+Mg) values range from 0.22 to 0.49. Mg and Ca contents in zonal tourmaline usually increase towards the grain peripheries but some zoning is reversed. In both cases, we assume a metamorphic origin of the tourmaline. X-vacancies vs. Ca ratio at the X position indicate low- to medium-grade conditions of metamorphism (Henry and Dutrow, 1996).

Some tourmalines consist of a detrital core of schorlitic-dravitic composition (Fe/(Fe+Mg) = 0.58), surrounded by asymmetrical overgrowths with inner and outer rims, which mark abrupt chemical discontinuities. Nevertheless, these tourmalines retain a dravitic composition.

Rare inherited tourmaline cores with slightly rounded edges (Fig. 6C) display schorlitic compositions with rather high Mn contents (0.14 wt% MnO) and no Cr, while rims of this zonal tourmaline show a dravitic compound. The molar Fe/(Fe+Mg) ratio of the inherited core is 0.71.

Unzoned tourmalines also show a dravitic composition.

**Detrital chromian spinels** are found as subhedral to euhedral grains (Fig. 6D), with rare fragments. According to Stevens's (1944) classification, which is based on Cr, Al and Fe<sup>3+</sup> contents, the spinels are represented by aluminium-chromite and chromian-spinel. Cr# and Mg# parameters classify them as chromite, magnesiochromite and rarely spinel s.s. (Deer et al., 1992).

Spinel is usually unzoned and homogeneous. The chemical composition shows different contents of the main oxides: Cr<sub>2</sub>O<sub>3</sub> (up to 50.50 wt.%) which dominates in most of the grains analysed; Al<sub>2</sub>O<sub>3</sub> content is within the range of 16.06 to 32.75 wt.%, while the high TiO<sub>2</sub> (0.70–2.30 wt.%) and the low [Fe<sup>2+</sup>/Fe<sup>3+</sup> = (1.0–2.15)] ratio suggests a volcanic origin of the bulk of the grains analysed (Lenaz et al., 2000; Kamenetsky et al., 2001), with a Mg# between 0.5 and 0.7. Grains containing TiO<sub>2</sub> up to 0.1 wt.%, Fe<sup>2+</sup>/Fe<sup>3+</sup> = (8.9–14.5) and Mg# = 0.42–0.48 are also present. Some grains are rich in TiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and FeO, which increase at the expense of Cr<sub>2</sub>O<sub>3</sub> towards the grain peripheries. Other grains show higher contents of MnO (0.49 wt.%) and ZnO (0.43 wt.%), which may indicate alteration processes, although these are not shown by grain textures. Also, Zhu et al. (2004) considered the content of MnO <0.5 wt.% as typical of spinels. A decrease in Al<sub>2</sub>O<sub>3</sub> content at the peripheries of grains, typical of alteration and/or metamorphic processes, does not occur.

According to the diagram used to determine Cr-spinel origin (Lenaz et al., 2000; Kamenetsky et al., 2001), the samples investigated lie in the field of suprasubduction zone peridotites and volcanic spinels, respectively. The volcanic spinels fall outside the boundaries of different tectonic settings (Fig. 7C).

**Zircon internal structure.** Zircon forms either euhedral to subhedral short-prismatic dipyramidal grains or long-prismatic

ones without signs of corrosion and cutting of edges. Both groups are colourless or pink. Rounded zircon shapes are also present. Their colour is the same – colourless, pink, rarely yellowish.

According to cathodoluminescence study, euhedral to subhedral short-prismatic zircons usually precipitate from nuclei or around inherited cores (Fig. 6E) and display a regular oscillatory zoning without marginal resorption. In some grains, local recrystallisation is observed. This may be associated with a late- to post-magmatic stage of zircon development. The presence of inclusions, such as apatite and melt, is common. The internal structure of the long-prismatic zircon also shows regular oscillatory zoning.

Rounded zircons indicate a polycyclic history. The occurrence of inherited cores is a common feature of many rounded zircons. For some zircons an irregular zoning is typical. Additional zircon grains are characterized by convolute zoning due to recrystallisation processes (Fig. 6F). Zircons have been found with inherited cores showing no zoning, on which a narrow zone with regular oscillatory zoning is present.

