

# Petrographic and heavy mineral analysis of the Upper Cretaceous – Paleocene turbiditic deposits of the Pupov Formation (Western Carpathians, Pieniny Klippen Belt, Terchová-Zázrivá area)

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## AGEOS

**Abstract:** Provenance study of the Upper Cretaceous – Paleocene turbiditic deposits of the Pupov Formation with uncertain tectonic affiliation exposed in the Varín sector of the Pieniny Klippen Belt is presented. Petrographic and heavy mineral analyses including geochemistry of detrital Cr-spinels, tourmalines and garnets were carried out. The studied deposits include litho-quartzose to feldspatho-litho-quartzose sandstones, fine-grained conglomerates and pebbly mudstones. They contain mostly lithoclasts of low- to medium-grade metamorphic rocks, basic volcanites and carbonates. Impoverished heavy mineral assemblage includes ultrastable tourmaline, zircon, rutile accompanied by Cr-spinel and apatite. Garnets, exclusively of almandinic composition, are rare, except of one locality from pebbly mudstone where they dominate. Cr-spinels of harzburgitic peridotite composition derived from supra-subduction zones dominate over Cr-spinels of volcanic origin. Detrital tourmalines include besides schorlitic-dravitic tourmalines also subhedral to angular distinctly zoned tourmalines of schorlitic-dravitic and magnesiofoititic to foititic compositions with fine intergrowth with quartz and tourmalines possessing magnesiofoititic-foititic cores and schorlitic-dravitic rims. The primary source for the Cr-spinels and complexly zoned tourmalines appears to be in Meliata ophiolite-bearing complexes feeding flysch deposits in the original Patric Zliechov Basin. After the Turonian emplacement of the Patric nappe system beyond the Tatric edge, the exotics-bearing formations of the Klape Unit formed the source of ophiolitic detritus in foreland basins of the developing accretionary wedge. Based on the specific structural position, age, lithological composition and impoverished heavy mineral assemblage, the Pupov Formation could represent a part of the wedge-top Gosau-type basin system developed during the meso-Alpidic (Coniacian – Eocene) tectonic epoch.

**Key words:** Pieniny Klippen Belt, Gosau, Upper Cretaceous, Pupov Formation, heavy minerals, provenance

## 1. INTRODUCTION

The Upper Cretaceous to Eocene synorogenic formations of the Western Carpathians provide, through their detrital material and ophiolitic detritus, crucial information for better understanding and constraining character and timing of the pre-Coniacian thrusting events. These were followed by the meso-Alpidic (Coniacian to Eocene) evolution of the trench-foredeep and wedge-top basin systems developed in front and on top of a dynamic accretionary wedge of the Pieniny Klippen Belt (PKB) and neighbouring Central Western Carpathians (CWC) (Plašienka & Soták 2015; Kováč et al. 2016; Plašienka 2018 and references therein).

The Upper Cretaceous flysch-like sequence exposed between Terchová and Zázriva villages in the Varín (Kysuce) sector of the PKB (Fig. 1) is formed by thick turbiditic deposits with unclear tectonic affiliation. The turbiditic deposits have been defined as the Pupov Beds already by Andrusov (Andrusov, 1938, 1945). The age determinations of the Pupov Beds differ by various authors covering the range from the Albian to Campanian/Maastrichtian (Andrusov 1938; Andrusov & Scheibner 1960;

Scheibner 1967; Andrusov & Samuel 1973; Haško 1977; Haško & Polák 1979; Potfaj et al. 2003). The Upper Cretaceous flysch sequence was considered by Andrusov (1938) as a part of the deep-water Kysuca (Pieniny) Unit of the PKB. Later Andrusov & Scheibner (1960) regarded all post-Turonian clastic sediments as a “third sedimentary cycle” lying transgressively on various Jurassic to Lower Cretaceous Klippen Belt successions. Scheibner (1967) attributed the Senonian clastic deposits to the Klape Unit that overrode the Kysuca Unit in the late Santonian and was subsequently transgressed by deposits of the Pupov Beds. In the 1970-ties, the Pupov Beds were assigned to the Manín Unit, but currently regarded as the Klape Unit (Potfaj et al. 2003; Bezák et al. 2004) and forming the upper level of the undivided Albian to Santonian–Maastrichtian flysch succession (Began & Samuel 1975; Haško 1977; 1978; Haško & Samuel 1977; Haško & Polák 1979).

Recently, some arguments, needing further verifications, allow to consider the Pupov Beds as a part of post-thrusting, late syn-orogenic sedimentary sequence correlated with the Gosau Group sediments overlying deep-water deposits of the Oravic Pieniny Unit or possibly the Klape Unit (Plašienka et al.

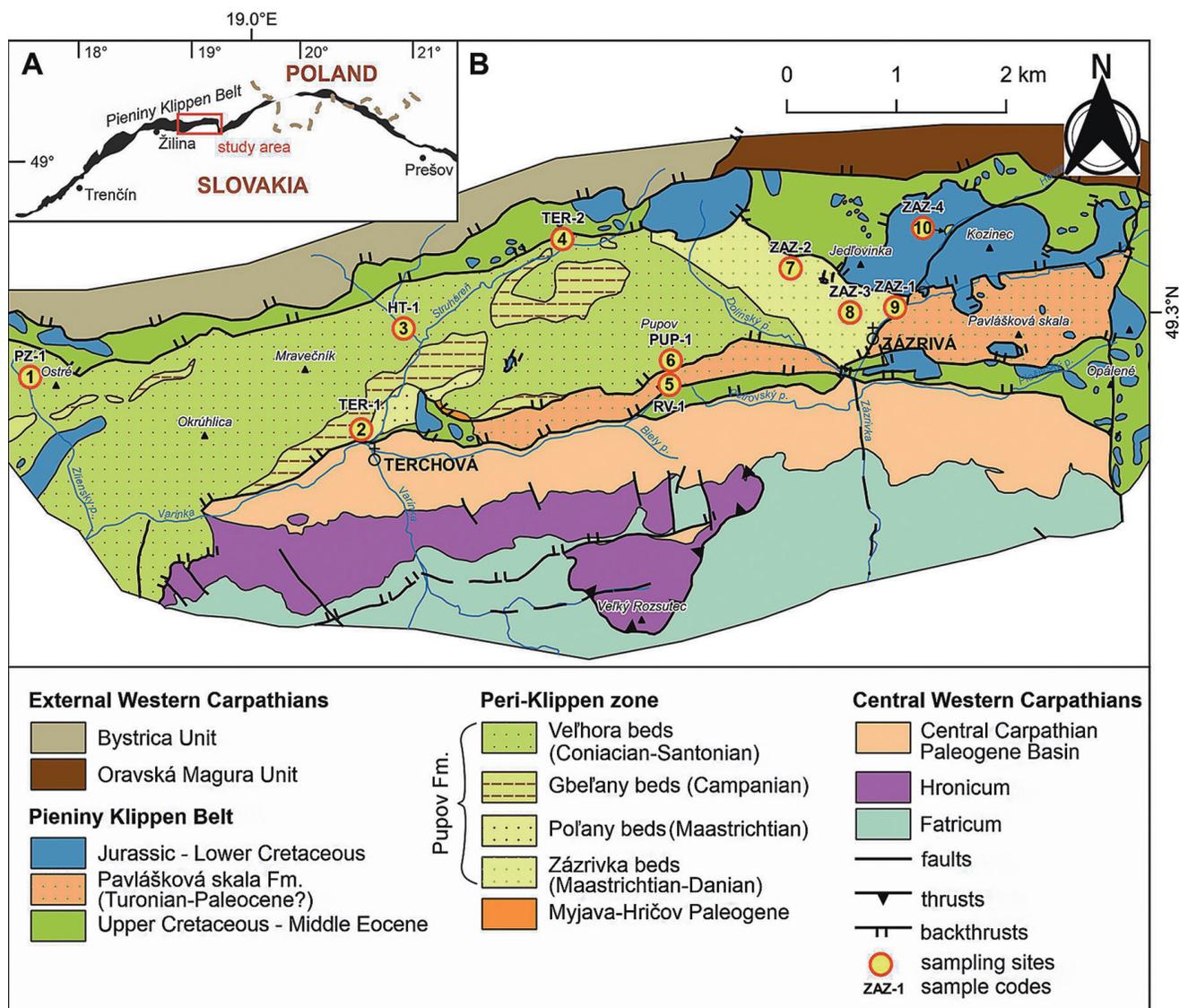


Fig. 1: Location (A) and simplified geological map of the studied area with sampling sites (B) (according to Haško & Polák 1978; Plašienka et al. 2021).

2021). These authors also redefined the Pupov Beds as a regional lithostratigraphic unit – the Pupov Formation (Fig. 1).

Hitherto, no heavy mineral data from the Pupov Fm. have been available, except of those from few localities studied recently by Aubrecht et al. (2021). This study presents new results of the petrographic and heavy mineral analyses including geochemical microprobe analyses of detrital Cr-spinels, tourmalines and garnets. The geochemistry aims at the composition of detrital minerals and comparison of obtained results with heavy mineral data from other Upper Cretaceous to Eocene syn-orogenic formations of the PKB and adjacent zones (Salata 2004; Aubrecht et al. 2009; 2021; Bellová et al. 2018; Madzin et al. 2019; Bónová et al. 2020), CWC nappe units (Bellová et al. 2018; Plašienka et al. 2019; Aubrecht et al. 2020a; 2020b) as well as from the deposits of the overstep Gosau Group basin system of the Northern Calcareous Alps and westernmost part of the Western Carpathians (Wagreich & Marschalko 1995; Stern & Wagreich 2013). Consequently, the study attempts to constrain the provenance of these sediments and shed some light on the tectonic evolution of the PKB and adjacent zones.

