

# Provenance and diagenesis of the Upper Miocene sandstones in the south-western part of the Pannonian basin system, Croatia

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**PROVENANCE AND DIAGENESIS OF  
THE UPPER MIOCENE SANDSTONES IN  
THE SOUTH-WESTERN PART OF THE  
PANNONIAN BASIN SYSTEM, CROATIA**

DOCTORAL DISSERTATION

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Supervisors:

Davor Pavelić, PhD, Tenured Professor  
Marijan Kovačić, PhD, Full Professor

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Sveučilište u Zagrebu

Rudarsko-geološko-naftni fakultet

Mario Matošević

**PROVENIJENCIJA I DIJAGENEZA  
GORNJOMIOCENSKIH PJEŠČENJAKA  
JUGOZAPADNOG DIJELA PANONSKOG  
BAZENSKOG SUSTAVA U HRVATSKOJ**

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*“Civilization exists by geological consent, subject to change without notice.”*

- Will Durant

## ABSTRACT

This study provides a thorough examination of the Upper Miocene sandstones in the North Croatian Basin, critical sandstone reservoirs for oil and gas in Croatia and the wider Pannonian Basin System in Central Europe. Despite their longstanding role in hydrocarbon production and potential for new energy initiatives like carbon capture and geothermal projects, their sedimentary provenance and diagenesis remained largely unexplored. The research focuses on the Upper Miocene sandstones from cores of exploration wells in the Sava and Drava depressions. Various methods, including petrographic, SEM-EDS, geochemical, heavy mineral, XRD, and Raman spectroscopic analyses, alongside petrophysical measurements, were employed to characterize the sediments. Results reveal a carbonaticlastic feldspatho-litho-quartzose (fLQ) composition, with carbonate, metamorphic, and plutonic rock fragments, among quartz and feldspar, primarily sourced from the Eastern Alps (ALCAPA tectonic mega-unit), with the predominant Austroalpine nappe. Heavy mineral assemblages are dominated by almandine-rich garnet, with apatite, epidote, tourmaline, rutile, zircon, staurolite, zoisite, and clinozoisite, indicating detrital supply from low to medium-grade metamorphic source rocks. Higher Ca and Mg concentrations than in the Upper Continental Crust (UCC) standard reflect abundant limestone and dolostone rock fragments and carbonate cements. Consistent grain sizes, modal compositions, and diagenetic alterations were observed across both depressions. Mechanical and chemical compaction, with cementation by carbonate minerals (particularly calcite and Fe-dolomite/ankerite), quartz, and clay minerals (including illite, chlorite, kaolinite, and mixed-layered species), significantly affect reservoir quality and hydrocarbon productivity. Primary intergranular porosity reduction with depth, coupled with secondary porosity, resulting from dissolution processes, and microporosity, influences overall reservoir quality. This research enhances understanding of source-to-sink model of the Late Miocene Lake Pannon drainage system and the evolution of the south-western Pannonian Basin System. Strikingly similar detrital signatures observed in both depressions suggest a shared origin within the lake's depositional setting characterized by the same sediment-dispersal system, possibly linked to Palaeo-Sava and/or Palaeo-Drava rivers. The study distinguishes the Upper Miocene reservoirs from other sedimentary units within the basin, informing about regional geological dynamics and supporting future exploration projects, with implications for hydrocarbon exploration, energy transition, and environmental sustainability efforts.

## PROŠIRENI SAŽETAK

Istraživanje pruža sveobuhvatnu analizu gornjomiocenskih pješčenjaka iz Sjevernohrvatskog bazena, ključnih pješčenjačkih ležišta nafte i plina u Hrvatskoj te širem Panonskog bazenskog sustava unutar Srednje Europe. Unatoč njihovoj dugogodišnjoj ulozi u proizvodnji ugljikovodika i potencijalu za nove energetske inicijative, poput zbrinjavanja ugljikovog dioksida i geotermalnih projekata, njihova provenijencija sedimenata i dijageneza nedovoljno su istraženi. Istraživanje se usredotočuje na gornjomiocenske pješčenjake iz jezgara istražnih bušotina u Savskoj i Dravskoj depresiji. Različite metode, uključujući petrografsku, SEM-EDS, geokemijsku, analizu teških minerala, XRD i Raman spektroskopsku analizu, uz petrofizikalna mjerenja, korištene su za karakterizaciju sedimenata. Rezultati otkrivaju karbonatno-klastični feldspatno-litično-kvarcni (fLQ) sastav s karbonatnim, metamorfnim i plutonskim fragmentima stijena koji, uz kvarc i feldspate, potječu iz područja Istočnih Alpa (tektonske mega-jedinice ALCAPA), uključujući dominantnu austroalpsku navlaku. Asocijacijom teških minerala dominiraju granati obogaćeni almandinskom fazom, uz apatit, epidot, turmalin, rutil, cirkon, staurolit, zoisit i klinozoisit, koji ukazuju na detritalni izvor iz metamorfnih stijena niskog do srednjeg stupnja metamorfizma. Povišene koncentracije Ca i Mg u odnosu na standard gornje kontinentalne kore (*Upper Continental Crust*, UCC) odražavaju obilje fragmenata vapnenaca i dolomita te karbonatne cimente. Uočene su postojane veličine zrna, modalni sastavi i dijagenetske promjene u obje depresije. Mehanička i kemijska kompakcija, uz cementaciju karbonatnim mineralima (posebno kalcitom i Fe-dolomitom/ankeritom), kvarcom i mineralima glina (uključujući ilit, klorit, kaolinit i miješano-slojne vrste), značajno utječu na kvalitetu ležišta i ugljikovodičnu produktivnost. Redukcija primarne međuzrnske poroznosti s dubinom, zajedno sa sekundarnom poroznosti, koja proizlazi iz procesa otapanja, te mikroporoznosti, utječe na ukupnu kvalitetu ležišta. Istraživanje omogućuje poboljšano razumijevanja *source-to-sink* modela jezera Panon iz kasnog miocena i razvoj jugozapadnog dijela Panonskog bazenskog sustava. Izrazito slični detritalni signali uočeni u obje depresije sugeriraju zajedničko podrijetlo pješčenjaka unutar taložnog okoliša jezera, koji je karakteriziran istovjetnim sustavom donosa i disperzije sedimentnog materijala, vjerojatno povezanog s paleo-Savom i/ili paleo-Dravom. Istraživanje omogućuje razlikovanje gornjomiocenskih pješčenjačkih ležišta od drugih sedimentnih jedinica unutar bazena, pružajući podatke o regionalnoj geološkoj dinamici i podržavajući buduće istraživačke projekte, s implikacijama na istraživanje nafte i plina, energetske tranziciju i nastojanja u vezi održivosti okoliša.

## **Ciljevi i hipoteze**

Glavni ciljevi ovog istraživanja bili su utvrditi provenijenciju i dijagenezu gornjomiocenskih pješčenjaka jugozapadnog dijela Panonskog bazenskog sustava, učiniti petrografsku, mineralošku i geokemijsku analizu gornjomiocenskih pješčenjaka iz odabranih istražnih bušotina Savske i Dravske depresije, odrediti detaljan sastav gornjomiocenskih pješčenjaka i njihove parametre od važnosti za karakterizaciju ležišta te usporediti gornjomiocenske pješčenjake Savske i Dravske depresije na temelju dobivenih rezultata analiza.

Ovi su se ciljevi temeljili na tri glavne hipoteze: (i) studija provenijencije sedimenata omogućuje rekonstrukciju i interpretaciju povijesti sedimenata od početne erozije izvorišnih stijena do konačnog zatrpavanja njihovog detritusa, (ii) petrografske analize, analize teških minerala, kemijske analize i analize pojedinačnih zrna zajednički omogućuju opsežno utvrđivanje provenijencije i dijageneze pješčenjaka i (iii) mogu se odrediti razlike u provenijenciji i dijagenezi gornjomiocenskih pješčenjaka iz Savske i Dravske depresije.

## **Znanstveni doprinos**

Rezultati ovog istraživanja omogućili su sveobuhvatno određivanje provenijencije gornjomiocenskih pješčenjaka Savske i Dravske depresije, definiranje sličnosti i različitosti u sastavu gornjomiocenskih pješčenjaka Savske i Dravske depresije, prvo detaljno određivanje dijagenetskih promjena u gornjomiocenskim pješčenjacima Savske i Dravske depresije te procjenu kvalitete gornjomiocenskih pješčenjaka Savske i Dravske depresije kao ležišnih stijena s porastom dubine.

## **Metode i postupci**

U svrhu ispitivanja i određivanja sastava, provenijencije i dijageneze gornjomiocenskih pješčenjaka iz Savske i Dravske depresije provedene su detaljne petrografske, mineraloške i geokemijske analize. Petrografske analize tankih izbrusaka stijena pomoću svjetlosnih polarizacijskih mikroskopa (Olympus BX51 i Leitz Orthoplan) korištene su za određivanje modalnog sastava uzoraka, veličine, zaobljenosti i sortiranosti zrna, ispuna međuzrnskog prostora, vrsta poroznosti te klasifikaciju pješčenjaka i zaključivanje o provenijenciji, tektonskim modelima i dijagenetskim procesima (**Wentworth, 1922; Powers, 1953; Evamy i Shearman, 1962; Krumbein i Sloss, 1963; Dickinson, 1970, 1985; Ingersoll i dr., 1984; Zuffa, 1985; Jerram, 2001; Weltje, 2006; Garzanti, 2016, 2019**). Analize pretražnim/skenirajućim elektronskim mikroskopom (SEM; JEOL JSM-6510 LV SEM) s energetski disperzivnom spektroskopijom X-zraka (EDS; Oxford INCA X-act system) korištene su za detaljno ispitivanje morfoloških karakteristika pješčenjaka, pojedinih



zrna i veziva, uključujući analizu kemijskog i mineraloškog sastava (**Welton, 1984; Severin, 2004**). Analize teških minerala izvršene su pomoću svjetlosnog polarizacijskog mikroskopa (Leitz Wetzlar Orthoplan) za određivanje vrsta izvorišnih stijena, procesa transporta, taloženja i dijageneze detritusa te za usporedbu rezultata s literaturom (**Mange i Maurer, 1992**). Korišten je konfokalni Raman mikroskop (Bruker Senterra II) za dobivanje Raman spektara granata na temelju kojih su određeni relativni udjeli njihovih krajnjih članova pomoću metode *Micro-Raman Garnets Evaluation Method* (MIRAGEM; **Bersani i dr., 2009; Karampelas i dr., 2023**) i interpretirane njihove izvorišne stijene (**Morton i Hallsworth, 2007; Garzanti i dr., 2018; Mange i Morton, 2007; Aubrecht i dr., 2009**). Geokemijske analize pješčenjaka provedene su u Bureau Veritas Mineral Laboratories (Vancouver, Kanada). Kvantifikacija glavnih elemenata, elemenata u tragovima i elemenata rijetkih zemalja pomoću emisijske spektroskopije i masene spektrometrije s induktivno spregnutom plazmom (ICP-ES i ICP-MS) korištena je za interpretaciju sastava, provenijencijskih, tektonskih i dijagenetskih signala pješčenjaka, učinaka trošenja, sortiranja i recikliranja sedimenata te za usporedbu rezultata s literaturom. Rendgenska difrakcija (XRD) na izdvojenim frakcijama iz uzoraka korištena je za određivanje minerala glina i kristaliniteta ilita (**Moore i Reynolds, 1997**). Petrofizikalna mjerenja uključivala su mjerenja poroznosti pomoću helijskog porozimetra na standardiziranim laboratorijskim valjčićima, oslanjajući se na Boyleov zakon (**American Petroleum Institute, 1998**). Statistička obrada rezultata omogućila je interpretaciju dobivenih podataka, trendova i odnosa između rezultata.

### **Rezultati i zaključci**

Gornjomiocenski pješčenjaci iz Savske i Dravske depresije Sjevernohrvatskog bazena pokazuje nekoliko ključnih karakteristika. Zrna unutar pješčenjaka uglavnom su uglasta do poluzaobljena, veličine od 70 do 160  $\mu\text{m}$  u uzorcima iz Savske depresije i od 90 do 220  $\mu\text{m}$  u uzorcima iz Dravske depresije, što ukazuje na dominaciju vrlo sitnozrnastog i sitnozrnastog pijeska. Zrna su općenito dobro do umjereno dobro sortirana, s povremenim slučajevima umjereno do loše sortiranih zrna. Kontakti zrna variraju od točkastih do dugih i konkavno-konveksnih kontakata, prelazeći čak i u suturirane kontakte u pješčenjacima zahvaćenim većom kompakcijom na većim dubinama. Petrografska analiza glavnih zrna otkriva sastav kojim dominira kvarc (Q), feldspati (F) i litični fragmenti (L), s manjim udjelima filosilikata (M) i teških minerala (HM). Prosječni modalni QFL sastav pješčenjaka iz Savske depresije je Q50.3%F15.6%L34.1% i Q51.4%F14.7%L33.9% za uzorke iz Dravske depresije. Prosječni modalni LmLvLs sastav pješčenjaka iz Savske depresije je Lm27.9%Lv10.0%Ls62.1% i Lm28.8%Lv9.7%Ls61.4% za one iz Dravske depresije. Pješčenjaci su klasificirani kao

karbonatno-klastični feldspatno-litično-kvarcni (fLQ) i rjeđe kao litično-kvarcni (LQ) ili feldspatno-kvarcno-litični (fQL) prema **Garzantiju (2016)**. Fragmenti sedimentnih stijena su gotovo isključivo dolomiti i vapnenci, s rijetkom pojavom čerta. Fragmenti metamorfnih stijena uključuju tinjčaste škriljavce, kvarcite, gnajseve, slejtove i filite. Fragmente magmatskih stijena sačinjavaju granitoidi te rijetko izmijenjeno vulkansko staklo ili fragmenti tufa. Također se pojavljuju i fragmenti *rip-up* klasta.

Koncentracija teških minerala kreće se od 0.2 % do 9.6 % u Savskoj depresiji i od 3.6 % do 9.1 % u Dravskoj depresiji, pri čemu je granat dominantan mineral, praćen apatitom, epidotom, turmalinom, rutilom i cirkonom. Granati su pretežno piralspiti, s almandinom kao primarnom fazom. Također su detektirani titanit, kloritoid, staurolit, zoisit, klinozoisit, brookit, kromit, kijanit i anatas.

Međuzrnski volumeni pješčenjaka u obje depresije su neispunjeni ili djelomično/potpuno ispunjeni sitnozrnastim matriksom i/ili cementom. Matriks se sastoji od čestica veličine silta i gline dobivenih razgradnjom i izmjenama glavnih mineralnih zrna tijekom transporta i taloženja. Autigeni cement uglavnom se sastoji od karbonatnih minerala (pretežno kalcita i Fe-dolomita/ankerita) i/ili kombinacije karbonatnih minerala s mineralima glina (uključujući ilit, klorit, kaolinit i miješano slojne vrste), kvarcom i feldspatom.

Pješčenjaci pokazuju raspon sadržaja  $\text{SiO}_2$  između 41 % i 63 %,  $\text{Al}_2\text{O}_3$  između 5.7 % i 12 %, i CaO između 7.6 % i 22 %. Mg i Ca su obogaćeni, a Na, K, Al i Fe osiromašeni u odnosu na standard gornje kontinentalne kore (UCC; **Taylor i McLennan, 1985**). Normalizacija elemenata rijetkih zemalja (REE) prema hondritu (**Boynnton, 1984**) pokazuje obogaćenje lakih REE ( $\text{La}_{\text{cn}}/\text{Yb}_{\text{cn}}$  8.1-9.6 za Savsku depresiju i 7.7-8.9 za Dravsku depresiju) i karakterističnu negativnu anomaliju Eu ( $\text{Eu}/\text{Eu}^*$  0.64-0.78 za Savsku depresiju i 0.62-0.74 za Dravsku depresiju). Generalno, geokemijski sastav pješčenjaka ukazuje na neznatne učinke trošenja i irelevantan utjecaj sortiranja i recikliranja sedimenata.

Primarna međuzrnska poroznost prevladava u svim uzorcima pješčenjaka. Promjeri pora obično variraju od 5-100  $\mu\text{m}$ , s prosječnom veličinom unutar raspona od 10-40  $\mu\text{m}$  u obje depresije. Iako većina pora pokazuje povezanost ili djelomičnu povezanost, uočljive su i nepovezane pore, posebno u uzorcima gdje karbonatni cement uspostavlja veze od zrna do zrna. Sekundarna poroznost, proizašla iz djelomičnog do potpunog otapanja detritalnih zrna i cementa, uglavnom je vidljiva u feldspatima i filosilikatima, sa sporadičnim pojavama unutar fragmenata stijena, a rijetko je povezana i s cementom. Mikroporoznost postoji u vidu sitnih pora (manje od 2  $\mu\text{m}$ ), uglavnom vezanih za matriks ili različito orijentirane detritalne i autigene minerale glina, što doprinosi ukupnoj poroznosti. Smanjenje poroznosti uzrokovano

kompakcijom, što se očituje razvojem kontakata zrna i tlačnim otapanjem, široko je rasprostranjeno, dovodeći do smanjenja poroznosti s dubinom. Mjerenja poroznosti potvrđuju ovaj trend, pri čemu vrijednosti u analiziranim uzorcima opadaju od 34.9 % do 3.0 % u Savskoj depresiji i od 28.7 % do 1.9 % u Dravskoj depresiji.

Gornjomiocenski pješčenjaci iz Savske i Dravske depresije derivirani su iz provenijencijskog područja recikliranog orogena, tj. ekvivalentnog eksternog terena bez značajnih lokalnih utjecaja - izvorne stijene sastoje se od karbonata, metamorfnih stijena i plutonskih stijena, uglavnom istočnoalpskog orogenetskog pojasa (tektonske mega-jedinice ALCAPA) s dominantnom austroalpskom navlakom. Gotovo uniformni sastav pješčenjaka iz obje depresije sugerira vremenski identične procese erozije izvornih stijena, transporta pjeskovitog materijala i taloženja, uglavnom kao relativno brz i masivan donos u bazen tijekom kasnog miocena. Izrazito slični detritalni signali zapaženi u obje depresije sugeriraju njihovo zajedničko podrijetlo unutar taložnog okoliša jezera, koje karakterizira isti sustav disperzije sedimenata povezan moguće s paleo-Savom i/ili paleo-Dravom, prateći uglavnom smjer transporta detritusa od SZ do JI, što je također karakteristično i za progradacijski sustav paleo-Dunava. Rezultati studije pokazuju da nisu postojale značajne morfološke barijere između depresija koje bi mogle ometati donos detritalnog materijala preko rijeka i delta u Panonsko jezero. Niski tHMC indeks bez prisutnosti feromagnezijskih minerala i umjerene količine otpornog cirkona, turmalina i rutila, ukazuje na značajne učinke selektivnog dijagenetskog otapanja unutar bazena.

Ovo istraživanje otkriva tijek dijagenetskih procesa unutar gornjomiocenskih jezerskih pješčenjaka u Sjevernohrvatskom bazenu, pružajući uvid u njihove ležišne karakteristike i novi energetski potencijal. Uniformni dijagenetski procesi utvrđeni su u obje depresije, pri čemu je karbonatna cementacija glavni uzrok smanjenja poroznosti, uz kompakciju s dubinom. Promatrani dijagenetski paragenetski procesi otkrivaju složeni odnos mineraloških transformacija i petrofizikalnih svojstava ležišta. Minerali glina, kako detritalni tako i autigeni, kompleksno djeluju s drugim dijagenetskim procesima, dodatno utječući na poroznost i propusnost. Autigeni minerali glina pojavljuju se na određenim dubinama, djelujući kao cementi koji ispunjavaju pore ili obavijaju mineralna zrna, te dodatno inhibiraju protok fluida. Ove spoznaje ističu složenost dijagenetskih interakcija i njihovih implikacija za kvalitetu i potencijalnu produktivnost ležišta. Razumijevanje ovih procesa ključno je za predviđanje kvalitete ležišta i putova migracije fluida pa studija pruža vrijedne uvide za održivi razvoj resursa u regiji.

Ovi uvidi, ključni za bolje razumijevanje sedimentnih procesa i geološke evolucije Panonskog bazenskog sustava, nude novu perspektivu za razlikovanje gornjomiocenskih ležišta od drugih sedimentnih jedinica unutar bazena te pružaju vrijedne informacije za industriju, tj. buduća istraživanja i razvoj resursa povezanih također s energetsom tranzicijom i održivošću okoliša. Unatoč opsežnoj istraživačkoj povijesti bazena, ležišne osobine pješčenjaka relativno su nedovoljno istražene, ističući važnost ovog istraživanja - razgraničenje petrografskih, mineraloških i geokemijskih signala omogućuje poboljšanu karakterizaciju ovih ležišta, olakšavajući također njihovu stratigrafsku (kemostratigrafsku) procjenu i prognozno modeliranje u podzemlju.

## **KEYWORDS**

*Sandstone Reservoirs*

*Sedimentary Provenance*

*Diagenesis*

*Late Miocene*

*Pannonian Basin System*

## **KLJUČNE RIJEČI**

*Pješčenjačka ležišta*

*Provenijencija sedimenata*

*Dijageneza*

*Kasni miocen*

*Panonski bazenski sustav*

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# 1. INTRODUCTION

## 1.1. Significance of sedimentary provenance and diagenesis analysis

The formation and subsequent behavior of sedimentary rocks in a basin, crucial for reservoir characterization, are deeply influenced by their provenance and diagenesis. Source rock composition shapes sediment composition and behavior during burial diagenesis (**Suttner et al., 1981; Bhatia, 1983; Pettijohn et al., 1987; Armstrong-Altrin & Verma, 2005; Garzanti, 2016, 2019**), impacting key petrophysical parameters like porosity and permeability (e.g., **Ehrenberg, 1990; Worden et al., 2018; Lawan et al., 2021**). Analyzing provenance and diagenesis is integral for assessing reservoir quality and development (**Worden et al., 1997, 2018; Worden & Burley, 2003; Gier et al., 2008; Rahman & Worden, 2016; Lawan et al., 2021**). Methods like petrographic, heavy mineral, and geochemical analyses are fundamental in sedimentary provenance study, providing insights into the geological processes governing sediment generation, transportation, deposition, and successive diagenesis (e.g., **Dickinson et al., 1983; Weltje et al., 1998; Garzanti, 2016, 2019; Augustsson, 2021**), aiding, alongside other geological methods, e.g., well-log and seismic data interpretation and integration, hydrocarbon exploration by facilitating better-quality correlation and modeling of subsurface sediments and their properties.

The main goal of sedimentary provenance analysis is to reconstruct and interpret the geology of a source area and to connect the final burial of a detritus with the initial erosion of parent rocks (**Ibbeken & Schleyer, 1991; Weltje & von Eynatten, 2004**). Detrital modes of sandstones can be related to the plate-tectonic setting of a sedimentary basin they were deposited in (**Dickinson & Suczek, 1979; Dickinson et al., 1983; Dickinson, 1985; Weltje & von Eynatten, 2004; Weltje, 2006**) and can reflect the nature of the source terranes, as well as the tectonostratigraphic level achieved by erosion in space and time (**Garzanti, 2016**).

In addition to provenance, diagenesis significantly affects reservoir quality and development by altering sediment post-deposition. It involves physical, chemical, and biological processes modifying sedimentary assemblages, ranging from subaerial weathering to low-temperature metamorphism (**Curtis, 1977; Burley et al., 1985; Worden and Burley, 2003**). Understanding burial diagenetic processes, influenced by factors such as depth, temperature, pressure, mineralogy, and pore fluid geochemistry, is important since they hugely shape reservoirs (**Morad et al., 2000; Worden and Burley, 2003**). This

understanding is crucial for predicting porosity distribution, fluid migration pathways, and reservoir productivity (**Hurst, 1987**), essential for accurate reservoir characterization and advanced recovery techniques (**Pittman and King, 1986; Kantorowicz et al., 1992**).

## **1.2. Importance of the Upper Miocene sandstones from the North Croatian Basin**

Main hydrocarbon deposits of the Pannonian Basin System are Neogene rocks accounting for more than 80 % of all reservoirs reported in this area. Of these, sandstones account for 95 %, with 90 % being Miocene in age (**Dolton, 2006**). The most important reservoir rocks of the Pannonian Basin in Croatia are the Upper Miocene sandstones, exhibiting notable porosity and permeability values in the North Croatian Basin, ranging from 10 % to over 35 % and from 0.01 to 1000 mD (**Lučić et al., 2001; Vrbanac et al., 2010; Novak Zelenika et al., 2010; Malvić & Velić, 2011; Velić et al., 2012; Kolenković Močilac et al., 2022**). They were formed during the post-rift phase as prograding systems of delta environments within brackish Lake Pannon, which were associated with turbidites in deeper parts of the lake (**Juhász, 1994; Basch et al., 1995; 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**).

The main hydrocarbon source rocks in the North Croatian Basin are represented by the Middle Miocene late syn-rift and early post-rift marine deposits (Badenian and Sarmatian mudrocks), as well as the Upper Miocene early post-rift lacustrine deposits (early Pannonian mudrocks; **Barić et al., 2000; Lučić et al., 2001; Saftić et al., 2003; Troškot-Čorbić et al., 2009; Zečević et al., 2010; Matošević et al., 2019**). Hydrocarbon generation was triggered by a high geothermal gradient during the initial and subsequent thermal subsidences of the basin (**Saftić et al. 2003**).

Despite decades of study by the petroleum industry, the reservoir properties of these sandstones remain incompletely understood, making them subjects of ongoing interest not only for oil exploration and development of existing oil fields but also for energy transition initiatives, including carbon capture and storage, as well as geothermal energy (**Sneider 1990; Kolenković Močilac et al., 2013; Horváth et al., 2015; Podbojec and Cvetković, 2016; Alcalde et al. 2019; Macenić et al., 2020; Davies & Simmons 2021; Willems et al. 2021; Tuschl et al., 2022; Vulin et al., 2023**). Despite their longstanding importance, a comprehensive understanding of their provenance and diagenesis has been lacking.



### 1.3. Geological background and overview of research conducted hitherto

The Sava and Drava depressions represent the two largest depressions of the North Croatian Basin, which covers almost the entire area of northern Croatia and belongs to the southwestern part of the Pannonian Basin System, a back-arc basin surrounded by the Alps, Carpathians and Dinarides, and paleogeographically belonging to the area of the Central Paratethys. These depressions represent river lowlands on the modern surface and, as border areas between Croatia and neighboring countries, are of vast ecological and economic importance. The maximum depth prior to the pre-Miocene bedrock is 5500 m in the Sava depression and 7000 m in the Drava depression (**Prelogović, 1975; Pavelić, 2001; Saftić et al., 2003; Horváth et al., 2006; Malvić, 2011; Malvić & Cvetković, 2013**).

