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The Alps as the main source of sand for the Late Miocene Lake Pannon (Pannonian Basin, Croatia)

Mario Matošević[1]*, Eduardo Garzanti[2], Sanja Šuica[1], Danilo Bersani[3], Frane Marković[4], Ivan Razum[5], Anita Grizelj[6], Krešimir Petrinjak[6], Marijan Kovačić[4] and Davor Pavelić[7]

[1] INA – Industrija nafte d.d., Exploration & Production, Field Development, Exploration & Production Laboratory, Lovinčićeva 4, 10000 Zagreb, Croatia; (*corresponding author: mario.matosevic@ina.hr)
[2] University of Milano-Bicocca, Department of Earth and Environmental Sciences, Laboratory for Provenance Studies, 20126 Milano, Italy
[3] University of Parma, Department of Mathematical, Physical and Computer Science, 43124 Parma, Italy
[4] University of Zagreb, Faculty of Science, Department of Geology, Division of Mineralogy & Petrology, Horvatovac 95, 10000 Zagreb, Croatia
[5] Croatian Natural History Museum, Demetrova 1, 10000 Zagreb, Croatia
[6] Croatian Geological Survey, Department of Geology, Sachsova 2, 10000 Zagreb, Croatia
[7] University of Zagreb, Faculty of Mining, Geology and Petroleum Engineering, Pierottijeva 6, 10000 Zagreb, Croatia

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Abstract

The provenance of the Upper Miocene sandstones from the Sava and Drava depressions of the North Croatian Basin was investigated using petrographic, geochemical, and heavy mineral analyses, including Raman spectroscopy. The study of these sandstones, which represent important oil and gas reservoirs in Croatia, allowed reconstruction of the Late Miocene source-to-sink model of the Lake Pannon drainage system and the evolution of the southwestern Pannonian Basin. The studied feldspatho-litho-quartzose sandstones consist of a mixture of sedimentary, metamorphic, and igneous detritus. Heavy-mineral assemblages are dominated by almandine-rich garnet with apatite, epidote, tourmaline, rutile, zircon, staurolite, and zoisite, indicative of low to medium-grade metamorphic source rocks. Higher concentrations of Ca and Mg than in the Upper Continental Crust standard (UCC) additionally reflect the abundance of limestone and dolostone rock fragments as well as carbonate cement. Geochemical compositional variations between sandstone samples from the Sava and Drava depressions primarily stem from diagenetic processes. CIX and alpha values indicate only minor weathering. Compositional features indicate an orogenic source located in the Eastern Alps and primarily represented by Austroalpine and Penninic nappes. This research offers a novel perspective to distinguish the Upper Miocene reservoirs from other sedimentary units within the basin, contributing to a more comprehensive understanding of the regional geological dynamics and supporting future exploration projects also related to energy transition.

Keywords: Sandstone Reservoirs, Sedimentary Provenance, Heavy Minerals, Geochemical Composition, North Croatian Basin, Late Miocene, Eastern Alps

1. INTRODUCTION

The Upper Miocene sandstones of the North Croatian Basin are major hydrocarbon reservoirs not only in Croatia but also in large parts of the Pannonian Basin. These rocks play a vital role in the energy sector (DOLTON, 2006). Despite decades of exploration, a comprehensive understanding of the sandstones source rocks remains unsatisfactory (e.g., ŠĆAVNIČAR, 1979; MATOŠEVIĆ et al., 2023). This knowledge is crucial for evaluating their potential as reservoir rocks, not only for traditional oil and gas exploration but also for emerging technologies such as carbon capture, utilization, storage, and geothermal energy initiatives (e.g., SNEIDER, 1990; HORVÁTH et al., 2015; MACENIĆ et al., 2020; ALCALDE et al., 2019; TUSCHL et al., 2022).

The composition of the hinterland source rocks influences sedimentary deposits in a basin (BHATIA, 1983; PETTIJOHN et al., 1987; GARZANTI, 2016, 2019), shaping sediment behaviour during burial diagenesis and impacting petrophysical parameters including porosity and permeability. Provenance analysis is integral to assessing reservoir development and quality (WORDEN et al., 1997, 2018; WORDEN & BURLEY, 2003; LAWAN et al., 2021). Notably, the Upper Miocene sandstones in the North Croatian Basin exhibit porosity values from 10% to over 30% and permeability values from 0.01 to 1000 mD (VRBANAC et al., 2010; NOVAK ZELENIKA et al., 2010; MALVIĆ & VELIĆ, 2011; VELIĆ et al., 2012; KOLENKOVIĆ MOČILAC et al., 2022).

This study focuses on the Upper Miocene sandstones derived from the two largest depressions within the North Croatian Basin – the Sava and Drava depressions (PAVELIĆ & KOVAČIĆ, 2018). The research delves into the geological history of the vast expanse of Lake Pannon, formed by the isolation of the Central Paratethys from the Late Miocene to the Early Pliocene (STEININGER & RÖGL, 1979; BÁLDI, 1980; RÖGL & STEININGER, 1983; RÖGL, 1998; MAGYAR et al., 1999; HARZHAUSER et al., 2007; PILLER et al., 2007; HARZHAUSER & MANDIC, 2008; KOVÁČ et al., 2017, 2018; MAGYAR, 2021).

The Miocene epoch in the investigated region, linked with the dynamic history of the Central Paratethys, witnessed the formation of the Pannonian Basin in the Early Miocene due to the subduction of the Eurasian plate beneath the African plate, leading to thermal perturbations, crustal weakening, and the basin's formation as a back-arc sedimentary basin (ROYDEN, 1988; KOVÁČ et al., 1998; PAVELIĆ, 2001; MATENCO & RADIVOJEVIĆ, 2012; HORVÁTH et al., 2015; PAVELIĆ & KOVAČIĆ, 2018). The subsequent evolution witnessed climate shifts, tectonic activity, and the formation of Lake Pannon, where the Upper Miocene sandstones were deposited by cyclic turbiditic currents fed by the progradation of delta systems (BASCH et al., 1995; MAGYAR et al., 1999, 2013; IVKOVIĆ et al., 2000; SAFTIĆ et al., 2003; KOVAČIĆ et al., 2004; KOVAČIĆ & GRIZELJ, 2006; VRBANAC et al., 2010; MALVIĆ & VELIĆ, 2011; SZTANÓ et al., 2013, 2015; BALÁZS, 2017; BALÁZS et al., 2018; SEBE et al., 2020; ANĐELKOVIĆ & RADIVOJEVIĆ, 2021; ŠPELIĆ et al., 2023). The primary contributor of detrital material to the Pannonian Basin can be traced back to the Alpine-Carpathian source region (e.g., KUHLEMANN et al., 2002). This is particularly evident in the ALCAPA tectonic mega-unit within the Eastern Alps and Western Carpathians – the region mostly comprising Mesozoic carbonates, Proterozoic to Palaeozoic low to medium-grade metamorphic rocks, and Palaeozoic granitoids (e.g., ASCH, 2003; SCHMID et al., 2008, 2020; MATOŠEVIĆ et al., 2023). Predominant transport trajectories align with W/NW to E/SE patterns in the Croatian and Hungarian sectors of the basin (IVKOVIĆ et al., 2000; SAFTIĆ et al., 2003; KOVAČIĆ et al., 2004; KOVAČIĆ & GRIZELJ, 2006; MAGYAR et al., 2013; SEBE et al., 2020). However, in the Serbian sector (SE tip of the Pannonian Basin) a smaller opposite SE to NW source-to-sink system from the Southern Carpathians was identified (RADIVOJEVIĆ et al., 2022).

Selected publications addressing diverse sediments in the Pannonian Basin and adjacent basins include KOVAČIĆ (2004), GRIZELJ et al. (2011, 2017), as well as more recent studies by ARATÓ et al. (2021), AMOROSI et al. (2022), and MATOŠEVIĆ et al. (2023), offer valuable insight for comparative analysis. This facilitates assessment of the provenance of the Upper Miocene sandstones and aids understanding of the geological dynamics involved in the North Croatian Basin and its broader regional context during the Late Miocene.