## 2. GEOLOGICAL SETTINGS

The PKB forms a narrow, internally intricate zone, which creates a dividing element between the External Western Carpathians and Central Western Carpathians (Froitzheim et al. 2008; Plašienka 2018) or between the External and Internal Western Carpathians sensu Hók et al. (2014; 2019). The study area belongs to the eastern part of the Varín (Kysuce) sector of the PKB in northern Slovakia (Fig. 1). The Varín sector of the PKB is approximately 3–5 km wide fault-bounded belt that stretches in the W–E directions from Žilina to Zázrivá village (Fig. 1). From the north, the PKB adjoins the Bystrica and Krynica (Oravská Magura) units (Potfaj et al. 2003), which belong to the Magura Nappe Unit of the Outer Carpathian Flysch Belt (e.g. Teták et al. 2019). On the south, the PKB is back-thrusted (Marko et al. 2005; Pešková et al. 2012) onto the post-emplacement sedimentary sequences of the Gosau Group (Plašienka & Soták, 2015) and fore-arc Central Carpathian Paleogene Basin (e.g. Soták et al. 2001).

The very complicated structure of the PKB in the studied area (Andrusov 1931; 1938; 1968; Haško & Polák 1979; Plašienka et



**Table 1: Geographic coordinates (WGS 84) and petrographic description of the sandstone samples used for heavy mineral analysis.**

No.	Locality	Sample	GPS	Formation	Age	Unit	Lithology
1.	Panské Zliene	PZ-1	49°16'11.1" 18°56'40.8"	Pupov Fm. Velhora beds	Coniacian- Santonian	Gosau	massive in the upper part of bed parallel laminated fine-grained sandstone
2.	Terchová – view point	TER-1	49°15'40.0" 19°01'39.0"	Pupov Fm. Gbelany beds	Campanian- Maastrichtian	Gosau	parallel to wavy laminated very fine-grained sandstones/siltstones
3.	Horná Tižina	HT-1	49°16'40.6" 19°02'18.6"	Pupov Fm. Velhora beds	Coniacian- Santonian	Gosau	fine-grained sandy matrix of pebbly mudstone/sandstone
4.	Terchová – Kvočkovci	TER-2	49°17'30.7" 19°04'40.4"	Pupov Fm. Velhora beds	Coniacian- Santonian	Gosau	structureless massive fine-grained sandstone
5.	Rovná Hora	RV-1	49°14'43.2" 19°06'14.1"	Pupov Fm. Velhora beds	Coniacian- Santonian	Gosau	medium-grained sandstone
6.	Pupov – Miškovci	PUP-1	49°16'20.0" 19°06'18.9"	Pupov Fm. Velhora beds	Coniacian- Santonian	Gosau	parallel laminated fine-grained sandstone
7.	Zázrivá 2 – Dolina	ZAZ-2	49°17'13.7" 19°08'06.3"	Pupov Fm. Zázrivka beds	Maastrichtian- ?Paleocene	Gosau	structureless massive fine-grained sandstone
8.	Zázrivá 3 – Cemetery	ZAZ-3	49°16'48.2" 19°08'58.5"	Pupov Fm. Zázrivka beds	Maastrichtian- ?Paleocene	Gosau	fine-grained sandstone
9.	Zázrivá 1 – Center	ZAZ-1	49°16'50.9" 19°09'40.6"	Pupov Fm. Zázrivka beds	Maastrichtian- ?Paleocene	Gosau	fine-grained sandstone
10.	Zázrivá 4 – Havrania	ZAZ-4	49°17'36.8" 19°10'31.8"	Poruba Fm.?	Albian-Santonian	Kozinec	fine-grained sandstone

Rovná Hora (locality no. 5, Fig. 1, Table 1) the sample for heavy mineral analysis was taken from calcareous sandstones (RV-1) resting directly on pelagic “biancone” limestones belonging to one of the deep-water sedimentary successions of the PKB. Additional two hand samples (RV-2A, B, only for thin sections) were taken from the fine-grained conglomerate body outcropping immediately above the calcareous sandstones.

The sample ZAZ-4 (locality no. 10, Fig. 1, Table 1) was taken from Albian-Santonian turbiditic sandstones outcropping in a problematic position interpreted either as a tectonic window of the Pupov Beds belonging to the Manín Unit (Haško 1977) or as the youngest member of the overturned special-type Oravic Kozinec succession (for more details see Aubrecht et al. 2020a; Plašienka et al. 2021 p. 41-42 and references therein).

From all samples standard and polished thin sections were produced for the petrographic examination. The petrographic composition of the sandstone samples was determined by ribbon-counting method by counting at least 250 grains in each thin section. The main framework components recorded included quartz, feldspars and lithic fragments including carbonates and cherts, bioclasts, matrix and cement.

The samples for heavy mineral analyses were cleaned, crushed and sieved under 250 µm. Then the 63–125 µm fraction obtained was soaked in 10 % diluted cold acetic acid for at least 24 hours in order to dissolve carbonate content. After acidic treatment the fraction was cleaned in an ultrasonic bath to remove remained carbonate or adhered clay minerals and dried in an oven. Such prepared fraction was ready for the heavy liquid separation in LST heavy liquid (2.85 g/cm<sup>3</sup>) using either a centrifuge or a separating funnel. After separation the heavy mineral fraction was cleaned with ethyl alcohol and dried in air. Associations of heavy minerals were studied under a petrographic microscope Zeiss Axiom Scope A.1 and a stereomicroscope Olympus SZ-61

in both transmitted and reflected light. At least 200 grains were counted using the ribbon counting method (Galehouse 1971) and are shown as number percentages (Table 2). Minerals selected for geochemical analysis, including Cr-spinels, tourmalines and garnets, were hand-picked, placed in an epoxy resin, polished and carbon coated for microprobe analyses.

Chemical compositions of the selected minerals were analysed using a JEOL JXA-8530FE microprobe (Earth Science Institute of the Slovak Academy of Sciences, Banská Bystrica, Slovakia) under the following conditions: accelerating voltage 15 kV, sample current 20 nA, probe diameter 2–5 µm, counting time 10 s – peak and 5 s for background, ZAF correction. The standards used, including lines and detection limits (in ppm) were: Ca (K $\alpha$ , 19–21) – diopside, Mn (K $\alpha$ , 49–62) – rodonite, Si (K $\alpha$ , 45–50) – quartz, Mg (K $\alpha$ , 35–37) – olivine, F (K $\alpha$ , 112–294) – fluorite, Na (K $\alpha$ , 31–36) – jadeite, Al (K $\alpha$ , 38–40) – kyanite, K (K $\alpha$ , 29–38) – orthoclase, Fe (K $\alpha$ , 43–57) – hematite, Ti (K $\alpha$ , 35–38) – rutile, Cr – (K $\alpha$ , 71–130) – Cr<sub>2</sub>O<sub>3</sub>, Cl (K $\alpha$ , 27–34) – tugtupite.

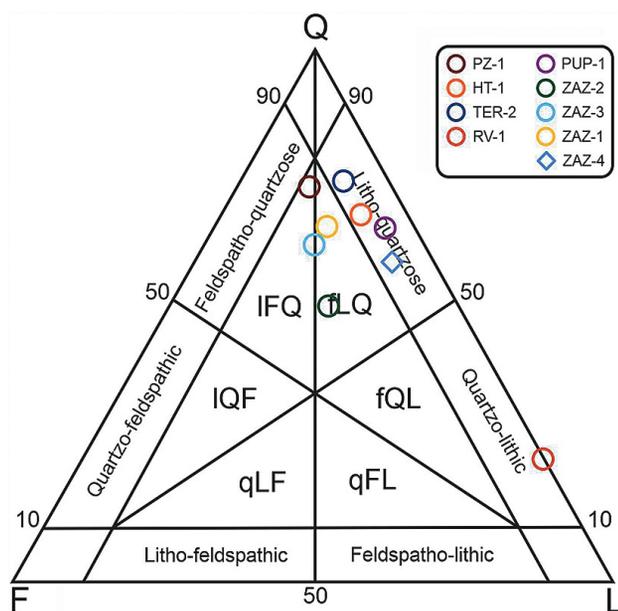
### 3. RESULTS

#### 3.1. Petrography

Based on the simple and pure descriptive classification proposed by Garzanti (2015; 2019), the turbiditic sandstones (localities no. 1, 3, 4, 6–10, Fig. 1, Table 1) can be classified mostly as litho-quartzose ( $Q > L > 10\% QFL > F$ ) to feldspatho-litho-quartzose ( $Q > L > F > 10\% QFL$ ) and occasionally as litho-feldspatho-quartzose sandstones ( $Q > F > L > 10\% QFL$ ) (Fig. 3, 4A). Matrix and cement content varied between 1–20 % and 10–28 %, respectively. Matrix is represented mostly by clay minerals. Cement is formed by calcite. Sandstones are well- to moderately well-sorted.