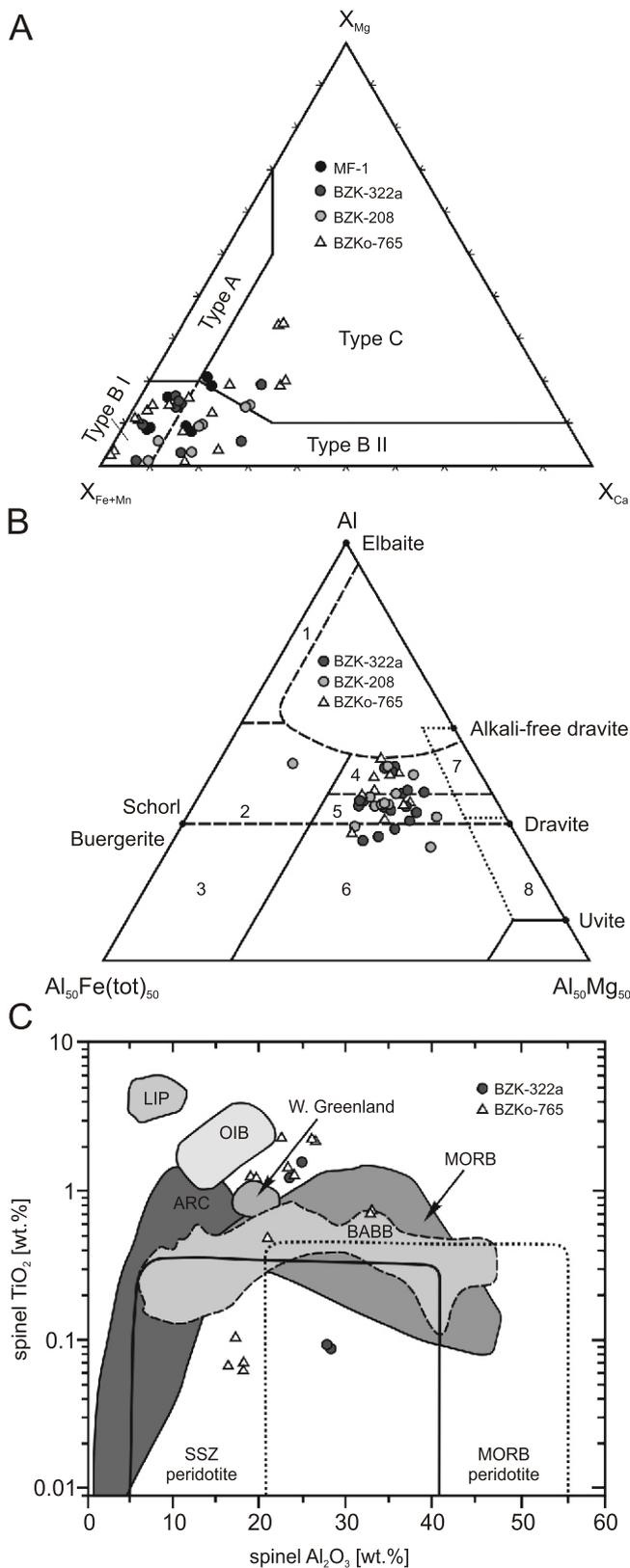
Dendritic grains shifting to polyhedral morphology with parallel zoning to the crystallographic c axis showing rapid growth of the zircon are also present.

## DISCUSSION

### SEDIMENTOLOGY AND PALAEOFLOW TRANSPORT

We infer that the Malcov Formation (together with the uppermost, Oligocene part of the Zlín Fm. in the Rača Unit) originated in several small-sized sub-basins the axes of which run NW–SE. Formation and isolation of these sub-basins are associated with Alpine tectonics and transformation of the remnant Outer Western Carpathians basin into a flexed foreland basin (e.g., Oszczypko, 2006). The sub-basins have characteristics of piggy-back basins (*sensu* Ori and Friend, 1984) developed on the top of the frontal part of the Western Carpathians orogenic wedge, which constitutes a wedge-top depozone (*sensu* DeCelles and Giles, 1996) of the foreland basin. The nature of the processes and sediments in the sub-basins was also affected by climate change at the boundary between the Eocene and Oligocene (Terminal Eocene Event) when significant cooling of the climate and glacio-eustatic regression started (e.g., Leszczyński, 1997; Soták, 2010; Oszczypko-Clowes and Zydek, 2012). From the Early Oligocene the Carpathian basins provided the first records of isolation from the open sea (Early Paratethys; Báldi, 1980, 1984). The palaeoenvironmental changes reflected the cooling of the climate and isolation of the Paratethys. The consequences of these changes are the Globigerina Marls, menilite facies and laminated limestones that commenced at the boundary between the Eocene and Oligocene (nannoplankton zone NP21). The shape and size of the sub-basins significantly affected the sedimentation of the clastic deposits.

Marginal parts of the Malcov sub-basins [in our case two sub-basins have been distinguished – the southern (inner) sub-basin in the Krynica Unit and the northern (outer) sub-basin in the Inner Rača Unit] were deformed and older successions of surrounding units of the Magura Nappe and Klippen Belt were uplifted. The deformation of the successions therefore occurred before and during sedimentation, and immediately afterwards, respectively (e.g., Świerczewska and Tokarski, 1998) and continued to the Early Miocene. We assume that the southern edge of the southern sub-basin was formed from uplifted units of the



PKB and the Magura Nappe (the Proč, Čergov and Strihovce fms.), whereas the northern edge of the sub-basin was formed from deformed strata of the Bystrica Unit sedimentation area. The structural elevation of the uplifted Bystrica Unit (the sedimentation area of this unit) was also the southern boundary of the northern sub-basin. The northern sub-basin was bounded by a slightly uplifted external part of forming Rača Unit on the northern side. We understand the individual structural elevations as submerged ridges parallel to the direction of the Magura Basin (NW–SE in recent coordinates of the study region). Older (Late Cretaceous to Eocene) flysch sediments may have been redeposited from these ridges via submarine slumps and various types of gravity currents to neighbouring Malcov sub-basins in a transverse direction (NE–SW). Deposition of the Malcov Fm. lithofacies in relatively small sub-basins is also indicated by the palaeoflow record. Variable palaeoflow data highlights the contribution of sedimentary material from several directions and/or diversion of gravity currents from the main flow direction. Besides the dominant flow direction from SE to NW [longitudinal transverse input of clastic sediments from peripheral parts (structural elevations)] of sub-basins also had a significant impact on final clastic composition.