This study thoroughly examines the petrographic, heavy-mineral, and geochemical composition of the Upper Miocene sandstones to reveal the nature of their source rocks, assesses the degree of weathering, traces detrital pathways, unravels mixing processes, and aids comprehension of the final mechanisms of deposition in the basin. These results will enhance future exploration and correlation of the Upper Miocene sandstones in the North Croatian Basin and the wider area of the Pannonian Basin and help in modeling and calibration of compositional data, incorporating methodologies including well-log and seismic interpretation.

2. GEOLOGICAL BACKGROUND

The North Croatian Basin is an elongated extensional basin covering ~32,000 km^2 in northern Croatia and belonging to the Pannonian Basin, surrounded by the Alps, Carpathians, and Dinaride mountains (Fig. 1A). The North Croatian Basin is characterized by various depressions and sub-depressions, with the Sava depression in the south and the Drava depression in the north being the largest (PAVELIĆ & KOVAČIĆ, 2018; Fig. 1B). The basin is filled by Lower to Upper Miocene age strata, deposited in a range of sedimentary environments, including marine, brackish, and freshwater (PAVELIĆ, 2001; LUČIĆ et al., 2001; SAFTIĆ et al., 2003; PAVELIĆ & KOVAČIĆ, 2018), resting unconformably on tectonized Palaeozoic to Palaeogene units (e.g., PAMIĆ, 1986, 1999; ŠUICA et al., 2022a, b).

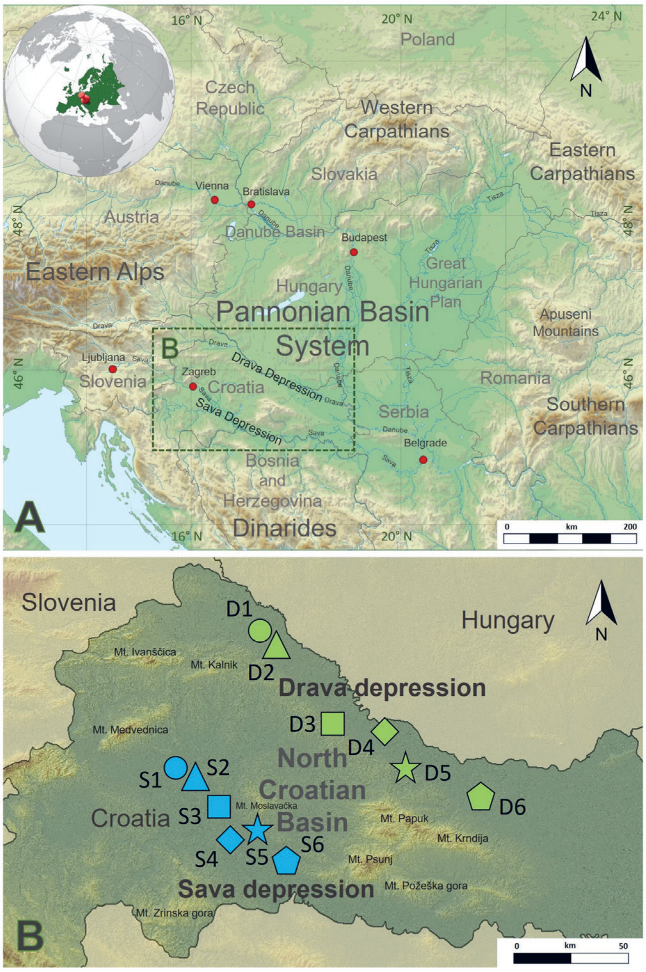

Figure 1. A – Geographical overview of the Pannonian Basin within the Alpine, Carpathian, and Dinaride mountain ranges in Central Europe. B – Spatial representation of the North Croatian Basin, encompassing the Sava and Drava depressions, indicating the locations of the Upper Miocene reservoir sandstones extracted from exploration wells.

The Miocene epoch in the Pannonian Basin is closely associated with the dynamic history of Central Parathetys, which repeatedly connected and disconnected with the open ocean until it was definitively isolated at ~11.6 Ma (STEININGER & RÖGL, 1979; BÁLDI, 1980; RÖGL & STEININGER, 1983; RÖGL, 1998; MAGYAR et al., 1999; HARZHAUSER et al., 2007; PILLER et al., 2007; HARZHAUSER & MANDIC, 2008; TER BORGH et al., 2013; KOVÁČ et al., 2017, 2018; MAGYAR, 2021).

The Pannonian Basin formed in the Early Miocene in connection with the subduction of the Eurasian plate beneath the African (Apulian) plate, involving several continental fragments and leading to thermal perturbations in the upper mantle, weakening and extension of the crust, and the formation of a back-arc sedimentary basin (ROYDEN, 1988; HORVÁTH, 1993, 1995; KOVÁČ et al., 1998; PAVELIĆ, 2001; MATENCO & RADIVOJEVIĆ, 2012; HORVÁTH et al., 2015; BALÁZS et al., 2016; PAVELIĆ & KOVAČIĆ, 2018).

In the initial "syn-rift" phase of Pannonian Basin development, crustal thinning and isostatic subsidence led to the transition from continental to marine environments (ROYDEN, 1988; TARI et al., 1992). In the North Croatian Basin, normal listric faulting (PAVELIĆ, 2001) formed half-grabens and these elongated sub-basins served as main depocentres, while the climate changed from semi-arid to humid, volcanism increased, and marine transgressions and regressions were associated with the activity of normal faults (PAVELIĆ, 2001; BIGUNAC, 2022; PAVELIĆ & KOVAČIĆ, 2018; RUKAVINA et al., 2023).

Subsequently, reduced tectonic activity led to lithosphere cooling and subsidence, and the whole Pannonian Basin was isolated from marine influences of the Central Paratethys ("post-rift" phase; ROYDEN, 1988; TARI et al., 1992), leading to the formation of the large and long-lived Lake Pannon (MAGYAR et al., 1999; HARZHAUSER & PILLER, 2007; PILLER et al., 2007; MANDIC et al., 2015). The Upper Miocene sandstones were deposited during this stage as prograding deltas in proximal settings and as turbidites in deeper areas of Lake Pannon (JUHÁSZ, 1994; MAGYAR et al., 1999; IVKOVIĆ et al., 2000; SAFTIĆ et al., 2003; KOVAČIĆ & GRIZELJ, 2006; VRBANAC et al., 2010; MALVIĆ & VELIĆ, 2011; SZTANÓ et al., 2013, 2015; BALÁZS et al., 2018; SEBE et al., 2020; ANĐELKOVIĆ & RADIVOJEVIĆ, 2021; ŠPELIĆ et al., 2023).

In the Late Miocene, the North Croatian Basin (as for the whole Pannonian Basin) experienced tectonic quiescence and thermal subsidence, while the surrounding Alpine-Carpathian-Dinaric fold belt underwent uplift and erosion. Along with the prevailing humid climate (SZTANÓ et al., 2013; BALÁZS et al., 2018), this resulted in considerable lake depths and deposition of mainly siliciclastic post-rift sediments reaching a thicknesses of several thousand metres in the central part of the depressions (JUHÁSZ, 1994; IVKOVIĆ et al., 2000; SAFTIĆ et al., 2003; KOVAČIĆ & GRIZELJ, 2006; MALVIĆ & VELIĆ, 2011; VELIĆ et al., 2012; SEBE et al., 2020; MATOŠEVIĆ et al., 2023; ŠPELIĆ et al., 2023).

In the Pliocene and Quaternary, the North Croatian Basin underwent structural inversion, strike-slip faulting, counter-clockwise rotation, and uplift of basement blocks leading to the formation of the present-day mountains (JAMIČIĆ, 1995; MÁRTON et al., 1999, 2002; PAVELIĆ, 2001; TOMLJENOVIĆ & CSONTOS, 2001).

3. METHODS

A total of 18 samples were carefully selected from exploration wells drilled by INA (Industrija nafte d.d.). Nine cored sandstone samples from six different exploration wells in the Sava depression and nine cored sandstone samples from six different exploration wells in the Drava depression were obtained to ensure the representativeness of the studied reservoir material (Fig. 1B), consistently choosing thick beds without significant sedimentary structures and considering their stratigraphic association validated by biostratigraphic analyses previously conducted by INA. Well-log correlation and seismic interpretation of key regional horizons also provided essential insight for the samples' selection.