**Table 2:** Percentage of the heavy mineral species in the studied samples. Abbreviations used after Whitney & Evans (2010). ZTR = ultrastable species including zircon, tourmaline and rutile (Hubert 1962).

No.	Locality	Sample	Zrn	Tur	Rt	Grt	Cr-spl	Ap	Mnz	St	ZTR	ZTR+ Cr-spl
1.	Panské Zliene	PZ-1	14.0	27.5	14.0	6.0	4.0	31.5	1.0	2.0	55.5	59.5
3.	Horná Tižina	HT-1	7.5	6.5	10.0	56.0	1.5	18.5	0.0	0.0	24.0	25.5
4.	Terchová-Kvočkovci	TER-2	28.5	30.5	11.5	6.0	23.0	0.5	0.0	0.0	70.5	93.5
5.	Rovná Hora	RV-1	33.2	17.1	20.1	5.0	16.1	6.0	1.0	1.5	70.4	86.4
6.	Pupov - Miškovci	PUP-1	25.0	30.5	11.5	6.5	14.0	11.0	1.5	0.0	67.0	81.0
7.	Zázrivá 2 - Dolina	ZAZ-2	38.0	21.0	14.0	1.0	20.0	6.0	0.0	0.0	73.0	93.0
8.	Zázrivá 3 - Cemetery	ZAZ-3	26.0	27.0	16.0	5.0	11.0	15.0	0.0	0.0	69.0	80.0
9.	Zázrivá 1 - Center	ZAZ-1	23.5	25.5	15.0	5.0	17.5	11.5	2.0	0.0	64.0	81.5
10.	Zázrivá 4 - Havrania	ZAZ-4	26.0	15.0	4.5	2.0	37.5	15.0	0.0	0.0	45.5	83.0



**Fig. 3:** Petrographic classification of the turbiditic sandstones studied (localities no. 1, 3, 4, 6–10, Fig. 1, Table 1). The classification is based on the relative abundance of their three main components (quartz, feldspars and lithic fragments). Q pole includes both mono- and polycrystalline quartz. F pole includes all feldspars. Lithic pole L includes all volcanic, ultramafic, metamorphic and sedimentary clasts including carbonates and cherts. If the main components exceed 10% QFL then are listed in order of abundance (e.g. in a feldspatho-litho-quartzitic sandstone  $Q > L > F > 10\% QFL$ , in a litho-feldspatho-quartzitic sandstone  $Q > F > L > 10\% QFL$ , in a litho-quartzose sandstone  $Q > L > 10\% QFL > F$ ) (from Garzanti 2015 and references therein).

Framework grains occur mostly in angular to subrounded shape (Fig. 4A).

Quartz is ubiquitous and most common constituent and occurs in both monocrystalline and polycrystalline form. Monocrystalline quartz frequently shows undulation. Polycrystalline quartz represents either fine-grained (<62 μm) or coarse-grained aggregates with or without feldspars and mica flakes.

Alkaline feldspars, such as orthoclase or microcline dominate over plagioclases. All feldspars suffered moderate to high degree of alteration, mostly sericitization.

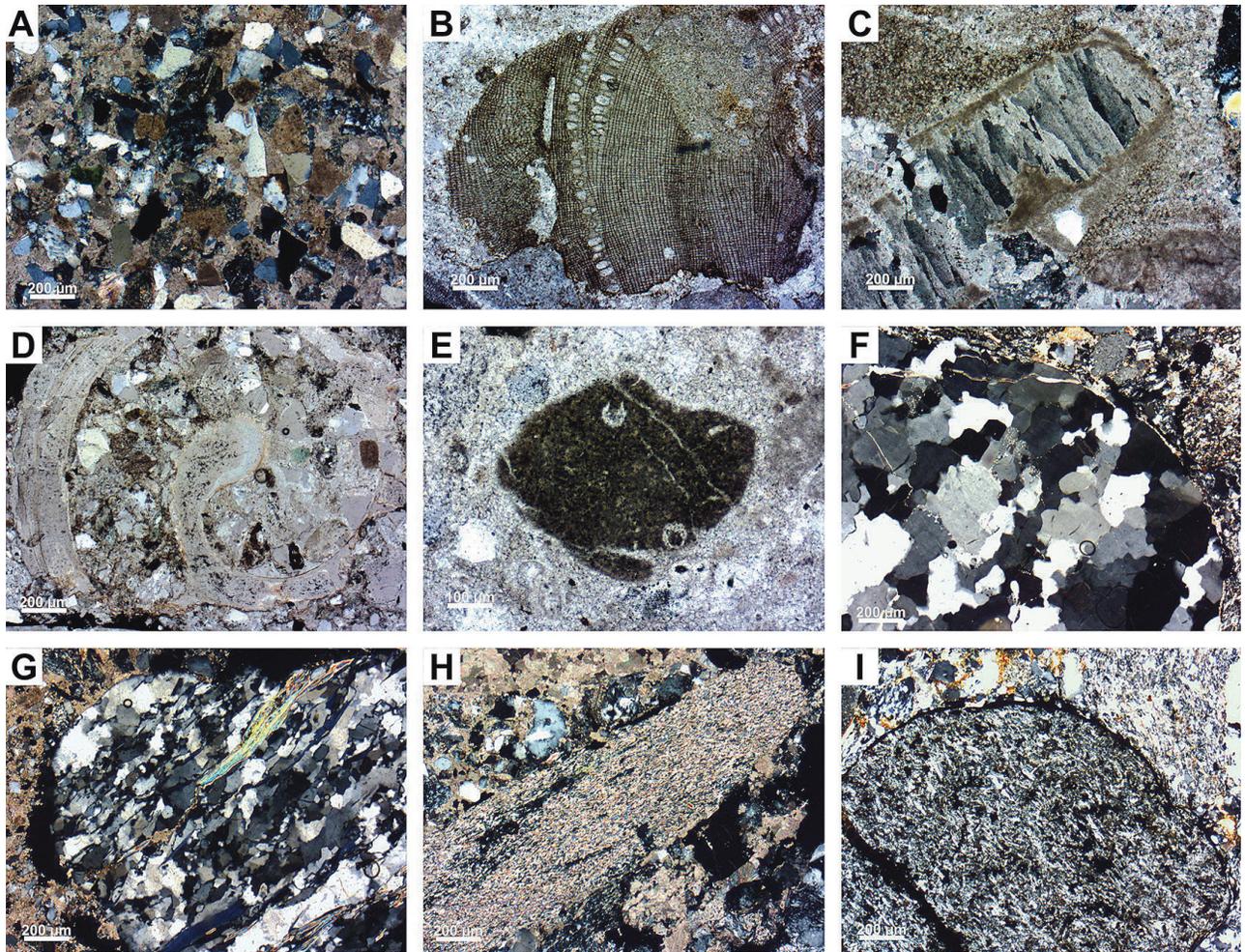
Lithic grains include metamorphic, volcanic and carbonate

rocks. Volcanic grains are represented by mafic volcanic rocks with intersertal texture. Metamorphic grains are represented mostly by low- to medium-grade mica schists and gneisses. Lithoclasts of low-grade chlorite-sericite schists were less abundant. Carbonate clasts include mainly micritic limestones. Bioclasts include planktonic and benthic foraminifera, bivalve and brachiopod fragments.

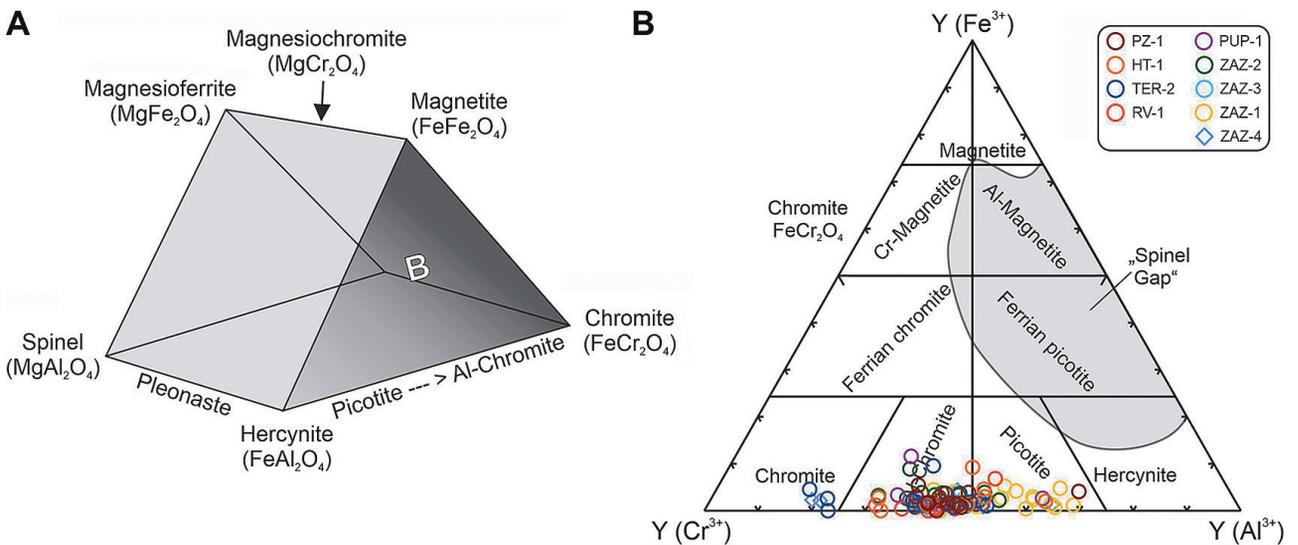
The sample RV-1 (locality no. 5, Fig. 1, Table 1) was taken from medium- to coarse-grained calcareous sandstones occurring in a direct contact with the Lower Cretaceous pelagic “biancone” limestones belonging most probably to one of the deep-water successions of the PKB. The calcareous sandstones are composed almost exclusively of carbonate lithoclasts with sporadic admixture of monocrystalline and polycrystalline quartz (Fig. 3). Two hand samples RV-2A, RV-2B were taken from fine-grained conglomerate body outcropping immediately above the calcareous sandstones. The sample RV-2A represents polymict clast-supported conglomerate which consists predominantly of shallow-water carbonates mixed with sporadic deep-water carbonate lithoclasts. Shallow-water carbonate lithoclasts include organodetrital aggregate clasts and bioclasts represented by abundant fragments of red coralline algae, inoceramid bivalves and bryozoans. Less frequently brachiopods, gastropods and echinoderms have been observed (Fig. 4B-D). Benthic agglutinated foraminifera are abundant as well. Planktonic foraminifera were rarely observed. Sporadic deep-water carbonate clasts comprise calpionella- and radiolaria-bearing wackestones (Fig. 4E). Cherts and clasts of basic volcanic rocks are rarely present. The sample RV-2B contains more variegated clastic material. Unlike the sample RV-2A, abundant lithoclasts of various igneous rocks, mostly granitoids and low- to medium-metamorphic rocks such as chlorite-muscovite gneisses, mica-schists and chloritic-sericite schists were observed (Fig. 4F-H). Volcanic rock fragments involve prevalingly basic volcanic rocks with various degree of alteration (Fig. 4I). Sedimentary rocks are represented by lithoclasts of shales, siltstones, quartzose sandstones and cherts. Lithoclasts of shallow- or deep-water carbonates are less frequent.