The development of the Pannonian Basin System began in the Early Miocene as a result of continental collision and subduction of the Eurasian plate under the African (Apulian) plate, including other continental fragments from the south that caused thermal perturbation of the upper mantle, thereby resulting in the weakening and extension of the crust and formation of a back-arc type 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**). The first stage of the Pannonian Basin System development was characterized by tectonic thinning of the crust and isostatic subsidence, with sedimentary environments changing from continental to fully marine (syn-rift phase; **Royden, 1988; Tari et al., 1992**). The second stage was characterized by reduced tectonic influence, termination of rifting, and subsidence caused by cooling of the lithosphere, with gradual isolation of the Pannonian Basin System from marine influences of the Central Paratethys (post-rift phase; **Royden, 1988; Tari et al., 1992**). The transition from the syn-rift to the post-rift phase was diachronous through the Pannonian Basin System (**Matenco & Radivojević, 2012**). The final isolation happened approximately 11.6 Ma ago when the large and long-lived Lake Pannon, which existed from the Late Miocene to the Early Pliocene, was formed (**Magyar et al., 1999; Harzhauser & Piller, 2007; Piller et al., 2007; Mandić et al., 2015**).

Relatively slow thermal subsidence of the Pannonian Basin System without significant tectonic activity in the Late Miocene (**Horváth & Royden, 1981; Ivković et al., 2000; Pavelić & Kovačić, 2018**) was coupled with uplift and erosion of the surrounding Alpine–Carpathian–Dinaric fold belt. Parts of the belt enabled a huge supply of sediment via large fluvial and deltaic systems to flow into the lake (**Bérczi et al., 1988; Szentgyörgyi & Juhász, 1988; Juhász, 1991, 1994; Saftić et al., 2003; Kovačić et al., 2004; Kovačić & Grizelj, 2006; Magyar et al., 2013; Pavelić & Kovačić, 2018; Sebe et al., 2020; Andelković &**

**Radivojević, 2021**). Great depths in the lake developed due to a high rate of subsidence of the basin and prevailing humid climate (**Sztanó et al., 2013; Balázs et al., 2018**). These factors led to the accumulation of post-rift deposits in the sedimentary succession of several thousand meters, including large sandstone bodies, with the greatest thickness identified in the central part of the depressions (**Juhász, 1994; Ivković et al., 2000; Saftić et al., 2003; Kovačić & Grizelj, 2006; Malvić & Velić, 2011; Sebe et al., 2020**). The thickness of the Upper Miocene deposits in the North Croatian Basin which, according to regional chronostratigraphy, belong to the Pannonian stage (**Hilgen et al. 2012; Mandić et al. 2015; Neubauer et al. 2015**), fluctuates between 2000 m in the western part of the Sava depression to 4000–5000 m along the SW edge of the Drava depression (**Saftić et al., 2003**). The Pannonian (Upper Miocene) deposits, situated below Cernikian (Pliocene to Pleistocene) lacustrine and alluvial deposits, overlie the Sarmatian (Middle Miocene) marine deposits. (**Pavelić, 2001; Mandić et al., 2015; Kurečić, 2017; Pavelić & Kovačić, 2018**). The thickness of the Upper Miocene sandstone bodies varies and is usually in the range of a few meters to several tens of meters, commonly intercalated with mudrocks (layers of siltstones, mudstones, claystones, marls, and shales). The sandstones appear as turbidite mass flow deposits on the slope and basin floor (deeper-water fan lobes, channels, and levees) and are subsequently covered by delta and fluvial system deposits that are morphologically related to regional progradation of the shelf (prodelta, delta front, delta plain and alluvial plain, including distributary channels and mouth bars; **Pogácsás, 1984; Saftić et al., 2003; Sztanó et al., 2013, 2015**). Turbidite systems are restricted to deeper and central parts of the depressions (**Sztanó et al., 2015**), while delta systems succession and subsequent fluvial deposits are associated with subsequent progradation of the shelf-margin slope (corresponding to clinoforms on seismic profiles; **Magyar et al., 2013**). The high material inflow in the Pannonian Basin System was mainly from the Alpine–Carpathian source areas, generally following a W/N-NW towards E/S-SE direction (**Šćavničar, 1979; Ivković et al., 2000; Saftić et al., 2003; Kovačić et al., 2004; Kovačić & Grizelj, 2006; Magyar et al., 2013; Sebe et al., 2020**). This applies especially to the sources in the ALCAPA tectonic mega-unit of the Eastern Alps and Western Carpathians, characterized by Mesozoic carbonates, Proterozoic to Palaeozoic low to medium-grade metamorphic rocks, and Palaeozoic granitoids (e.g., **Kuhlemann et al., 2002; Asch, 2003; Schmid et al., 2008, 2020**). Some explorations of the Upper Miocene sandstones outcropping on the surface in the North Croatian Basin show that their detritus is of uniform composition, as well as mineralogically and texturally mature (**Kovačić et al., 2004, 2011; Kovačić & Grizelj, 2006**).

The Pliocene and Quaternary are characterized by a transition towards overall compression and structural inversion in the entire North Croatian Basin (**Pavelić, 2001; Tomljenović & Csontos, 2001**). This change is reflected in the formation of many compressional structures and the uplift of basement blocks that formed the present mountains in the area. The uplift was accompanied by strike-slip faulting and counter-clockwise rotation of blocks (**Jamičić, 1983; Márton et al., 1999, 2002**).

#### **1.4. Objectives and hypotheses of research**

Main objectives of this research were: (1) determination of provenance and diagenesis of the Upper Miocene sandstones of the south-western part of the Pannonian Basin System, (2) petrographic, mineralogical, and geochemical analysis of the Upper Miocene sandstones from selected exploration wells of the Sava and Drava Depression, (3) determination of the detailed composition of the Upper Miocene sandstones and their parameters of importance for the reservoir characterization, and (4) comparison of the Upper Miocene sandstones of the Sava and Drava Depressions based on the obtained results of analyses.

These objectives were based on three fundamental hypotheses:

1. Study of sediment provenance enables the reconstruction and interpretation of sediment history from the initial erosion of parent rocks to the final burial of their detritus.
2. Petrographic analyses, heavy mineral analyses, chemical analyses and analyses of individual grains allow determination of the composition and the provenance of sandstones.
3. Differences in provenance and diagenesis of the Upper Miocene sandstones from Sava and Drava Depressions can be determined.

#### **1.5. Scientific contribution**

The results of this research provide a comprehensive determination of the provenance and diagenesis of the Upper Miocene sandstones in the Sava and Drava depressions. This includes defining the similarities and differences in the composition of these sandstones, the first detailed determination of diagenetic changes in the sandstones, and an assessment of their quality as reservoir rocks with increasing depth.

## 2. ORIGINAL SCIENTIFIC PAPERS

*Paper 1: Matošević, M. Marković, F., Bigunac, D., Šuica, S., Krizmanić, K., Perković, A., Kovačić, M. & Pavelić, D. (2023): Petrography of the Upper Miocene sandstones from the North Croatian Basin: Understanding the genesis of the largest reservoirs in the southwestern part of the Pannonian Basin System. *Geologica Carpathica*, 74 (2), 155-179.*

# Petrography of the Upper Miocene sandstones from the North Croatian Basin: Understanding the genesis of the largest reservoirs in the southwestern part of the Pannonian Basin System

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**Abstract:** This paper presents a petrographic study of the Upper Miocene sandstones from exploration wells in the Sava and Drava Depressions in the North Croatian Basin (SW of the Pannonian Basin System), Central Europe. These sandstones represent the most important reservoir rocks for oil and gas in Croatia. A total of 130 core samples from depths of more than 3000 m were examined. The sandstones generally have a feldspatho–litho–quartzose (fLQ) composition. The modal composition of samples from the Sava Depression is  $Q_{39.2-61.0}F_{8.9-26.0}L_{26.1-42.3}$ , and  $Q_{40.6-63.5}F_{6.6-23.3}L_{20.9-42.3}$  for those from the Drava Depression. Lithic fragments are dominated by extrabasinal carbonates with subordinate metamorphic grains (mostly schists) and less frequent magmatics (granitoids). Garnet, tourmaline, apatite, rutile, epidote, clinozoisite, zoisite, titanite, zircon, staurolite, and opaque minerals form the heavy mineral association and imply an area of provenance dominated by metamorphic rocks. The tectonic setting of the sandstones corresponds to recycled orogen, i.e., a subduction complex or fold-thrust belt. There are no significant compositional differences between the sandstone samples from the two depressions, thus indicating a common main source area and similar diagenetic processes in the subsurface. The sandstones originated from eroded parts of the uplifted Alpine–Carpathian fold belt and are distinct from the Lower and Middle Miocene sandstones in the North Croatian Basin. Observed variations in the sandstones' composition with regards to geographic location and depth are associated with variations in the erosion of parent rocks, including subsequent modification of detrital sediment during transport, mixing, and deposition, as well as burial diagenesis. The Sava and Drava Depressions were part of Lake Pannon in the Late Miocene with no significant topographical obstacles between them for the inflow of the detritus. The study will therefore help with the correlation of the Upper Miocene reservoir rocks in various parts of the North Croatian Basin (as well as adjacent parts of the Pannonian Basin System) and thereby assist with future hydrocarbon exploration in this area.

**Keywords:** petrography, sedimentary provenance, sandstone reservoirs, Late Miocene, Pannonian Basin System

## Introduction

Petrography is a fundamental method in sedimentary provenance studies and remains the prevailing geological approach for understanding the generation of sediments, i.e., their modifications during erosion, transport, deposition, and subsequent diagenesis (e.g., Dickinson et al. 1983; Weltje et al. 1998; Garzanti 2016, 2019; Augustsson 2021). Since the primary composition of source rocks in hinterlands affects the final petrographic composition of sediment in a sedimentary basin (Suttner et al. 1981; Bhatia 1983; Pettijohn et al. 1987; Armstrong-Altrin & Verma 2005; Garzanti 2016, 2019) and plays a major role in sediment behaviour during burial diagenesis affecting its petrophysical parameters (e.g., Ehrenberg 1990; Worden et al. 2018; Lawan et al. 2021), petrography

is crucial for estimating reservoir development and quality (Worden et al. 1997, 2018; Worden & Burley 2003; Gier et al. 2008; Rahman & Worden 2016; Lawan et al. 2021). In addition to well-log and seismic data interpretation and integration, petrography persists as an indispensable method in hydrocarbon exploration, contributing to better-quality correlation and modelling of sediments and their properties in the subsurface.

The main goal of sedimentary provenance analysis is to reconstruct and interpret the geology of a source area and to connect the final burial of a detritus with the initial erosion of parent rocks (Ibbeken & Schleyer 1991; Weltje & von Eynatten 2004). Petrography is the only method used in sandstone provenance analysis that simultaneously provides both mineralogical and textural data of sandstones and their forming

constituents (small-scale originals of their parent rocks), thereby allowing direct visualization and reconstruction of the source area (Garzanti 2016). Detrital modes of sandstones can be related to the plate-tectonic setting of a sedimentary basin they were deposited in (Dickinson & Suczek 1979; Dickinson et al. 1983; Dickinson 1985; Weltje & von Eynatten 2004; Weltje 2006) and can reflect the nature of the source terranes, as well as the tectonostratigraphic level achieved by erosion in space and time (Garzanti 2016).

Neogene rocks are the main hydrocarbon deposits of the Pannonian Basin System (PBS). They account for more than 80 % of all reservoirs reported in this area – sandstones constitute 95 % of this, with 90 % being of Miocene age (Dolton 2006). Hydrocarbon generation was triggered by a high geothermal gradient during the initial and subsequent thermal subsidences of the basin (Saftić et al. 2003). The most important reservoir rocks of the PBS in Croatia are the Upper Miocene sandstones, which had been formed through the mechanism of lacustrine cyclic turbidite deposition associated with the progradation of delta systems in Lake Pannon (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). In the North Croatian Basin (NCB), these sandstones exhibit high values of porosity and permeability. The porosity is usually in the range of 10 % to more than 35 %, with permeability between 0.01 to 1000 mD (Vrbanac et al. 2010; Velić et al. 2012; Kolenković Močilac et al. 2022).

This paper presents a petrographic study of the Upper Miocene sandstones in the NCB (SW part of the PBS), the largest reservoirs of oil and gas discovered in Croatia. Although the basin is a relatively mature petroleum province that has been studied for several decades (e.g., Lučić et al. 2001; Saftić et al. 2003; Dolton 2006; Vrbanac et al. 2010; Malvić & Velić 2011; Velić et al. 2012; Matošević et al. 2019b,c, 2021; Kolenković Močilac et al. 2022), the reservoir properties of the sandstones themselves are not well-known. These sandstones not only continue to be of great interest to the oil industry both in terms of new exploration and the development of existing oil fields, but also in terms of the energy transition, including carbon capture, utilization, and storage, and geothermal energy (cf. Sneider 1990; Alcalde et al. 2019; Davies & Simmons 2021; Willems et al. 2021). Understanding of the primary composition of sandstone reservoir rocks and the factors which influence their petrophysical characteristics is the first step toward reservoir quality evaluation and prediction (Worden et al. 1997, 2018; Worden & Burley 2003; Rahman & Worden 2016; Lawan et al. 2021).

For this study, petrographic analyses were performed on sandstone samples from exploration wells in the Sava and Drava Depression, which are the two largest depressions in the NCB. The paper aims to determine the petrographic composition of the sandstones, compare the samples from the two depressions, interpret their provenance areas, and assess their variability in terms of geographic location and

depth. The results will contribute to further exploration of the area and, in particular, to the improved correlation of the Upper Miocene sandstones in the Croatian and adjacent parts of the PBS.

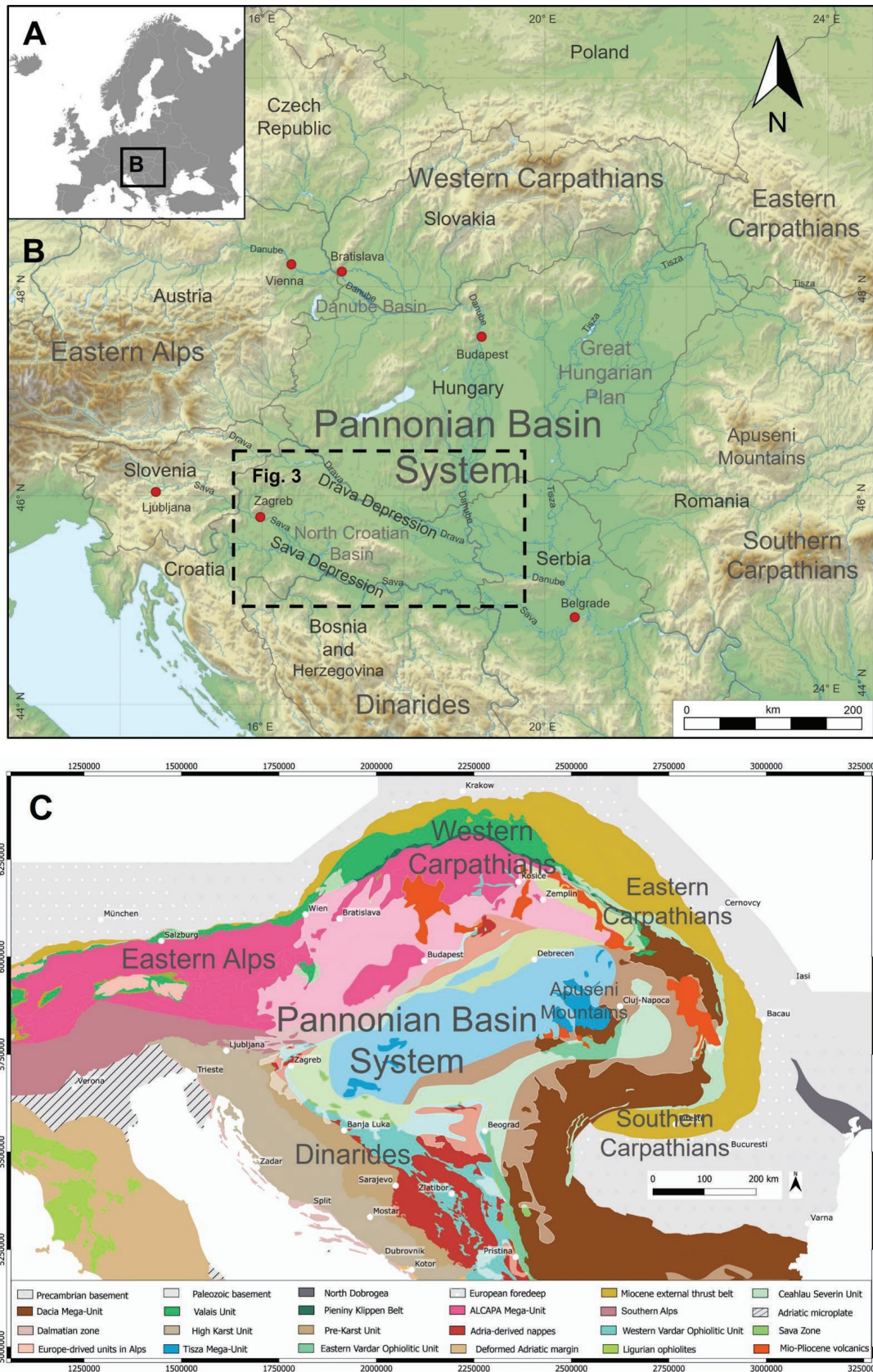
## Geological background

The Miocene PBS, which is surrounded by the Alps, the Carpathians, and the Dinarides (Fig. 1), is paleogeographically related to the area of the Central Paratethys. During the Miocene, the connection of the Central Paratethys with the world ocean had been established and ceased several times. The final isolation of the Central Paratethys commenced some 11.6 Ma ago (Steininger & Rögl 1979; Rögl & Steininger 1983; Rögl 1998; Magyar et al. 1999; Harzhauser et al. 2007; Piller et al. 2007; Harzhauser & Mandić 2008; Kováč et al. 2017, 2018; Magyar 2021). The nature of the Central Paratethys evolution and the occurrences of endemic faunas have thus necessitated the establishment of regional Miocene stages (Fig. 2).

The SW part of the PBS, which covers almost the entire area of northern Croatia, is represented by the NCB (Figs. 1 and 3). The NCB is elongated, with a WNW–ESE-trending direction and includes several depressions which are bordered by mountains (Pavelić & Kovačić 2018; Figs. 1 and 3). The two largest are the Sava Depression (SD) along the southern edge of the basin, and the Drava Depression (DD) along the northern edge of the basin. These depressions represent river lowlands on the modern surface and, as border areas between Croatia and neighbouring countries, are of vast ecological and economic importance. The maximum depth prior to the pre-Miocene bedrock is 5500 m in the SD and 7000 m in the DD (Prelogović 1975; Pavelić 2001; Saftić et al. 2003; Horváth et al. 2006; Malvić 2011; Malvić & Cvetković 2013). Miocene siliciclastic sediments of the NCB unconformably cover strongly tectonized Paleozoic, Mesozoic, and Paleogene basements, and were deposited from the Early to the Late Miocene in marine, brackish, and freshwater environments (Pamić 1999; Pavelić 2001; Saftić et al. 2003; Pavelić & Kovačić 2018; Šuica et al. 2022a,b).

The development of the PBS began in the Early Miocene as a result of continental collision and subduction of the Eurasian plate under the African (Apulian) plate, including other continental fragments from the south that caused thermal perturbation of the upper mantle, thereby resulting in the weakening and extension of the crust and formation of a back-arc type 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). The first stage of the PBS development was characterized by tectonic thinning of the crust and isostatic subsidence, with sedimentary environments changing from continental to fully marine (syn-rift phase; Royden 1988; Tari et al. 1992). The second stage was characterized by reduced tectonic influence, termination of rifting, and subsidence caused by





**Fig. 1.** **A** — Europe with the geographic position of the Pannonian Basin System. **B** — The location of the Sava and Drava Depressions in the SW part of the Pannonian Basin System (North Croatian Basin) surrounded by the mountain ranges of the Alps, the Carpathians, and the Dinarides. **C** — Modified schematic presentation of the main tectonic units of the region (after Schmid et al. 2008, 2020).

cooling of the lithosphere, with gradual isolation of the PBS from marine influences of the Central Paratethys (post-rift phase; Royden 1988; Tari et al. 1992; Fig. 2). The transition from the syn-rift to the post-rift phase was diachronous through the PBS (Matenco & Radivojević 2012). The final isolation happened approximately 11.6 Ma ago when the large and long-lived Lake Pannon, which existed from the Late Miocene to the Early Pliocene, was formed (Magyar et al. 1999; Harzhauser & Piller 2007; Piller et al. 2007; Mandić et al. 2015). In the NCB, the main hydrocarbon source rocks are represented by the Middle Miocene late syn-rift and early post-rift marine deposits (Badenian and Sarmatian mudrocks), as well as the Upper Miocene early post-rift lacustrine deposits (early Pannonian mudrocks; Barić et al. 2000; Lučić et al. 2001; Saftić et al. 2003; Troskot-Čorbić et al. 2009; Zečević et al. 2010; Matošević et al. 2019a).

The sandstones that later became significant oil and gas reservoirs were formed during the post-rift phase as prograding systems of delta environments within Lake Pannon, which were associated with turbidites in deeper parts of the lake (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). Relatively slow thermal subsidence of the PBS without significant tectonic activity in the Late Miocene (Horváth & Royden 1981; Ivković et al. 2000; Pavelić & Kovačić 2018) was coupled with uplift and erosion of the surrounding Alpine–Carpathian–Dinaric fold belt. Parts of the belt enabled a huge supply of sediment via large fluvial and deltaic systems to flow into the lake (Bérczi et al. 1988; Szentgyörgyi & Juhász 1988; Juhász 1991, 1994; Saftić et al. 2003; Kovačić et al. 2004; Kovačić & Grizelj 2006; Magyar et al. 2013; Pavelić & Kovačić 2018; Sebe et al. 2020; Anđelković & Radivojević 2021). Great depths in the lake developed due to a high rate of subsidence of the basin and prevailing humid climate (Sztanó et al. 2013; Balázs et al. 2018). These factors led to the accumulation of post-rift deposits in the sedimentary succession of several thousand meters, including large sandstone bodies, with the greatest thickness identified in the central part of the depressions (Juhász 1994; Ivković et al. 2000; Saftić et al. 2003; Kovačić & Grizelj 2006; Malvić & Velić 2011; Sebe et al.

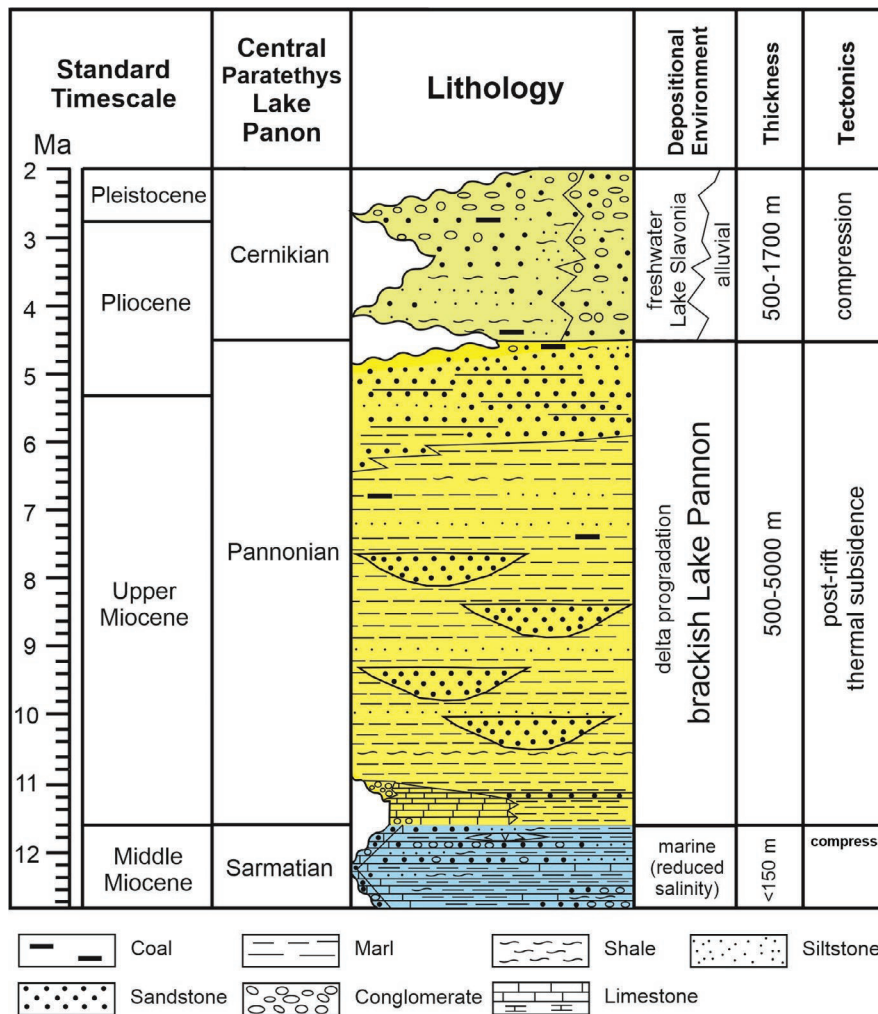
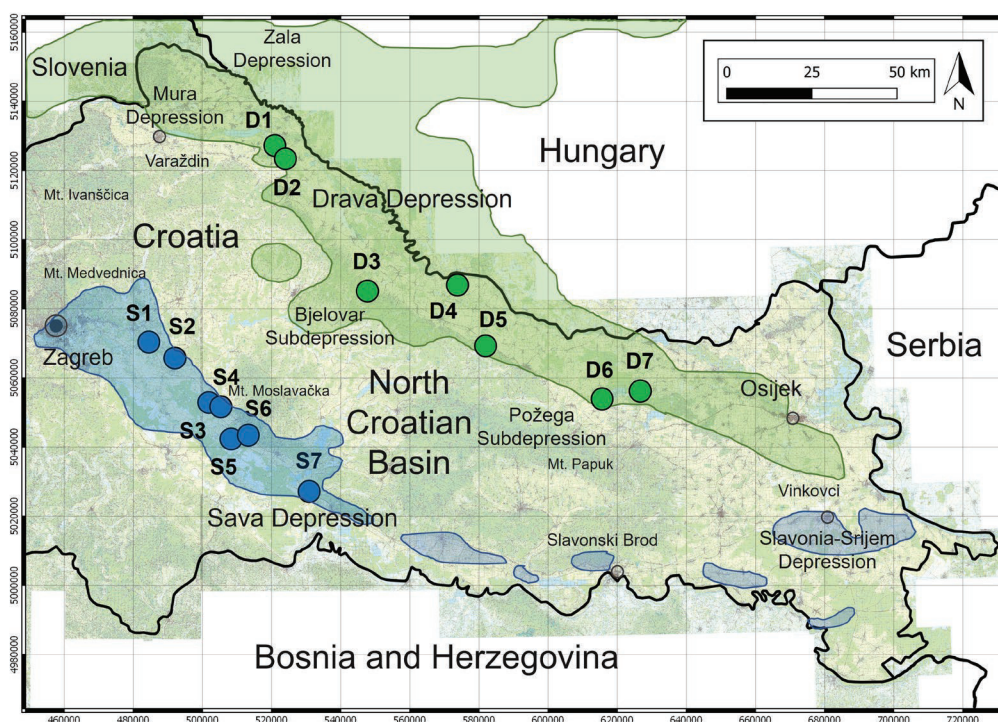


Fig. 2. Stratigraphic scheme of the North Croatian Basin, modified after Saftić et al. (2003), Mandić et al. (2015), and Pavelić & Kovačić (2018).