3.1. Petrography and Heavy Minerals

Petrographic analyses of the Upper Miocene sandstones from the Sava and Drava depressions, coupled with scanning electron microscope (SEM) and energy dispersive X-ray spectroscopy (EDS) analyses, were conducted, and documented by MATOŠEVIĆ et al. (2023). Full and detailed results are provided in the Electronic Supplements, accessible online via Suppl. 1. Sandstone classification is after GARZANTI (2016, 2019).

Sandstone samples were crushed into pieces > 5 mm and calcite cement was subsequently dissolved using a 5% acetic acid (CH_3COOH) solution, followed by treatment with 15% H_2O_2 to eliminate organic matter. The processed samples were then sieved, and the more dense grains were separated from the 63-125 µm fraction using sodium polytungstate (SPT; density 2.90 g/cm^3), followed by centrifugation at 2500 rpm for 5 minutes. Over 300 transparent heavy minerals (tHM) were counted on each grain mount by the ribbon method (MANGE & MAURER, 1992). Results are provided in Suppl. 2.

3.2. Garnet Raman Spectroscopy

The Raman spectra of 104 garnet grains from seven sandstone samples from both the Sava and Drava depressions, encompassing various depth intervals within the same exploration well, were acquired using a Bruker Senterra II confocal Raman microscope. A 532 nm laser with 12 mW output power and a x50 LD objective were employed. Spectra spanning the 50-1410 cm^{-1} range were recorded using a high-resolution grating with 1200 lines/mm, with acquisition times between 9 and 15 s. Calibration was performed automatically using SureCAL^{TM} technology. Peak positions were discerned *via* Lorentzian curve fitting facilitated by Opus software. The six peaks characteristic of garnets were harnessed as input data for an updated version of the Matlab Routine MIRAGEM (Micro-Raman Garnets Evaluation Method) to estimate the relative abundance of garnet end members (BERSANI et al., 2009; KARAMPELAS et al., 2023). The new version of the software improves the reliability of the results working on a five end members basis (uvarovite is excluded). The complete dataset is provided in Suppl. 3.

3.3. Geochemistry

Whole rock chemical analyses on all 18 sandstone samples from the Sava and Drava depression were conducted at Bureau Veritas Mineral Laboratories (Vancouver, Canada) by lithium metaborate/tetraborate fusion and nitric acid digestion. Major elements were quantified using inductively-coupled-plasma emission spectroscopy (ICP-ES) and trace elements by inductively-coupled-plasma mass spectrometry (ICP-MS) (for detailed information on adopted analytical protocol, standards, and precisions see https://acmelab.com). Handling and processing of geochemical data were carried out using *GCDkit*

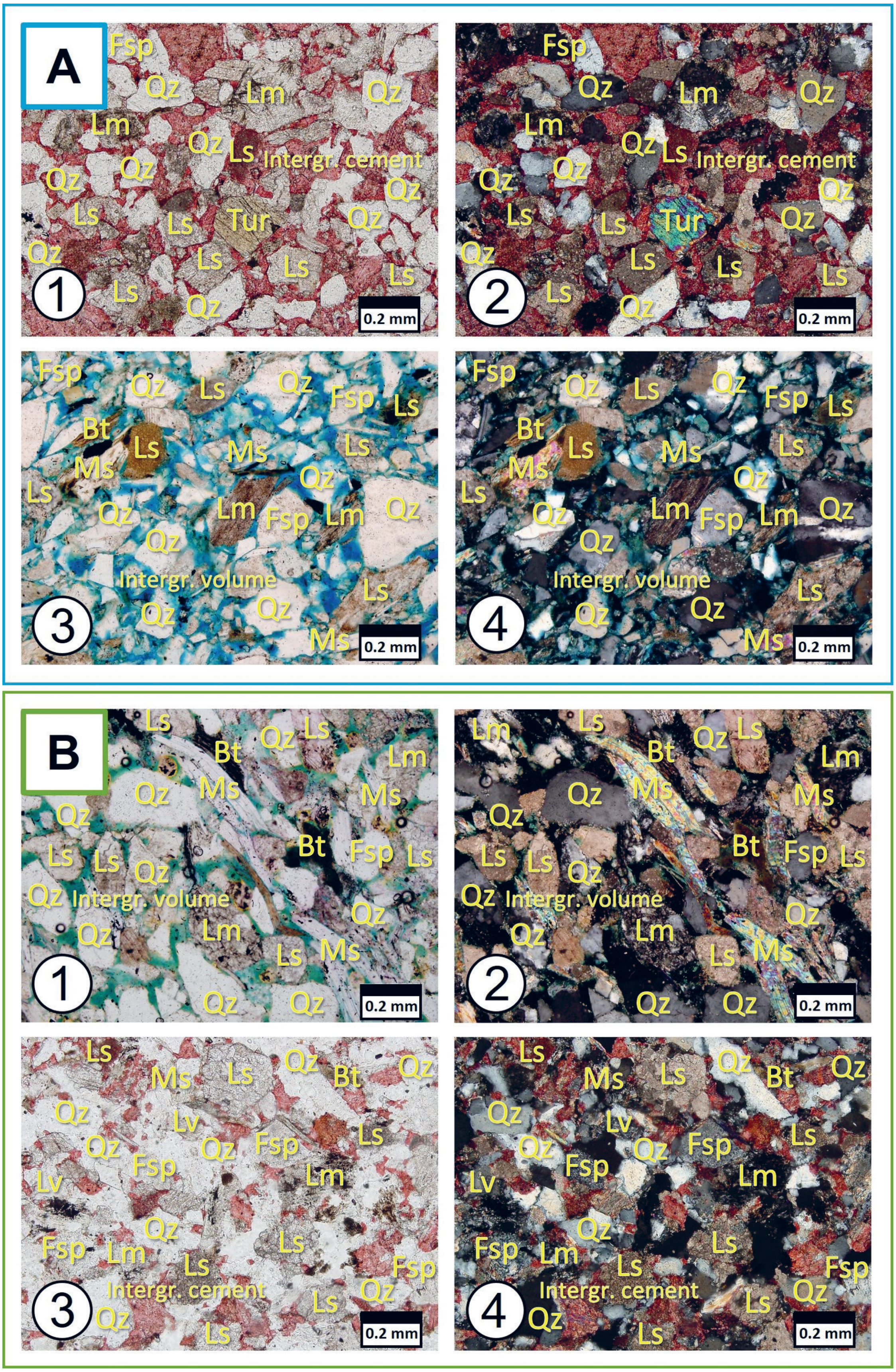

Figure 2. Thin-sections of the Upper Miocene sandstones from the Sava depression (Plate A): 1 – Main sandy grains and intergranular volume filled with carbonate (calcite) cement (S1[1], PPL), 2 – The same as 1, under XPL, 3 – Intergranular volume between sandy grains filled with blue-dyed epoxy showing primary porosity of the sandstone (S4[1], PPL), 4 – The same as 3, under XPL; and thin-sections of the Upper Miocene sandstones from the Drava depression (Plate B): 1 – Main sandy grains and primary intergranular porosity (intergranular volume filled with blue-dyed epoxy) (D2, PPL), 2 – The same as 1, under XPL, 3 – Intergranular volume between sandy grains filled with carbonate (calcite) cement (D6, PPL), 4 – The same as 3, under XPL. Qz – quartz, Fsp – feldspar, Lm – metamorphic rock fragment, Lv – magmatic rock fragment, Ls – sedimentary rock fragment, Ms – muscovite, Bt – biotite, Tur – tourmaline. PPL = plane-polarized light, XPL = cross-polarized light.