### 3.2. Heavy mineral analysis

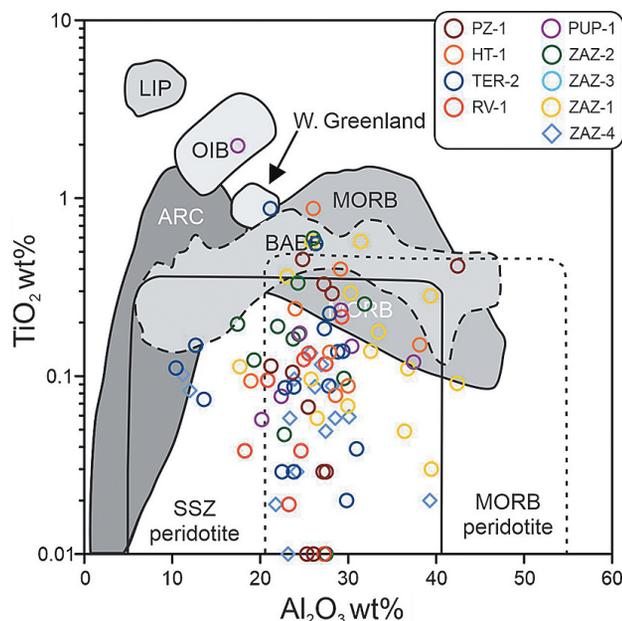
Heavy mineral spectra are dominated by ultra-stable zircon-tourmaline-rutile (ZTR) trinity accompanied by Cr-spinel and



**Fig. 4:** Microphotographs of representative lithoclasts in the deposits studied. A) characteristic sandstone facies of the turbiditic sandstones, litho-quartzitic sandstone, sample PUP-1, locality no. 6, Pupov-Miškovci; B–I lithoclasts in the calcareous sandstones and fine-grained conglomerates at locality no. 5 Rovná Hora (samples RV-1, RV-2A, RV-2B): B) fragment of coralline red algae, C) inoceramid bivalve fragment, D) gastropod shell *Acteonella* sp.?, E) calpionella-bearing wackestone clast, F) well-rounded clast of polycrystalline quartz, G) well-rounded clast of chlorite-muscovite schist, H) fragment of chlorite-sericite schist, I) well-rounded clast of altered basic volcanic rock. Pictures taken in XPL, except B and E which are in PPL.



**Fig. 5:** Chemical classification diagrams for the spinel group minerals (after Gargiulo et al. 2013). A) Spinel prism for multi-component system: spinel-hercynite-chromite-magnesiochromite-magnesioferrite-magnetite (after Deer et al. 1992). Letters „B“ on the lateral face represent the diagram in B. B) Triangular classification ( $Cr^{3+}-Al^{3+}-Fe^{3+}$ ) diagram. „Spinel gap“ field after Barnes & Roeder (2001). Field contours after Stevens (1944), Haggerty (1991) and Deer et al. (1992).



**Fig. 6:**  $\text{Al}_2\text{O}_3$  versus  $\text{TiO}_2$  diagram with Cr-spinel discrimination fields after Kamenetsky et al. (2001). Explanations: SSZ = supra-subduction zone, MORB = mid-ocean ridge basalts, BABB = back-arc basin basalts, ARC = island-arc magmas, OIB = ocean-island basalts, LIP = large igneous province.

apatite. Garnets occur in subordinate amounts, exception is the sample from pebbly mudstone/sandstone at locality no. 3 (sample HT-1) (Fig. 1), where garnets dominate, reaching 56 % of the total. Monazite and staurolite were observed in amounts <1 % (Table 2). Sample TER-1 (locality no. 2, Fig. 1) from very fine-grained sandstones/siltstones (Polany beds, Fig. 2) contained a very low amount of heavy minerals including only ultra-stable ZTR and has not been included in counting.

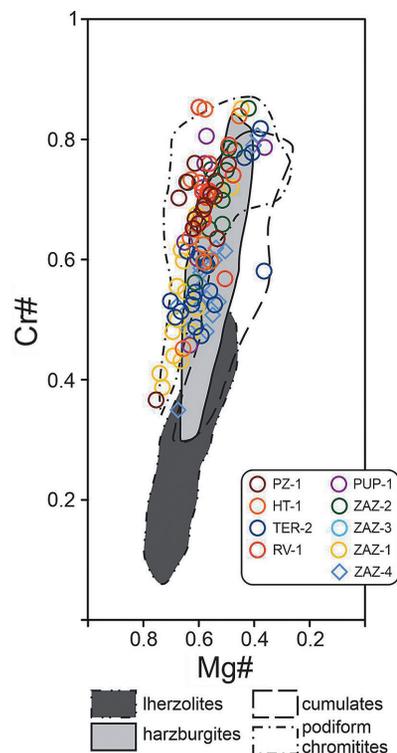
Sample ZAZ-4 taken from Albian–Santonian turbiditic sandstones at locality Zázrivá–Havrania is compositionally similar to samples from the Pupov Fm. However, it contains the highest Cr-spinel content (37.5 %) of the all samples studied (Table 2).

### 3.2.1. Geochemistry of detrital Cr-spinels

Detrital Cr-spinels are represented by angular fragments of subhedral to anhedral grains. Texturally, the analysed spinels do not show signs of alteration or zonation. In total, 106 detrital Cr-spinels were analysed (Supplementary Table S.1). Mineral inclusions, observed in one case, consists of Mg-rich olivine. In the chemical variation diagrams (Fig. 5) constructed by Gargiulo et al. (2013), based on the previous diagrams published by Stevens (1944), Haggerty (1991) and Deer et al. (1992), the analysed spinels plot in the triangular Cr-Al-Fe diagram (Fig. 5B) mostly in the fields of Al-chromite and picotite. Less often they fall in to the chromite field.

The  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  contents vary in the range of 8.63 to 40.65 and 0.00 to 1.97 wt %, respectively. The  $\text{Cr}_2\text{O}_3$  content ranges from 25.12 to 58.75 wt %. Contents of MnO and ZnO oxides are low, except in one grain where MnO content is higher than 0.5 wt %, which is considered as an indication of alteration (Zhu et al. 2004).

**Fig. 7:** Cr# versus Mg# diagram for Cr-spinels. Fields after Pober & Faupl (1988).



Majority of the analysed spinels (78 %) have the  $\text{Fe}^{2+}/\text{Fe}^{3+}$  ratio higher than 4, what is characteristic for mantle peridotites (Lenaz et al. 2000; Kamenetsky et al. 2001). In the  $\text{Al}_2\text{O}_3$  vs.  $\text{TiO}_2$  diagram (Fig. 6) these spinels plot mostly in the supra-subduction zone peridotite field (Lenaz et al. 2000; Kamenetsky et al. 2001). Spinel grains having the  $\text{TiO}_2$  concentrations higher than 0.2 wt %, originating from volcanic rocks (Lenaz et al. 2000; Kamenetsky et al. 2001), constitute 22 % of the analysed grains. The volcanic spinels correspond mostly to the compositions of MORB-type basalts and back-arc basin basalts. Occasionally, they fall in to the ocean-island basalts field (Fig. 6).

The Cr# and Mg# range from 0.29 to 0.82 and from 0.36 to 0.76, respectively. In the Cr# vs. Mg# diagram (Fig. 7) (Pober & Faupl 1988) they best match the harzburgite field and/or podiform chromitite field.