Palaeoflow analysis shows that the main clastic source area was situated to SE of the Malcov sub-basins. We propose the Marmarosh Massif as the main clastic source, despite some considerations about its NE position (Oszczypko, 2004; Oszczypko et al., 2005b, 2015) or its position between the Dukla and Magura basins (Oszczypko and Oszczypko-Clowes, 2009; Gagala et al., 2012), respectively. The presence of the intra-basinal Marmarosh Ridge between the Magura and Dukla basins was suggested by Leszczyński and Malata (2002); Ślęczka et al. (2006), Warchoł (2007) and Gagala et al. (2012). It uplifted along the forebulge during the Late Eocene and drowned in Early Oligocene due to tectonic loading (Gagala et al., 2012). On the other hand, Koráb and Ďurkovič (1973, 1978) demonstrated the existence of a single sedimentary basin for the Magura and Dukla units during the Middle Cretaceous to Early Oligocene in Eastern Slovakia, i.e. these units were sedimented in a basin which was not divided by a ridge.

#### HEAVY MINERALS AND THEIR POSSIBLE ORIGIN

The ZTR index (percentage of the combined zircon, tourmaline and rutile grains among the transparent, nonmicaceous, detrital heavy minerals *sensu* Hubert, 1962), which reflects sediment maturity, is within the range of 47–56% for the Rača Unit, and of 36–50% for the Krynica Unit. The ZTR index suggests

**Fig. 7A** – composition of detrital garnets from the Malcov Fm. in a Fe + Mn-Mg-Ca ternary diagram (Morton et al., 2004); **B** – Al-Fe-Mg diagram for tourmalines (Henry and Guidotti, 1985); **C** – composition of analysed Cr-spinels in a  $Al_2O_3$  vs.  $TiO_2$  diagram (Kamenetsky et al., 2001)

Type A – Grt from granulites, type B I – Grt from intermediate to acid igneous rocks, type B II – Grt from metasedimentary rocks of amphibolite facies, type C – Grt from metabasic rocks; 1 – Li-rich granites, 2 – Li-poor granites and aplites, 3, 6 –  $Fe^{3+}$ -rich quartz-tourmaline rocks, 4 – metapelites and metapsammities co-existing with Al-rich phases, 5 – metapelites and metapsammities not co-existing with Al-rich phases, 7 – low-Ca metaultramafic rocks, Cr- and V- rich metasedimentary rocks, 8 – metacarbonates and metapyroxenites; LIP – large igneous provinces, OIB – ocean-island basalt, MORB – mid-ocean ridge basalt, ARC – island-arc basalt, SSZ – peridotite of supra-subduction zone, BABB – back-arc basin basalt (after Lenaz et al., 2000)

the erosion of crystalline massifs, excluding multi-cycle sediments, for which ZTR >90% is typical (Hubert, 1962). A medium value of the ZTR index indicates a relatively close source, and insignificant role of intrastratal dissolution, which is influenced by burial depth (Morton, 1987).

The garnet/zircon index (GZi; Morton and Hallsworth, 1994), the value of which could be affected by diagenetic processes and intrastratal dissolution of garnet, varies from 65 to 74% for the Krynica Unit and from 60 to 66% for the Rača Unit. The chromian-spinel/zircon index (CZi; Morton and Hallsworth, 1994), the value of which varies from 6 to 9% and from 17 to 24% for the Rača and Krynica units, respectively, provides a good reflection of source area characteristics because these minerals are comparatively immune to alteration during the sedimentary cycle. This index could be used to directly match sediment with the source material, even for suites of first-cycle origin (Morton and Hallsworth, 1994). Its higher value indicates that an important proportion of ophiolite detritus was supplied chiefly for the Krynica Unit.

Index minerals such as *staurolite*, *kyanite* and *sillimanite* are valid only for certain settings, i.e. Barrovian-type metamorphism of Al-rich pelitic rocks. The presence of these rocks in the source area is evident from tourmaline and garnet geochemistry and pebbles such as clasts of gneiss and mica schist. The existence of staurolite in sandstone-conglomerate facies of the Malcov Fm. (Rača Unit) is confirmed, while kyanite is rare (Appendix 2). Āurkovič (1965) described a “relatively low content of staurolite in Malcov layers” within the Rača Unit. It is relatively abundant in the subjacent Beloveža Fm. (Fejdiová, 1990). Kyanite is present in the flysch facies of the Krynica Unit, while staurolite is uncommon. Despite the conclusions resulting from the study of the transformation of smectite to illite (Świerczewska, 2005) or from the study of fluid inclusions in hydrothermal quartz, which involve the loss of 5.4–7.4 km of sediment by erosion in the Magura Basin (Hurai et al., 2006), a decrease in staurolite and kyanite concentration or their dissolution influenced by sediment burial is not assumed, because less stable minerals such as pyroxenes and amphiboles (Appendix 2) are retained in the heavy mineral spectra. As regards intrastratal dissolution, these minerals are extremely susceptible due to sediment burial depths besides other factors affecting dissolution (Scavnicar, 1979; Morton, 1984; Morton and Hallsworth, 1999, 2007). The transformation of smectite to illite, which is directly dependent on fluid circulation, excludes a low permeability which could protect the sediment from circulation of pore fluids thus to prevent the disintegration of unstable heavy minerals. The presence of amphibole suggests that significant post-depositional dissolution is unlikely to have taken place. Pyroxenes and rare amphiboles are also present in the subjacent formations (Fejdiová, 1990). The existence of Ca-rich garnets, which are less stable than Ca-poor ones during diagenesis (Morton and Hallsworth, 2007), was also noticed. In general, dissolution of garnet is not supported because of the high GZi value (Morton and Hallsworth, 1994, 1999). A decrease of mineral diversity with increasing burial depth is also not obvious (cf. Fejdiová, 1990). Etched garnet co-exists with garnet grains that show no sign of corrosion, indicating that the etching did not take place *in situ*.