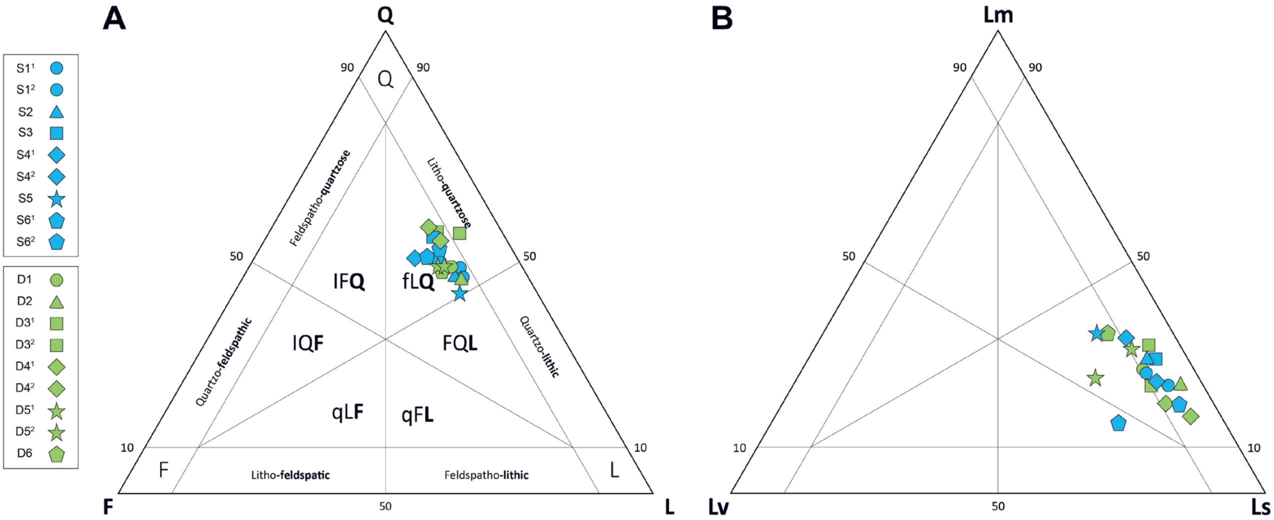

Figure 3. Petrographic classification of the Upper Miocene sandstones from the Sava and Drava depressions presented in the QFL diagram (A) and the LmLvLs diagram (B) according to GARZANTI (2016, 2019).

4.1 (JANOUŠEK et al., 2006) and the R *Provenance* package (VERMEESCH et al., 2016). Chemical data are provided in Suppl. 4.

3.4. Statistical Analyses

Petrographic, heavy-mineral, and geochemical data carry compositional information stored in ratios between components (AITCHISON, 1986; PAWLOWSKY-GLAHN et al., 2015; VERMEESCH, 2018). Sample space for compositional data is called simplex, defined as:

$$s^D = \left\{ x = [x_1, x_2, \ldots x_D] \middle| x_1 > 0, i = 1, 2, \ldots D; \sum_{i=1}^{D} x_i = K \right\}, \quad (1)$$

where K is usually 100. To perform log-ratio transformations and enable data treatment in Euclidian space, we eliminated zero values from the dataset by using Bayesian multiplicative zero replacement (MARTÍN-FERNÁNDEZ et al., 2015). Isometric log-ratio transformation (ILR; EGOZCUE et al., 2003) was used for both heavy-mineral and geochemical data. Heavy-mineral data were transformed through a sequential binary partition (SBP; EGOZCUE & PAWLOWSKY-GLAHN, 2005), by which minerals are continuously split into two groups until all groups consist of one mineral only. Through this procedure, variables (called balances) are constructed as invariant (i.e., they do not change during transport or diagenesis and thus preserve the original source signal; RAZUM et al., 2021, 2023). This was of the utmost importance since the selective dissolution of heavy minerals (HM) is expected and even documented in this particular geological setting (ŠĆAVNIČAR, 1979; MATOŠEVIĆ et al., 2023).

4. RESULTS

4.1. Petrography

The studied sandstones are very fine to fine grained (ranging from 80 to 130 µm in the Sava depression and from 100 to 220 µm in the Drava depression), well to moderately-well sorted, and mainly show tangential grain-to-grain contacts (Suppl. 1; Fig. 2A & B). All samples except one are classified as feldspatho-litho-quartzose carbonaticlastic, with average composition almost identical in the Sava depression (Q50 F14 L36, Lm26 Lv10 Ls64; Suppl. 1; Fig. 3A & B) and in the Drava depression (Q52 F13 L35, Lm26 Lv10 Ls64; Suppl. 1; Fig. 3A & B). Sedimentary grains are virtually all dolostones and limestones (mudstone to packstone or grainstone), with rare chert. Metamorphic rock fragments include mica schist, quartzite, gneiss, slate, and phyllites. Igneous rock fragments include granitoids and altered volcanic glass or tuff. Rip-up clasts also occur.

Porosity is mostly primary intergranular (Fig. 2). The intergranular volume may be partially or completely filled by carbonate cement (calcite and ankerite), micrite, small quartz, mica or feldspar grains, phyllosilicates (including detrital and authigenic clay minerals), or even silica cement in the form of quartz overgrowths.

4.2. Heavy Minerals

Heavy mineral concentration (HMC) ranges from 0.2% to 9.6% (average: 4.8%) in the Sava depression and from 3.6% to 9.1% (average: 5.5.%) in the Drava depression (STab. 2). The very poor to rich tHM suites (0.1 ≤ tHMC ≤ 5) are garnet-dominated (28-79%, average 59% in the Sava depression, 30-84%, average 54% in the Drava depression), with apatite (average 15% in the Sava depression, 27% in the Drava depression), epidote (average 10% in the Sava depression but negligible in the Drava depression), tourmaline (2-10% in the Sava depression, 2-15% in the Drava depression), rutile (1-10% in the Sava depression, 3-12% in the Drava depression), and zircon (2% in the Sava depression, 3% in the Drava depression) (Suppl. 2; Fig. 4). Clinozoisite occurs in the Sava depression (average 2%) but is negligible in the Drava depression. Titanite, chloritoid, staurolite, zoisite, brookite, chromite, kyanite, and anatase were also detected (Suppl. 2; Fig. 4).

4.3. Garnet Raman Spectroscopy

The studied garnets are mostly pyralspites (Suppl. 3). Almandine is the main component in the majority of the analyzed

Figure 4. Heavy mineral association of the Upper Miocene sandstones from the Sava and Drava depressions. A – Anhedral garnet with conchoidal fractures, B-F – Garnet grains showing small to large-scale etch facets due to advanced dissolution, G & H – Apatite grains, I-K – Tourmaline grains, L & M – Rutile grains, N – Zircon, O – Opaque mineral grain, P – Epidote, Q – Clinozoisite, R – Zoisite, S – Chlorite, T – Biotite, U – Titanite, V – Chloritoid, W – Staurolite, X – Chromite, Y – Brookite, Z – Anatase, A' – Kyanite.

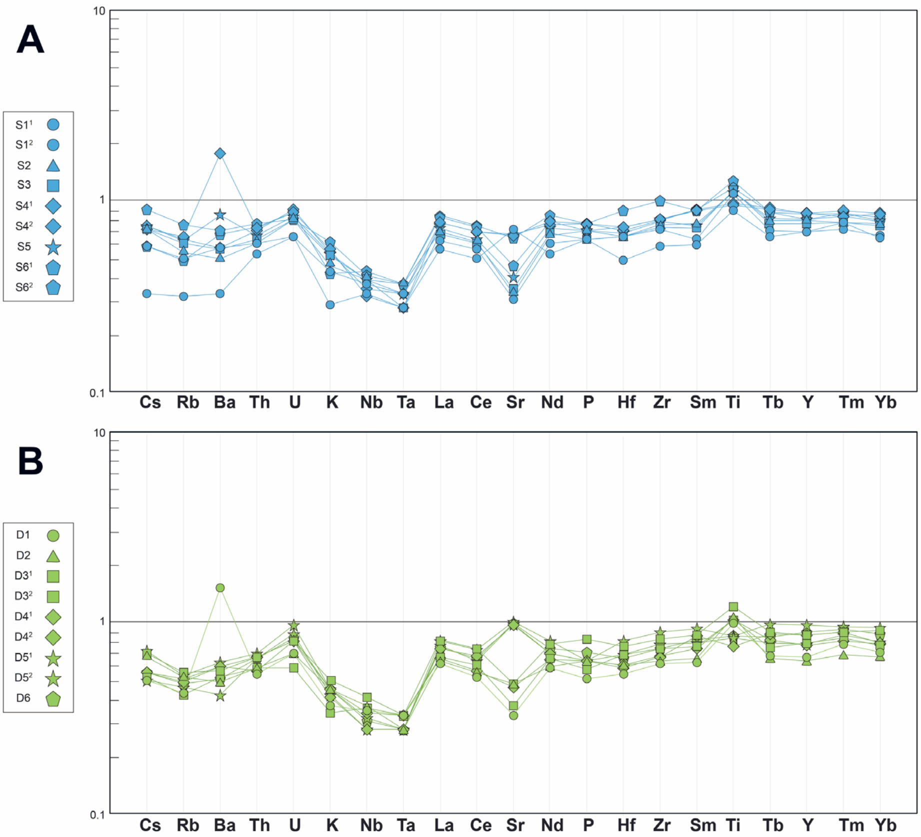

Figure 5. Trace element plots of the Upper Miocene sandstones from the Sava depression (A) and the Drava depression (B), normalized to the Upper Continental Crust (UCC, TYLOR & McLENNAN, 1985).