### 3.2.2. Geochemistry of detrital tourmalines

Detrital tourmalines occur as subhedral to anhedral, less frequently as subrounded grains (Fig. 8). Tourmalines often display distinct optical zonation (Fig. 8), but unzoned grains were observed as well. Altogether 158 analyses, representing spot analyses placed either in centres or in distinct optical zones of detrital tourmalines, were carried out in order to reveal possible changes in chemical compositions indicative of their evolution (Supplementary Table S.2). Some grains contain mineral inclusions represented mostly by quartz, rutile, zircon, ilmenite, apatite, albite and Fe-oxides (Fig. 8A). Based on the dominant occupancy of X-site two groups of tourmalines were distinguished. In the first group the dominant cation is Na and detrital tourmalines can be classified as alkali group tourmalines (Fig. 9). In the second, smaller but significant, group vacancies dominate

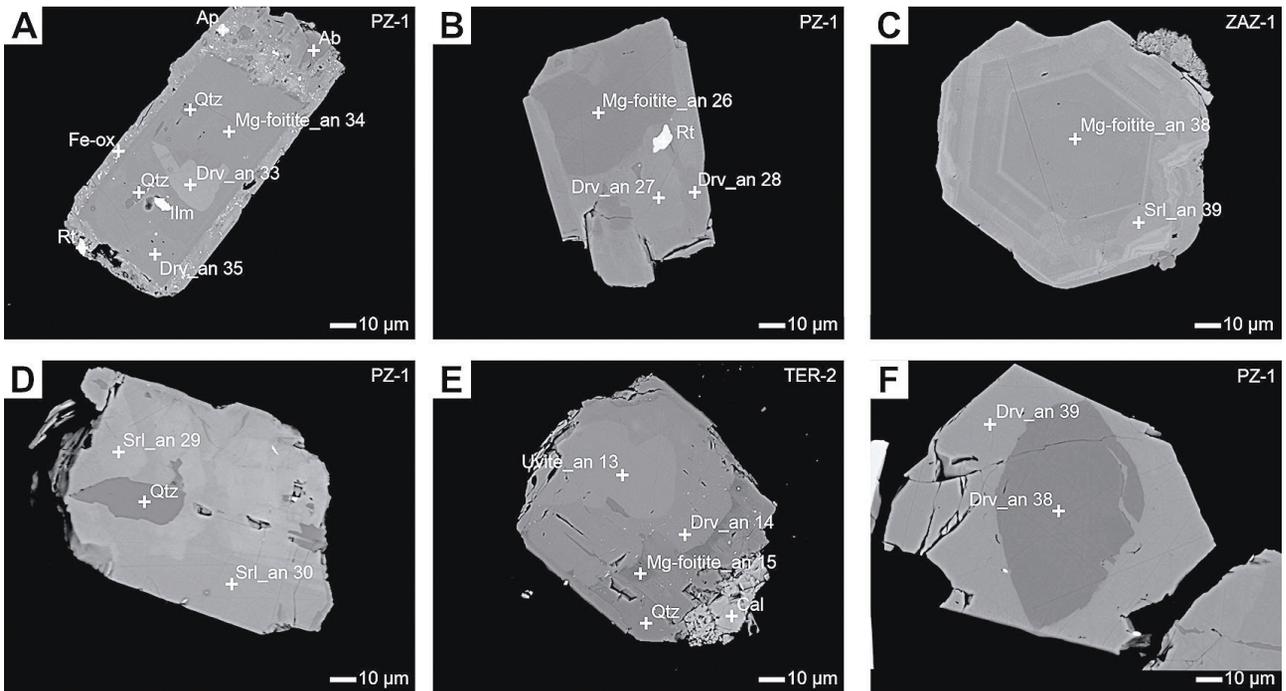


Fig. 8: BSE images of detrital tourmalines studied showing distinct optical zonations exhibiting compositional changes.

at the X-site and the analysed tourmalines are classified as X-vacant group tourmalines (Fig. 9). Calcic group tourmalines were rarely observed only in sample TER-2 at locality no. 4 (Fig. 1). Most of the analysed tourmalines have schorlitic-dravitic, less magnesiofoititic-foititic and in one case uvitic composition (Fig. 10, Supplementary Table S.2). The latter group forms mostly cores of the grains or optically distinct zones, while the former occurs at rims (Fig. 8B-E). Some grains with distinct zones of schorlitic-dravitic and Mg-foititic to foititic compositions exhibit fine intergrowths with quartz (Fig. 8A, D), while other distinctly

zoned grains do not show compositional variations (Fig. 8F).

In the triangular  $Al_{50}Fe_{50}-Al_{50}Mg_{50}-Al$  discrimination diagram (Fig. 11A) of Henry & Guidotti (1985) the analysed tourmalines cover a broad range of various types of source rocks. Tourmalines come mostly from metasedimentary rocks both coexisting and not coexisting with Al-saturation phase. Lesser amount of analysed tourmalines could have been derived from iron-rich quartz-tourmaline, calc-silicate rocks and also from Li-poor granitoids and associated pegmatites and aplites. Some grains show compositions of tourmalines coming from low-Ca metaultramafic rocks or from Cr-, V-rich metasediments. In the  $Fe_{tot}-Mg-Ca$  diagram (Fig. 11B) (Henry & Guidotti 1985) the analysed tourmalines plot mostly in the fields of Ca-poor

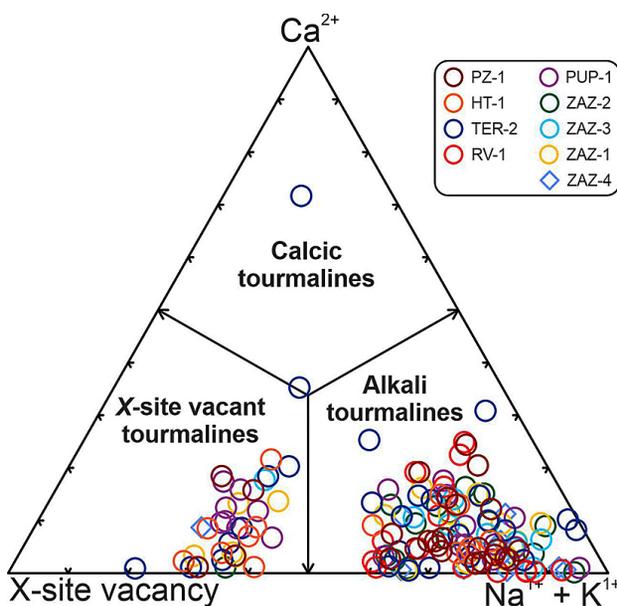


Fig. 9: Classification  $Ca^{2+}$ -X-site vacancy  $-Na^{+}+K^{+}$  diagram of primary tourmaline groups based on the dominant cation occupancy at the X-site (according to Henry et al. 2011).

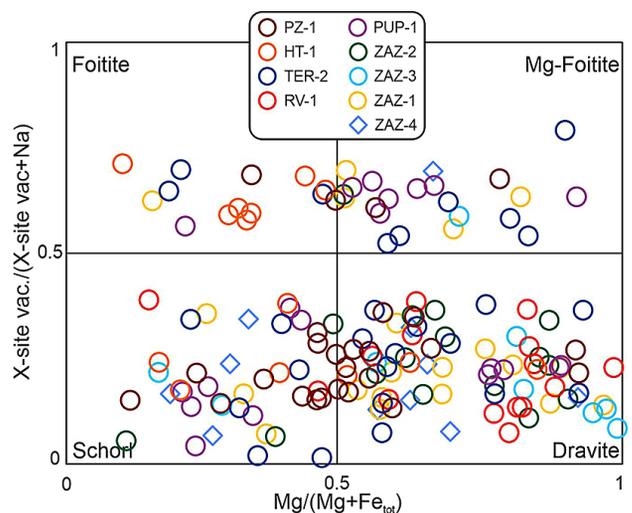
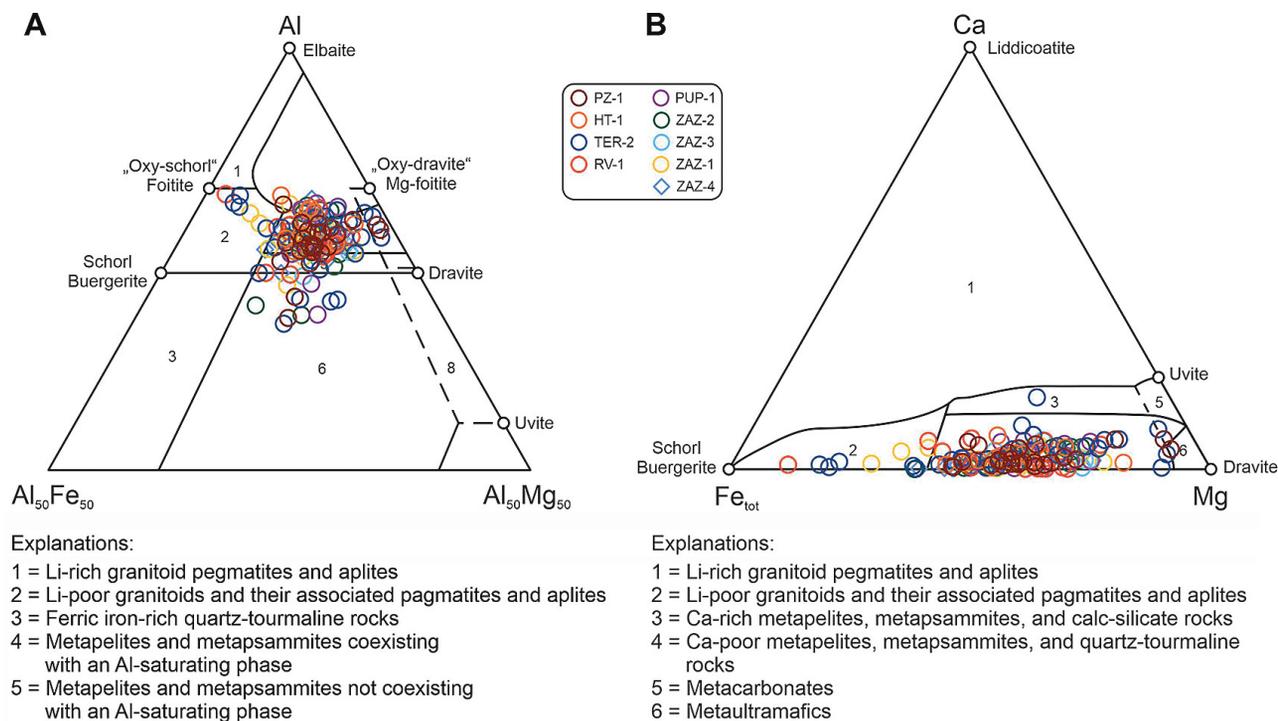