**Fig. 3.** Position of the North Croatian Basin and locations of exploration wells, which are the subject of this study within the approximate areas of the Sava (S1–S7) and Drava Depression (D1–D7), including neighboring Neogene depressions and sub-depressions.

2020). The thickness of the Upper Miocene deposits in the NCB which, according to regional chronostratigraphy, belong to the Pannonian stage (Hilgen et al. 2012; Mandić et al. 2015; Neubauer et al. 2015; Fig. 2), fluctuates between 2000 m in the western part of the SD to 4000–5000 m along the SW edge of the DD (Saftić et al. 2003). The Pannonian (Upper Miocene) deposits, situated below Cernikian (Pliocene to Pleistocene) lacustrine and alluvial deposits, overlie the Sarmatian (Middle Miocene) marine deposits. (Pavelić 2001; Mandić et al. 2015; Kurečić 2017; Pavelić & Kovačić 2018; Fig. 2). The thickness of the Upper Miocene sandstone bodies varies and is usually in the range of a few meters to several tens of meters, commonly intercalated with mudrocks (layers of siltstones, mudstones, claystones, marls, and shales). The sandstones appear as turbidite mass flow deposits on the slope and basin floor (deeper-water fan lobes, channels, and levees) and are subsequently covered by delta and fluvial system deposits that are morphologically related to regional progradation of the shelf (prodelta, delta front, delta plain and alluvial plain, including distributary channels and mouth bars; Pogácsás 1984; Saftić et al. 2003; Sztanó et al. 2013, 2015). Turbidite systems are restricted to deeper and central parts of the depressions (Sztanó et al. 2015), while delta systems succession and subsequent fluvial deposits are associated with subsequent progradation of the shelf-margin slope (corresponding to clinofolds on seismic profiles; Magyar et al. 2013). The high material inflow in the PBS was mainly from the Alpine–Carpathian source areas, generally following a W/N-NW towards E/S-SE

direction (Šćavničar 1979; Ivković et al. 2000; Saftić et al. 2003; Kovačić et al. 2004; Kovačić & Grizelj 2006; Magyar et al. 2013; Sebe et al. 2020). This applies especially to the sources in the ALCAPA tectonic mega-unit of the Eastern Alps and Western Carpathians (e.g., Kuhlemann et al. 2002; Asch 2003; Schmid et al. 2008, 2020). Some explorations of the Upper Miocene sandstones outcropping on the surface in the NCB show that their detritus is of uniform composition, as well as mineralogically and texturally mature (Kovačić et al. 2004, 2011; Kovačić & Grizelj 2006).

The Pliocene and Quaternary are characterized by a transition towards overall compression and structural inversion in the entire NCB (Pavelić 2001; Tomljenović & Csontos 2001). This change is reflected in the formation of many compressional structures and the uplift of basement blocks that formed the present mountains in the area. The uplift was accompanied by strike-slip faulting and counter-clockwise rotation of blocks (Jamičić 1983; Márton et al. 1999, 2002).

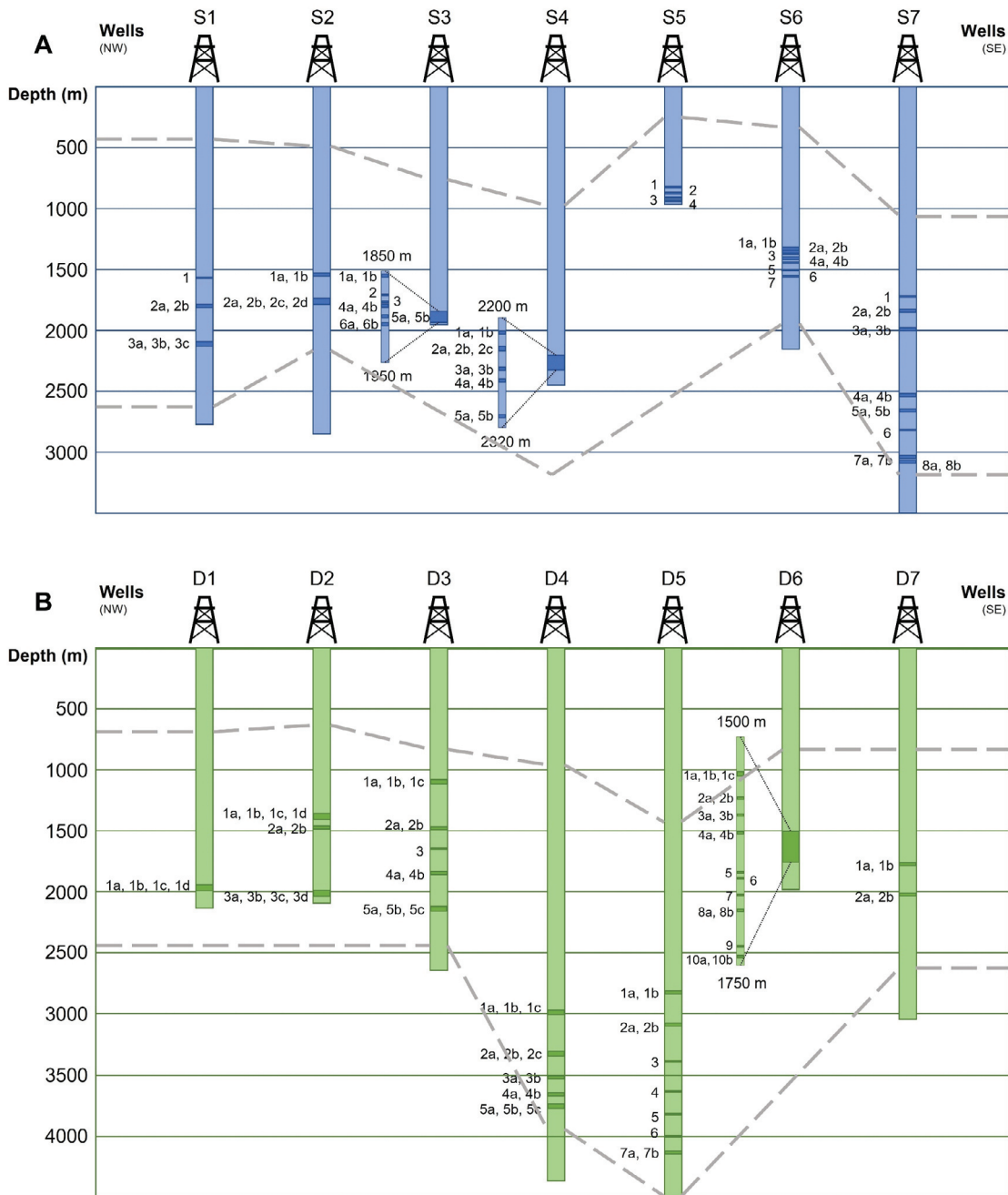
## Materials and methods

### Materials

A total of 130 core samples from the exploration wells drilled by INA – Industrija nafte d.d. were selected and prepared for analyses. The samples included 61 sandstones from seven wells in the SD (Figs. 3 and 4) and 69 sandstones from

seven wells in the DD (Figs. 3 and 4). In order to investigate variations in the sandstones' composition and provenance in both space and time, core samples were taken from wells that covered almost the entire area of the two depressions and were derived from depths ranging from 827.70 m to 3073.90 m in the SD and from 1098.95 m to 4141.15 m in the DD, i.e., covering the entire Upper Miocene succession. The stratigraphic affiliation of the samples was verified by previous biostratigraphic (palynological) analyses, well-log correlation,

and seismic interpretation of key horizons representing regional stratigraphic tops, performed in the INA – Industrija nafte d.d. The sampled sandstones were macroscopically homogeneous and massive and were selected from relatively thick beds which represent major reservoir units. Samples without significant sedimentary structures (such as erosional, depositional, and post-depositional structures, and bioturbations) and which were of similar grain size were chosen to avoid significant inter-sample variations.



**Fig. 4.** Positions of the Upper Miocene sandstones' samples from the exploration wells of the Sava (A) and Drava (B) Depressions. The stratigraphic correlation of the wells is based on the well-log data boundaries between the Upper Miocene (Pannonian) and Pleistocene/Pliocene (Cernikian) in shallower intervals (upper dashed line) and the Upper Miocene (Pannonian) and Middle/Lower Miocene or the Neogene basement in deeper intervals (lower dashed line). For the geographic location of wells, see Fig. 3.

## Methods

### *Petrography*

A total of 130 thin sections of sandstones were prepared and analyzed under Olympus BX51 and Leitz Orthoplan polarizing microscopes with an Olympus DP 70 camera and analySIS Five imaging software. The samples were stained with Alizarin Red S according to Evamy & Shearman (1962) to distinguish carbonate minerals (calcite and dolomite), while some were impregnated with blue dye epoxy. At least 100 grains per thin section were measured to determine grain size ranges and the average grain size of each sandstone using the Wentworth (1922) scale. The grain sizes were manually measured (both length (the largest diameter) and width (average diameter)), from which the arithmetic means of the grain sizes were calculated. Based on these measurements, an approximation of the average grain size for each sample was deduced. For the grain-matrix size, a cut-off of 20  $\mu\text{m}$  was applied. The measured grain sizes were generally rounded to whole numbers (expressed in intervals of 10  $\mu\text{m}$ ). The roundness of grains was visually estimated using graphical determination charts (Powers 1953; Krumbein & Sloss 1963). Grain sorting was estimated according to Jerram (2001). The textural maturity of the sandstones was estimated according to Folk (1951).

The modal composition of the sandstones was determined by examining the relative proportion of the three main components: quartz (Q), feldspars (F), and lithics or rock fragments (L). At least 300 grains per sample were analyzed (except for the D7-2a, due to the small amount of the material). To classify provenance lithotypes, emphasis was placed on detailed microscopic analyses of rock fragments and their textures (Garzanti 2016). The amounts of phyllosilicates (mica and chlorite) and heavy minerals present were also considered; however, they were not included in the classification of the sandstones since they are mostly conditioned by hydraulic sorting and depositional environment. These components were, however, considered part of the abundance of detrital modes. Sandstone classifications were based on modal compositions (Garzanti 2016, 2019), thereby allowing provenance models to be inferred (Dickinson 1985; Weltje 2006; Garzanti 2016; Augustsson 2021).

The QFL detrital mode analyses were performed according to the modification of the Gazzi-Dickinson point-counting method (Dickinson 1970; Ingersoll et al. 1984; Zuffa 1985). Total quartz content (Q or Qt) included all types of monocrystalline (Qm) and all types of polycrystalline quartz (Qp). Polycrystalline quartz (Qp) included polycrystalline quartz grains, all types of polycrystalline tectonic quartz (quartzite), and polycrystalline microcrystalline quartz (cherty grains). Feldspar content (F) included twinned plagioclase feldspar (Pt) and all potassium (alkali) feldspar (i.e., microcline, orthoclase, perthite, and sanidine), non-twinned plagioclase feldspar, sericitized plagioclase feldspar, albitized potassium (alkali) feldspar, and other altered and partly-dissolved plagioclase or

potassium (alkali) feldspar, all represented as a single value (K+P). Rock fragments or lithic grains (L) included metamorphic (Lm), magmatic (igneous; Lv), and sedimentary (Ls) rocks. Rock fragments were considered as such only if individual minerals forming the rock fragments were less than 63  $\mu\text{m}$  (e.g., quartz, feldspar, and mica forming a granitoid fragment). Carbonatic clasts were considered rock fragments, regardless of the sizes of individual minerals and/or fossils forming the clast. The share of total rock fragments (Lt; Dickinson 1985) included all types of polycrystalline quartz (Qp), together with tectonic quartz and cherty grains. The modal composition of rock fragments (L), excluding polycrystalline quartz (Qp; Garzanti 2019), along with tectonic quartz and cherty grains, included the content of metamorphic rocks (Lm) with all types of quartz–mica tectonites (TQM) and polycrystalline mica (Mp), the content of magmatic (igneous) rocks (Lv) with extrusive (volcanic) rocks (Lex) and intrusive (plutonic) rocks (Lp), and the content of sedimentary rocks (Ls) with all types of carbonatic (Lcarb) and siliciclastic (Lncarb) sediments. Quartz–mica tectonite grains (TQM) included all types of metapsammite grains, with or without feldspar, and all types of metaigneous grains, along with gneiss and some types of mica schist (including those with more than 25–30 % share of quartz). Polycrystalline mica grains (Mp) included all types of metapelite grains, along with slate, phyllite, some types of mica schist (including those with less than 25–30 % share of quartz), and metamorphic rocks of possible primary volcanic origin as well. Extrusive (volcanic; Lex) grains included all types of extrusive (effusive) grains, along with volcanoclastic rocks (and/or tuffs) and volcanic glass. Intrusive (plutonic; Lp) grains included granitoid rocks, along with microlithic granites, and all types of other intrusive rocks (e.g., diorite, gabbro, etc.). Sedimentary carbonatic grains (Lcarb) included all types of limestones and dolomites (micrite, sparite, grainstone, packstone, wackestone, etc.) and evaporites, along with recrystallized carbonatic grains and metacarbonatic grains, since it was difficult to distinguish diagenetically altered and recrystallized carbonatic rock fragments from e.g., rock fragments of marble, as well as all types of intrabasinal or recycled carbonate bioclasts. Sedimentary siliciclastic grains (Lncarb) included all types of mudrocks (siltstones mudstones, claystones, marls, shales, etc.). Phyllosilicates were grouped separately in the M group, including mica (muscovite, biotite, etc.) and chlorite. Accessory heavy minerals and opaque minerals were grouped in the HM group. Miscellaneous and unidentified grains were grouped in the Misc group, including hardly-recognizable, deformed, and altered (usually dissolved) grains, as well as those that were indeterminable due to other reasons (e.g., knowledge, size, etc.).

### *SEM & EDS*

Scanning electron microscope (SEM) analyses were performed on selected sandstone samples to confirm the features observed using polarizing light microscopes. The morphological characteristics of the sandstones and individual grains,



including their chemical and mineralogical composition, were studied. Particular attention was given to the determination of heavy minerals and rock fragments that were hard to determine by optical microscopes due to the small dimensions or to alteration (e.g., dissolution and recrystallization). SEM analyses were also used to determine types of cement and clay minerals present in intergranular space. The mineral determination relied on the combination of morphological characteristics of a single crystal, which had been acquired with a secondary electron (SE) image, back-scattered electron (BSE) image, and energy-dispersive X-ray spectroscopy (EDS) spectra. X-ray spectra were compared to literature data of known spectra, and a mineral name was assigned to the point of interest (Severin 2004). Measurements were performed on gold-coated samples. Analytical conditions included a high vacuum and a variety of acceleration voltages (5–25 kV), spot sizes (30–50), and magnifications (25–20,000 $\times$ ). Analyses were performed using a JEOL JSM-6510 LV SEM apparatus (JEOL, Tokyo, Japan) equipped with an energy-dispersive X-ray spectroscope using Oxford INCA X-act system (Oxford Instruments, High Wycombe, UK).

## Results

### *Modal composition*

In the following section, the results of petrographic analyses of the Upper Miocene sandstones from the wells in the SD and DD are presented. Tables with detailed results of modal compositions for each sample are provided in the Electronic Supplement, which can be found online ([Supplementary Tables S1–S14](#)).

The Upper Miocene sandstones from 35 cores from the SD and 33 cores from the DD were examined (Fig. 4). The sandstones have angular to sub-rounded grains with average sizes from 70 to 160  $\mu\text{m}$  in samples from the SD and from 90 to 220  $\mu\text{m}$  in samples from the DD ([Suppl. Tables S1–S14](#)). The grains correspond to very fine and fine-grade sand (grain sizes range from 20 to 1200  $\mu\text{m}$  in the SD and from 20 to 1400  $\mu\text{m}$  in the DD). Grains are predominately well- to moderately well-sorted, rarely moderately-well or well-sorted, and in extreme cases, moderately-well to poorly-sorted. They have tangential (point) and, to a lesser extent, concavo-convex and sutured contacts. Sandstones are texturally mature to sub-mature, and rarely exclusively mature or sub-mature.

The framework petrography (QFL modal composition) of the sandstones in the samples from the SD matches the content of Q in the range of 39.2–61.0 % (average: 50.3 %), F 8.0–26.0 % (average: 15.6 %), and L 26.1–42.3 % (average: 34.1 %; Table 1; Figs. 5 and 6). In samples from the DD, the content of Q is 40.6–63.5 % (average: 51.4 %), F 6.6–23.3 % (average: 14.7 %), and L 20.9–42.3 % (average: 33.9 %; Table 1; Figs. 5 and 6). Qm in the SD is in the range of 28.5–48.6 % (average: 37.2 %) and the share of Lt is between 37.6–57.8 % (average: 47.2 %; Table 1). In the DD, Qm is in the range

of 28.1–48.7 % (average: 36.5 %) and the share of Lt is between 37.7–60.6 % (average: 48.8 %; Table 1).

The modal composition of rock fragments in samples from the SD is characterized by Lm ranging from 11.6 % to 42.9 % (average: 27.9 %), Lv 4.9–22.1 % (average: 10.0 %), and Ls 43.2–77.7 % (average: 62.1 %; Table 2; Figs. 7 and 8). For samples from the DD, Lm range from 14 % to 41.9 % (average: 28.8 %), Lv 2.2–28.5 % (average: 9.7 %), and Ls 37.4–80.9 % (average: 61.4 %; Table 2; Figs. 7 and 8). In the total modal grain composition of samples from the SD, M accounts for 4.5–23.4 % (average: 15.5 %) and HM for 1.2–6.7 % (average: 3.4 %; Table 1). M accounts for 6.4–26.3 % (average: 15.5 %) and HM for 0.8–6.2 % (average: 3.4 %) in samples from the DD (Table 1).

If considering only the most abundant QFL component (Garzanti 2019), all analyzed sandstones can be classified as quartzose sandstones, except for one sample from S6 which responds to lithic sandstone. Based on the two most abundant QFL components, the sandstones belong to the LQ group and, to a lesser extent, to the QL group (some samples from S6). Similarly, according to the sixfold subdivision (Weltje 2006), the sandstones can be classified as quartzolithic (Ql), and only one sample from S6 as lithoquartzose (Lq) sandstone. According to the expanded scheme of Garzanti (2016), the sandstones can generally be classified as feldspatho–litho–quartzose (fLQ) and less often as litho–quartzose (LQ; some samples from S4, and some samples from D1, D3, and D4) and feldspatho–quartzo–lithic (fQL; one sample from S6; Fig. 6).

### *Rock fragments*

A significant presence of fragments of numerous rock types in the analyzed sandstones was determined in both depressions (Tables 1 and 2; Figs. 5, 7 and 8). The most frequently observed fragments are those of Ls, followed by Lm and Lv (Table 2; Figs. 5, 7 and 8). Lcarb are the dominant type of sedimentary rock fragments, constituting on average 20.8 % of the sum of QFL composition in the SD and 20.5 % in the DD (Figs. 5 and 7; [Suppl. Tables S1–S14](#)). They include different types of dolomites and limestones, usually in the form of altered and recrystallized sparite, micrite (mudstone), packstone to grainstone, etc. Lcarb also include possible metamorphosed carbonates (metacarbonate and marble), as well as rarely observed recycled bioclasts, such as fragments of undetermined foraminifera shells, mollusks, and red algae (Fig. 7). The carbonate fragments are more rounded in comparison to other grains (Figs. 5 and 7). Lcarb can be observed to a lesser degree, with an average of only 0.7 % in the sum of QFL composition in both depressions ([Suppl. Tables S1–S14](#)). They are usually in the form of mudrocks (mostly marls, claystones to mudstones, shales rich with organic matter, and siltstones) and generally represent deformed rip-up clasts (Figs. 5 and 7). Although imperceptible through the modal composition, fragments of sandstones were also observed during the analyses, albeit in extraordinarily small quantities. Cherty grains are extremely rare (Fig. 7). Lm are very frequent and mostly

**Table 1:** Modal composition of the Upper Miocene sandstones from the exploration wells of the Sava (S1–S7) and Drava Depression (D1–D7) with minimal, average, and maximal values for the individual component: Q – quartz; Qm – monocrySTALLine quartz; F – feldspar; L – rock fragments; Lt – total rock fragments (including polycrystalline quartz); M – phyllosilicates; HM – heavy minerals.

Exploration Well (Nr. of Samples)	Q (%)			Qm (%)			F (%)			L (%)			Lt (%)			M (%)			HM (%)		
	Min	Average	Max	Min	Average	Max	Min	Average	Max	Min	Average	Max	Min	Average	Max	Min	Average	Max	Min	Average	Max
S1 (6)	42.4	47.6	51.4	28.5	32.1	34	11.1	13.4	18	34.6	39	41.5	53.1	54.5	57.8	4.5	11.4	17.8	2	3.2	4.6
S2 (6)	47.4	51.7	54.6	37.3	40.2	42.9	11.2	13.3	15.8	31.6	34.9	39.6	43.3	46.4	49.7	6.4	10	14.6	1.7	3.7	6.7
S3 (10)	43	49.1	54.2	30.5	37.5	41.4	15.8	17.4	18.8	27	33.5	39.4	41.5	45.1	52	10.2	15.7	19.7	2.4	3.6	5.9
S4 (11)	48.7	56.3	61	34.4	42.4	48.6	8	11.5	13.4	28.2	32.2	40.3	40.6	46.1	54.6	9.3	15.8	22.6	1.2	2.9	5.8
S5 (4)	46	49.7	51.7	33.1	37.1	40.4	14.2	17.7	22.7	30.1	32.6	34.9	40.4	45.2	52.7	11.1	16.1	20.5	3.8	5.4	6.6
S6 (10)	39.2	46.2	49.4	31.2	34.2	39.1	13.1	17.1	21	29.6	36.6	42.3	42.5	48.6	52.7	10.9	18.6	22.7	1.4	3.4	6.6
S7 (14)	45.9	50	57.9	31.9	36	39.5	12.7	17.8	26	26.1	32.2	38.1	37.6	46.1	53	9.2	16.8	23.4	1.6	3	5
D1 (4)	47.3	48.9	51.4	31.6	33.4	35	9.9	13.9	19	33.7	37.4	42.3	48.1	52.7	55.9	13	15.4	18.7	1.6	2.8	3.8
D2 (10)	46.2	51.2	53.8	29.2	33.9	37.8	10.2	12.3	14.6	32.8	36.5	41	47.6	53.8	60.6	7	13	20.3	1	3.5	6.2
D3 (11)	56.3	58	62.2	37.5	42.7	48.7	6.6	10	14.7	27.1	32	36.7	41.9	47.3	54.5	11.2	16.1	22.6	1.3	2.3	3.7
D4 (13)	49.3	54.3	63.5	33.4	36.7	42.3	9.8	13.3	17.7	20.9	32.4	38	44.5	50	56.8	6.4	14	26.3	1.3	2.8	4.3
D5 (10)	43.5	49.1	52.9	31.4	36.4	40	13.9	16.6	22.2	30.2	34.2	39.7	41.4	46.9	52.8	10.8	15.2	21.2	0.8	2	3.9
D6 (17)	40.6	46.5	49.5	28.1	33.6	37.7	15.4	18.8	23.3	27.8	34.7	39.4	40.5	47.6	54.8	6.8	17.1	21.5	0.8	2.4	4.1
D7 (4)	49.8	53.7	57	39.3	41.2	44.2	13.5	16.7	20.4	24	29.6	34.2	37.7	42.1	47.1	11.5	13.6	16.3	1.1	2.4	5

include mica schists, quartzites, and gneisses, but also metapelites (slates and phyllites) and in extremely rare circumstances, metasandstones (Figs. 5 and 7). TQM group fragments on average constitute 6.1 % of the sum of QFL composition in the SD and 5.7 % in the DD (Suppl. Tables S1–S14). The Mp group on average constitutes 3.5 % of the sum of QFL composition in the SD and 3.7 % in the DD (Suppl. Tables S1–S14). Lv are present to a lesser extent and mainly represented by granitoids (Fig. 7). Due to different intersections of grains, they may in part correspond to metamorphic rocks (gneisses and/or schists). They constitute on average 3.0 % of the sum of QFL composition in the SD and 2.9 % in the DD (Suppl. Tables S1–S14). Lex are far less commonly observed than Lp and include altered volcanic rocks with accompanying volcanoclastics (tuffs) and volcanic glass (Fig. 7). They constitute on average only 0.3 % of the sum of QFL composition in the SD and 0.2 % in the DD (Suppl. Tables S1–S14).