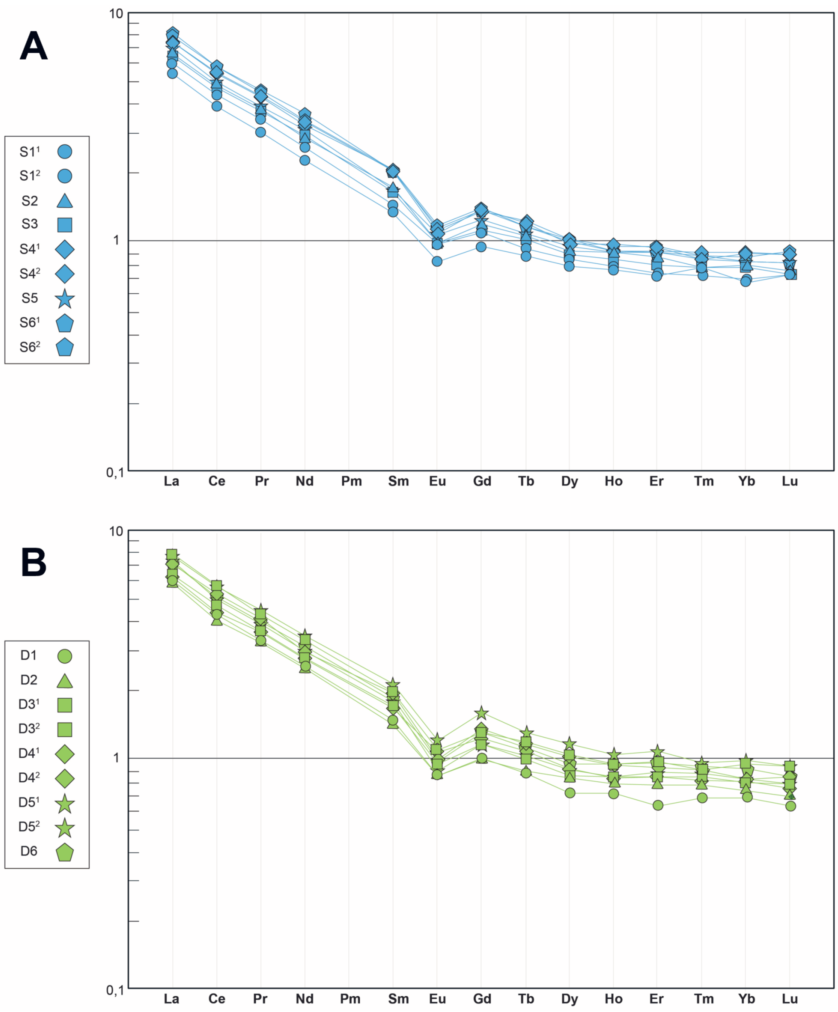

Figure 6. Rare earth element plots of the Upper Miocene sandstones from the Sava (A) and the Drava (B) depressions, normalized to chondrite according to BOYNTON (1984).

garnets, ranging from ~38 to ~96%. Other components include spessartine (<46%), pyrope (<26%), and grossular (<26%). Spessartine garnet, with spessartine 40-62%, almandine <48%, pyrope <26%, and grossular <14%, is present in most samples although less abundant (12% in the Sava depression, 4% in the Drava depression).

4.4. Geochemistry

The studied samples contain 41-63% SiO_2, 5.7% to 12% Al_2O_3 (generally lower in the Drava depression), and 7.6% to 22% CaO (generally higher in the Drava depression) (Suppl. 4). Al_2O_3 correlates best with Fe_2O_3 (r 0.94) and K_2O (r 0.92), moderately well with Na_2O (r 0.64) and TiO_2 (r 0.62), weakly with SiO_2 (r 0.45), insignificantly with MgO (r 0.20), and negatively with CaO (r -0.75) (STab. 4). In the Drava depression samples, MgO correlates positively with Fe_2O_3, Al_2O_3, and TiO_2, but negatively with CaO. Mg and Ca are enriched, and Na, K, Al, and Fe depleted relative to the Upper Continental Crust standard (UCC; TAYLOR & MCLENNAN, 1985). The lack of correlation between Ba and K_2O and very high Ba content in some samples were observed (Fig. 5). Rare earth element (REE) patterns normalized to chondrite (BOYNTON, 1984; Fig. 6) show light REE enrichment (La_{cn}/Yb_{cn} 8.1-9.6 for the Sava depression and 7.7-8.9 for the Drava depression) and a distinct negative Eu anomaly (Eu/Eu* 0.64-0.78 for the Sava depression and 0.62-0.74 for the Drava depression; Suppl. 4).

In the case of sandstones with very high carbonate content, the CIA index (NESBITT & YOUNG, 1982) widely used to assess weathering effects in sediments, requires a very large correction for calcium not hosted in silicates. We thus preferred to use the CIX index, a simple modification of the CIA that excludes CaO in the calculation (GARZANTI et al., 2014). Virtually identical CIX values characterize both the Sava and Drava depression sandstones (73-77).

The effect of weathering is however far better detangled from other controls if mobile elements (Mg, Ca, Na, K, Tb, Sr, and Ba) are considered one by one. This is done by using alpha indices ($\alpha^{Al}E$ values) – defined as $(Al/E)_{sample} / (Al/E)_{standard}$ (GARZANTI et al., 2013) –, which compare the concentration of any mobile element E with reference to non-mobile Al in our samples versus an appropriately selected standard composition (e.g., UCC). Aluminium, hosted in a wide range of rock-forming minerals with diverse density, shape, and size, including phyllosilicates (concentrated in mud) and feldspars (concentrated in sand), is used as a reference for all elements. Alpha indices are very low for Ca and Mg and low for Sr, whereas they range between 1 for Rb and 1.5 for Na.

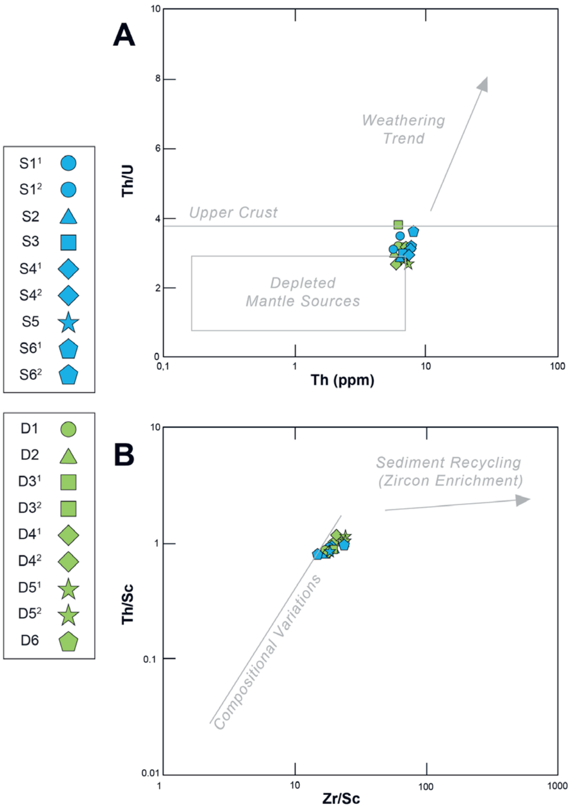

Figure 7. A – Plot of Th/U versus Th indicating insignificant intensity of weathering for the Upper Miocene sandstones from the Sava and Drava depressions, with values similar to the upper crust, according to McLENNAN et al. (1993). B – Plot of Th/Sc versus Zr/Sc indicating negligible influence from sedimentary sorting or recycling for the Upper Miocene sandstones from the Sava and Drava depressions according to McLENNAN et al. (1993).