Fig. 10: Binary X-site vacant/(X-site vacant +  $Na^{+}+K^{+}$ ) versus  $Mg/(Mg+Fe)$  classification diagram of generalized tourmaline species (according to Henry et al. 2011).



**Fig. 11:** Triangular Al- $Al_{50}Fe_{50}$ - $Al_{50}Mg_{50}$  in A) and Ca- $Fe_{tot}$ -Mg in B) diagrams (in molar proportions) discriminating tourmalines coming from various rock types (after Henry & Guidotti 1985).

metasedimentary rocks, less in the field of Li-poor granitoids and related pegmatites and aplites. The tourmalines with higher content of Mg could also have been derived from metaultramafic rocks (Henry & Dutrow 1996).

### 3.2.3. Geochemistry of detrital garnets

Detrital garnets dominate the heavy mineral fraction only in pebbly mudstone/sandstone at Horná Tižina locality (sample HT-1, locality no. 3, Fig. 1). Their chemical composition was analysed on 24 grains, exclusively from this locality (Supplementary Table S.3). In other samples garnets occur in subordinate amounts (Table 2). Garnets occur as angular or subrounded fragments and show signs of slight to moderate dissolution, represented by etch pits and small-scale facets. The garnets do not exhibit distinctive zonation. Mineral inclusions observed, exclusively of quartz, are rare. The analysed garnets are almandine-rich with various pyrope, spessartine and grossular molecule proportions. Almandine-pyrope-spessartine ( $Alm_{68-75}-Prp_{12-17}-Sp_{7-12}$ ) and almandine-pyrope-spessartine-grossular garnets ( $Alm_{65-75}-Prp_{13-19}-Sp_{5-10}-Grs_{3-9}$ ) are the most frequent types. Almandine-pyrope ( $Alm_{83-84}-Prp_{11-13}$ ) and almandine-pyrope-grossular-spessartine types z ( $Alm_{74-75}-Prp_{-16}-Grs_{4-5}-Sp_{3-4}$ ) are less frequent and one almandine-spessartine-pyrope garnet ( $Alm_{63}-Sp_{20}-Prp_{13}$ ) was observed. In the almandine-pyrope-grossular and almandine-pyrope-spessartine discrimination diagrams (Fig. 12, Méres, 2008; Aubrecht et al. 2009) the analysed garnets group in the

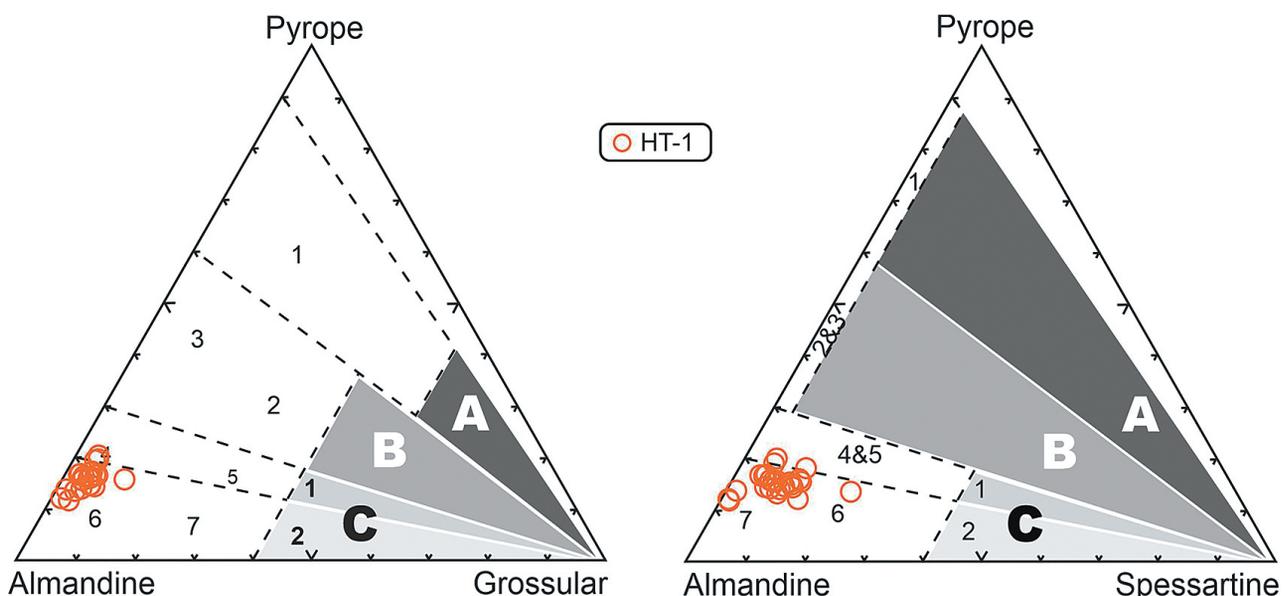
field C2 or at the boundary with the field C1 corresponding compositionally to rocks originating in the amphibolite facies conditions or in the transitional conditions between granulite and amphibolite facies conditions, respectively.

## 4. DISCUSSION

### 4.1. Petrography and heavy mineral associations

In the Dickinson's provenance diagrams (Dickinson 1985), the investigated sandstones of the Pupov Fm. plot into the field of recycled orogen (Fig. 13). Composition of lithic clasts in the sandstones indicates that the source areas were composed mostly of low- to medium-grade metamorphic rocks. Mafic or ultramafic source rocks are indicated by sporadic lithoclasts of basic volcanic rocks.

Wealth of provenance information provide the fine-grained conglomerates at the locality Rovná Hora (samples RV-2A, B, locality no. 5, Fig. 1). The conglomerates contain predominantly clasts of shallow-water carbonates mixed with less abundant clastic sedimentary rocks and rare deep-water carbonates. Lithoclasts of low- to medium-grade metamorphic rocks, acid magmatic rocks and basic volcanic rocks are abundant as well. Presence of dominant shallow-water carbonate lithoclasts and littoral biotritus infer the proximity of shallow-water shelf environment. Mixture of more stable metamorphic and igneous lithoclasts with less stable sedimentary lithoclasts may suggest



**Fig. 12:** Triangular pyrope-almandine-grossular and pyrope-almandine-spessartine discrimination diagrams for garnets (after Méres 2008; Aubrecht et al. 2009). Explanations: A – Grt derived from UHP/HP metamorphic conditions; position around number 1 – Grt derived from UHP eclogites, garnet peridotites and kimberlites; B – Grt derived from granulite and eclogite facies conditions; position around number 2 – Grt derived from HP eclogites and HP mafic granulites; position around number 3 – Grt derived from HP felsic and intermediate granulites; C – Grt derived from amphibolite facies conditions; C1 – Grt derived from transitional high amphibolite to granulite facies conditions; position around number 4 – Grt derived from gneisses metamorphosed under transitional high amphibolite to granulite facies conditions; position around number 5 – Grt derived from amphibolites metamorphosed under transitional high amphibolite to granulite facies conditions; C2 – Grt derived from amphibolite facies conditions; position around number 6 – Grt from gneisses metamorphosed under amphibolite facies conditions; position around number 7 – Grt from amphibolites metamorphosed under amphibolite facies conditions. Grey fields – immiscibility gap of end-members composition.

short transport and proximity of parental rocks, or more likely two distinct but still local sources, one providing shallow-water carbonates, the other delivering a more stable well-rounded material possibly recycled from older flysch deposits with polymict conglomerates of the Klape Unit (Plašienka 2012; Plašienka & Soták 2015).