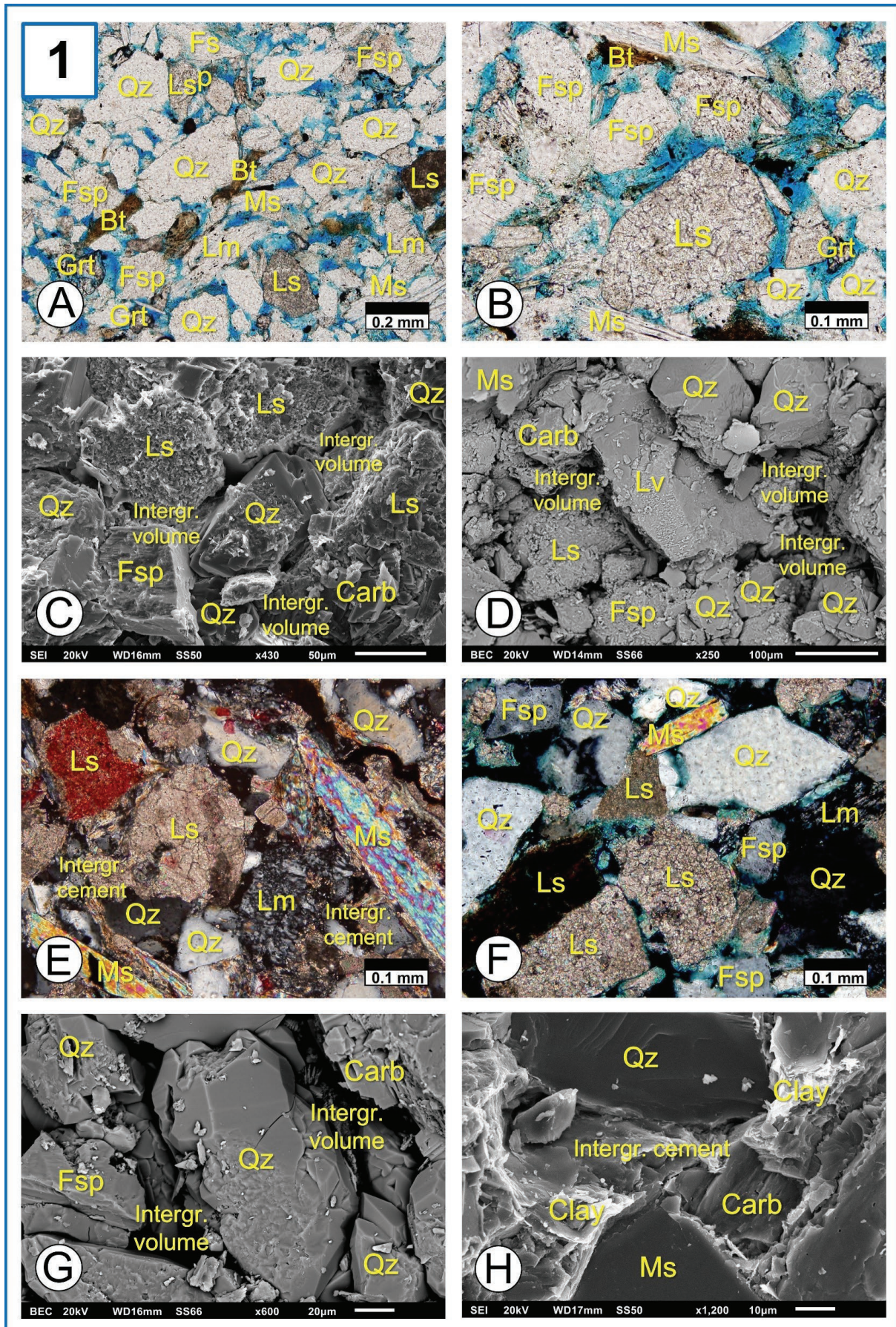
*Heavy minerals*

Almost the same association of accessory minerals was identified in both depressions. Garnet is the most abundant heavy mineral in the sandstones and is present in all analyzed samples in greater quantities (Figs. 5 and 9). It is followed by apatite, tourmaline, rutile, and zircon (Fig. 9). Minerals of epidote, clinozoisite, and zoisite, as well as staurolite and in rare circumstances, amphibole, are more significantly present in shallower intervals of wells (up to 1000–1500 m), especially in the SD. A small amount of chloritoid and titanite was also observed. SEM and EDS analyses confirmed the heavy mineral association (Fig. 5) detected via optical microscopy and determined additional heavy minerals in extremely small quantities – monazite and barite. The EDS of garnet grains frequently matches the almandine phase. Numerous opaque minerals were also detected with optical microscopy. SEM and EDS analyses of these opaque minerals confirmed that they mostly correspond to iron oxides, hydroxides, and sulfides (magnetite, hematite, goethite, and pyrite).

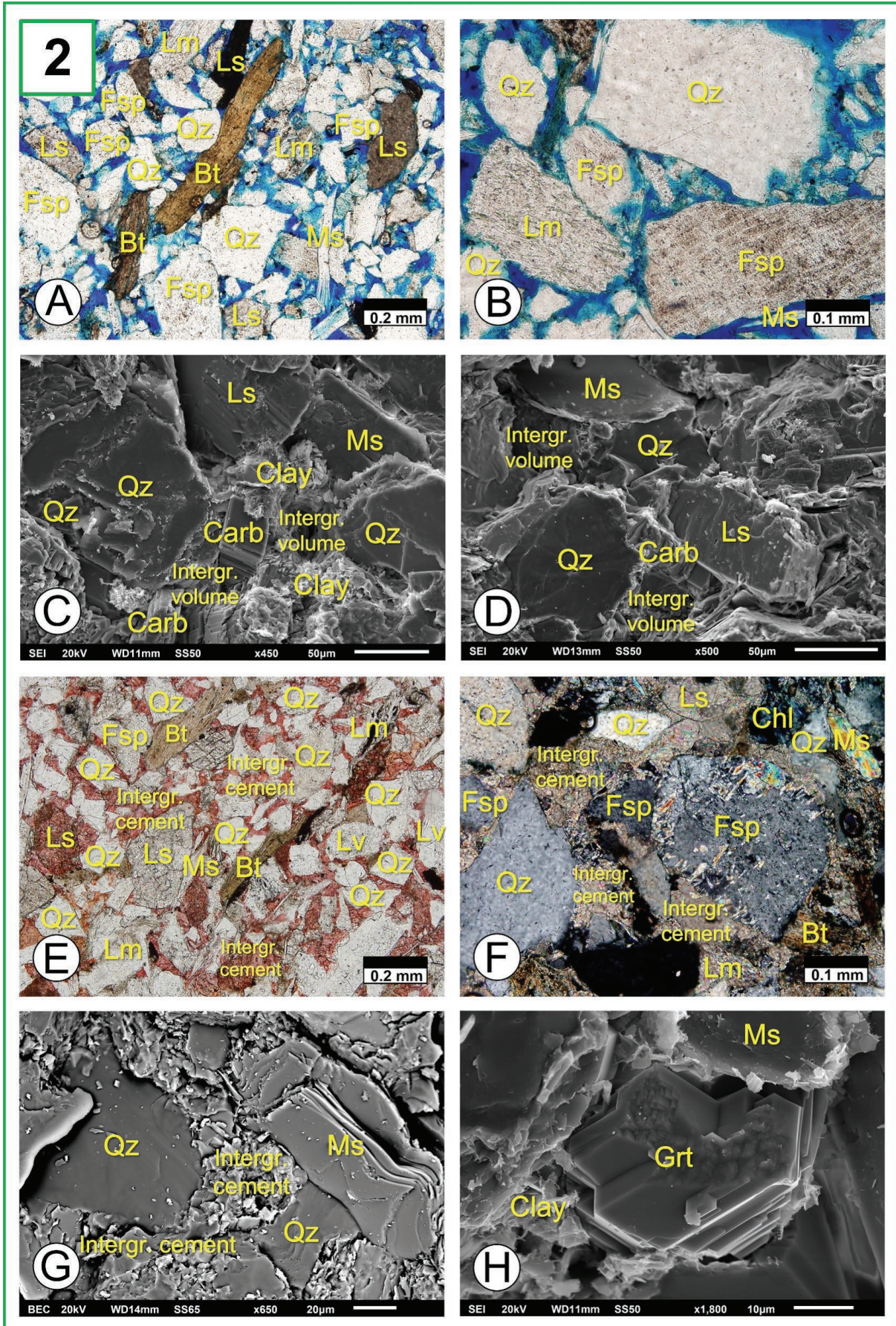
*Compositional changes with the geographic position and depth*

Petrographic analyses of the Upper Miocene sandstones revealed trends of relative changes in the modal composition with a change of geographic position and depth (Supplementary Figs. S1–S7). From the W/NW towards the E/SE, both depressions exhibit an increase in F (Suppl. Fig. S1), Lm (Suppl. Fig. S3), and Lv (Suppl. Fig. S3) components, as well as a decrease in Q (Suppl. Fig. S1), L (Suppl. Fig. S1), Lt (Suppl. Fig. S2), and Ls (Suppl. Fig. S3) components. Subgroups of Q, F, Lm, Lv, and Ls further display uniform trends of changes from the W/NW towards the E/SE in both depressions: a decrease in Qp, Lex, Lcarb, and TQM, as well as an increase in all types of K+P, Lp, Lncarb, and Mp. However, a slight decrease in Pt can be detected in the SD, while an increase can be observed in the DD. The decreasing trends of











**Fig. 5.** The Upper Miocene sandstones of the Sava Depression under polarizing light and scanning electron microscope. **(1):** **A** — Main grains and intergranular volume of the sandstone filled with blue-dyed epoxy showing primary porosity (S3-4b, PPL); **B** — Mineral grains and rock fragments in the sandstone (with rounded carbonatic Ls in the middle; S6-2b, PPL); **C** — Secondary electron image of the sandstones with main grains and intergranular volume (S2-2c, SEM SE); **D** — Backscattered electron image of the sandstone with main grains and intergranular volume (S6-5, SEM BE); **E** — Carbonatic Ls and Lm rock fragments and other grains in the sandstone with carbonate cement in intergranular space (S4-3b, XPL); **F** — The sandstone with primary intergranular porosity under cross-polarized light (S7-1, XPL); **G** — Qz grains and Qz overgrowths on grains and within pores (S1-3a, SEM SE); **H** — Intergranular space of the sandstone filled with cement (combination of carbonate minerals and clays; S7-8a, SEM SE); and the Upper Miocene sandstones of the Drava Depression under polarizing light and scanning electron microscope. **(2):** **A** — Main grains and intergranular volume of the sandstone filled with blue-dyed epoxy showing primary porosity (D6-10a, PPL); **B** — Mineral grains and rock fragments in the sandstone (Lm on the left; D6-1b, PPL); **C** — Secondary electron image of the sandstones with main grains and intergranular volume partly filled with carbonate minerals and clays (D1-1c, SEM SE); **D** — Main grains in the sandstone and the intergranular space partly filled with carbonate cement (D4-2b, SEM BE); **E** — Mineral grains and rock fragments in the sandstone with carbonate cement in intergranular space (D7-2b, PPL); **F** — The sandstone with carbonate cement precipitated between grains (S5-1a, XPL); **G** — Backscattered electron image of the sandstone with main grains and cement (combination of carbonate minerals and clays) in intergranular space (D5-6, SEM BE); **H** — Etched garnet (D1-1c, SEM SE). Qz – quartz, Fsp – feldspar, Lm – metamorphic rock fragment, Lv – magmatic rock fragment, Ls – sedimentary rock fragment, Ms – muscovite, Bt – biotite, Chl – chlorite, Grt – garnet, Carb – carbonate cement. PPL = plane-polarized light, XPL = cross-polarized light, SE = secondary electron image, BE = backscattered electron image. Photo: M. Matošević

Q in both depressions are not overly pronounced (Suppl. Fig. S1). Coincidentally, the SD exhibits almost no changes in Qm from the W/NW towards the E/SE, while a slight increase can be observed in the DD (Suppl. Fig. S2). Increasing trends of F and Lm, as well as the decreasing trends of Lt and Ls are more prominent in the DD (Suppl. Figs. S1–S3). A more pronounced increase in Lv values was detected in D5 (Suppl. Fig. S3). M shows increasing trends from the W/NW towards the E/SE in both depressions. A very slight increasing trend of HM in the SD and a decreasing trend of HM in the DD can also be noticed. The sandstones' grain size from the W/NW towards the E/SE has a decreasing trend in the SD and an increasing trend in the DD.

An increase in Q and Lt components, as well as a decrease in F, L, Lm, and Ls components can be observed in both depressions with the increase of relative depth (Suppl. Fig. S4 and S6). However, a slightly decreasing trend of Lv can be observed in the SD, while an increasing trend can be detected in the DD (Suppl. Fig. S4 and S6). In addition, some sub-groups of Q, F, Lm, and Ls display uniform trends of changes with depth in both depressions: a decrease in K+P, TQM, and Lcarb, as well as an increase in Qp. Trends of Qm, Pt, Mp, and sub-groups of Lv do not coincide between the two depressions. There are no indications of changes with the depth of Lcarb in the SD, while the DD exhibits a decreasing trend of Lcarb. The increasing trend of Q and the decreasing trend of F are more prominent in the SD (Suppl. Fig. S4 and S6). HM shows decreasing trends with depth in both depressions (Suppl. Fig. S5 and S7); however, in the SD, the trend is more noticeable. While the SD exhibits an increase in M with depth, the DD exhibits a decrease in M (Suppl. Fig. S5 and S7). The sandstones' grain size with depth has a decreasing trend in the SD (Suppl. Fig. S5) and an increasing trend in the DD (Suppl. Fig. S7). Furthermore, a mutual connection and dependence of ratios of M and Mp values can be observed in most samples. Changes in HM in relation to changes in M, in the form of an increased share of one in relation to a decreased share of the other and vice versa, were noticed in the samples as well (Suppl. Fig. S5 and S7). A strong bond between

the HM component and grain size is also confirmed in the sandstones (Suppl. Fig. S5 and S7) – the smaller the average grain size of the sample, the higher the concentration of HM.

#### *Matrix and cement composition*

For samples from both depressions, the intergranular volume of the sandstones is either unfilled, i.e., with empty pores between grains, or is partly or fully filled with carbonate cement and a fine-grained matrix, as well as different types of clay minerals (Fig. 5).

SEM and EDS analyses confirmed the matrix and cement composition of the sandstones. Carbonate cement largely matches calcite and ankerite (Fig. 5). The fine-grained matrix occurs as a combination of carbonate minerals (including calcite, dolomite, and ankerite), quartz, mica, and feldspar (Fig. 5). Different types of detrital and authigenic clay minerals were determined as well in the samples – kaolinite, illite, chlorite, and different types of interstratified clays (Fig. 5). Silica cement was also frequently observed, usually in the form of quartz overgrowths on the main quartz grains (Fig. 5).

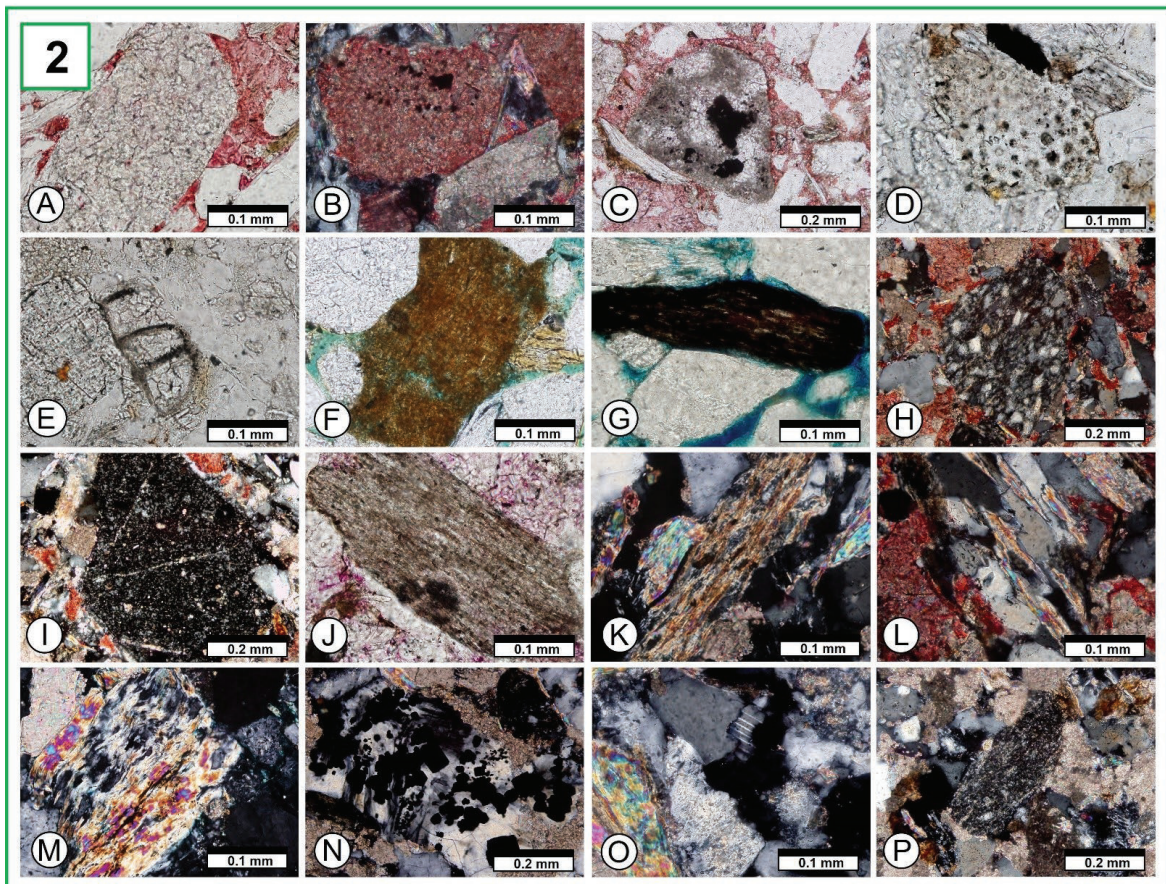
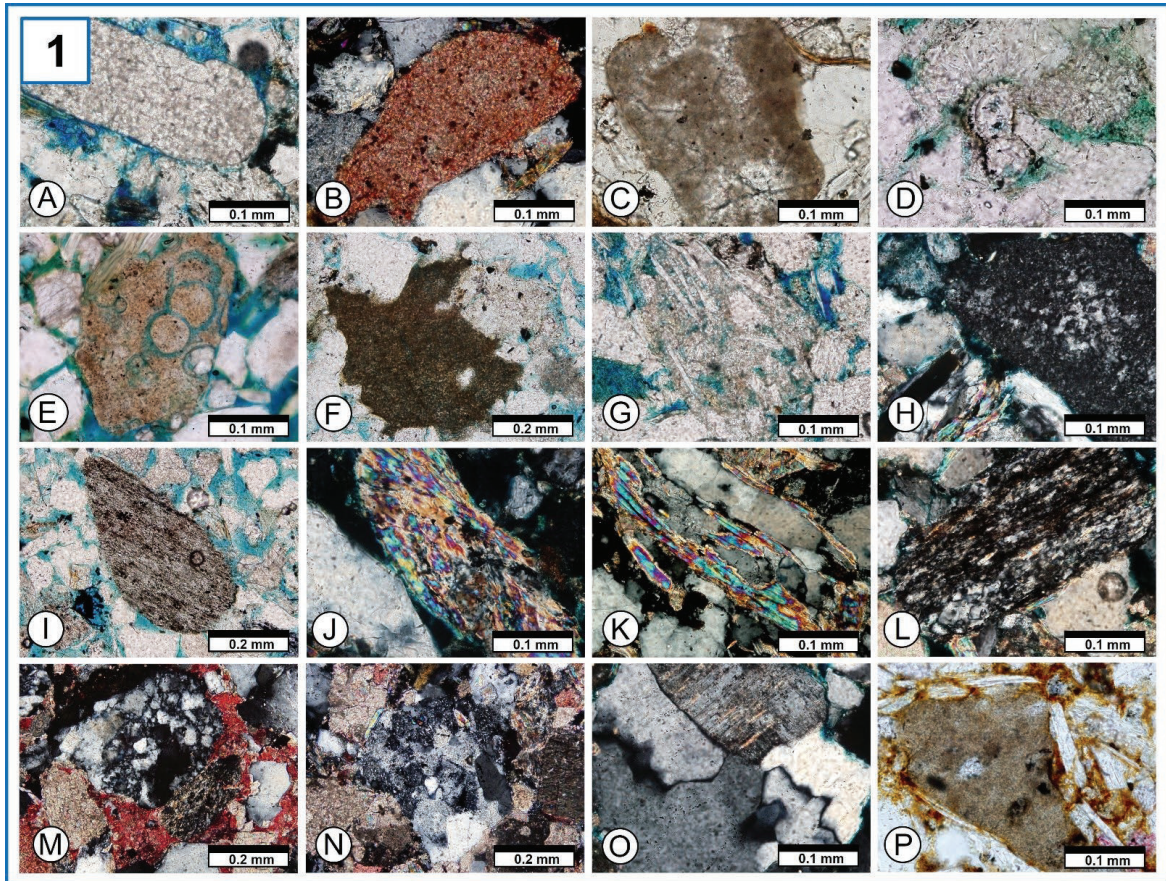
## **Discussion**

The Upper Miocene sandstones from the SD and DD are compositionally very similar. The sandstones' most abundant rock fragments are carbonate sedimentary rocks, mica schists, quartzites, gneisses, and granitoid rocks. Although the composition of the HM assemblage could have been modified by diagenesis, similar HM association of the Upper Miocene sandstones was earlier detected on the surface and in deep exploration wells in the NCB (Šćavničar 1979; Kovačić & Grizelj 2006). Therefore, it can be proposed that HM, along with rock fragments, reflects the composition of the source area. The majority of HM in the sandstones could have been derived from two groups of parent rocks: low and medium-grade dynamo-thermal metamorphic and felsic to











**Fig. 7.** Rock fragments in the Upper Miocene sandstones from the Sava Depression with sedimentary (A–H), metamorphic (I–M), and magmatic rock fragments (N–P). **(1):** **A** — Recrystallized dolomite (S4-4a, PPL); **B** — Recrystallized limestone (sparite) stained with Alizarin Red S (S7-3b, PPL); **C** — Limestone of packstone/grainstone type (S6-7, PPL); **D** — Recycled carbonatic bioclast (planktonic foraminifera shell) (S7-3b, PPL); **E** — Fossiliferous limestone (mudstone/wackestone; S5-1, PPL); **F** — Deformed unstable mudrock (marl rip-up clast; S6-2b, PPL); **G** — Siltstone (S4-3a, PPL); **H** — Chert (S1-3b, XPL); **I** — Metapelite/slate (S7-1, PPL); **J** — Fragment of mica schist under cross-polarized light (S5-2, XPL); **K** — Gneiss/schist (S7-2b, XPL); **L** — Microcrystalline mica schist (S1-3a, XPL); **M** — Quartzite (S1-1, XPL); **N** — Granitoid fragment (S7-5b, XPL); **O** — Granite (S4-5a, XPL); **P** — Altered volcanic rock (S6-1, PPL); and rock fragments in the Upper Miocene sandstones from the Drava Depression with sedimentary (A–I), metamorphic (J–N), and magmatic rock fragments (O & P). **(2):** **A** — Recrystallized dolomite (D5-2b, PPL); **B** — Recrystallized limestone (mudstone) stained with Alizarin Red S and dolomite (D7-2b, XPL); **C** — Limestone of grainstone type (D3-3, PPL); **D** — Recycled undetermined carbonatic bioclast (D4-2a, PPL); **E** — Fragment of carbonatic bioclast (bentic foraminifera shell; D5-1b, PPL); **F** — Deformed unstable rip-up clast of mudrock (claystone/marl; D2-1d, PPL); **G** — Mudrock rich in organic matter (shale; D7-1b, PPL); **H** — Siltstone under cross-polarized light (D4-3a, XPL); **I** — Chert (D3-1b, XPL); **J** — Metapelite/slate (D5-1a, PPL); **K** — Mica schist under cross-polarized light (D3-5a, XPL); **L** — Gneiss/mica schist (D7-2b, XPL); **M** — Fragment of mica schist (D2-3c, XPL); **N** — Quartzite with euhedral pyrite minerals (D4-5a, XPL); **O** — Granitoid rock fragment (D3-5b, XPL); **P** — Altered volcanoclastic rock (tuff; D2-3d, XPL). PPL = plane-polarized light, XPL = cross-polarized light. Photo: M. Matošević

have provenance mainly from carbonate, followed by low to medium-grade metamorphic and, to a lesser extent, granitoid rocks.

Detected small-scale originals of source rocks point toward recycled carbonate-clastic, axial belt metamorphic complex, and continental-block orogenic provenance (Garzanti 2016). Based on the QFL or QtFL modal compositions, according to Dickinson (1985) and Weltje (2006), the analyzed sandstones correspond to the tectonic setting of recycled orogen (subduction complex or fold-thrust belt; Fig. 10). This complex tectonic setting is the result of collisional processes that put together different tectonic units of the Alpine–Carpathian–Dinaridic orogenic system, including the basement of the PBS, which all represent potential source areas of the material in the Upper Miocene sandstones of the NCB. Furthermore, the tectonic units of Schmid et al. (2008, 2020) and the geological map of Europe by Asch (2003) were used to describe the regional geological situation (Fig. 1). The Southern Alps are mainly composed of Mesozoic carbonates, while the Eastern Alps and the Western Carpathians (ALCAPA mega-unit) are comprised of both Mesozoic carbonates and Proterozoic to Paleozoic low to medium-grade metamorphic rocks, with Paleozoic granitoids mainly confined to the Western Carpathians. Similarly, the Tisza mega-unit is predominantly composed of Paleozoic low to medium-grade metamorphic rocks and granitoids, with Upper Paleozoic and Mesozoic siliciclastic and carbonate sedimentary rocks. The Eastern and Southern Carpathians (Dacia mega-unit) are comprised of both Paleozoic medium to high-grade metamorphic rocks and Mesozoic ophiolitic rocks. At the same time, the Dinarides are mainly composed of Mesozoic carbonates (High Karst Unit and Dalmatian zone), partially metamorphosed Paleozoic siliciclastic rocks, and Mesozoic ophiolites (Pre-Karst Unit and Adria-derived nappes with Western Vardar Ophiolitic Unit). The innermost unit of the Dinarides is the Sava Zone, which is composed of various igneous, metamorphic, and sedimentary rocks of the Late Cretaceous. Additionally, various Mesozoic and younger siliciclastic flysch basins, as well as younger granitoid intrusions, volcanic rocks, volcanoclastics, and sedimentary rocks of the Late Cretaceous, Paleogene, and

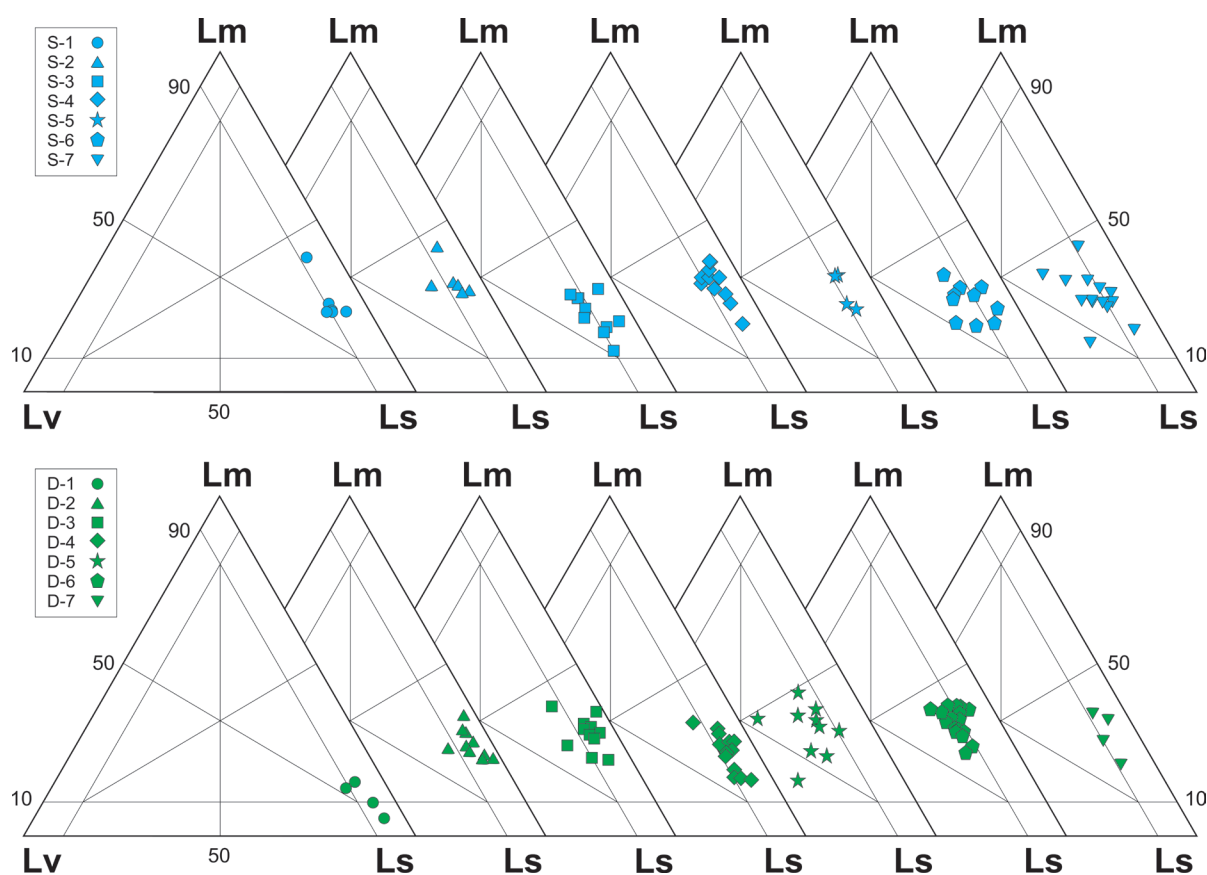
Neogene ages can be detected in different tectonic units in this region.