The Th/U ratio for both depressions ranges from 2.7 to 3.8 (MCLENNAN et al., 1993; Fig. 7A) and the Zr/Sc ratio from 14.9 to 24.1 (MCLENNAN et al., 1993; Fig. 7B).

5. PROVENANCE ANALYSIS

The studied Upper Miocene sandstones from the Sava depression and the Drava depression are mostly feldspatho-litho-quartzose carbonaticlastic indicating provenance from cover strata with contribution from metamorphic and igneous rocks (Fig. 2 & Fig. 3). The virtually identical composition of the Sava and Drava depression sandstones indicates they are part of the same sedimentary system with a common dispersal path from source to sink, typical of a recycled orogen (DICKINSON, 1985; Fig. 8). MATOŠEVIĆ et al. (2023) envisaged a provenance from Mesozoic carbonates, Proterozoic to Palaeocene low to medium-grade metamorphic rocks, and Palaeozoic granitoids of the Eastern Alps and Southern Alps, possibly with minor additional detritus from the Western Carpathians, but excluding significant supply from the Dacia tectonic mega-unit of the Eastern and Southern Carpathians or from the Dinarides, although the latter is geographically closer to the North Croatian Basin than the Eastern Alps. This inference is supported by dominant palaeoflow directions from W/NW, as widely indicated by measurements at the outcrops and by the interpretation of seismic profiles in both the Sava depression and the Drava depression (IVKOVIĆ et al., 2000; SAFTIĆ et al., 2003; KOVAČIĆ et al., 2004; KOVAČIĆ & GRIZELJ, 2006; SEBE et al., 2020; ŠPELIĆ et al., 2023).

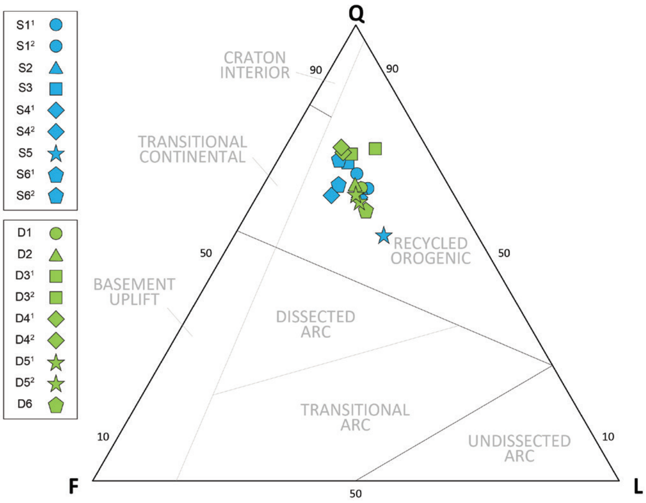

Figure 8. Tectonic setting discrimination diagram of the Upper Miocene sandstones from the Sava and Drava depressions according to DICKINSON (1985).

Provenance from metamorphic units of the Eastern Alps is supported by tHM suites dominated by mostly almandine-rich garnet and including chloritoid, biotite, kyanite, epidote, staurolite, and zoisite (Suppl. 2; Fig. 4). Such an assemblage closely resembles that found in modern river sediments sourced from metamorphic rocks of the Eastern Alps, dominated by the ALCAPA (Adria-derived) tectonic mega-unit with the Austroalpine and Peninic nappes (e.g., Drava and Mura; SCHMID et al., 2008, 2020; GARZANTI et al., 2010; ARATÓ et al., 2021; MATOŠEVIĆ et al., 2023). The differential source influence can be attributed mostly to the Upper and Lower Austroalpine basement, as well as the Upper Austroalpine cover (c.f., SCHUSTER et al., 2013; BOUSQUET et al., 2012; MENCIN GALE et al., 2019a, 2019b; HAUKE et al., 2019; JANÁK et al., 2004). Though the moderately high ZTR index (15 ± 8) might indicate significant recycling from older sandstones (HUBERT, 1962; GARZANTI, 2017), in this case, it is primarily attributed to the substantial diagenetic dissolution of unstable HM in the subsurface (Fig. 9). Statistical discrimination of the most prevalent HM in the sandstones from both depressions indicates their shared source (Fig. 9). The almost complete absence of chemically labile ferromagnesian minerals indicates that tHM assemblages were significantly affected by selective intrastratal dissolution, more extensive in the Drava depression, and thus do not represent the original mineral suite. For this reason, we focused on garnet, one mineral that proves to be relatively resistant during burial diagenesis (MORTON & HALLSWORTH, 2007; GARZANTI et al., 2018). According to the classification of MANGE & MORTON (2007), garnet grains in the Upper Miocene sandstones are mostly of Type B (typical of amphibolite-facies metasedimentary rocks and granitoids), with a minority of Type C (found in high-grade metamafic rocks and quartz-biotite gneisses) and Type A minerals (found in granulite-facies metasediments). This is consistent with the garnet variation diagram by AUBRECHT et al. (2009)