Apart from the occurrence of specific minerals, single grain characteristics are used to deduce the metamorphic origin of detrital grains and to further unravel the type of metamorphic host-rocks. The compositional variation of garnet is large, making it a candidate for source rock discrimination based on single-grain geochemistry.

Chemical composition of detrital *garnets*, and also the distribution of inclusions in these grains suggest that most of them

were derived from metamorphic rocks that originated within conditions of amphibolite or even epidote-amphibolite facies. Their metamorphic origin is likewise demonstrated by the high garnet/zircon index (GZi), which sensitively reflects provenance characteristics of heavy minerals in sediments (Morton and Hallsworth, 1994, 1999).

Garnets with significant Fe and Mn contents may be derived from low- to medium-grade metasediments or from intermediate to acidic gneisses (Deer et al., 1992; Morton et al., 2004; Suggate and Hall, 2013). Garnets from low-grade metapelites are characterized by zoning, and a zoned pattern of some detrital garnets is visible (Appendix 3). Zoned grossular-almandines obviously originated from source rocks of low-grade amphibolite facies. The presence of low- to medium-grade metamorphic rocks (phyllites, mica schists) in the source area is supported by their existence in the form of lithic fragments in the deposits studied. Spessartine-almandine garnets, in which chemical zoning is scarcer, though pyrope (around 10–12 mol%) and grossular (around 15 mol%) molecules are relatively abundant, might have originated in migmatites (?) or gneisses. Spessartine-almandines (with 1–3 mol% Grs) showing the least chemical changes in the core – periphery direction point to a genesis in granitic pegmatites or granites. They occur in deposits of both the Rača and Krynica units. According to Mange and Morton's (2007) criteria, low Ca and Mg contents in garnet indicate their provenance in intermediate to acidic igneous rocks (Fig. 7A). Garnets with significant grossular molecule (up to 27 mol%) and a slightly higher content of pyrope (up to 33 mol%), which increases at the expense of almandine (45–62 mol%) component, indicate their derivation from basic metamorphic rocks (amphibolites or mafic granulites).

Spessartine-rich garnet appears as a distinct indicator of its background. Spessartines with low amounts of grossular content are known to originate from low-grade regionally metamorphosed rocks such as metapelites, metacherts, especially those in thermal aureoles (Miyashiro, 1955; Deer et al., 1982; Spišiak et al., 1989; Méres, 2008), or from Mn carbonate-silicate rocks (Matkovskiy, 1971; Deer et al., 1992; Mohapatra and Nayak, 2005; Matkovskiy et al., 2011; Kanungo et al., 2014), moreover, from garnetites (often with high andradite component, Kropáč, 2012) and gondites (Melcher, 1995; Matkovskiy et al., 2011; Vrána, 2011). They have rarely been found in granite pegmatites (Manning, 1983; Baldwin and Von Knorring, 1983; Királi and Török, 2003), and in blueschists, with an absence of zoning (Martínek and Štolfová, 2009). Normal zoning of spessartines denotes their formation under conditions of progressive metamorphism. Ca enrichment, coupled with depletion of Mn at the rims of the garnets, may have been caused by pressure increase (Green, 1977) or by recrystallisation through the medium of Ca-rich fluids (Királi and Török, 2003).

Generally, there are minimal differences in the chemical composition between detrital garnets from the Rača and Krynica units, respectively. However, the source rocks are heterogeneous. Certain proportion of garnets in the Malcov deposits could have resulted from resedimentation from the older formations of the Magura Nappe.

**Tourmaline** is primarily derived from metamorphic, mostly metapelitic rocks. Detrital tourmalines, according to the diagram from Henry and Guidotti (1985; Fig. 7B), which is used for determination of environment for tourmaline origin, were derived from Al-poor and Al-rich metasedimentary rocks. Their metamorphic origin is also indicated by relatively high Ti (up to 0.3 a.p.f.u.) contents.

The tourmaline investigated represents mainly polycyclic grains – they are either (1) zoned with inherited schorlitic-dravitic cores derived from metapelite (metapsammite) co-existing with

Al-saturated phases and with rims derived from Al-saturated phases-free metapelite (Henry and Guidotti, 1985); or (2) zoned tourmalines with inherited schorlitic cores derived from granite (granite differentiates). The inner margins of zoned tourmalines from the second group indicates their origin in Al-saturated phase-free metapelites, while their outer rim was formed in an Al-rich metapelite environment. The presence of the inner and outer rims separated by compositional discontinuities around some inherited cores suggests punctuated evolution of the overgrowths. This implies that boron was sporadically available during diagenesis and metamorphism (Henry and Dutrow, 1992).

Tourmalines originated from Fe<sup>3+</sup>-rich quartz-tourmaline rocks bordered by zones formed in a metapelite environment.

The scarce euhedral acicular tourmalines were derived from metapelites with Al-saturated phases, most probably representing first-cycle delivery from a nearby source. The very low contents (often zero value) of tetrahedral Al and the  $X_{vac}$  values around 0.2–0.3 a.p.f.u. suggest medium-grade metamorphic conditions during their precipitation (Henry and Dutrow, 1996).

Most studies have reported a strong predominance of tourmaline from metasedimentary rocks, although tourmaline is very common in granitoids and their differentiates (von Eynatten and Gaupp, 1999; Morton et al., 2005; von Eynatten and Dunkl, 2012). This might be caused by (1) inherited grain size effects and/or (2) the possibly higher stability of Mg-rich tourmaline against extreme weathering (Van Loon and Mange, 2007; von Eynatten and Dunkl, 2012). More importantly, there might be a direct relationship between grain size and composition, i.e. specific mineral phases may show systematically contrasting grain-size distributions in the same or different source rocks (von Eynatten and Dunkl, 2012). Ultimately, the grain size distribution in the source rocks controls the availability of specific mineral grain sizes in the sediment (Morton and Hallsworth, 1999). These possibilities may have caused the dravitic tourmaline dominance in the Malcov Fm. deposits.