Parts of the PBS basement outcropping on the surface in the NCB today (e.g., Kalnik, Medvednica, Moslavačka Gora, Slavonian Mts.; e.g., Pamić & Lanphere 1991; Horvat et al. 2018; Starijaš et al. 2010; Biševac et al. 2010, 2011; Balen & Petrinc 2011; Balen et al. 2013, 2020; Belak et al. 2022; Schneider et al. 2022) or found deep in exploration wells in the depressions inside the NCB (e.g., Pamić 1986, 1999; Matošević et al. 2015; Matošević & Šuica 2017; Šuica et al. 2022a,b) also include the aforementioned types of rocks and belong to different tectonic units. They mostly incorporate tectonized magmatic, metamorphic, and sedimentary rocks belonging to the Tisza mega-unit, as well as the Sava zone and the Adria-derived nappes with Western Vardar Ophiolitic Unit of the Dinarides.

The massive inflow of detritus into the Lake Pannon during the Late Miocene (Bérczi et al. 1988; Juhász 1994; Magyar et al. 2013; Pavelić & Kovačić 2018; Anđelković & Radivojević 2021) produced considerable thicknesses of the sediments in the SD and DD (Ivković et al. 2000; Saftić et al. 2003; Kovačić et al. 2004; Kovačić & Grizelj 2006; Pavelić & Kovačić 2018; Malvić & Velić 2011). It is thus not likely that local parts of the NCB basement, with their relatively small volumes, served as the main sources of the material. Some islands within the lake and peninsulas that surrounded it at some point could have contributed to a smaller amount of the detritus (Bérczi 1988; Kovačić & Grizelj 2006; Sztanó et al. 2015). It is, however, not likely that these sources contributed greater amounts of detritus, since the lake reached its largest extension around 10.5 Ma (Magyar et al. 1999). Moreover, due to a high rate of subsidence of the basin and wet climate, great water depths developed (of up to 1000 m locally) leaving all the islands under water (Pogácsás 1984; Sztanó et al. 2013; Balázs et al. 2018). Finally, the Upper Miocene sediments cover some parts of the mountains in the NCB today and therefore signal their major uplift after the Late Miocene, i.e., during the later compressional phase of the basin evolution (e.g., Pavelić 2001; Tomljenović & Csontos 2001; Márton et al. 2002; Pavelić & Kovačić 2018).

The Lower and Middle Miocene rocks stratigraphically underlying the Upper Miocene sediments in northern Croatia include different types of sediments deposited in freshwater and marine environments, including fossiliferous carbonates and different types of siliciclastic sediments, as well as volcanic and volcanoclastic rocks (Tadej 2011; Kovačić et al. 2011; Matošević et al. 2015, 2019a; Pavelić & Kovačić 2018; Hemitz Kučenjak et al. 2018; Avanić et al. 2018, 2021; Brlek et al. 2020, 2023; Grizelj et al. 2020, 2023; Bigunac 2022; Kopecká et al. 2022; Premec Fuček et al. 2022; Sremac et al. 2022). Similar types of Lower and Middle Miocene rocks were also found in other basins of the PBS (cf. Rybár et al. 2016, 2019; Sant et al. 2020; Nyíri et al. 2021; Bordy & Sztanó 2021; Csibri et al. 2022). As a consequence, the possible local

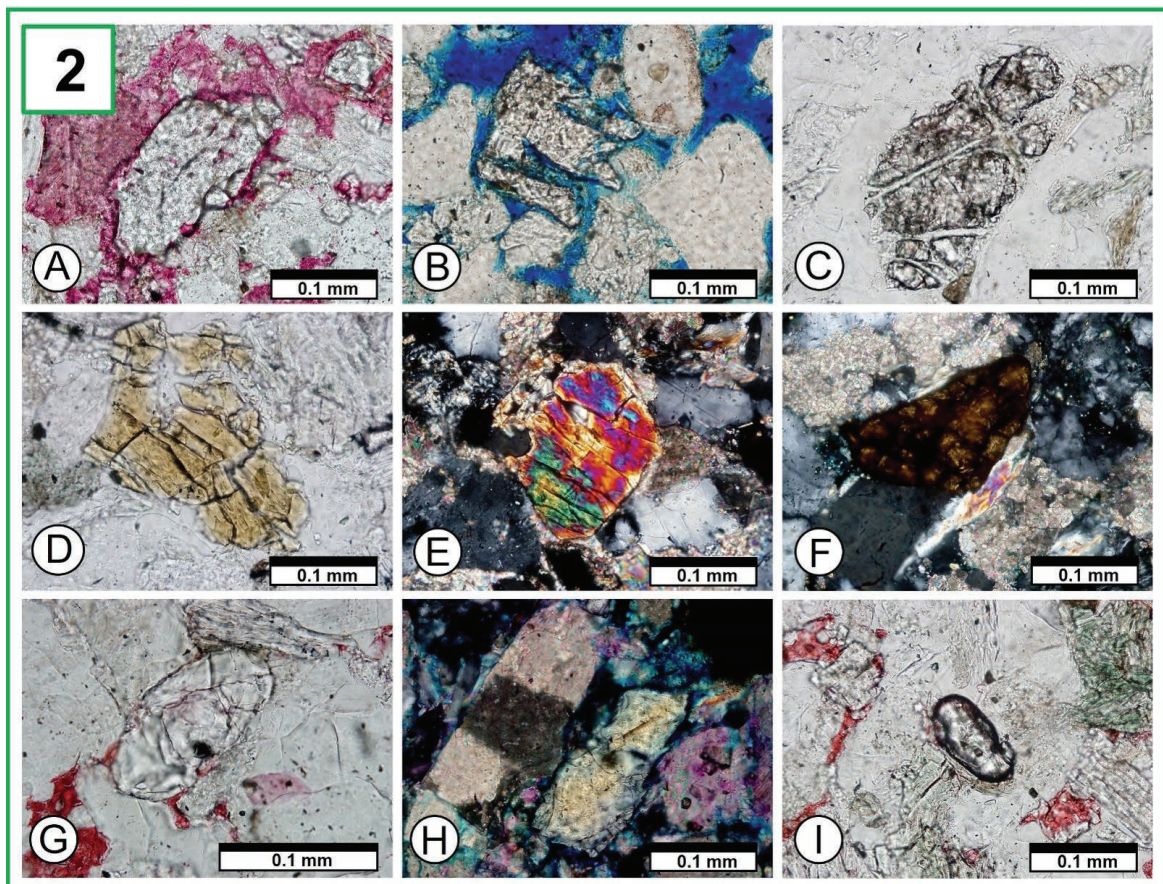
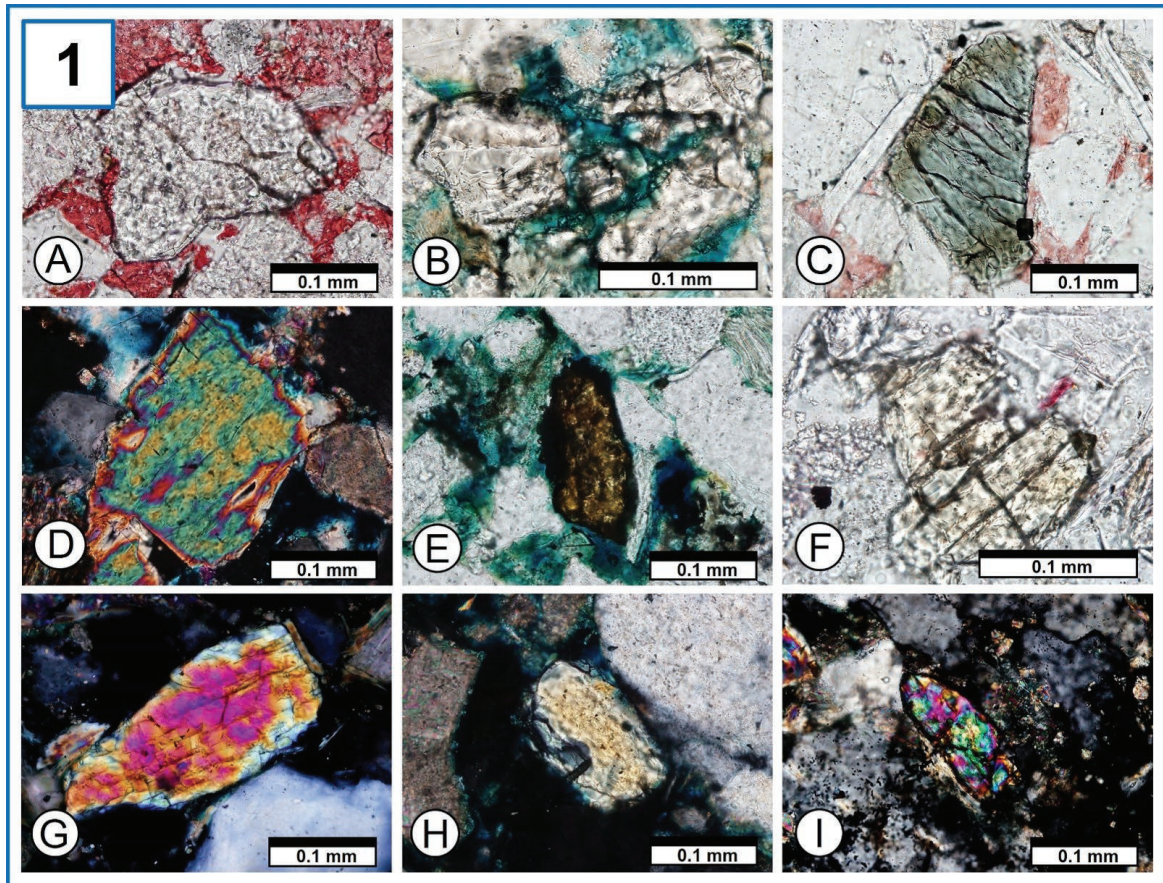
origin of some of the carbonatic fragments (e.g., micritic limestones and recycled carbonate bioclasts) or volcanic/volcanoclastic rocks in the Upper Miocene sandstones, which were possibly eroded from the underlying Lower and Middle Miocene rocks in the Late Miocene, cannot be completely ruled out. The same applies to siliciclastic sediments (e.g., mudrocks in the form of intrabasinal rip-up clasts) that could have been delivered from different parts of the PBS into the depressions and be partly of the same Late Miocene age. Nevertheless, a large share of fragments of carbonates in the sandstones does not bear any physical (textural) resemblance to those of the Early and Middle Miocene. They are principally non-fossiliferous and largely recrystallized. Even carbonatic bioclasts are found extremely rarely in the samples.



**Fig. 8.** LmLvLs diagrams of the Upper Miocene sandstones from exploration wells of the Sava (S1–S7) and Drava Depression (D1–D7) according to Garzanti (2019).

**Fig. 9.** Heavy mineral association in the Upper Miocene sandstones from the Sava Depression. (1): **A** — Garnet with inclusions and mechanical fractures surrounded by calcite cement (S1-1, PPL); **B** — Skeletal garnet (S1-3a, PPL); **C** — Rounded triangular cross-section of color-zoned tourmaline with irregular fractures (S7-3a, PPL); **D** — Tourmaline under cross-polarized light (S3-4b, XPL); **E** — Subhedral rutile (S2-2a, PPL); **F** — Epidote (S6-7, PPL); **G** — Clinozoisite (S5-1, XPL); **H** — Staurolite with inclusions (S5-1, PPL); **I** — Fractured zircon with inclusions under cross-polarized light (S7-3b, XPL); and heavy mineral association in the Upper Miocene sandstones from the Drava Depression. (2): **A** — Garnet within calcite-filled intergranular space (D4-4a, PPL); **B** — Etched garnet due to dissolution (D6-4b, PPL); **C** — Fractured skeletal garnet (D3-5c, PPL); **D** — Fractured and partly dissolved tourmaline (D3-1b, PPL); **E** — Trigonal cross-section of tourmaline under cross-polarized light (D4-2b, XPL); **F** — Fractured rutile under cross-polarized light (D2-3b, XPL); **G** — Prismatic apatite (D5-3, PPL); **H** — Staurolite (D6-5, XPL); **I** — Small zircon crystal with inclusions (D4-1c, PPL). PPL = plane-polarized light, XPL = cross-polarized light. Photo: M. Matošević







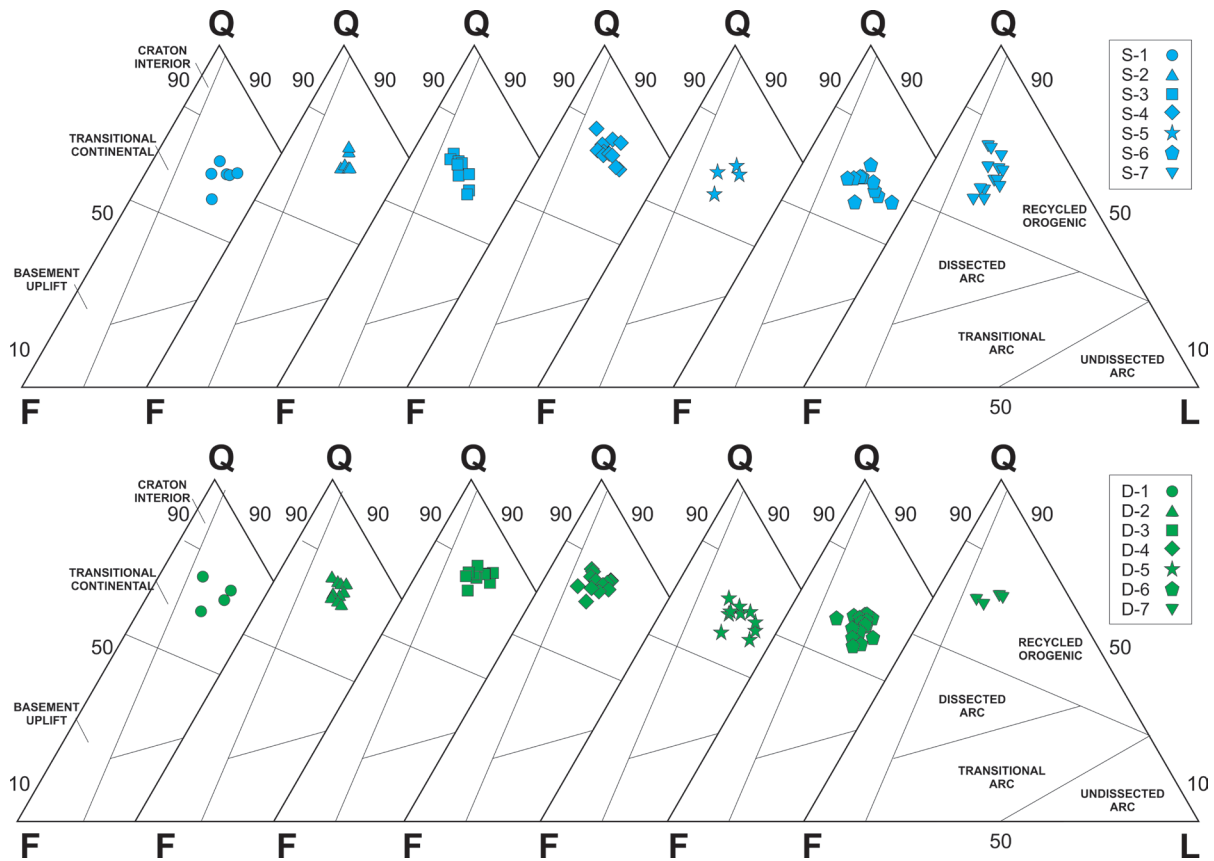


Fig. 10. QFL of the Upper Miocene sandstones from exploration wells of the Sava (S1–S7) and Drava Depression (D1–D7) showing their tectonic setting diagrams according to Dickinson (1985).

It is therefore possible to conclude that the fragments of carbonates correspond rather to those of the Paleozoic and Mesozoic age found in the surrounding area of the NCB and the PBS as a whole.

Furthermore, fragments of mudrocks, on average, form less than 0.7 %, while extrusive (volcanic) rocks form less than 0.3 % in the total modal composition. They are, in comparison to other rock fragments, insignificant for determining and interpreting main sources. According to von Eynatten (2003) and his modified conceptual model of the petrographic data based on Weltje et al. (1998), the weathering index for the Upper Miocene sandstones in the SD and DD corresponds to the field value '0' and indicates the early stages of weathering of plutonic and metamorphic/sedimentary source rocks in the high mountain relief area.

Earlier studies have shown that accumulated sediments in the PBS had mostly been eroded from the Eastern Alps and Carpathians between the Late Miocene (ca. 11–10 Ma) and Late Pliocene (Ščavničar 1979; Kuhlemann et al. 2002; Kovačić et al. 2004; Kovačić & Grizelj 2006; Magyar et al. 2013; Sebe et al. 2020). Nevertheless, it is often difficult to distinguish the Alpine and Carpathian sediment provenance, since they share a similar evolution history with similar bed-rock composition (Arató et al. 2021). Since the Upper Miocene

sandstones of the SD and DD do not contain any significant amounts of mafic and ultramafic rocks or radiolarites, nor any heavy minerals derived from these rocks (e.g., chrome spinel), the Dinarides, as well as the Southern and the Eastern Carpathians can be ruled out as main source areas (cf. Lužar-Oberiter et al. 2009, 2012; Ustaszewski et al. 2009, 2010). The exclusively Western Carpathian provenance would have resulted in a larger content of volcanic or volcanoclastic material (e.g., Seghedi & Downes 2011). Based on the previously mentioned lithic fragments determined in the sandstones, the Eastern Alps are proposed as the most probable major source area. Even studies of the present-day Drava and Mur River sediments, those sourced in the Eastern Alps with dominantly metamorphic lithologies, show similarities in the HM composition with the Upper Miocene sandstones from the SD and DD, with garnet as the major component, followed by epidote, amphibole, rutile, apatite, tourmaline, and titanite (Arató et al. 2021).

Progradation of the Upper Miocene sandstones' clinoforms from seismic profiles indicates high material inflow from N/NW and W directions into the SD and DD (Ivković et al. 2000; Saftić et al. 2003; Magyar et al. 2013; Špelić et al. 2019; Sebe et al. 2020). The same general directions were detected on the surface in the NCB (Kovačić & Grizelj 2006; Kovačić

et al. 2011). As inferred from the seismic data, parts of the DD depositional system are also locally characterized by sets of clinoform sediments with shelf-margin slope progradation from a N/NE towards S/SW direction. Similar findings in the Bjelovar sub-depression (Fig. 3) were recently demonstrated by Ružić (2021). Altogether, these findings point towards the Alps (Eastern Alps and potentially Southern Alps) as major source areas of the Upper Miocene sandstones, with a minor addition from the Carpathians (Western Carpathians and potentially Eastern Carpathians; Šćavničar 1979; Kovačić & Grizelj 2006; Kovačić et al. 2011; Magyar et al. 2013; Sztanó et al. 2013).

Additionally, there is no progradation of the sandstones' clinoforms observed from seismic profiles indicating main material inflow from S, SW, or SE directions into the SD and DD. Compositional equivalence, which was detected by petrographic analyses of the samples and paleotransport of the sandstones, implies that the two depressions were connected in the Late Miocene with no significant topographical obstacles for the inflow of the sandy detritus. This additionally supports the concept that the main uplift of the mountains in the NCB occurred during the Pliocene and Quaternary (Prelogović et al. 1998; Tomljenović & Csontos 2001; Pavelić 2001; Márton et al. 2002; Saftić et al. 2003; Pavelić & Kovačić 2018; Kurečić et al. 2021).

Almost all trends of relative changes in the sandstones' modal composition with the change of geographic position and depth are correspondent between the two depressions. They can be associated with characteristics and variations of the exhumation in the source area – in the form of potential primary erosion of covering sedimentary (carbonate) rocks and subsequent erosion of underlying metamorphic and magmatic (plutonic) rocks. This is indicated by the decrease of L and Lt values, supported by the decrease of Ls values, from the W/NW to the E/SE in both depressions, i.e., in the direction of the progradation of the sandstones. The same assumption is indicated by the increase of Lv and Lm values, supported by the increase of F values, from the W/NW to the E/SE, but also by the decrease of L and F values with depth. The observed trends could additionally be caused by the mixing of detritus before the final influx in the basin (e.g., a possible admixture of material from local islands and peninsulas or other rivers and tributaries from the northern and north-eastern parts of the PBS, such as the paleo-Danube and paleo-Tisza; e.g., Magyar et al. 2013). Further causes could also embrace weathering and alteration processes (including dissolution of carbonate rock fragments with time and length of the transport from source to sink), as well as the consequences of diagenetic processes within the basin (including stronger or weaker resistance of some grains to burial depth). The higher increase of Lv in D5 could indicate an increased inflow of plutonic (granitoid) detritus from other, possibly local source(s) or a possible existence of a minor geographic (topographical) barrier for the main inflow of detritus in this area (e.g., an island or an underwater elevation of the lake bottom). Changes in Ls with depth are primarily the result of diagenesis since they strongly

depend on deformation, alteration, and dissolution during the burial. The same applies to the HM component – a significant decrease or complete absence of some of the minerals (e.g., epidote, clinozoisite, zoisite, staurolite, amphibole) in deeper intervals of the wells (generally deeper than 1000 m) is a direct consequence of their dissolution with depth, i.e., instability with increasing temperature, pressure, and interaction with pore fluids (Pettijohn 1957; Šćavničar 1979; Morton & Hallsworth 2007; Andò et al. 2012; Garzanti 2016). Some of the grains (e.g., F, Lv, and Lm, including their sub-groups) have been better preserved with depth in the DD than in the SD. The prevention of deformation and chemical alterations of the grains is a consequence of early carbonate cement precipitation in intergranular space in deeper parts of the DD. Similarly, changes in Q values with depth are largely the result of diagenesis, since quartz is more resistant to burial processes than other grains. Seeing that the sandstones from both depressions are exclusively very fine and fine-grained, it can be assumed that the grain size does not play a significant role in the distribution of trends of individual components. The changes in the HM share in relation to changes in the M share strongly support the theory of the influence of hydraulic sorting on the deposition of grains of the same provenance (Casalho & Fradique 2007; Kompar 2007; Garzanti et al. 2008; Garzanti 2016).

Additionally, the NCB Upper Miocene sandstones largely differ from those from the Lower and Middle Miocene, which are subordinate oil and gas reservoirs in the area (Lučić et al. 2001; Saftić et al. 2003; Pavelić & Kovačić 2018). The sandstones from the two depressions are fine-grained, generally well- to moderately well-sorted, and texturally mature, in accordance with the results of previous studies (Kovačić et al. 2004, 2011; Kovačić & Grizelj 2006). They contain fragments of magmatic, metamorphic, and sedimentary rocks, including fragments of extrabasinal carbonates from further regional sources. The Lower and Middle Miocene sandstones are, by contrast, principally of coarser grain sizes, poorly-sorted, and texturally less mature. They were formed during the syn-rift phase of the PBS development and are frequently interbedded with breccias and conglomerates generated along normal faults associated with the initial rifting of the NCB (Pavelić et al. 1998; Saftić et al. 2003; Pavelić 2005; Matošević et al. 2015; Brlek et al. 2016; Pavelić & Kovačić 2018; Bigunac 2022). They are intermittently interbedded with volcanoclastic sediments and mudrocks (e.g., fossiliferous marls) associated with marine transgressions occurring during the existence of the Central Paratethys (Matošević et al. 2019a; Brlek et al. 2020, 2023; Marković et al. 2021; Grizelj et al. 2020, 2023; Premec Fuček et al. 2022; Sremac et al. 2022). As can be observed from seismic profiles, the paleo-transport of their detritus is from locally uplifted parts of the NCB basement with no preferable main transport directions, primarily perpendicular to the normal faults. The Lower and Middle Miocene sandstones form facies of alluvial fans and related fluvial and deltaic environments, with detritus being further transported into deeper parts of the depressions (Tadej 2011;

Pavelić & Kovačić 2018; Bigunac 2022). Aside from a much larger ratio of fragments of extrusive volcanic and volcanoclastic rock, they also include a much larger ratio of intrabasinal carbonatic rock fragments and abundant and diverse quantities of carbonate bioclasts (this applies especially to the Middle Miocene sandstones formed in marine environments; e.g., Brlek et al. 2016, 2023; Kovačić et al. 2011; Matošević et al. 2019a; Sremac et al. 2022; Premec Fuček et al. 2022; Grizelj et al. 2023). The findings of this study demonstrate that this is atypical for the Upper Miocene sandstones.

### Conclusions

For the most part, the Upper Miocene sandstones from the SD and the DD in the NCB correspond to feldspatho–litho–quartzose sandstones and are interpreted to have been derived from a provenance area of a recycled orogen. Sandstones' modal compositions and detailed analyses of rock fragments and heavy minerals suggest an equivalent external source terrain without major local influences – parent rocks consist of carbonates, metamorphic rocks, and plutonic rocks, mainly of the Alpine–Carpathian fold belt (ALCAPA tectonic mega-unit). The uniform composition of the sandstones from both depressions suggests time-identical processes of source rock erosion, sandy material transport, and deposition, mainly as a relatively fast and massive inflow into the basin during the Late Miocene. Both depressions show an increase in the F, Lm, and Lv components, and a decrease in the Q, L, Lt, and Ls components from the W/NW towards the E/SE. These changes in the sandstones' composition reflect a combination of changes in the source area (erosion of parent rocks), alterations during transport, and mixing of the detrital material prior to deposition. An increase in the Q, Qp, and Lt components, as well as a decrease in F, K+P, L, Lm, TQM, Ls, Lncarb, and HM with depth was noticed in both depressions. In general, these changes in sandstone composition reflect diagenetic factors, such as the stability or instability of individual components to post-depositional alteration. Diagenetic alteration can be explained by compaction, the progressive increase of temperature and pressure with burial depth, and the presence of pore fluids. The Upper Miocene sandstones from the NCB differ in terms of their texture and composition from the Lower and the Middle Miocene sandstones in this area, which are of more local provenance. The results of the study show that no major morphological barriers were present between the SD and DD that could have obstructed the inflow of detrital material via rivers and deltas into the Lake Pannon. And so, the main uplift of the mountains in the NCB is inferred to have taken place after the Upper Miocene, which is consistent with previous studies.