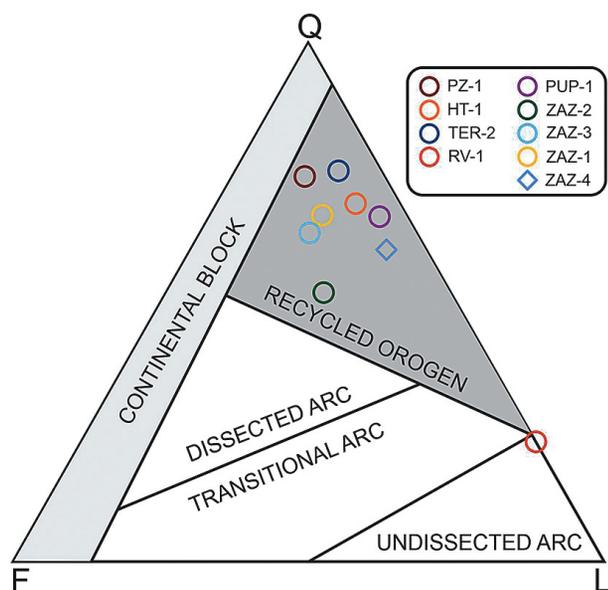
The heavy mineral association in the analysed sandstones is characterized by impoverished ultrastable ZTR trinity accompanied by Cr-spinels and apatites. Other metamorphic semistable to unstable heavy mineral species are very rare. Impoverished heavy mineral assemblage is very common in the Late Cretaceous to Early Tertiary syn-orogenic deposits of the PKB and CWC. On the other hand, surprisingly high amount of unstable pyroxenes, blue amphiboles and kyanites were preserved locally in exotics-bearing mid-Cretaceous turbidites of the Klape and Tatric cover units (Bellová et al. 2018; Aubrecht et al. 2020a). Consequently, the impoverishment was likely caused by mechanical breakdown of unstable minerals during their transport and/or intrastatal dissolution during subsequent burial (Morton & Hallsworth 2007). In both cases most of the original provenance information has been lost.

In addition to the samples from the Pupov Fm. we sampled also Albian–Santonian turbiditic sandstones at locality Zázrivá-Havrania (sample ZAZ-4, Table 1, 2, locality no. 10, Fig. 1). It is supposed that this mid-Cretaceous flysch sequence belongs to the so-called Kozinec Unit (Plašienka et al. 2021), which is suspected to belong to the “non-Oravic” units of the Fatric affiliation – i.e. analogous to the Klape or Manín units. Unlike

Aubrecht et al. (2020a, their sample “Havranský vrch”), we did not observe less stable sillimanite, pyroxene and particularly abundant kyanite in this sample. The high content of Cr-spinels would correspond rather to the mid-Cretaceous flysch formations too, since its general decrease in younger strata has been indicated by Aubrecht et al. (2020a; 2021).

#### 4.2. Provenance of Cr-spinels

Cr-spinels of harzburgitic peridotite composition derived from supra-subduction zones dominate over the analysed grains. The identical, almost exclusively, harzburgitic composition of Cr-spinels has been reported from the Albian to Cenomanian exotics-bearing flysch deposits of the Fatric and Tatric units of the CWC (Bellová et al. 2018; Aubrecht et al. 2020a), from the Klape Unit (Bellová et al. 2018; Plašienka et al. 2019; Aubrecht et al. 2020a) and from the Turonian to Coniacian–Santonian turbiditic sandstones of the Pieniny Unit (Salata 2004; Aubrecht et al. 2021). The Albian deposits of the Chmielowa Fm. of the Czersztyn succession (Aubrecht et al. 2009) and the Maastrichtian to Lower/Middle Eocene flysch deposits of the Jarmuta-Proč Fm. of the Šariš Unit (Salata 2004; Madzin et al. 2019) contain more heterogeneous population of Cr-spinels, including besides still dominant harzburgitic Cr-spinels also more Al-rich lherzolitic Cr-spinels coming from abyssal MORB peridotites. The Coniacian–Santonian deposits of the Gosau Group in the Northern Calcareous Alps (Stern & Wagerich 2013) contain Cr-spinels derived mostly from harzburgitic peridotites and



**Fig. 13:** Ternary provenance diagrams of the sandstones studied. Discrimination fields after Dickinson (1985). Q – quartz including both mono- and polycrystalline quartz, F – feldspars including plagioclase and K-feldspar, L – lithoclasts including lithoclasts of carbonates and cherts.

detrital garnets showing higher amount of pyrope molecule, which suggests derivation of both minerals from obducted ophiolites including sub-ophiolitic metamorphic soles occurring in the inner zones of the Northern Calcareous Alps (Stern & Wagneich 2013). The increasing amounts of Iherzolitic Cr-spinels has been observed in the Campanian deposits, while Maastrichtian to Paleocene deposits show decreasing amounts of Cr-spinels, again exclusively of supra-subduction provenance, and the heavy mineral associations become to be dominated by almandine-rich garnets derived from low- to medium-grade metamorphic rocks of the continental crust (Stern & Wagneich 2013). In the Slovak part of the Gosau Basin system, such a trend is not well discernible and the impoverished ultrastable heavy mineral suites with upward decreasing Cr-spinel contents are characteristic (Salaj & Prieckhodská 1987; Wagneich & Marschalko 1995; Stern & Wagneich 2013).

The provenance of the ophiolitic detritus including Cr-spinels, glaucophanite pebbles and other index minerals of the HP/LT metamorphism in the coarse-clastic deposits of the Western Carpathians is a topic of long-lasting debates, but their source in the Meliata ophiolite-bearing mélange complexes situated in the southern Western Carpathian zones has been generally accepted (Mišík & Marschalko 1988; Dal Piaz et al. 1995; Kissová et al. 2005; Aubrecht et al. 2009; Plašienka 2012; Plašienka & Soták 2015; Plašienka et al. 2019). There exist two conceptual models for derivation of the ophiolitic debris into the Cretaceous to Paleogene clastic deposits of the PKB and adjacent units.

1) A multiple recycling model, where the ophiolitic material is primarily deposited in pre- to syn-orogenic Albian-Cenomanian flysch deposits of the Poruba Fm. in the Fatric Zliechov Basin. The Poruba Fm. was, as a part of the Fatric Krížna nappe, emplaced by thrusting during the Late Turonian into a false

accretionary wedge position behind the outer Tatric margin, thereby becoming a part of the Klape Unit in the Peri-Klippen Zone (e.g. Plašienka 1995; 2012; 2019; Prokešová et al. 2012). In this position, the flysch complexes with large conglomerate bodies formed the main source of pebble material and ophiolitic detritus for still younger syn-orogenic deposits in the foredeep basins of the PKB (Plašienka 2012; Plašienka & Soták 2015; Plašienka et al. 2019). In such a way, the mid-Cretaceous flysch formations of the Klape Unit, already incorporated into the developing accretionary wedge, could also form the source of Cr-spinels for the turbiditic deposits of the Pupov Fm. deposited in a wedge-top Gosau-type basin (cf. Salaj & Prieckhodská 1987; Plašienka 2012; Plašienka & Soták 2015).

2) The Oravic crustal segment (Czorsztyn Ridge) originally formed a lateral continuation of the CWC, situated north of the Meliata oceanic realm (e.g. Michalík 1994; Aubrecht et al. 2009, their Fig. 12). After the closure of the Meliata Ocean in the Late Jurassic, the European Platform collided with the South Alpine/Dinaridic parts and the Meliata ophiolite-bearing suture zone, welded with both the CWC and Oravic segments, formed. During Cretaceous the Meliata ophiolite complex was secondarily doubled by a lateral shift of the Oravic segment along the northern margin of the CWC due to a clockwise rotation of the CWC amalgamated units (Aubrecht & Túnyi 2001). An elevated ridge, the so-called exotic Andrusov Ridge (e.g. Birkenmajer 1988), formed in a complex shear zone between the External and Internal Carpathians, which fed both the CWC and PKB units with pebble material and ophiolitic detritus for a prolonged time period (Marschalko 1986; Aubrecht et al. 2009; 2020a; Bellová et al. 2018).

We prefer here the recycling model for derivation of the Meliata ophiolitic material to the more external units of the Western Carpathians. Although, moderate to large clockwise rotations of the CWC nappe units, related to the thrusting in the Late Turonian, have been well documented (Márton et al. 2016; 2020), there is a lack of structural or sedimentary record clearly supporting the huge lateral shift of the Oravic crustal fragment during the Cretaceous. On the other hand, there exists a wealth of sedimentary, biostratigraphical and structural data documenting collision of the Oravic crustal fragment with the front of the already consolidated CWC units after elimination of the Vahic oceanic realm around Cretaceous/Tertiary boundary and an advanced propagation of the accretionary wedge involving still more external units of the PKB up to the Middle Eocene (for review see Plašienka 2012; 2018; 2019; Plašienka & Soták 2015; Plašienka et al. 2020).

### 4.3. Provenance of tourmalines

Detrital tourmalines represent one of the most frequent transparent heavy mineral in the deposits studied (Table 2). Their chemical compositions show that they were derived dominantly from low- to medium-grade metamorphic rocks with some contribution from granitoid and metaultramafic rocks (Fig. 11).