**Detrital zircons** exhibit variations not only in external morphology but also in internal textures. Euhedral zircons originate from primary igneous rocks. Zircons crystallising from nuclei showing fine regular oscillatory zoning, often without signs of resorption, may have formed in “early” melts (Hoskin and Schaltegger, 2003). Zircon elongation is considered as a factor reflecting grain crystallisation velocity (e.g., Corfu et al., 2003). “Stubby” and equant forms are associated with slowly cooled intrusions, whereas elongate acicular shapes occur as a result of rapid crystallization and are traditionally interpreted as of volcanic origin (Hoskin and Schaltegger, 2003). Complete euhedral shapes of zircons indicate their igneous origin without prolonged transport from the source region. The presence of inclusions, such as of apatite and melt, supports the igneous origin of this zircon group.

The origin of rounded grains in metasedimentary rocks, possibly in older recycled sediments, is documented not only by the zircon shapes, but also by their internal textures (Fig. 6F). The polymict character of the zircons indicates multiple source rocks.

**Chromian spinels** in the heavy mineral spectra signal some contribution of mafic to ultramafic rocks. The lithic fragments originated in these rocks are preserved in small amounts due to their easy disintegration during transport or diagenesis (BZKo-765, BZG-407 samples). Starobová (1962), Winkler and Ślącza (1992, 1994) and Oszczypko and Salata (2005) noted the significant amount of Cr-spinels in deposits from the south-easternmost areas of the Magura Basin. Consideration of reworked or re-eroded sediments from the Pieniny Klippen Belt (*sensu lato*) or more precisely from the Czorsztyn ridge realm (Winkler and Ślącza, 1994), as a possible source of Cr-spinels occurring in the Magura Basin deposits, cannot be excluded completely.

According to Lenaz et al. (2000), we discriminate between peridotitic and volcanic Cr-spinels on the basis of their TiO<sub>2</sub> contents and FeO/Fe<sub>2</sub>O<sub>3</sub> ratios. Lenaz et al. (2000) pointed out that peridotitic spinels show TiO<sub>2</sub> contents <0.2 wt.% and FeO/Fe<sub>2</sub>O<sub>3</sub> ratios >3, while spinels crystallised from basaltic magmas show TiO<sub>2</sub> contents >0.2 wt.% and FeO/Fe<sub>2</sub>O<sub>3</sub> ratios <4. Cr-spinels originating from cumulate and extrusive volcanic rocks tend to have higher TiO<sub>2</sub> (Kamenetsky et al., 2001). High Fe<sup>3+</sup> and TiO<sub>2</sub>, along with lower Mg#, suggest that the Cr-spinels could have been the products of fractional crystallisation and re-equilibration at lower temperatures due to slow cooling (Arai, 1992; Kamenetsky et al., 2001; Lužar-Oberiter et al., 2009), and thus probably derived from cumulate members of an ophiolite source. The increased contents of TiO<sub>2</sub> in most Cr-spinels from the Malcov Fm. are noticeable. Compared to Cr-spinels from the Czorsztyn Unit (Aubrecht et al., 2008, 2009), perhaps even the Jarmuta Fm. (PKB) and the Szczawnica Fm. (Magura Nappe, Krynica Unit, Salata, 2002a), there are some differences: Cr-spinels originated from volcanic rocks are predominant in the Malcov Fm., nevertheless peridotite spinels dominate in the Czorsztyn flysch. Increased amounts of oxides could have been caused by alteration. However, the majority of the grains analysed have optically and chemically homogeneous compositions (Fig. 6D). Significant decreases in Al<sub>2</sub>O<sub>3</sub>, MgO and Cr<sub>2</sub>O<sub>3</sub>, which might result from the alteration of Cr-spinels (Burkhard, 1993; Power et al., 2000; Spišiak et al., 2000, 2001; Mikuš et al., 2006; Mikuš and Spišiak, 2007), has not been observed.

Power et al. (2000) imputed the high Ti content in spinels to the solidus-solidus reactions between spinel and plagioclase in ophiolites. Spišiak et al. (2001) considered the Cr-spinels from the Šambron flysch zone (Central-Carpathian Paleogene Basin) to have been derived from two sources at least, although they included the possibility of re-sedimentation. According to Spišiak et al. (2001), the Cr-spinels investigated resemble the Cr-spinels derived from ultrabasic rocks from the Zbudza locality (Iňačovce–Kričovo Unit; Soták et al., 1990, 1991, 1995), or from ultrabasic rocks cropping out in Gemericum, as well as the spinels from the PKB, or Magura flysch units of the Polish Western Carpathians. However, low Ti contents in these Cr-spinels are notable (cf. Spišiak et al., 2001). The heterogeneous character of Cr-spinels in the Šambron Zone sediments was also shown by Lenaz et al. (2001, 2009), who described the similarity with spinels from the Vardar Zone. Peridotite spinels are prevalent (representing 90% of the spinels investigated, derived from peridotite of type II *sensu* Dick and Bullen, 1984). Scattered volcanic spinels have OIB character (Lenaz et al., 2009). Cr-spinels coupled with leucoxene are dominant in heavy mineral spectra, while garnets are entirely absent (Spišiak et al., 2001). Serpentinite clasts predominate in sandstones of the Šambron flysch zone, so it shows the clear dominance of basic sources in the Paleogene (especially the Late Oligocene; Soták and Bebej, 1996). These are deposits of perisuture basins, which were originally connected with trench-like flysch deposits of the Magura Unit (Soták and Bebej, 1996). Preliminary geochemical data of Cr-spinels from the Magura Formation of the Polish Western Carpathians indicate their affinity to the Vardar Zone (Lenaz et al., 2001).