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

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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., ŠČAVNIČAR, 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 ZELENKA 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 & MANDIĆ, 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; KOVAČIĆ 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<sup>2</sup> in northern Croatia and belonging to the Pannonian Basin, surrounded by the Alps, Carpathians, and Dinaride mountains (Fig. 1A). The North Croatian Basin



**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.

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).

The Miocene epoch in the Pannonian Basin is closely associated with the dynamic history of Central Paratethys, 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 & MANDIĆ, 2008; TER BORGH et al., 2013; KOVAČIĆ 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; KOVAČIĆ 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; MANDIĆ 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; TOMLJENIĆ & 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<sub>3</sub>COOH) solution, followed by treatment with 15% H<sub>2</sub>O<sub>2</sub> 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<sup>3</sup>), 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.

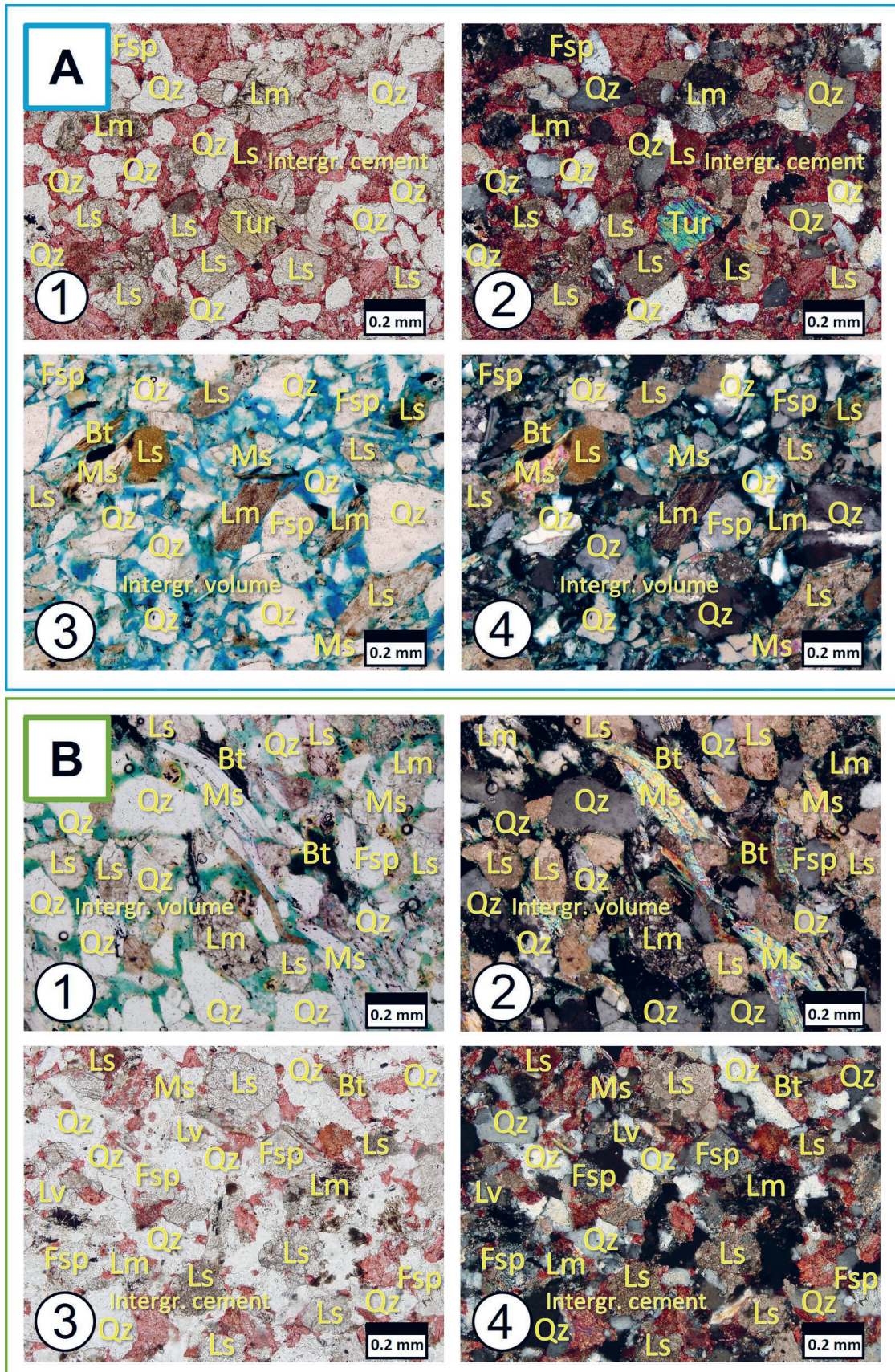
### 3.2. Garnet Raman Spectroscopy

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<sup>-1</sup> 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™ 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; KARAMELAS 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

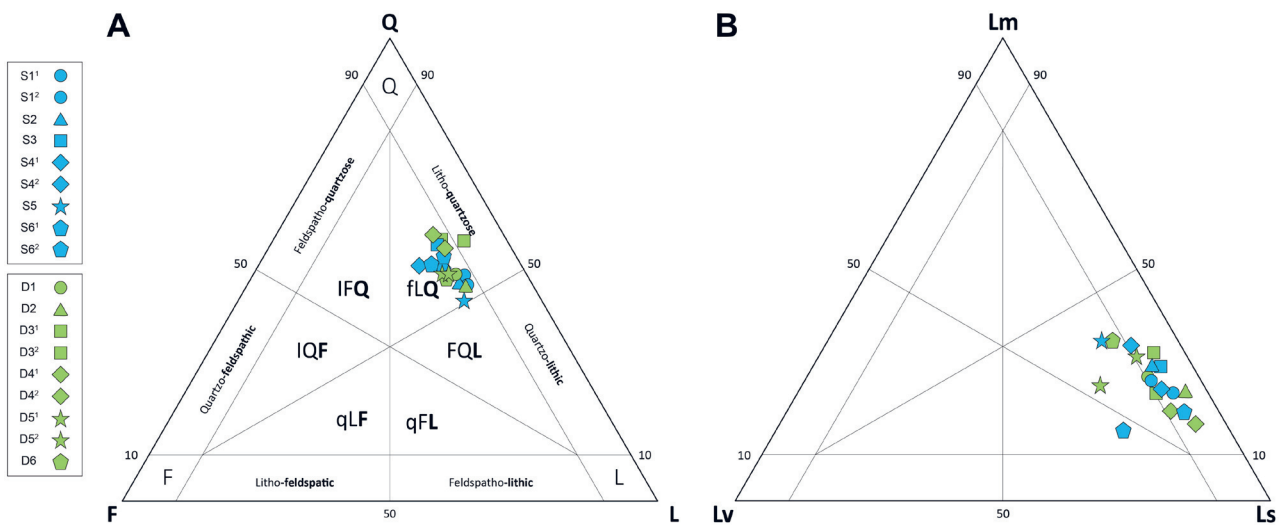
Whole rock chemical analyses on all 18 sandstone samples from the Sava and Drava depression were conducted at Bureau Veritas Mineral Laboratories (Vancouver, Canada) by lithium metaborate/tetraborate fusion and nitric acid digestion. Major elements were quantified using inductively-coupled-plasma emission 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', 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', 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 (JANOŠEK 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:

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

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 (ŠČAVNIČAR, 1979; MATOŠEVIĆ et al., 2023).

## 4. RESULTS

### 4.1. Petrography

The studied sandstones are very fine to fine grained (ranging from 80 to 130  $\mu\text{m}$  in the Sava depression and from 100 to 220  $\mu\text{m}$  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

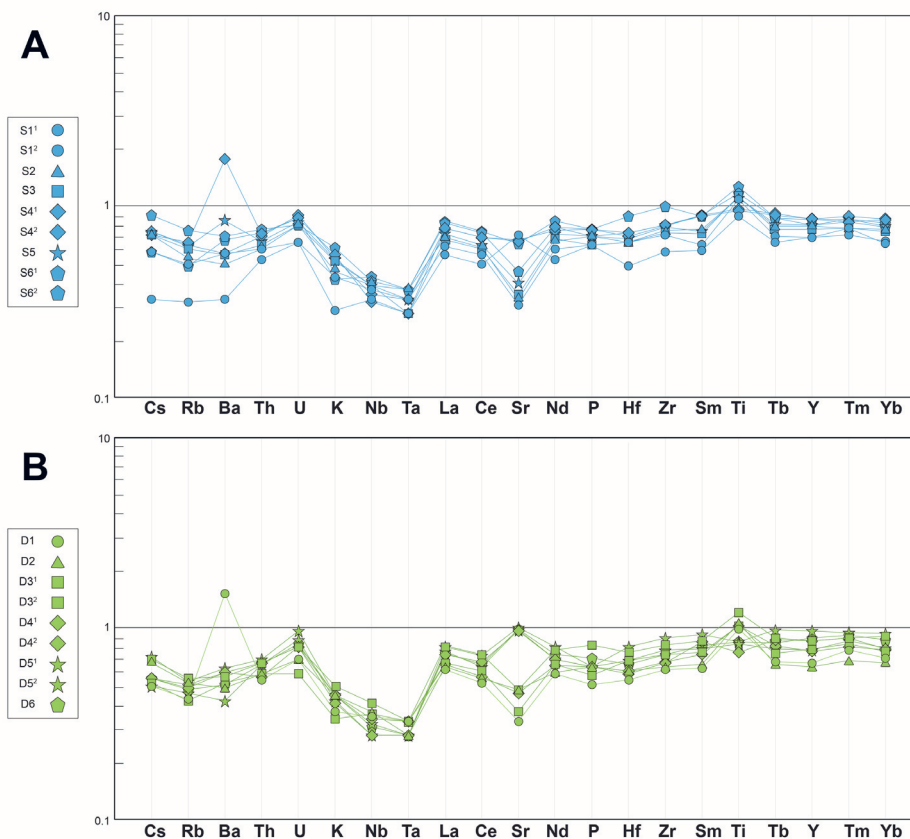
Heavy mineral concentration (HMC) ranges from 0.2% to 9.6% (average: 4.8%) in the Sava depression and from 3.6% to 9.1% (average: 5.5%) in the Drava depression (STab. 2). The very poor to rich tHM suites ( $0.1 \leq \text{tHMC} \leq 5$ ) are garnet-dominated (28-79%, average 59% in the Sava depression, 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). Clinzoisite 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

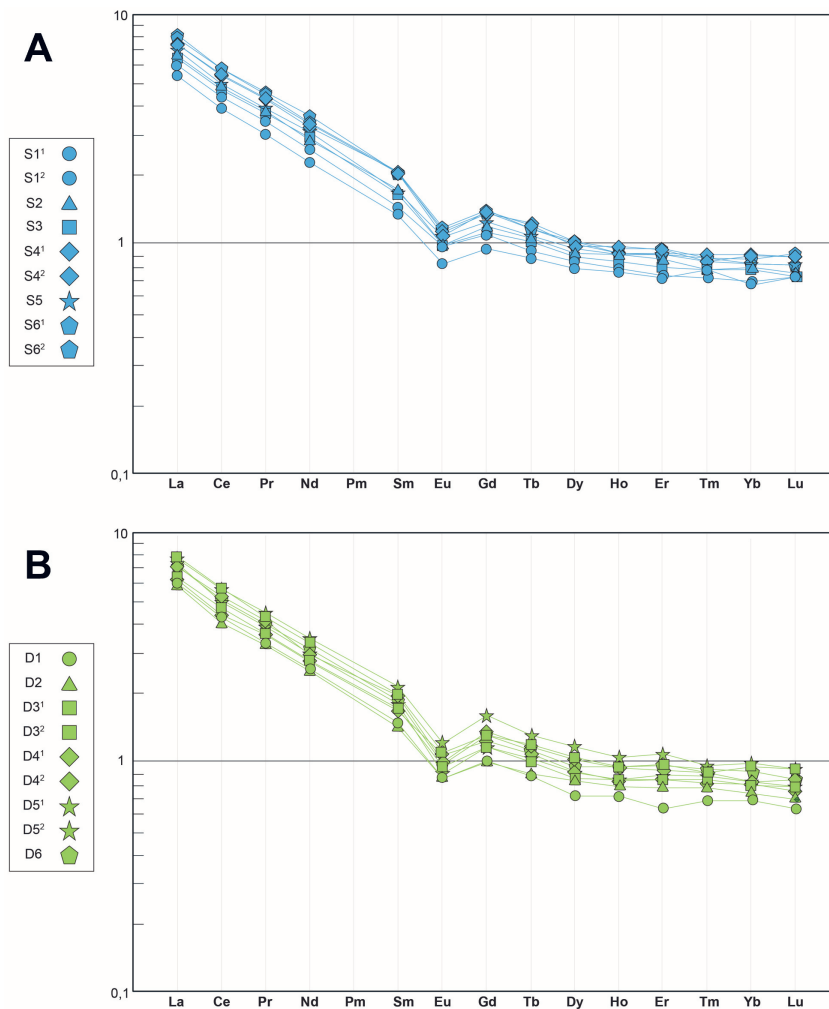
The studied garnets are mostly pyralspites (Suppl. 3). Almandine is the main component in the majority of the analyzed



**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

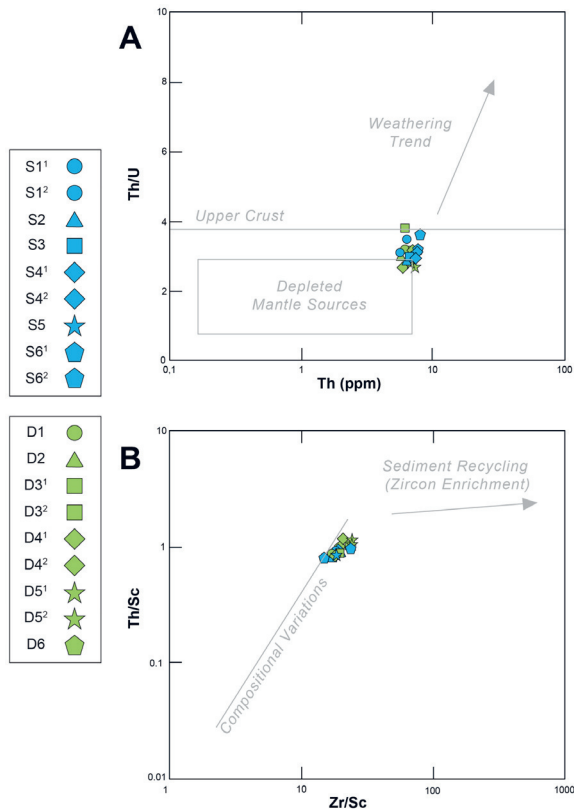
The studied samples contain 41-63% SiO<sub>2</sub>, 5.7% to 12% Al<sub>2</sub>O<sub>3</sub> (generally lower in the Drava depression), and 7.6% to 22% CaO (generally higher in the Drava depression) (Suppl. 4). Al<sub>2</sub>O<sub>3</sub> correlates best with Fe<sub>2</sub>O<sub>3</sub> (r 0.94) and K<sub>2</sub>O (r 0.92), moderately well with Na<sub>2</sub>O (r 0.64) and TiO<sub>2</sub> (r 0.62), weakly with SiO<sub>2</sub> (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<sub>2</sub>O<sub>3</sub>, Al<sub>2</sub>O<sub>3</sub>, and TiO<sub>2</sub>, 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<sub>2</sub>O 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<sub>cn</sub>/Yb<sub>cn</sub> 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  $(Al/E)_{\text{sample}} / (Al/E)_{\text{standard}}$  (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.



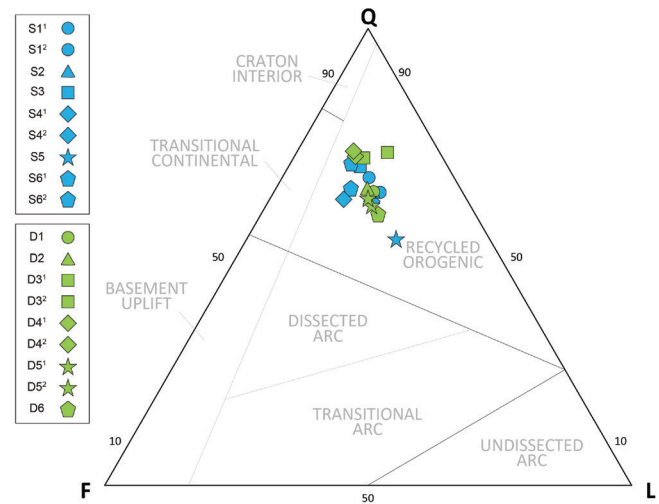


**Figure 7.** A – Plot of Th/U versus Th indicating insignificant intensity of weathering for the Upper Miocene sandstones from the Sava and Drava depressions, with values similar to the upper crust, according to McLENNAN et al. (1993). B – Plot of Th/Sc versus Zr/Sc indicating negligible influence from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

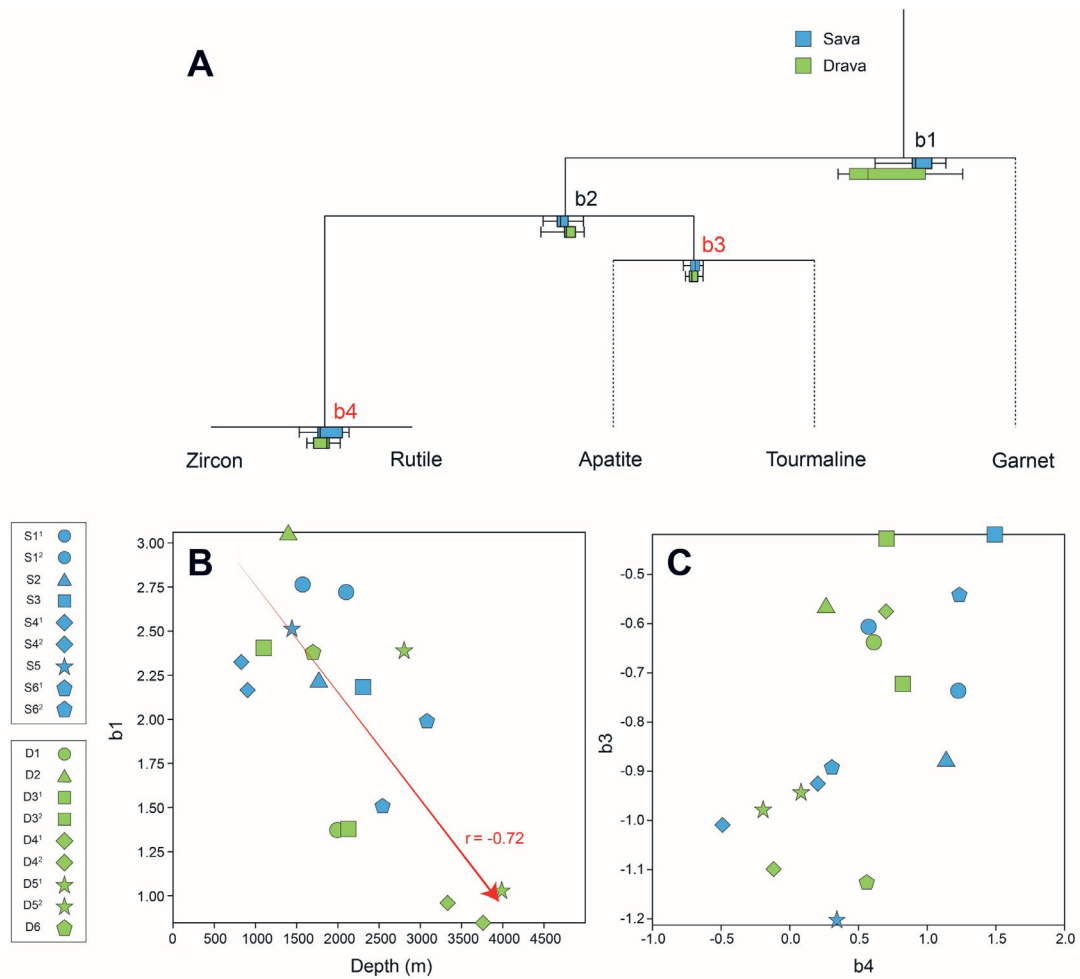
The studied Upper Miocene sandstones from the Sava depression and the Drava depression are mostly feldspatho-litho-quartzose carbonaticlastic indicating provenance from cover strata with contribution from metamorphic and igneous rocks (Fig. 2 & Fig. 3). The virtually identical composition of the Sava 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



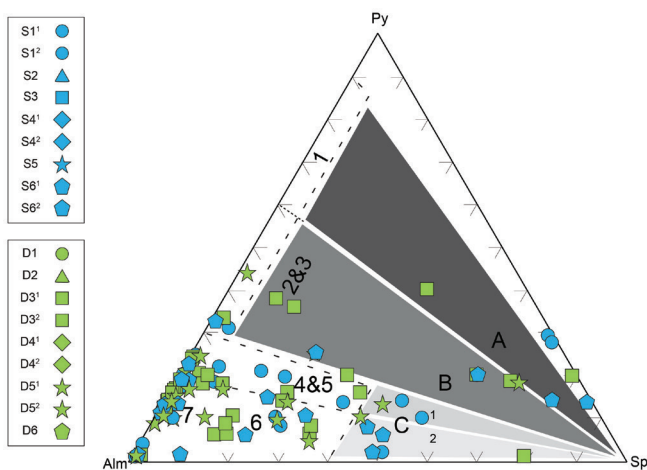
**Figure 8.** Tectonic setting discrimination diagram of the Upper Miocene sandstones from the Sava and Drava depressions according to DICKINSON (1985).

al., 2000; SAFTIĆ et al., 2003; KOVAČIĆ et al., 2004; KOVAČIĆ & GRIZELJ, 2006; SEBE et al., 2020; ŠPELIĆ et al., 2023).

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 ( $15 \pm 8$ ) 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 metamorphic 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)



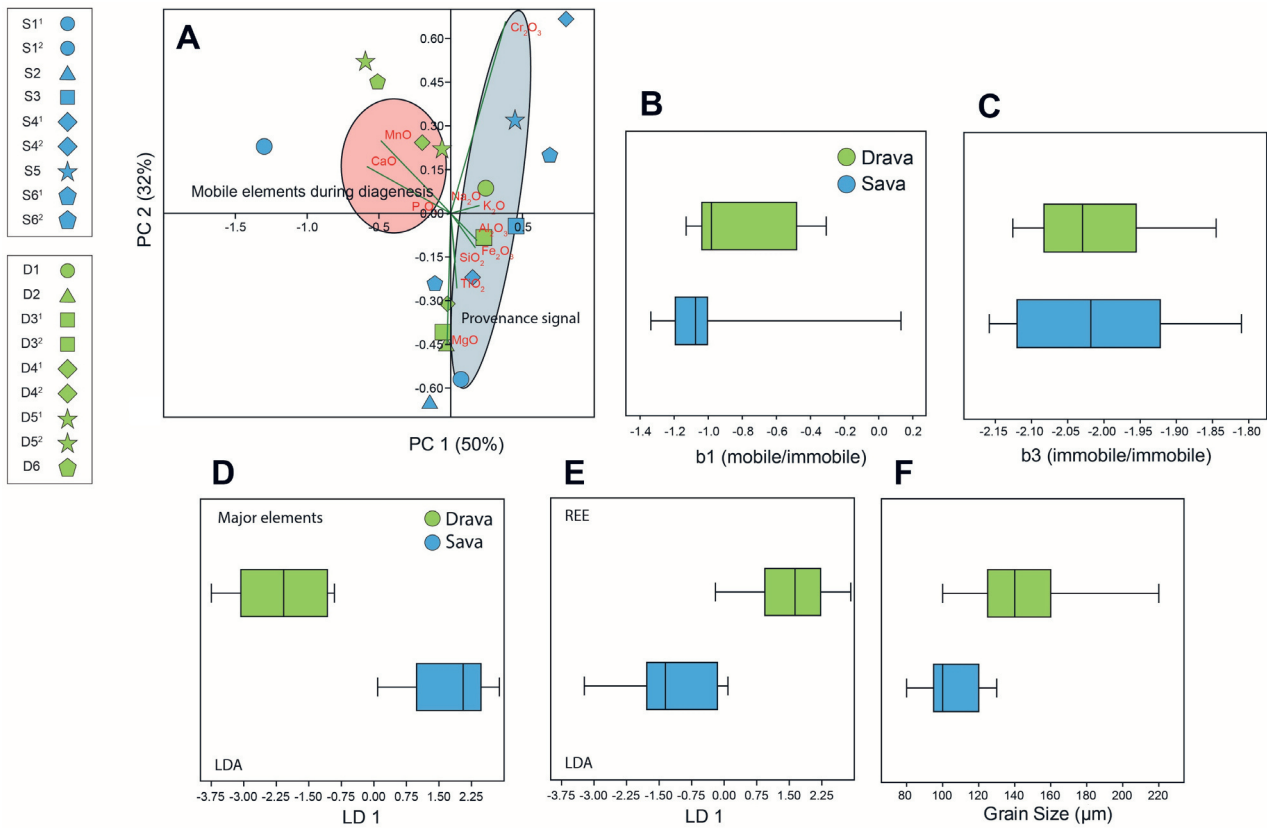
**Figure 9.** Discrimination of heavy mineral assemblages in the Upper Miocene sandstones between the Sava and Drava depressions. A – Balance dendrogram illustrating modeled balances (b1 = log ratio of garnet over other heavy minerals, sensitive to selective dissolution; b2 = log ratio of apatite and 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.