Recently, heavy mineral analyses of Albian to Cenomanian exotics-bearing formations of the CWC and Peri-Klippen Zone revealed considerable amounts of complex-zoned tourmalines

that could have been derived from decompressed (exhumed) HP/LT ultramafic rocks of the Meliata ophiolite-bearing complexes (Bellová et al. 2018; Plašienka et al. 2019; Aubrecht et al. 2020a, b). The complex-zoned “mosaic” tourmalines with obvious bosiite trend (Bačík et al. 2008; Aubrecht et al. 2020b) have not been observed in our sampled material. Exceptional schorlitic tourmalines derived from Li-poor granitoid rocks (Aubrecht et al. 2020a, their sample “Havranský vrch”) has not been proved in our sampled material as well (sample ZAZ-4, locality no. 10, Fig. 1, 11). However, besides frequent unzoned tourmaline grains, tourmalines of schorlitic-dravitic and/or magnesiofoititic-foititic compositions with quartz intergrowths (Fig. 8), similar to the second type of complex-zoned tourmalines described in Aubrecht et al. (2020a; b), have been observed. Such complex-zoned tourmalines were reported from a wide range of environments including hydrothermal deposits, evaporitic formations, HP-UHP eclogites, HP/LT blueschist facies rocks or other types of medium to high-grade metamorphic rocks, where they formed due to hydrothermal fluid interaction processes either during prograde or, more frequently, retrograde (exhumation) metamorphism (see Aubrecht et al. 2020b and references therein).

Tourmaline grains with magnesiofoititic to foititic inherited cores and schorlitic-dravitic rims represent another peculiar group of zoned tourmalines in the deposits studied (Fig. 8C). Such, quite unusual, magnesiofoititic-foititic tourmalines were described from alpine-type hydrothermal veins cutting the crystalline basement rocks of the Tatric Unit (Uher et al. 2009) or, more frequently, the Gemic Unit (Bačík et al. 2017; 2018). The distribution of the hydrothermal veins has been, however, considered to be limited as an important regional source for the tourmalines in the Upper Cretaceous flysch deposits (Aubrecht et al. 2020a; b). The primary source for the complex zoned tourmalines seems to be the same as for the Cr-spinels, i.e. the Meliata ophiolite-bearing complexes feeding flysch formations deposited in the original Fatric Zliechov Basin. Prevailing subhedral to anhedral tourmaline grains (Fig. 8) did not experience a long transport, which supports their proximal source. After the Turonian emplacement of the Fatric nappe system to its secondary position north of the Tatric edge, the coarse-grained deposits, already as a part of the Klape Unit, could feed ophiolitic detritus as well as complex-zoned tourmalines to the foreland basins in front and atop the developing accretionary wedge (Plašienka 2012; Plašienka & Soták 2015; Plašienka et al. 2019).

#### 4.4. Provenance of garnets

Composition of detrital garnets was studied only at one locality (sample HT-1, locality no. 3, Fig. 1), where almandine-rich garnets prevail. Almandine is the most frequent garnet type observed in Cretaceous to Early Tertiary clastic formations throughout the entire PKB (Salata 2004; Aubrecht et al. 2009; 2020a; 2021; Bellová et al. 2018; Madzin et al. 2019; Bónová et al. 2020). Almandine garnets could originate in a wide range of rocks, mostly in rocks metamorphosed under amphibolite to lower granulite facies conditions (e.g. Méres 2008). Consequently,

the analysed garnets do not provide more specific provenance information and their chemical composition suggests their derivation from common metamorphic or granitoid rocks forming upper levels of the continental crust (cf. Salata 2004; Aubrecht et al. 2021).

#### 4.5. Paleotectonic implications

The tectonic affiliation of the Pupov Fm. has not been unified and there have been published several interpretations (summarized in the Introduction and Geological settings sections). Currently, the turbiditic deposits of the Pupov Fm. are regarded as a part of the Klape Unit (Potfaj et al. 2003; Bezák et al. 2004). The nappe position of the Pupov Fm. was documented by assumed tectonic windows of the Campanian variegated marls of the Gbelany Beds assigned to the Kysuca (Pieniny) succession (Haško 1978). Recently, an alternative interpretation regarding the Campanian Gbelany Beds, outcropping in tight overturned synclines, as an integral constituent of the Pupov Fm. has been proposed (Plašienka et al. 2021). If true, the Pupov Fm. could represent a part of the post-Turonian to late Middle Eocene sequence of the Gosau basin system (Plašienka & Soták 2015; Plašienka et al. 2021).

Predominance of shallow-water carbonate lithoclasts and littoral biodetritus in the calcareous sandstones and fine-grained conglomerates, resting directly on one of the deep-water successions of the PKB at the locality Rovná Hora (Fig. 1-3), infer their deposition near a shallow-marine environment. Since the flysch-sequence of the Pupov Fm. occurs mostly in an overturned position, the transgressive character of the fine-grained conglomerates, suggested by Plašienka et al. (2021), cannot be definitely confirmed. The fine-grained conglomerates might indicate rather a shallowing cycle followed by the deposition of the Maastrichtian shallow-water siltstones of the Polány beds outcropped near Terchová village (Plašienka et al. 2021) (Fig. 1, 2). Conglomerates containing resedimented algal-coral reefs and other shallow-marine material represent a common feature repeatedly occurring in the sedimentary sequences of the overstep Gosau-type basins (Plašienka & Soták 2015; Plašienka 2019 and references therein). The very poor heavy mineral associations observed in the Polány beds, similar to those in the Upper Cretaceous to early Tertiary deposits of the Gosau-type basins (Wagreich & Marschalko 1995; Stern & Wagreich 2013), could also support such an interpretation. The heavy mineral assemblages reported from the exotics-bearing turbiditic Albian to Cenomanian formations of the Klape Unit and Tatric/Fatric Poruba flysch (Bellová et al. 2018; Plašienka et al. 2019; Aubrecht et al. 2020a) are also generally impoverished, although a bit more diverse and locally enriched of unstable blue amphiboles, pyroxenes and kyanites. The primary source for the Cr-spinels and complex zoned tourmalines in both Klape and Gosau Group deposits, seems to be the same Meliata ophiolite-bearing complexes feeding the flysch formations deposited in the original Fatric Zliechov Basin.

The impoverishment of the heavy mineral suite in the Pupov Fm. might be caused by a combination of several factors. 1) Distinct depositional areas of the studied deposits relative to the

exposed source of ophiolitic detritus, i.e. foredeep Fatric Poruba/Klape flysch basin system vs. wedge-top Gosau basin system, 2) Impoverishment due to extensive intrastratal dissolution, which obviously affected also the Albian–Cenomanian flysch complexes of the Klape and related units, where the contrast between a variety of pebble material in exotics-bearing conglomerates and generally impoverished heavy mineral suites in the intercalated sandstones is clearly visible (Bellová et al. 2018; Aubrecht et al. 2020a; 2021).

## 5. CONCLUSIONS

The petrographic and heavy mineral analysis of turbiditic deposits including fine-grained sandstones, fine-grained conglomerates and pebbly mudstones/sandstones of the Coniacian-Maastrichtian/Paleocene Pupov Fm. from the Varín (Kysuce) sector of the PKB were carried out. The sandstones are classified as litho-quartzose to feldspatho-litho-quartzose and occasionally as litho-feldspatho-quartzose. The fine-grained polymict conglomerates (locality Rovná Hora) contain predominantly clasts of shallow-water, less deep-water carbonates, shales, siltstones, quartzitic sandstones and cherts. Besides sedimentary rocks the conglomerates contain abundant clasts of granitoid rocks, mica-schists, gneisses, metasedimentary pelitic rocks and basic volcanic rocks. Presence of shallow-water carbonate lithoclasts and littoral biodetritus in the fine-grained conglomerates infer shallowing sedimentary cycle, a common feature repeatedly occurring in the Gosau-type basin sequences.

Heavy mineral spectra are dominated by ultra-stable tourmaline, zircon and rutile accompanied by Cr-spinel and apatite. Garnets are rare, except one locality where garnets dominate the heavy mineral spectra in the sample from pebbly mudstone/sandstone. The garnets exhibit almandine-rich composition inferring their derivation from common metamorphic or granitoid rocks of upper levels of the continental crust.

Cr-spinels of harzburgitic peridotite composition derived from supra-subduction zones dominate over Cr-spinels of volcanic origin. They were most probably recycled from Albian to Cenomanian polymict conglomerates of the Klape Unit, thus coming originally from Meliata ophiolite-bearing complexes situated in the southern Western Carpathian zones.

Detrital tourmalines represent one of the most frequent heavy mineral in the deposits studied. The presence of subhedral to angular, distinctly zoned tourmalines either of schorlitic-dravitic and magnesiofoititic-foititic compositions with fine intergrowth with quartz, or tourmalines with inherited magnesiofoititic-foititic cores and schorlitic or dravitic rims is an important feature. The primary source for the complex zoned tourmalines seems to be the same as for the Cr-spinels, i.e. the Meliata ophiolite-bearing complexes feeding flysch formations deposited in the original Fatric Zliechov (foredeep Poruba/Klape flysch) Basin. After the Turonian emplacement of the Fatric nappe system beyond the Tatric edge, the exotics-bearing formations of the Klape Unit formed the source of ophiolitic detritus and complex zoned tourmalines in foreland basins of the developing accretionary wedge.

Based on a specific structural position, age, lithological composition of the fine-grained conglomerates and impoverished heavy mineral assemblages in the turbiditic sandstones, the Pupov Fm. could represent a part of the sedimentary sequence of the wedge-top Gosau-type basin system developed in the Carpathian accretionary wedge during meso-Alpine (Coniacian – Eocene) epoch.

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