The association of heavy minerals, their geochemical composition and petrographic analysis point to a heterogeneous source for the Malcov sediments. There is a combination of material, in particular of supra-crustal origin derived from metasediments recrystallised at the conditions of amphibolite and green-schist facies, partly from older strata, granitoid rocks and carbonates.

The petrographic study of the Malcov deposits revealed the predominance of monocrystalline quartz over polycrystalline

quartz. This indicates that the sediments may have been partially derived from a granitic source (Basu et al., 1975). The influence of igneous rocks on the sandstone composition is documented (1) by the presence of feldspars (K-feldspars), (2) by granite (and volcanic) fragments in the deposits and (3) by the existence of magmatic zircon in the heavy mineral spectra.

Dickinson's (1985) schemes suggest that the Malcov sediments were derived from recycled orogens shedding material of continental affinity into the basin. Evidence for partial recycling is apparent from bulk sediment composition (high percentages of quartz) indicating upward petrographical maturity ( $Q_m > Q_p$ ), as well as from a zircon and chromian spinel enrichment due to their high stability within the sedimentary cycle. Heavy mineral ratios (CZi and GZi; Morton and Hallsworth, 1994) reflect minimal alteration of the Malcov deposits and/or invariant transport. The medium to high ZTR value, coupled with the heterogeneous character of the heavy mineral spectra, may point to a mixed source which is characterized by first-cycle detritus (e.g. acicular tourmaline) from a metamorphic basement representing remnants of continental margins, as well as by polycyclic detritus recycled from orogeny-derived clastic wedges (Garzanti et al., 2007).

#### PALAEOGEOGRAPHIC IMPLICATIONS

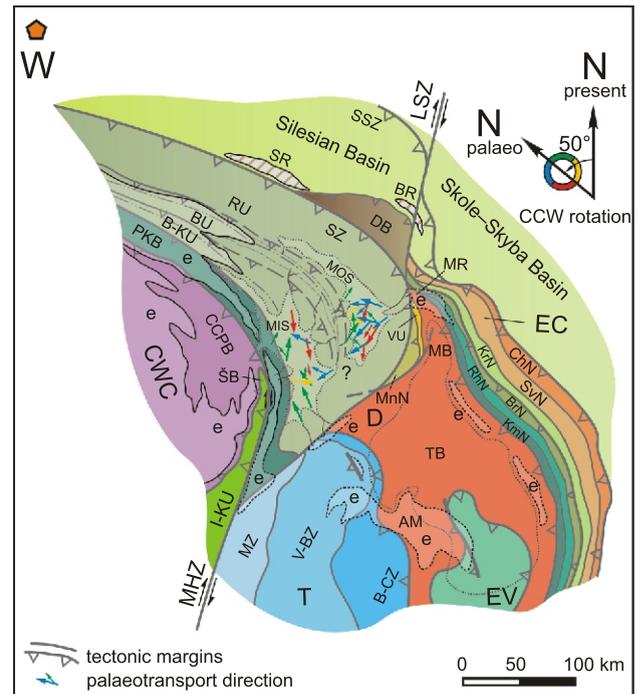
Minerals from rocks of supra-crustal origin dominate in the heavy mineral associations analysed. Some involvement of basic sources is indicated by Cr-spinel occurrence. Considering their character and occurrence within subjacent formations, their redeposition may be inferred.

We do not consider the Iňačovce–Kričovo Unit as a possible (original) source of basic material, since its outcrop and exhumation are datable to the Lower Miocene based on the FT zircon age (Soták et al., 2000, 2005). Additionally, Malcov detrital Cr-spinels show some geochemical differences compared with the spinels from this unit (Zbudza locality).

Cr-spinels may have been derived from the Pieniny Klippen Belt (PKB). However, we note the predominance of volcanic Cr-spinels in the Malcov deposits which are in a minority in the Czorsztyn Unit (Aubrecht et al., 2008, 2009) or the Jarmuta Fm. (Salata, 2002a). Sporadic volcanic Cr-spinel compositions appear to be more heterogeneous and change from ARC to MORB in the PKB (Lenaz et al., 2009). Cr-spinels from the Malcov Fm. show some compositional similarity (volcanic origin) with those from the Poruba Fm. (cf. Mikuš et al., 2006). Mikuš et al. (2006) inferred an affinity to the Meliata Ocean. Aubrecht et al. (2009) proposed the same origin for Cr-spinels from the Czorsztyn Unit considering as a source the exotic Andrusov Ridge, representing the Meliata melange supplying the Central-Carpathian Zone in the south, and Oravicum (PKB) in the north by ophiolite detritus simultaneously.