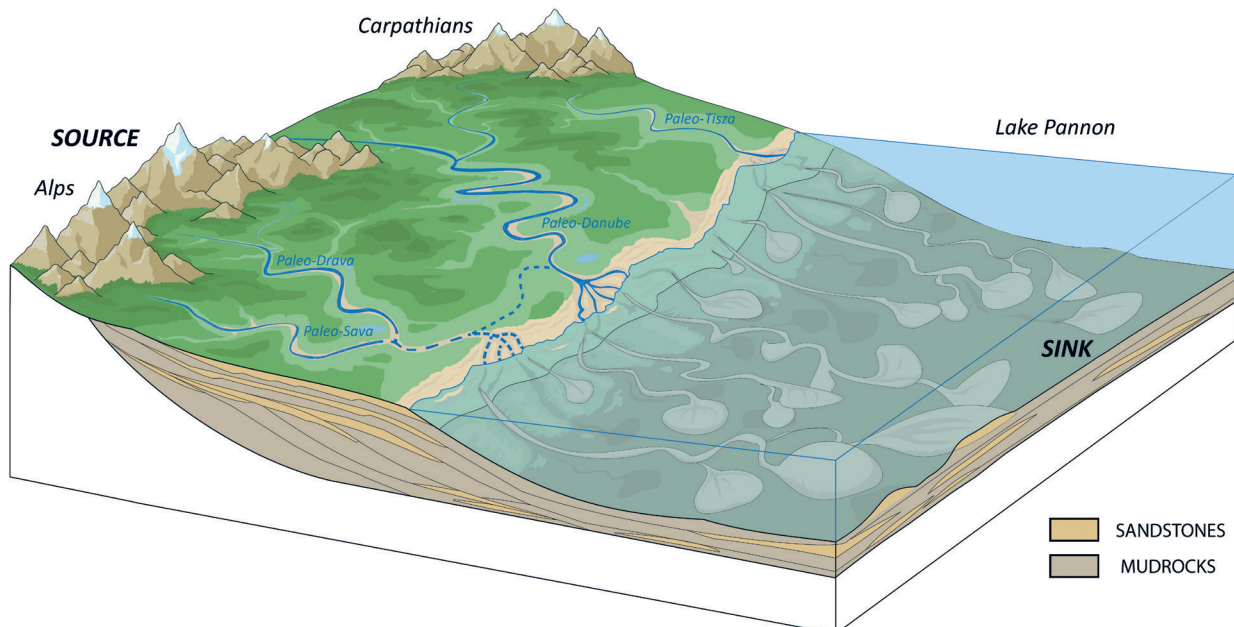
**Figure 10.** Composition of garnets in the Upper Miocene sandstones from the Sava and Drava depressions, illustrated in the classification diagram “pyrope (Py)-almandine (Alm)-spessartine (Sp)” according to Aubrecht (2009). A – Garnets from HP/UHP conditions, B – Garnets from granulite and eclogite 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<sub>2</sub>O, 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 ( $b1 = \text{ratio of MnO-CaO/Al}_2\text{O}_3\text{-TiO}_2$ ) 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 ( $b3 = \text{TiO}_2/\text{Al}_2\text{O}_3$ ). 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 (ŠČAVNIČAR, 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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# Reservoir Quality Evaluation: Unveiling Diagenetic Transformations through Mineralogical and Petrophysical Analyses of the Upper Miocene Lacustrine Sandstones in the Pannonian Basin System, Croatia

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## Abstract

The Upper Miocene lacustrine sandstones of the North Croatian Basin, located in the southwestern Pannonian Basin System, represent significant reservoirs for hydrocarbon exploration, yet their diagenetic evolution remains poorly understood. This study offers a comprehensive investigation into the diagenesis of these sandstones, analyzing samples from 14 exploration wells in the Sava and Drava depressions. Using petrographic analyses, scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS), X-ray diffraction (XRD), and petrophysical measurements, we aimed to explain the diagenetic processes affecting reservoir quality and hydrocarbon productivity. Our results reveal a consistent grain size distribution, modal composition, and diagenetic alterations across both depressions. Compaction, evidenced by evolving grain contacts and pressure dissolution, leads to a depth-dependent reduction in porosity. Carbonate cements, notably calcite and Fe-dolomite/ankerite, are primary contributors to reduction of primary intergranular porosity, alongside clay minerals, quartz, feldspar, etc. Secondary porosity resulting from dissolution and redistribution processes also significantly influences overall porosity evolution. Clay minerals, detrital and authigenic, exhibit a complex interplay with other diagenetic processes, further reducing porosity and permeability. Authigenic clay minerals, including illite, chlorite, and kaolinite, act as pore-filling cement or coatings, hindering fluid flow. Paragenetic processes delineate the intricate relationship between mineralogical transformations and petrophysical properties, defining reservoir quality. Understanding diagenetic dynamics is essential for predicting reservoir quality, fluid migration pathways, and hydrocarbon productivity. This study fills a crucial knowledge gap regarding the diagenesis of the Upper Miocene lacustrine sandstones in the southwestern part of the Pannonian Basin System, providing insights vital for the energy sector and supporting sustainable resource development in the region.

## Keywords:

sandstone reservoirs; diagenesis, porosity; North Croatian Basin; Late Miocene

## 1. Introduction

The Upper Miocene lacustrine sandstones of the North Croatian Basin are pivotal reservoir rocks for oil and gas exploitation, not only locally but also across the broader Pannonian Basin System (Lučić et al., 2001; Saftić et al., 2003; Dolton, 2006; Vrbanac et al., 2010; Malvić and Velić, 2011; Velić et al., 2012; Cvetković

et al., 2018; Matošević et al., 2019a, b, 2021, 2023a, 2024; Kolenković Močilac et al., 2022).

They were deposited during the Late Miocene in the brackish Lake Pannon, which was formed due to the isolation of the Central Parathetys from marine influences (Steininger and Rögl, 1979; Báldi, 1980; Harzhauser and Piller, 2007; Harzhauser and Mandić, 2008; Ter Borgh et al., 2013; Mandić et al., 2015; Kováč et al., 2018; Magyar, 2021). The sandstones were primarily deposited through deltaic system progradation associated with turbidites in deeper basin regions (e.g., Magyar

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et al., 1999, 2013; Ivković et al., 2000; Saftić et al., 2003; Kovačić et al., 2004; Kovačić and Grizelj, 2006; Vrbanac et al., 2010; Malvić and Velić, 2011; Sztanó et al., 2015; Balázs et al., 2018; Sebe et al., 2020; Andelković and Radivojević, 2021). The detritus originated from the Eastern Alps orogenic belt, with sedimentary, metamorphic, and igneous rocks of the ALCA-PA tectonic mega-unit, following W/NW to E/SE trajectories (Šćavničar, 1979; Kovačić and Grizelj, 2006; Matošević et al., 2023a, 2024).

In addition to provenance, diagenesis profoundly affects reservoir development and quality, by altering sediment post-deposition, significantly impacting petrophysical parameters (Guohua, 1982; Civitelli et al., 2023). It involves physical, chemical, and biological processes altering sedimentary assemblages, ranging from subaerial weathering to low-temperature metamorphism (Curtis, 1977; Burley et al., 1985; Worden and Burley, 2003). Understanding burial diagenesis is crucial in predicting porosity distribution, fluid migration pathways, and reservoir productivity (Hurst, 1987). With the increasing demand for petroleum resources, advanced recovery techniques necessitate a comprehensive grasp of diagenetic processes (Pittman and King, 1986; Kantorowicz et al., 1992). For instance, understanding mineral cementation, such as quartz, carbonate minerals, and clay minerals, is essential for predicting reservoir quality within a basin (Curtis, 1983; Burley et al., 1985). Integrating diagenetic processes throughout the sediment column is paramount for accurate reservoir characterization.

Despite longstanding importance of the Upper Miocene sandstones from the North Croatian Basin, a comprehensive understanding of their diagenesis has been lacking (e.g., Tadej et al., 1996; Matošević et al., 2019a, 2021). Diagenetic processes, influenced by factors such as burial depth, temperature, pressure, mineralogy, and pore fluid geochemistry, significantly shape such reservoirs (Morad et al., 2000; Worden and Burley, 2003). Filling this information void is crucial for the energy sector, facilitating further hydrocarbon exploration and production activities, as well as initiatives related to energy transition and environmental conservation, including carbon capture, utilization, and storage, along with expanding investments in regional geothermal energy (c.f., Sneider, 1990; Kolenković et al., 2013; Horváth et al., 2015; Podbojec and Cvetković, 2016; Macenić et al., 2020; Alcalde et al., 2019; Tuschl et al., 2022; Vulin et al., 2023).

In continuation of the preceding research by Matošević et al. (2023a, 2024), which provided insights into the provenance of the sandstones, this study aims to further elucidate the diagenetic processes affecting these reservoirs. This paper presents a comprehensive study on the diagenesis of the Upper Miocene sandstones in the North Croatian Basin, focusing on samples from exploration wells in the Sava and Drava depressions. Em-

ploying various methods, including petrography, scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS), X-ray diffraction (XRD), and petrophysical measurements, our objective was to identify diagenetic processes with increasing depth, including mineralogical alterations and compaction, influencing primary and secondary porosity. Additionally, we aimed to discern paragenetic sequences of diagenetic processes crucial for understanding reservoir property evolution. These insights enhance exploration and reservoir modelling efforts within the Pannonian Basin System, supporting sustainable energy resource development of the region.

## 2. Geological setting

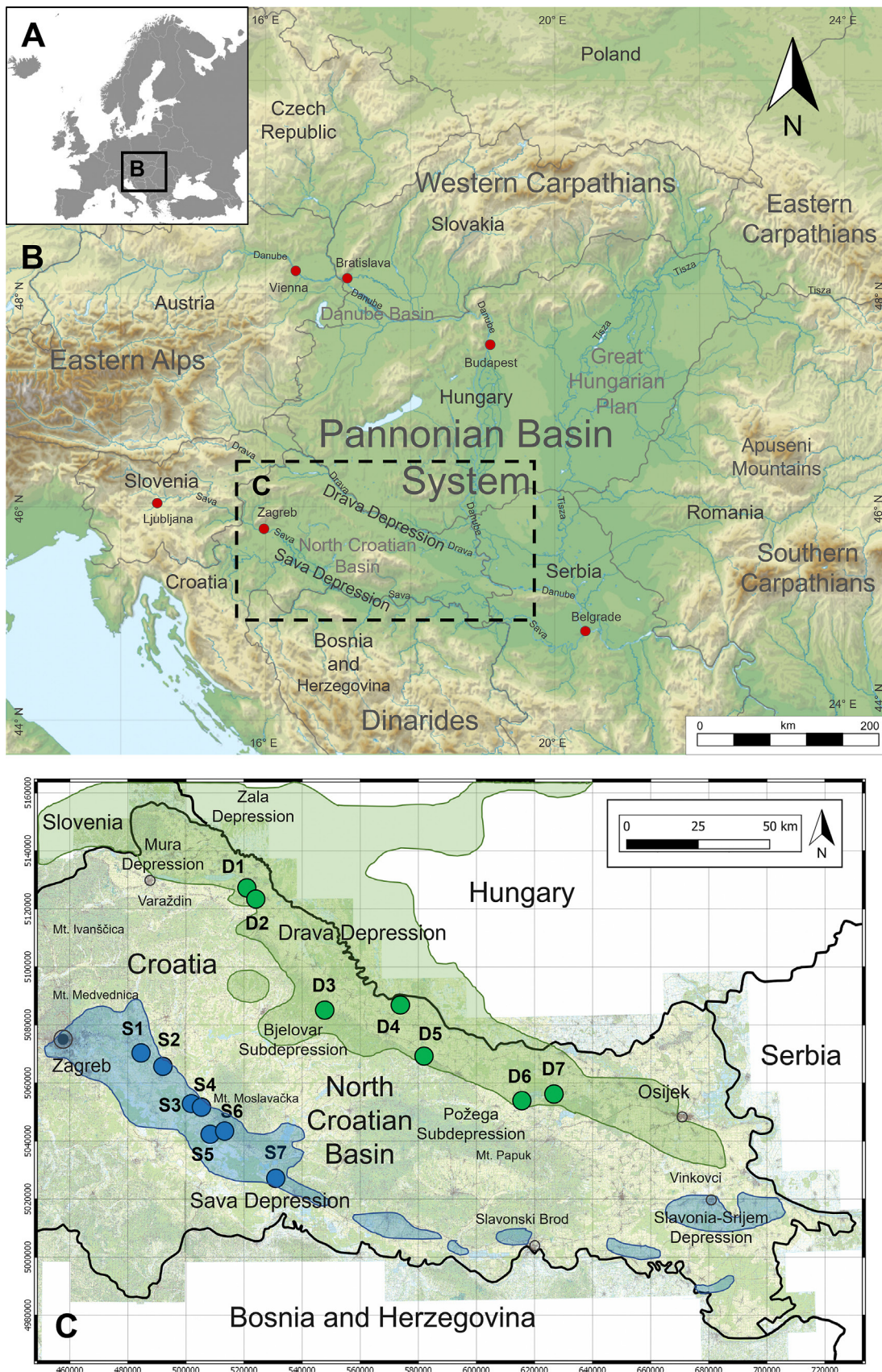
The North Croatian Basin, situated in northern Croatia, is part of the Pannonian Basin System, spanning approximately 32,000 km<sup>2</sup> (Figure 1). Notably, it comprises the Sava and Drava depressions, tectonically induced and crucial depocenters during the Neogene (Pavelić and Kovačić, 2018; Figure 1).

This SW part of the Pannonian Basin System witnessed the deposition of Lower to Upper Miocene strata over tectonized Paleozoic to Paleogene basement rocks (Pamić, 1999; Pavelić, 2001; Saftić et al., 2003; Matošević et al., 2015; Pavelić and Kovačić, 2018; Matošević and Šuica, 2017; Šuica et al., 2022a, b; Rukavina et al., 2023), marked by significant geological changes driven by tectonic shifts, climatic fluctuations, and volcanic events (Pavelić and Kovačić, 2018; Grizelj et al., 2020, 2023; Premec Fuček et al., 2022; Matošević et al., 2019c, 2023b). The evolution of the Pannonian Basin System is distinguished into syn-rift and post-rift phases (Royden, 1988; Tari et al., 1992; Matenco and Radivojević, 2012).

The post-rift phase, characterized by diminished tectonic activity, led to lithospheric cooling, subsidence, and the isolation of the Central Paratethys from global oceans, forming Lake Pannon (Rögl and Steininger, 1983; Royden, 1988; Tari et al., 1992; Rögl, 1998; Harzhauser et al., 2007; Piller et al., 2007; Ter Borgh et al., 2013; Kováč et al., 2017). Intense basin subsidence and humid conditions with significant lake depths (Sztanó et al., 2013; Balázs et al., 2018) fostered large accumulation of post-rift siliciclastic deposits, including sandstone bodies, particularly in the central part of the depressions (Ivković et al., 2000; Saftić et al., 2003; Kovačić and Grizelj, 2006; Malvić and Velić, 2011; Matošević et al. 2023a, 2024).

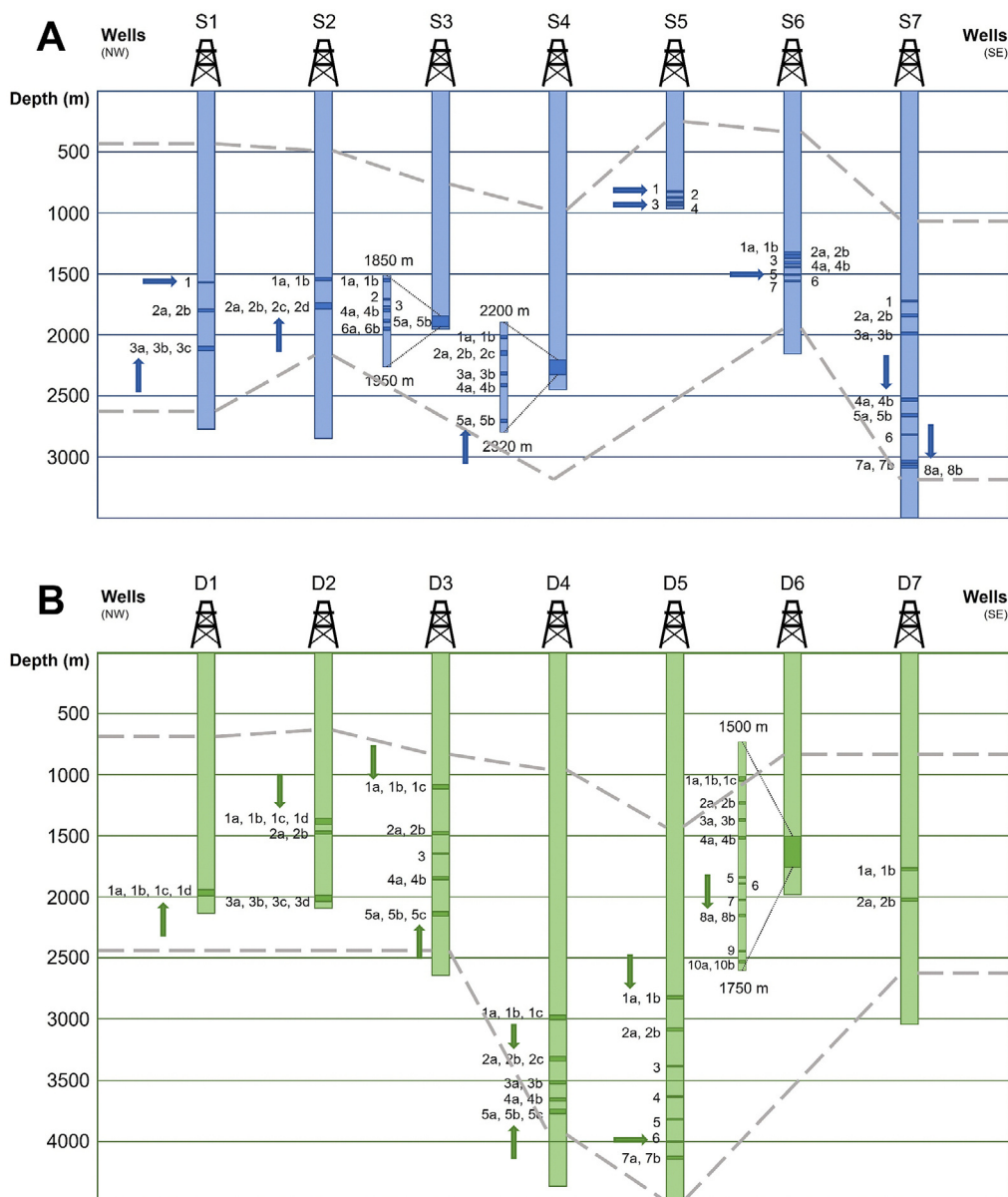
The Upper Miocene deposits, referred to as the Pannonian in the regional Central Paratethys time scale (Hilgen et al., 2012), underlie Pliocene and Pleistocene (Cernikian) lacustrine and alluvial sediments and overlie Middle Miocene (Sarmatian) marine deposits (Pavelić, 2001; Mandić et al., 2015; Pavelić and Kovačić, 2018; Kurečić et al., 2021). They predominantly formed as





**Figure 1:** A – Europe with the position of the Pannonian Basin System. B – Geographical overview of the Pannonian Basin System in Central Europe, surrounded by the mountain ranges of the Alps, the Carpathians, and the Dinarides, with the North Croatian Basin in its SW part. C – Locations of the Upper Miocene reservoir sandstones from exploration wells in the North Croatian Basin within the Sava (S1-S7) and Drava depression (D1-D7).





**Figure 2:** Positions of the Upper Miocene sandstone cores from the exploration wells in the Sava (A) and Drava (B) depressions which are the focus of the research. The stratigraphic correlation of the wells is based on the well-log data boundaries between the Upper Miocene and Pleistocene/Pliocene in shallower intervals (upper dashed line) and the Middle/Lower Miocene or the Neogene basement in deeper intervals (lower dashed line).

For the geographic location of wells, see **Figure 1**. The arrows indicate the positions of core samples that were selected for detailed petrographic, SEM-EDS and XRD analyses.

turbidite mass flow deposits and later covered by delta and fluvial system deposits (Ivković et al., 2000; Saftić et al., 2003; Magyar et al., 2013; Sztanó et al., 2015; Špelić et al., 2019, 2023; Sebe et al., 2020).

These Upper Miocene sandstones, known for their high porosity and permeability, are key reservoir rocks in Croatia (Lučić et al., 2001; Saftić et al., 2003; Vrbanac et al., 2010; Velić et al., 2012; Matošević et al., 2019a, b; Kolenković Močilac et al., 2022). These sandstones were deposited in various environments including deeper-water fan lobes, channels, and levees; as well as prodelta, delta front, delta plain, and alluvial

plain settings, including distributary channels and mouth bars (Pogácsás, 1984; Juhász, 1994; Basch et al., 1995; Magyar et al., 2013; Kovačić et al., 2004; Kovačić and Grizelj, 2006; Sztanó et al., 2015; Sebe et al., 2020; Anđelković and Radivojević, 2021; Špelić et al., 2023). Originating from a recycled orogen provenance area, these sandstones contain a diverse array of rock fragments, suggesting a complex provenance from the Eastern Alps, including the predominant Austroalpine nappe (Ščavničar, 1979; Kovačić and Grizelj, 2006; Matošević et al., 2024). Similar detrital signatures in both depressions suggest a shared origin within the

lake's depositional setting, possibly associated with Paleo-Sava and/or Paleo-Drava, following NW to SE transport direction of detritus (Matošević et al., 2024).

### 3. Methods

The dataset comprises 14 exploration wells from the Sava and Drava depressions, drilled by INA – Industrija nafte d.d., and a total of 130 sandstones samples from cored intervals (Figure 2), out of which 18 samples were analyzed for petrography, SEM-EDS, and XRD. These samples were chosen based on their representation of the Late Miocene succession of the basin infill and their suitability for comprehensive analysis of diagenetic alterations across different depth intervals. All 130 samples were utilized for petrophysical measurements, focusing on porosity. (Figure 2).

#### 3.1. Petrography and SEM-EDS

The petrographic examinations involved detailed analyses of the 18 selected sandstone samples impregnated with blue-dyed epoxy and stained with Alizarin Red S. The samples were analyzed by an Olympus BX51 polarizing microscope to discern grain contacts, porosity types, and diagenetic alteration processes in thin sections.

SEM-EDS analyses provided detailed insight into microstructural features and mineralogical compositions of the 18 selected sandstone samples. The samples underwent coating with a thin layer of Au for enhanced conductivity. SEM imaging was performed using a JEOL JSM-6510 LV SEM at acceleration voltages ranging from 5 to 25 kV, revealing surface topography at magnifications ranging from 25 to 20,000x. Analysis of individual minerals, considering morphological features and chemical composition, was conducted using secondary electron (SE) and back-scattered electron (BSE) images, alongside energy-dispersive X-ray spectra (EDS) from an Oxford INCA X-act system (Oxford Instruments, High Wycombe, UK). Mineral identification relied on comparing X-ray spectra with literature data (Welton, 1984; Severin, 2004). SEM-EDS analyses were performed in the Exploration & Production Laboratory in the INA – Industrija nafte d.d.

#### 3.2. XRD

XRD was employed for the determination of clay mineral assemblage in the 18 studied sandstone samples. The samples were crushed in jaw crusher followed by pulverization in agate mortar until passing sieve opening of 0.02 mm. Prior to separation of clay fraction, the samples were treated by acetic acid to dissolve carbonates, then by hydrogen peroxide to remove organic matter and finally by Tamm solution (mixture of oxalic acid and ammonium oxalate) to eliminate Fe-Mn-Al oxides/hydroxides. The clay fraction was separated by gravitation in a

centrifuge, and then deposited from suspension on glass mounts in order to obtain oriented samples for the phase analysis. Four sets of the samples were prepared: air-dried, glycolated, and heated for half an hour at 400°C and 550°C. XRD data were collected using Philips X'Pert PRO diffractometer PW 3040/60 at the Department of Geology, Faculty of Science, University of Zagreb, with CuK $\alpha$  radiation generated at 40 kV and 40 mA. Recording parameters were as follows: divergence slit 1/8°, anti-scatter slit 1/4°, sample mask of 10 mm, Soller slits inserted into primary and diffracted beam path, scanning step 0.026°2 $\theta$ , and measuring time 128.27 s/step. Pixel detector was employed for acquisition of data, which were later processed using X'Pert Highscore (Panalytical, 2004). Clay minerals were identified applying the following criteria: **illite** was distinguished by 10 Å peak that does not shift after glycolation and heating treatments; **chlorite** was confirmed by 14 Å and 7 Å peaks that do not shift and do not change intensity significantly after glycolation and heating treatments; **kaolinite** was identified by 7 Å peak that disappears after heating at 550°C; mutual discrimination of chlorite and kaolinite was done by spotting chlorite 004 (around 3.54 Å) and kaolinite 002 (around 3.57 Å) diffraction maxima in glycolated samples; mixed layered **illite/smectite** phases were recognized by a maximum between 10 and 14 Å and its shift toward higher d-values in glycolated samples, as well as by asymmetry of 10 Å peak; illite content in mixed-layered illite/smectite was estimated by observing the position of illite/smectite 001/002 and 002/003 diffraction maxima in glycolated samples; **illite/chlorite** was determined by 10-14 Å peak that does not change during glycolation and heating treatments; **kaolinite/smectite** was identified by 7.2-7.5 Å peak that slightly shifts and broadens to a higher d-value during glycolation while it further increases and broadens at 400°C but disappears at 550°C; **dioctahedral vermiculite** was identified by a slight increase of 14 Å peak during glycolation and its collapse to lower d-values after heating treatments (to around 12 and 11 Å); **dioctahedral illite/vermiculite** was determined by 10-14 Å peak that does not change or slightly increases during glycolation but discretely and stepwisely decreases during heating treatments (Moore and Reynolds, 1997). Illite crystallinity was determined by measuring the width at half maximum of 10 Å peak of glycolated samples. The method employs standardless approach that allows estimation of the illite crystallinity as described in the provenance and weathering studies (e.g., Liu et al., 2007; Griffiths et al., 2019, and references therein), thus enabling mutual comparison of the investigated samples in this respect. The experimental conditions for XRD recording in this case were the same as previously mentioned, and the XRD pattern profile around 10 Å peak was fitted using Panalytical X'Pert HighScore software with additional visual inspection for possible interferences of neighbouring diffraction maxima.

### 3.3. Porosity Measurements

Porosity measurements of sandstone samples were conducted following precise methodologies to ensure accuracy and reliability. Cylinder plugs were extracted from 130 sandstone core samples, each measuring 3.81 cm in diameter and 6 cm in length, ensuring sample uniformity. Before measurements, chloroform and methanol were used in the sample cleaning process to remove petroleum, salt, and other impurities. This step aimed to eliminate potential interference with the results. Subsequently, the samples were dried at 105°C in a conventional laboratory until they reached a constant weight, ensuring residual moisture removal without sample damage. The drying process was monitored until samples reached a constant weight.

Porosity measurements were conducted using a helium gas expansion porosimeter, relying on Boyle's law to govern gas expansion from a reference cell with a known volume to a sample cell at a constant temperature. The effective pore volume and total sample volume were determined, allowing calculation of porosity as the ratio of the pore volume to the total sample volume, expressed as a percentage (American Petroleum Institute, 1998).

These procedures ensured accurate determination of porosity in the sandstone samples, facilitating comprehensive analyses and interpretation of reservoir properties. However, it is essential to acknowledge potential limitations or sources of errors in the porosity measurement process, such as sample heterogeneity or gas adsorption effects, which could affect the precision of the results.