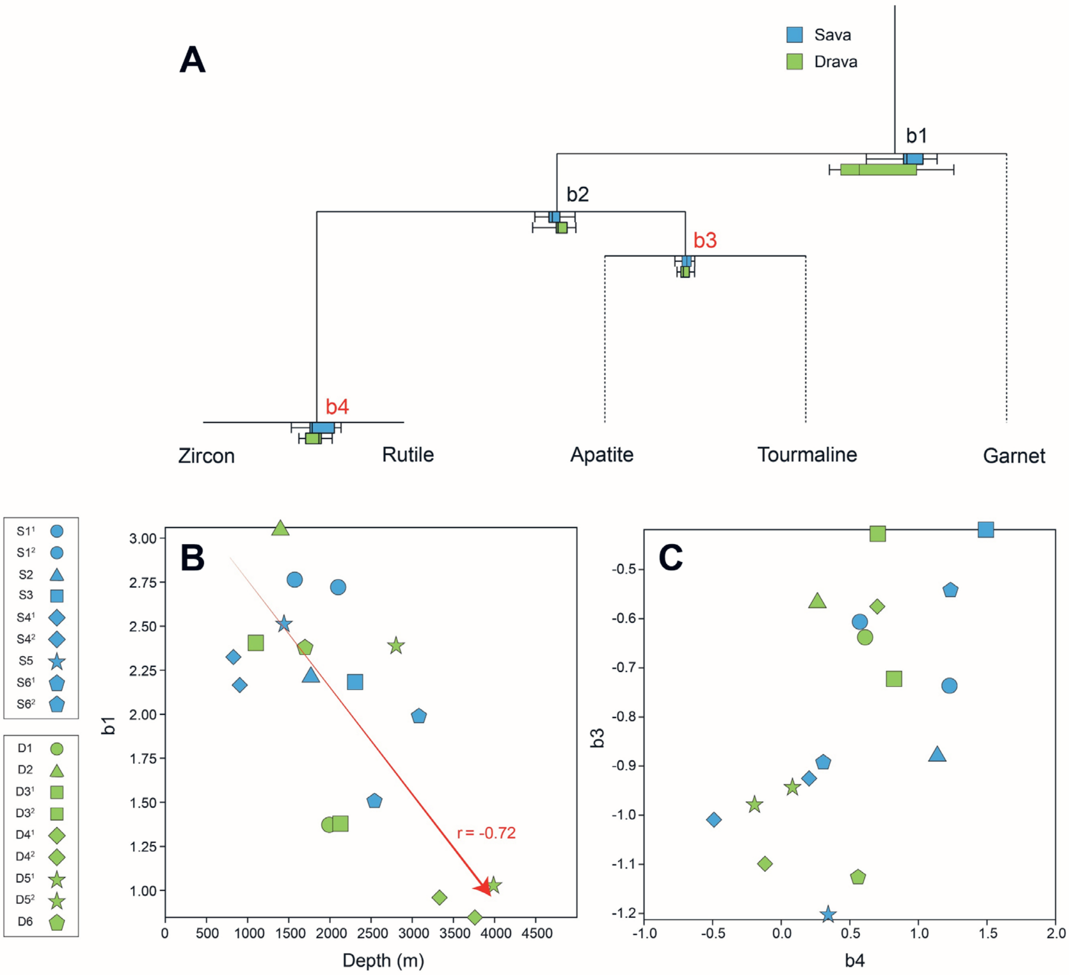

Figure 9. Discrimination of heavy mineral assemblages in the Upper Miocene sandstones between the Sava and Drava depressions. A – Balance dendrogram illustrating modeled balances (b1 = log ratio of garnet over other heavy minerals, sensitive to selective dissolution; b2 = log ratio of apatite and tourmaline over zircon and rutile, ultra-stable in burial diagenesis but sensitive to selective hydraulic sorting; b3 = log ratio of tourmaline over apatite; b4 = log ratio of rutile over zircon; both b3 and b4 include ratios of minerals with the same properties, making them transport- and dissolution-invariant and thus perfect provenance signals). B – Balance b1 is decreasing with depth, indicating instability (selective dissolution) of garnet over other heavy minerals during burial diagenesis. C – Scatterplot of balances b3 and b4, indicating no differences in heavy mineral composition between the Sava and Drava depressions, indicating the same provenance.

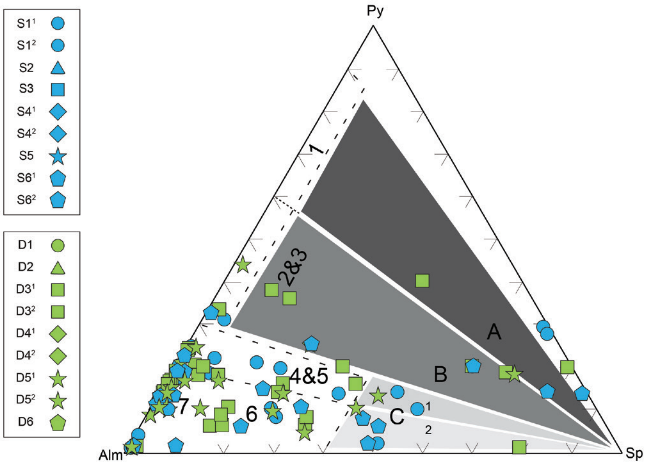

Figure 10. Composition of garnets in the Upper Miocene sandstones from the Sava and Drava depressions, illustrated in the classification diagram "pyrope (Py)-almandine (Alm)-spessartine (Sp)" according to Aubrecht (2009). A – Garnets from HP/UHP conditions, B – Garnets from granulite and eclogite facies conditions, C1 – Garnets from high amphibolite to granulite facies conditions, C2 – Garnets from amphibolite facies conditions, No. 1-7: source rocks of the individual garnets (refer to the original paper for details).

revealing most garnets align within amphibolite-facies conditions (Group 4, 5, 6, and 7; Fig. 10).

The geochemical composition of the studied sandstones reflects the mineralogy of both detrital and authigenic components (e.g., enrichment in CaO and MgO is an effect of the abundance of both detrital and diagenetic calcite, dolomite, and ankerite; Fig. 11). Because of extensive carbonate cement, Ca and Mg are enriched, and Na, K, Al, and Fe depleted relative to the Upper Continental Crust standard (UCC; TAYLOR & MCLENNAN, 1985). Calcite cement is more common in deeper-water sediments especially in the Drava depression, although fully cemented sandstones also occur in shallow-water deposits (MATOŠEVIĆ et al., 2023; Fig. 2). For this reason, limited provenance information can be obtained from geochemical data. The lack of correlation between Ba and K_2O, coupled with a very high Ba content in some samples (Fig. 5), reflects the presence of barite identified by SEM-EDS analyses (MATOŠEVIĆ et al., 2023). A comparison of the geochemical signature of the Upper Miocene sandstones in the North Croatian Basin with sediments from neighboring basins in the Po-Adriatic region, particularly focused on elements

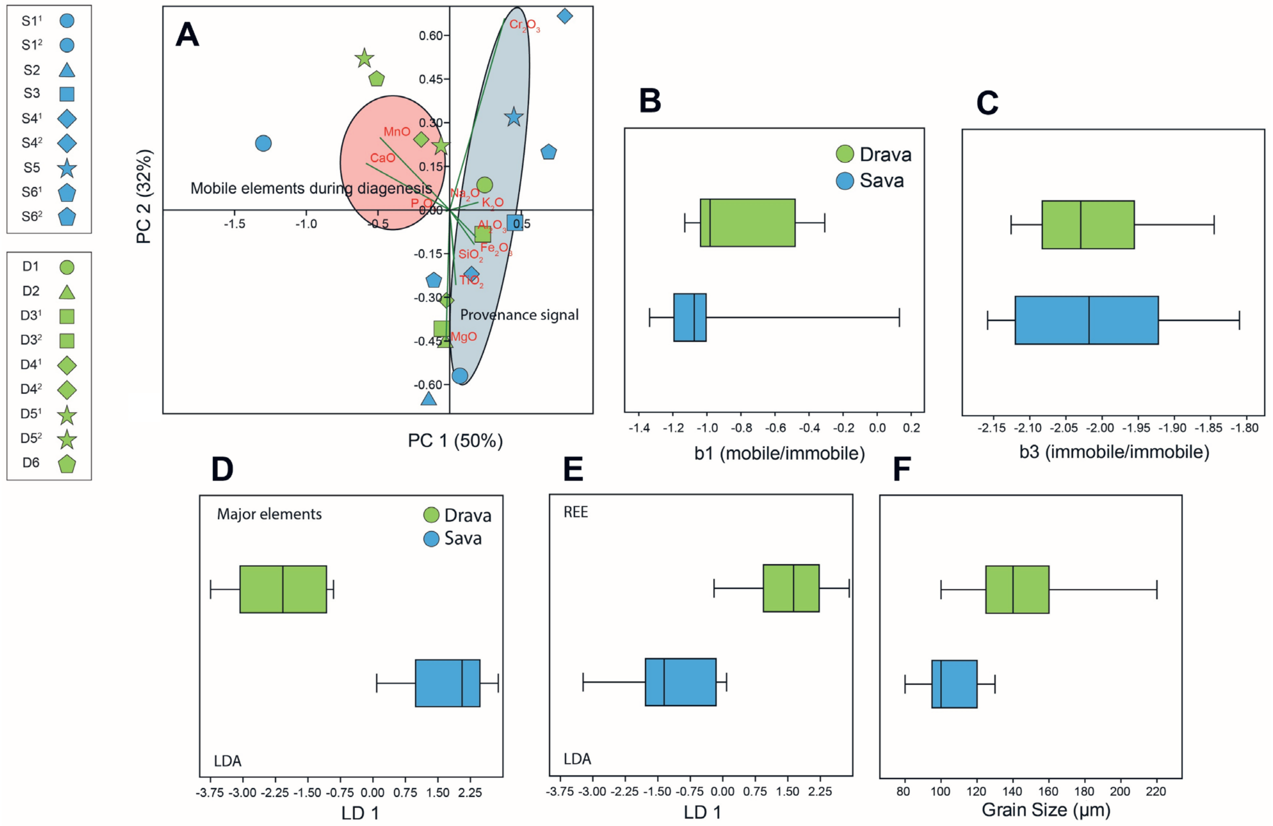

Figure 11. Discrimination of geochemical composition in the Upper Miocene sandstones between the Sava and Drava depressions. A – Compositional biplot explaining over 80% of total data variation, with 50% by the first principal component (PC1). Variable loadings indicate that diagenesis is the key factor in the dataset's variation, with CaO and MgO showing negative loadings, while other major elements exhibit positive loadings on PC1. B – Ratio of mobile to immobile elements (b1 = ratio of MnO-CaO/Al_2O_3-TiO_2) indicating that the geochemical difference is a result of diagenetic processes. C – Box plot of balances indicating that, when considering only immobile elements (associated with the detrital component, not the cement), there is no difference between the Sava and Drava depressions (b3 = TiO_2/Al_2O_3). D – Linear discrimination based on major oxides. E – Linear discrimination based on rare earth elements. D and E clearly separate the Sava and Drava depressions. However, as previously concluded, this separation results from diagenesis and/or sediment dispersal (i.e., sorting), as indicated by the grain size presented in F.