Communication between the Magura and Central-Carpathian Paleogene basins (CCPB; Leško, 1960; Nemčok, 1961; Leško and Samuel, 1968; Birkenmajer, 1986; Soták and Bebej, 1996; Oszczypko et al., 2005a, 2010; Jurewicz, 2005; Oszczypko and Oszczypko-Clowes, 2009; Soták, 2010; Oszczypko-Clowes and Zydek, 2012) during the Late Eocene to Oligocene may be considered. Malcov flysch sediments developed in the PKB depressions (Nemčok, 1961, 1990; Oszczypko et al., 2005a; Plašienka and Mikuš, 2010; Plašienka, 2011), and in depressions between the PKB and the Šambron–Kamenica Zone of the CCPB (Plašienka and Mikuš, 2010), also point to this. Nevertheless, we do not suggest their total interconnection (Jurewicz, 2005).

Following the analyses of palaeocurrents and petrography, heavy mineral spectra and geochemical composition of the detrital minerals, we consider the Marmarosh Massif as the most possible source of material for the Malcov deposits, especially in the Rača Unit (Fig. 8). This provenance [Marmarosh Ridge (Cordillera)/Marmarosh (Rakhov) Massif] was proposed by Leško and Samuel (1968), Potfaj (1998), Oszczypko (2004), Oszczypko et al. (2005b), Oszczypko and Oszczypko-Clowes (2006) for Magura sediments in different intervals of the Paleogene. The rocks of the Marmarosh crystalline basement (currently cropping out along the Ukrainian–Romanian border) are composed of metamorphic rocks of green-schist facies,



**Fig. 8. Schematic palaeogeographic situation of the Magura Basin and adjacent tectonic units during the initial Late Oligocene (based on Koráb et al., 1962; Stráňík, 1965; Contescu et al., 1966; Nemčok and Ďurkovič, 1989; Kováč et al., 1994; Leszczyński, 1997; Csontos and Vörös, 2004; Ślącza, 2005; Oszczypko and Oszczypko-Clowes, 2006, 2009; Márton et al., 2007, 2013; Schmid et al., 2008; Ustaszewski et al., 2008; Merten, 2011; Merten et al., 2011; Handy et al., 2014; Hnylko and Generalova, 2014; Plašienka and Soták, 2015; Hnylko et al., 2015a, b; Hnylko and Hnylko, 2016)**

Magura Basin: SZ – Siary Zone, RU – Rača Unit (zone), BU – Bystrica Unit (zone), B-KU – Biele Karpaty–Krynica Unit (zone); MnN – Monastrets Nappe; DB – Dukla Basin, SR – Silesian Ridge (Cordillera); BR – Bukowiec Ridge (Cordillera); CWC – Central Western Carpathians; PKB – Pieniny Klippen Belt; CCPB – Central-Carpathian Paleogene Basin; ŠB – Šambron Basin; I-KU – Iňačovce–Kričovo Unit; Tisza Mega-Unit (T); MZ – Mecsek Zone, V-BZ – Villány–Bihar Zone, B-CZ – Békés–Codru Zone; Dacia Mega-Unit (D); TB – Transylvanian Basin (land and epicontinental area), MB – Maramures Basin (trough), AM – Apuseni Mts.; VU – Vezhany Unit; EV – Eastern Vardar ophiolitic unit; Eastern Carpathians (EC): MR – Marmarosh (Rakhov) Ridge, Fore-Marmarosh Suture (Ceahlau); KmN – Kamyanyi Potik Nappe, RhN – Rakhiv Nappe, BrN – Burkut Nappe; KrN – Krasnoshora Nappe, SvN – Svydovets Nappe, ChN – Chornohora Nappe; MHZ – Mid-Hungarian Zone; LSZ – Latorytsa–Stryi strike-slip Zone; W – Wien, e – emergent areas

epidote-amphibolite and amphibolite facies (mica schists with garnet, sporadically with staurolite, gneisses, quartzites, amphibolites and amphibolite gneisses, schists) and granites (Săndulescu and Bercia, 1974; Zlatogurskaya et al., 1976; Rastochinskaya et al., 1981; Burov et al., 1986). The presence of rocks with lenses of rodonite-rodochroite mineralisation (with spessartine) in Paleozoic complexes is also observed (Zlatogurskaya et al., 1976; Matkovskiy et al., 2011), and the zoned spessartine garnet (~85 mol% Sps) could have originated from it. Spessartines with subordinate content of grs and alm components are described in garnet-magnetite-quartzite, garnet-quartzite rocks (gondites) and calcareous-siliceous manganese ores within the northern part of the Marmarosh Massif (Chivchiny Mountains; Matkovskiy, 1971; Matkovskiy et al., 2011). Spessartines are also known from the Marmarosh Massif attached to the Romanian Carpathians (Preluca Massif), usually with a higher molar content of andradite component (Udubaşa et al., 1996).

Mesozoic gabrodiabases and diabases in the Marmarosh Massif are a potential source of the basic rock relics (Cr-spinels) in the Malcov deposits. Blocks of basic volcanic material are found embedded in coarse-grained clastic deposits of Late Jurassic and Cretaceous age in the north of Romania and in Ukraine, in the continuation of the Ceahlău Black Flysch (Fore-Marmarosh Suture) units. The Severin-Ceahlău ophiolites and basalts, respectively, are regarded as remnants of an intracontinental oceanic basin situated within the European continental margin (Hoeck et al., 2006, 2009). The Chyvchyn Formation adherent to the Kamynnyi Potik Nappe (Fore-Marmarosh Suture) consists of Late Jurassic basic volcanic rocks (Hnylko et al., 2015b). Tectonic klippen of Jurassic–Early Cretaceous mafic volcanic rocks of both oceanic and continental origin are present at the base of the Burkut Nappe (Lyashkevich et al., 1995). The lower part of the Vezhany Nappe located to the north-west of the Marmarosh Crystalline Massif is represented by an Early Cretaceous ~1.000 m thick olistostrome with olistoliths of Proterozoic(?)–Paleozoic–Mesozoic rocks derived from the Marmarosh Massif as well as olistoliths of Mesozoic mafic-ultramafic rocks and of Early Cretaceous organogenic Urgonian-type limestones (Hnylko and Hnylko, 2016).