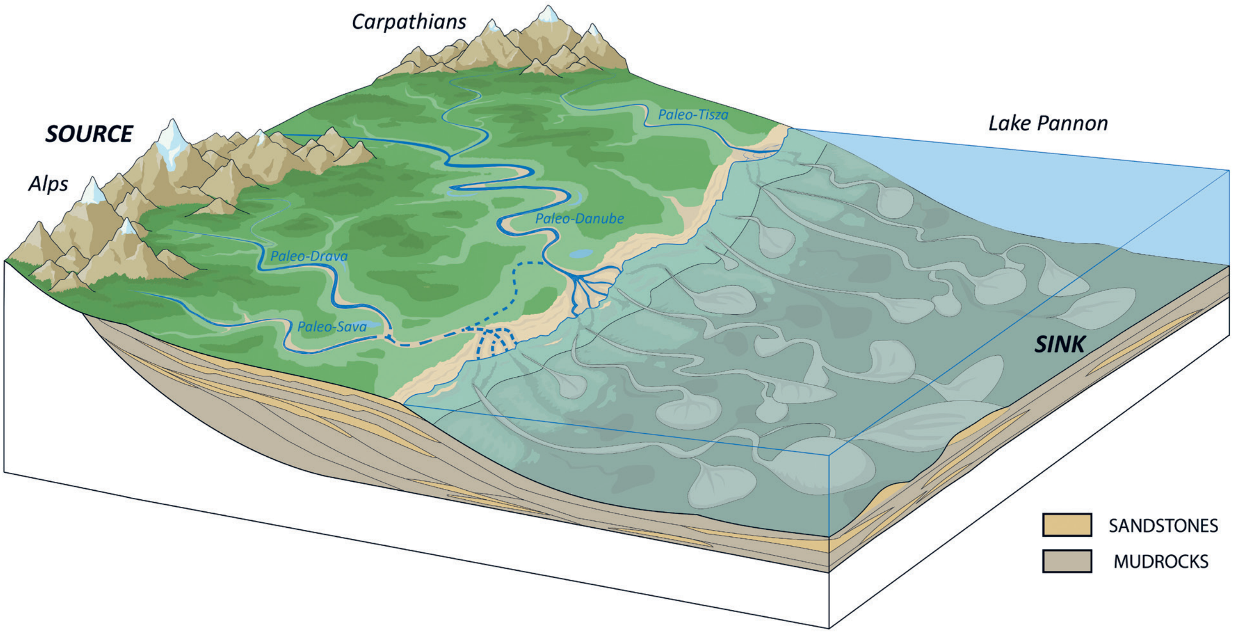

Figure 12. Simplified schematic source-to-sink model of the Upper Miocene sandstones from the North Croatian Basin. This model shows a broader view of the Late Miocene Lake Pannon region. The sediments from the Sava and Drava depressions exhibit virtually identical compositions, suggesting a common source in the Alps during the Late Miocene. Rapid subsidence within the Pannonian Basin facilitated substantial fluvial sediment transfer, potentially via the Palaeo-Sava and/or Palaeo-Drava River systems, from mountains to the lake. However, the connection with the Palaeo-Danube remains uncertain. The model is based on insights gleaned from previous studies (e.g., MAGYAR et al., 2013; SZTANÓ et al., 2013; PAVELIĆ & KOVAČIĆ, 2018; SEBE et al., 2020; MATOŠEVIĆ et al., 2023; ŠPELIĆ et al., 2023) and the findings of this research. It is important to note that the model represents a simplified overview in the Late Miocene, and possible islands and underwater elevations in the lake are not outlined.

such as Ni, Cr, or V diagnostic of mafic-ultramafic source rocks (e.g., MCLENNAN et al., 1993; AMOROSI et al., 2022), indicates a close affinity with river sediments derived from the Eastern Alps rather than the Dinarides or the Carpathians. The similar CIX and alpha values suggest only minor weathering effects. Very low alpha indices for Ca and Mg and low values for Sr reflect the abundance of carbonates, whereas the range between 1 for Rb and 1.5 for Na indicates a very low weathering intensity. Furthermore, the Th/U ratio analysis also dismisses the presence of significant weathering processes (MCLENNAN et al., 1993; Fig. 7A). Similarly, the Zr/Sc ratio indicates the inconsequential impact of sedimentary sorting or recycling (MCLENNAN et al., 1993; Fig. 7B), aligning with findings from prior research by MATOŠEVIĆ et al. (2023).

During the Late Miocene, the Eastern Alps underwent significant uplift concurrent with the rapid subsidence of the Pannonian Basin, thus creating the conditions for massive fluvial sediment transfer from the mountains to the basin (Fig. 12). Previous investigations in the North Croatian Basin are consistent with this scenario (ŠĆAVNIČAR, 1979; IVKOVIĆ et al., 2000; SAFTIĆ et al., 2003; KOVAČIĆ, 2004; KOVAČIĆ et al., 2004, 2011; KOVAČIĆ & GRIZELJ, 2006; GRIZELJ et al., 2007, 2017; PAVELIĆ & KOVAČIĆ, 2018; ŠPELIĆ et al., 2023). The virtually identical detrital signatures in the Sava depression and the Drava depression indicate that the two depressions were connected within Lake Pannon during the Late Miocene, being part of the same dispersal system, possibly represented by the Palaeo-Sava and/or Palaeo-Drava rivers (Fig. 12). The connection with the Palaeo-Danube remains uncertain (MAGYAR et al., 2013; SEBE et al., 2020) and in need of further provenance studies from the Hungarian part of the Pannonian Basin.

6. CONCLUSIONS

The Upper Miocene feldspatho-litho-quartzose carbonaticlastic sandstones from the Sava and Drava depressions, that belong to the North Croatian Basin, include limestone, dolostone, mica schist, quartzite, gneiss, phyllite, and granitoid rock fragments. Generally poor to moderately poorly preserved, transparent heavy-mineral assemblages are dominated by almandine-rich garnet, associated with epidote, staurolite, and zoisite sourced from low to medium-grade metamorphic source rocks. The low tHMC index with a lack of ferromagnesian minerals and moderate amounts of durable zircon, tourmaline, and rutile, indicate the significant effects of selective diagenetic dissolution and of the addition of detritus recycled from older siliciclastic deposits. Integrated petrographic, heavy-mineral, garnet Raman spectroscopic, and geochemical signatures concur to indicate a major provenance from the Eastern Alps orogenic belt including the sedimentary, metamorphic, and igneous rocks of the Austroalpine and Penninic nappes. The Late Miocene uplift of the Alps and subsidence of the Pannonian Basin created the conditions that led to massive fluvial sediment transfer from the orogen into Lake Pannon. The strikingly similar detrital signatures observed in both the Sava and Drava depressions suggest their shared origin within the lake's depositional setting, characterized by the same sediment-dispersal system associated possibly with Palaeo-Sava and/or Palaeo-Drava rivers, following mainly the NW to SE transport direction of detritus, which is also characteristic of the progradation system of the Palaeo-Danube but opposite to the system in the SE part of the basin (RADIVOJEVIĆ et al., 2022). These insights, crucial for a better understanding of sedimentary processes and the geological evolution of the Pannonian Basin, offer a novel perspective to distinguish the Upper Miocene reservoirs from other sedimentary units within the basin and provide valuable information for the industry, i.e., future resource exploration and development also related to energy transition and environmental sustainability. Despite the basin's extensive exploration history, the reservoir properties of the sandstones remained relatively unexplored, highlighting the significance of this study - the delineation of mineralogical and geochemical signatures enables improved reservoir characterization, facilitating chemo-stratigraphic assessment and prognosticative modeling in the subsurface.

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