The investigation of heavy mineral associations as well as their chemical composition show that the nature of the source material for the Magura Basin deposits (the Malcov Fm.) during the Eocene to Oligocene did not change significantly. Crystalline rocks still play an important role in the source region. During this period, crystalline complexes of the Dacia Mega-Unit were elevated (e.g., Hnylko, 2011a, c). Albeit burial is indicated for the NW part of the Transylvania Basin (including the Preluca Massif), East and SE Carpathians during Middle Eocene to Oligocene (Merten, 2011; Gröger et al., 2013), small parts of crystalline basement in the northern part of the Eastern Carpathians (as shown by the AHe ages of apatite – cf. Merten, 2011: fig. 6.4D) and Apuseni Mts. (Tisza Mega-Unit) were exhumed (Merten, 2011; Merten et al., 2011). However, it should be noted that the rocks from higher levels of the metamorphic complexes were exposed, and owing to the increased content of carbonate clasts along with the cover sequences or nappes of the Fore-Marmarosh Suture – Kamynnyi Potik, Rakhiv and Burkut (Porkulets), relatively rich in carbonates.

The PKB may have been the source of carbonate detritus (Đurkovič, 1966; Olszewska and Oszczypko, 2010) but Birkenmajer (1986) inferred that it was buried during the Late Eocene–Early Oligocene. Triassic and Jurassic carbonate rocks included in the Marmarosh Massif and Fore-Marmarosh

Suture (Andrusov, 1936; Lashmanov and Zaydis, 1971; Burov et al., 1986; Hnylko, 2011b, 2012; Hnylko et al., 2015b) appear to be a more probable source, but nevertheless part of the carbonate detritus was derived from Klippen Belt.

The Tisza Mega-Unit is proposed as a potential source for flysch sediments of the Magura Basin (e.g., Oszczypko et al., 2006; Oszczypko and Oszczypko-Clowes, 2009), its lithological composition substantially corresponding to the assumed source rocks – gneisses, mica schists, amphibolites, granitoids and minor calc-silicate rocks and eclogites. The position of the Tisza Mega-Unit during the Eocene and Oligocene (cf. Csontos et al., 1992; Kováč et al., 1994; Csontos, 1995; Csontos and Vörös, 2004; Kovács et al., 2007) and also during the Early Miocene (Kováč et al., 1994; Kovács et al., 2007; Ustaszewski et al., 2008), as well as palaeoflow directions (Koráb and Đurkovič, 1966; Stráník, 1965; Nemčok and Đurkovič, 1989; data from this work), indicate this unit as a possible source area mainly for the Krynica Unit. The chemical composition of detrital garnets from the Krynica Unit is similar to that of garnets from crystalline rocks of the Variscan metamorphic basement of the Görcsöny Ridge and/or Békés–Codru Zone which continues to the Apuseni Mountains (Romania) (Árkai et al., 1999; Horváth, 2007); these units are also possible sources for part of the staurolite and tourmaline. High-grade metamorphic complexes (with eclogite occurrences) of the Villány–Bihar Zone seem to be a source for detrital garnets with higher pyrope content. The chemical composition of garnets from eclogite occurrences within orthogneiss host rock in the Jánoshalma High (middle part of the crystalline basement of the Tisza Mega-Unit, Zachar et al., 2007) is the same.

## CONCLUSIONS

1. The Malcov Formation originated in several small-sized sub-basins the axis of which runs in NW–SE (present coordinates in the Eastern Slovakia). The shape and size of sub-basins significantly affected the sedimentation of the clastic deposits. The bulk of the clastic deposits has the character of classic turbidites. These lithofacies were deposited from turbidity currents or concentrated density flows. Very thick beds of greywacke quartz-carbonaceous sandstones with composite lamination originated from large-volume concentrated gravity currents, which were reflected or deflected in flow direction at the edges of sub-basins. Coarse-grained lithofacies were deposited mostly from concentrated density currents.

2. The palaeoflow record is very varied and highlights the contribution of sedimentary material from several directions and/or diversion of gravity currents from the main flow direction (SE–NW). A significant impact was also input of clastic sediments from peripheral parts of sub-basins – structural elevations, which at the time of sedimentation were still below sea level.

3. Analysis of the heavy mineral associations showed a dominance of garnet over zircon, rutile and tourmaline. The heterogeneous chemical composition of garnets and tourmalines is significant as regards metamorphic source rock origin, these being especially low- to medium-grade (rarely high-grade) metasedimentary rocks. Zoned spessartine garnets were derived from low-grade metasedimentary rocks with Mn mineralisation. Cr-spinels indicate a specifically volcanic origin. Some heavy minerals originated from resedimentation.

4. Following the heavy mineral spectra and palaeoflow analyses, we conclude that the main part of the detrital material forming the Malcov Formation deposits mainly in the Rača Unit was derived from the Marmarosh Massif and Fore-Marmarosh

Suture. Besides, we infer for the Krynica Unit a significant contribution of detrital material from medium- to high-grade metamorphic complexes of the Villány–Bihor and Békés–Codru zones (crystalline basement of Tisza Mega-Unit; Fig. 8). The Pieniny Klippen Belt probably also supplied the Malcov sedimentary basin. Part of the clastic material was redeposited from older flysch formations.

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