## 4. Results

### 4.1. Grain size distribution and modal composition

The Upper Miocene sandstones from the Sava and Drava depressions exhibit angular to sub-rounded grains, occasionally tabular, with average sizes of 110 µm and 150 µm, respectively (Figure 3; Supplementary Table 1 and Supplementary Table 2), which was already described in previous investigations (Matošević et al., 2023a, 2024), corresponding to very fine to fine-grained sand (Wentworth, 1922). Grain contacts vary from point to long and concavo-convex contacts, transitioning even to sutured contacts in more compacted sandstones at greater depths. However, point contacts are primarily contacts in all samples throughout investigated depth intervals (Figure 3). Grain deformations, such as bending of mica and rock fragments (particularly sedimentary and metamorphic fragments), are regularly observed in samples (Figure 3 and Figure 4 A, B, C). Dissolution of feldspar grains and carbonate rock fragments is evident in a good number of samples, noticeable already in shallower intervals (above 1000 m) and continuing in deeper intervals. This dissolution is closely associated with the reduced presence of carbonate (early calcite) cements

in the intergranular volume (Figure 4 D, E, F, G, H). The dissolution of heavy minerals has also been detected (e.g., the dissolution of staurolite and minerals belonging to the epidote group in shallower intervals and the dissolution of garnets in deeper intervals; Figure 4 I). Rarely, some fractured quartz grains can be found in deeper intervals. The sandstones are predominantly well- to moderately well-sorted (Figure 3, Supplementary Table 1). Framework petrography analyses by Matošević et al. (2023a, 2024) reveal quartz content ranging from 43.2% to 55.6% in the Sava depression and 46.2% to 57.7% in the Drava depression (Supplementary Table 1 and Supplementary Table 2). Feldspar content ranges from 11.9% to 19.1% in the Sava depression and 8.0% to 15.6% in the Drava depression, while rock fragments constitute 30.1% to 42.3% and 29.1% to 41.0% in the respective depressions (Supplementary Table 1 and Supplementary Table 2). Sedimentary rock fragments, notably carbonate types, dominate the rock fragment composition (Supplementary Table 1; Figure 3 and Figure 4), classifying the sandstones as carbonaticlastic feldspatho-litho-quartzose. Mica, chlorite, and accessory heavy minerals contribute to the overall composition. Results are provided in full detail in the Supplementary Table 1.

### 4.2. Intergranular volume

Sandstone intergranular volumes in both depressions are either unfilled or partly/entirely filled with fine-grained matrix and/or cement (Figures 3-6). Descriptive information about the filling of the intergranular volume for each sample can be found in the Supplementary Table 2.

#### 4.2.1. Matrix

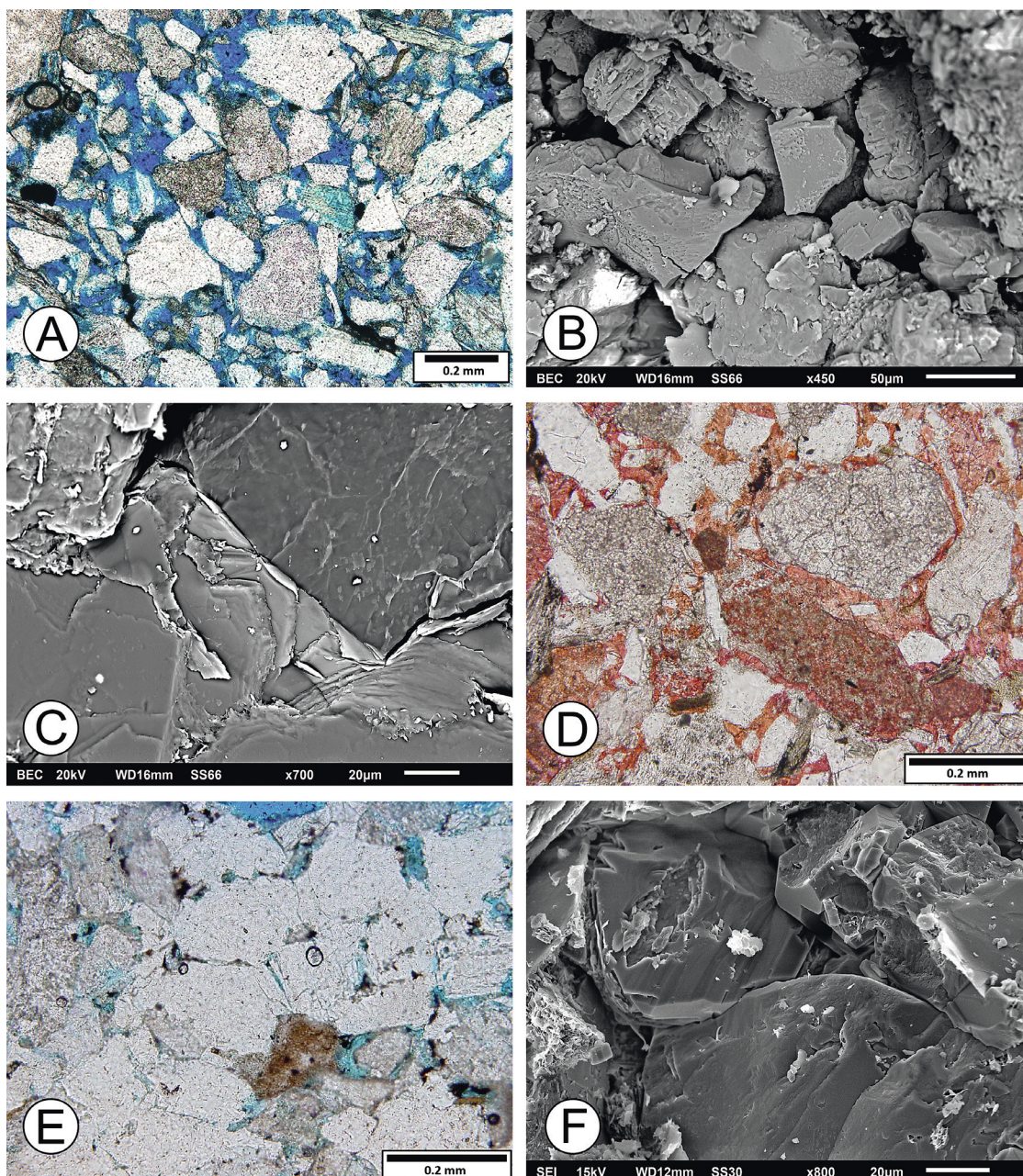
The matrix consists of silty and clayey particles derived from the breakdown and alteration of main mineral grains during transport and deposition. Phyllosilicates (mica, chlorite, and detrital clay minerals) predominate, with carbonate minerals, quartz, and feldspar also present. The matrix is in general more prominent in samples from shallower intervals in both depressions. Pseudomatrix, resulting from compaction and deformation of unstable grains during diagenesis, is occasionally observed, particularly from rock fragments like mudrocks (e.g., rip-up clasts of marl) and metasediments (Figure 4 C).

#### 4.2.2. Cement

Authigenic cement mainly comprises carbonate minerals and/or a combination of carbonate minerals with clay minerals, quartz, and feldspar (Supplementary Table 2; Figure 5 and Figure 6).

*Carbonate:* Predominantly composed of calcite and Fe-dolomite/ankerite, rarely dolomite and Fe-calcite, and extremely rarely siderite (Supplementary Table 2; Figure 3 C, D and Figure 5 A, B, C). It often forms



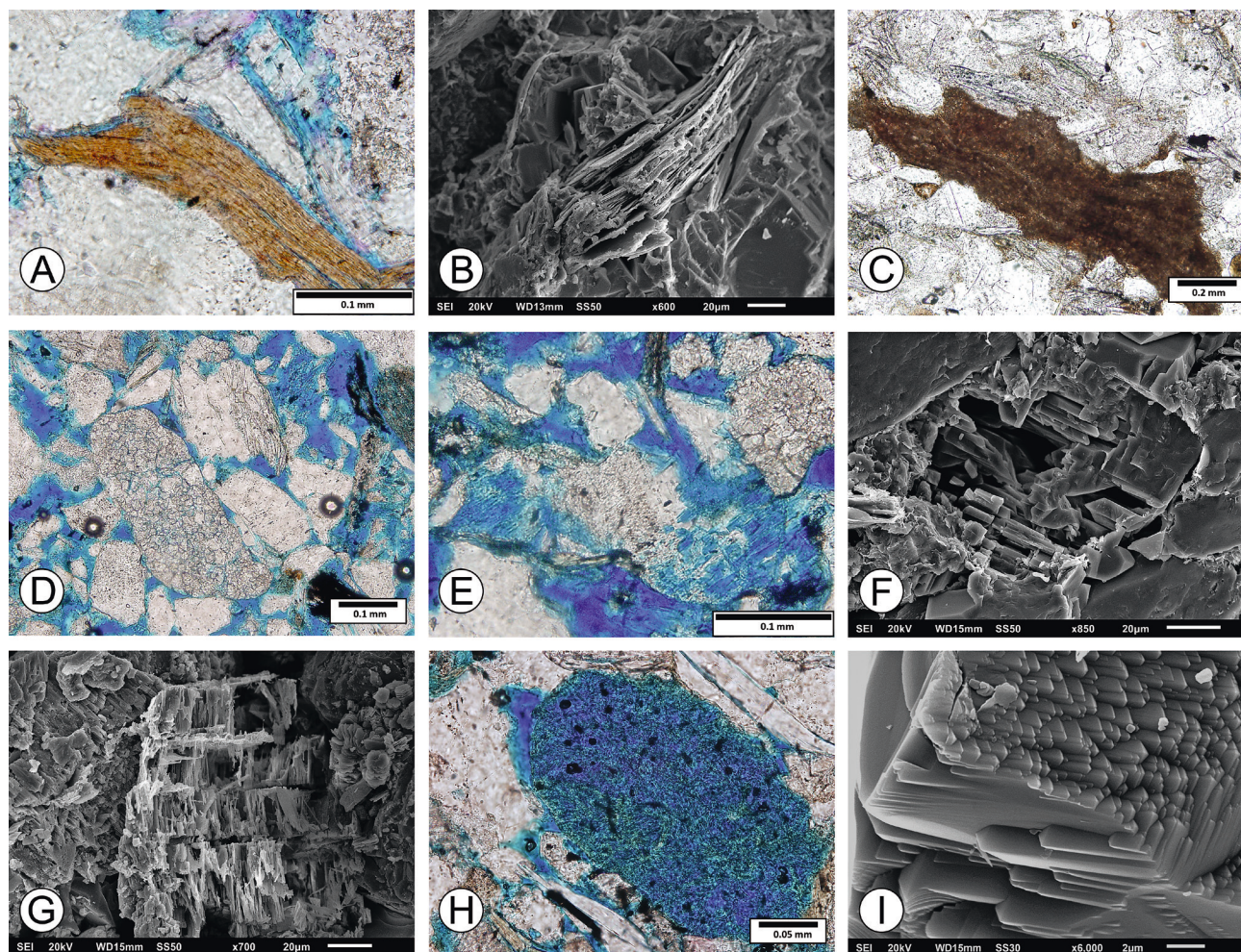


**Figure 3:** The Upper Miocene sandstones of the Sava and Drava depressions at low magnifications in thin sections and SEM. **A** – Sandstone impregnated with blue-dyed epoxy showing very fine to fine, angular to sub-rounded detrital grains, predominantly in point contacts, and primary intergranular porosity (S5\_1, PPL); **B** – Sandstone with main grains and mostly unfilled intergranular volume with connected pores (S5\_3, SEM BE); **C** – Detail of more compacted sandstone, showing densely packed grains with long contacts, grain deformations, and carbonate cement, filling the intergranular volume (S7\_4b, SEM BE); **D** – The main grains in the sandstone consist of quartz, feldspars and rock fragments (mostly sedimentary rock fragments in the form of recrystallized carbonates), and the cement is calcite, stained with Alizarin Red S (S1\_1, PPL); **E** – Rearrangement of grains in the sandstone due to overlying pressure (compaction and deformation), reducing primary intergranular porosity with precipitation of cement on grain surfaces and within pores, but also dissolving some grains with formation of secondary porosity (D3\_5c, PPL); **F** – Grains in mutual penetrant (concavo-convex) contacts, partly due to later cement overgrowing on primary detrital grains (D1\_1c, SEM SE). PPL = plane-polarized light, SEI = secondary electron image, BEI = backscattered electron image. Photo: M. Matošević.

rhombohedral crystals in intergranular space (**Figure 5 B**), sometimes in conjunction with other authigenic minerals. Authigenic carbonates create euhedral overgrowths on detrital carbonate grains and partly/completely fill pores as well (in some samples early calcite

sparry cement extends from grain to grain; **Figure 3 C, D and Figure 5 A**). It does not clearly depend on depth, although tightly cemented sandstones were mainly found in deeper intervals of the depressions, especially in the Drava depression. Fe-dolomite/ankerite generally





**Figure 4:** Diagenetic alterations of grains in the Upper Miocene sandstones of the Sava and Drava depressions in thin sections and SEM. **A** – Deformation (bending) of biotite due to compaction, locally making obstacles for fluid flow (D3\_5c, PPL); **B** – Simultaneous deformation and decomposition of muscovite and formation of secondary dissolution porosity (S1\_1, SEM SE); **C** – Formation of pseudomatrix by compaction and deformation of unstable grains (mudrock rock fragment) (S6\_5, PPL); **D** – Intragranular porosity stemming from partial dissolution of recrystallized carbonate rock fragment (D2\_1c, PPL); **E** – Secondary pores impregnated with blue-dyed epoxy in partly dissolved alkali feldspar contributing to overall porosity (D2\_1c, PPL); **F** – Pore formation due to dissolution of Na-plagioclase (S4\_5a, SEM SE); **G** – Resorption of Na-Ca plagioclase with preferred orientation of the remnants signifying that dissolution of the grain was crystallographically controlled (S6\_5, SEM SE); **H** – Complete dissolution of detrital grain (possibly volcanic rock fragment) with clay mineral replacement, promoting secondary porosity and microporosity (S4\_5a, PPL); **I** – Skeletal garnet with deeply etched faceted grain surface as a result of dissolution (D3\_1a, SEM SE). PPL = plane-polarized light, SEI = secondary electron image. Photo: M. Matošević.

occurs in samples from intervals deeper than 1000 m in both depressions.

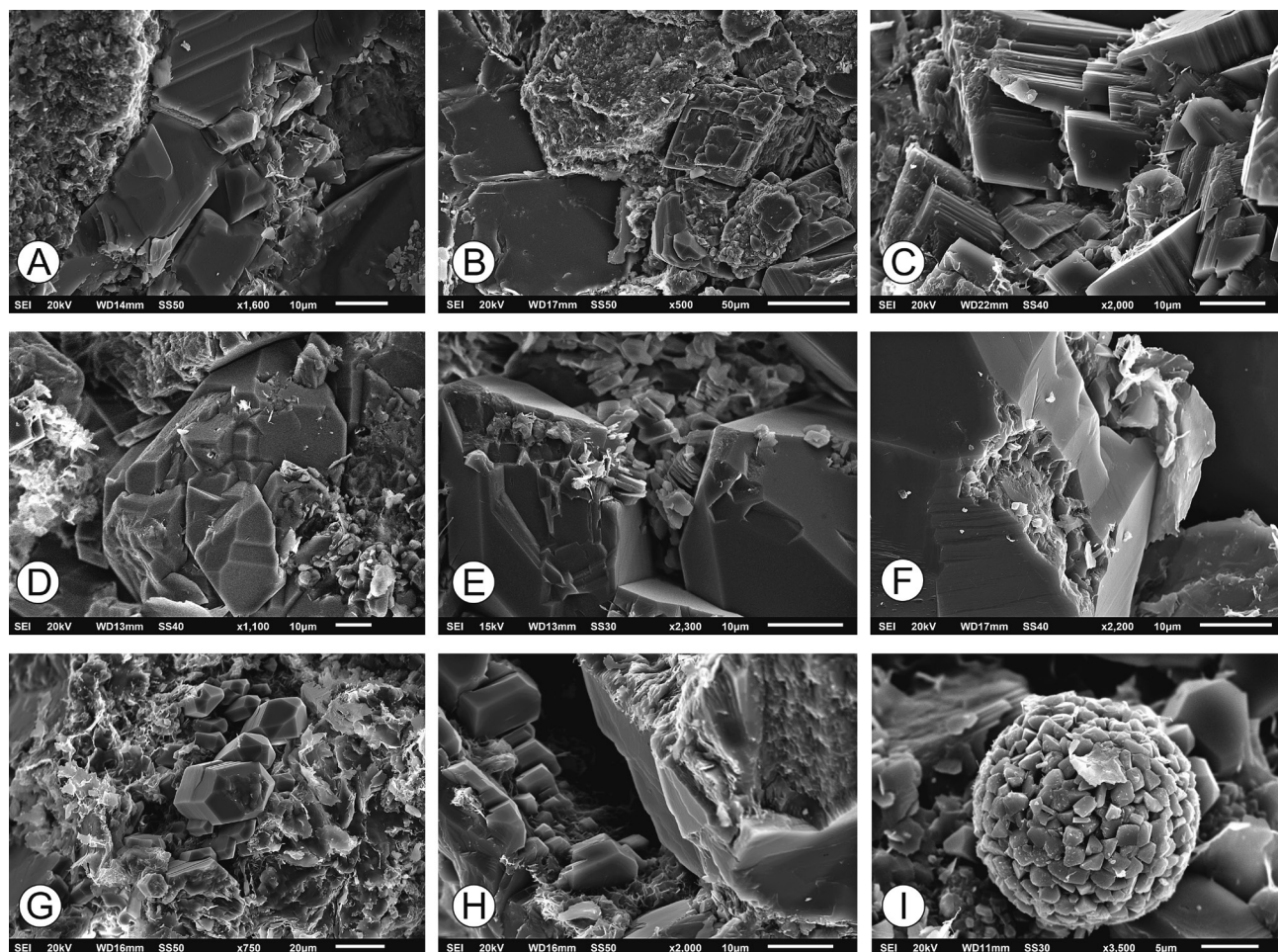
**Quartz:** Largely manifested as authigenic quartz overgrowths on detrital quartz grains. It primarily forms well-developed crystals with smooth euhedral faces, partly/completely surrounding quartz grains (**Supplementary Table 2; Figure 5 D, E, F**). Occasionally, it also occurs as pore-linings and pore-fillings with other minerals (primarily clay minerals), predominantly in the form of microcrystalline quartz, with a multitude of small bipyramidal crystals generally less than 10  $\mu\text{m}$  length (**Figure 5 G**). In general, quartz cement arises in samples from intervals deeper than 1000 m in both depressions.

**Feldspar:** Relatively sporadic, appearing as authigenic overgrowths of alkali feldspar and plagioclase (mainly Na-plagioclase) in the form of small poorly to well-developed crystals (**Figure 5 H**), sporadically also corroded. Na-plagioclase cement is observed from shallower intervals in both depressions, but with notable amount mostly in samples deeper than 2000 m.

**Pyrite:** Exceptionally rare, manifested usually as microcrystalline subhedral to euhedral framboids in clusters, together with other authigenic minerals (**Figure 5 I**). It appears mainly in samples from deeper intervals in both depressions, usually far deeper than 2000 m.

**Clay Minerals (detrital and authigenic):** Mainly illite, chlorite, kaolinite, and mixed-layered clays, observed as



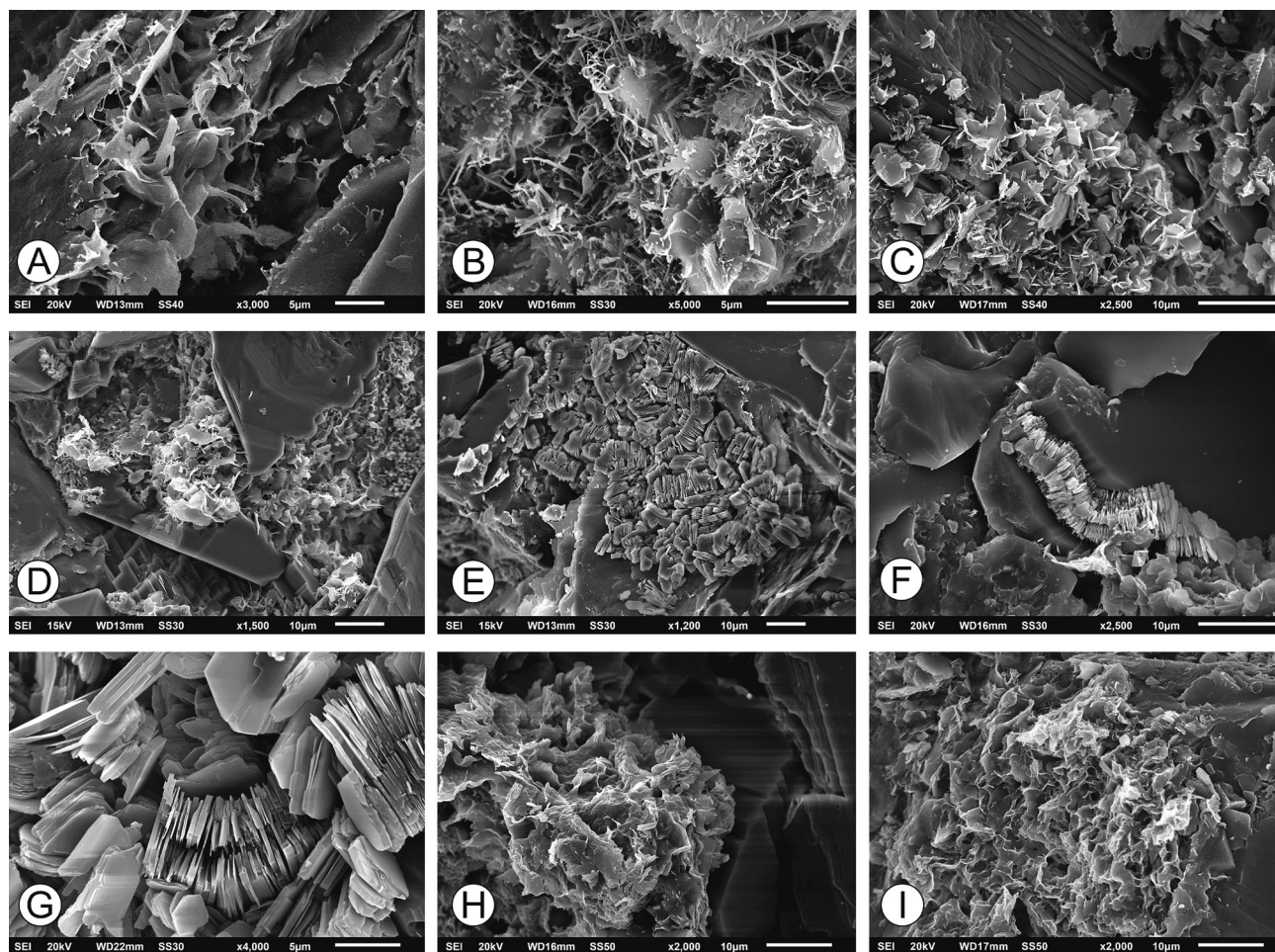


**Figure 5:** Cements in the Upper Miocene sandstones of the Sava and Drava depressions in SEM. **A** – Authigenic calcite in combination with clay minerals expanding from grain to grain (D<sub>4\_2b</sub>, SEM SE); **B** – Rhombohedral crystals of Fe-dolomite/ankerite in pore space contributing to the reduction of porosity and local permeability (S<sub>4\_5a</sub>, SEM SE); **C** – Calcite crystal overgrowths on detrital carbonate grain enlarging to intergranular space (S<sub>7\_8a</sub>, SEM SE); **D** – Quartz cement manifesting as authigenic quartz overgrowth completely surrounding detrital quartz grain (D<sub>1\_1c</sub>, SEM SE); **E** – Euhedral smooth quartz overgrowth on detrital quartz grain with clay minerals in intergranular volume (D<sub>1\_1c</sub>, SEM SE); **F** – Quartz grain enclosed in incomplete well-developed quartz overgrowth (S<sub>4\_5a</sub>, SEM SE); **G** – Microcrystalline quartz crystals with clay minerals lining intergranular pores (D<sub>3\_5c</sub>, SEM SE); **H** – Pore-lining microcrystalline Na-plagioclase overgrowths with clay minerals (D<sub>3\_5c</sub>, SEM SE); **I** – Framboidal pyrite (D<sub>1\_1c</sub>, SEM SE). SEI = secondary electron image. Photo: M. Matošević.

coatings, pore fillings, or matrix components (**Supplementary Table 2 and Supplementary Table 3; Figure 6 and Figure 7**). Illite is principally observed as flakes and fibrous structures on the surface of detrital grains, at the edges of phyllosilicates and rock fragments, dispersed through the matrix, or as pore-filling clusters (**Figure 6 A, B**). Detrital illite typically exhibits irregular, flake-like platelets oriented parallel to each other, while authigenic illite appears mostly in the form of filaments and/or ribbons, often lining and bridging pores (**Figure 6 A, B**). Authigenic illite appears mainly from 2000 m deeper in both depressions. Chlorite is found as detrital particles, usually as individual flakes with parallel face-to-face orientations, formed through alterations of mica and chlorite in shallower intervals, or as authigenic minerals, frequently as clusters of elongate to disc-like crystals partly filling pores and rarely as pore-lining, forming thin rims around detrital grains with euhedral crystals differently

oriented and perpendicular to the grain surface (**Figure 6 C, D**). Authigenic chlorite significantly occurs in samples from 2500 m deeper in the Sava depression and samples from 2000 m deeper in the Drava depression. Kaolinite predominantly appears in clusters within pores or adhering to detrital grains, originating from the decomposition of the grains (generally feldspars), occasionally exhibiting a vermiform structure (**Figure 6 E, F**). Kaolinite was predominantly recognized as authigenic, well-crystallized face-to-face stacks of hexagonal and pseudo-hexagonal plates or booklets (**Figure 6 G**). It occurs in samples from shallower intervals, above 1000 m, but continues in samples from deeper intervals as well in both depressions. Correspondingly to occurrence of feldspar dissolution, it is mainly found in samples which have no pronounced early calcite cementation. In samples from deeper intervals, dickite (blocky kaolinite) is also observed. Among mixed-layered and other clays, most





**Figure 6:** Clay cements in the Upper Miocene sandstones of the Sava and Drava depressions in SEM. **A** – Illite as flakes and fibrous structures on the surface and at the edges of muscovite, expanding to intergranular space (D1<sub>1c</sub>, SEM SE); **B** – Filamentous pore-lining and pore-bridging authigenic illite (D3<sub>5c</sub>, SEM SE); **C** – Cluster of authigenic chlorite to illite/chlorite partly filling pore (S7<sub>4b</sub>, SEM SE); **D** – Chlorite forming rims around detrital quartz grain preventing quartz overgrowth (D1<sub>1c</sub>, SEM SE); **E** – Cluster of kaolinite minerals within pore (D1<sub>1c</sub>, SEM SE); **F** – Vermiform kaolinite adhering to detrital grain (S2<sub>2c</sub>, SEM SE); **G** – Detailed view of authigenic kaolinite with face-to-face stacks of pseudo-hexagonal plates (S1<sub>3a</sub>, SEM SE); **H** – Cluster of illite/smectite to illite in intergranular space (S2<sub>2c</sub>, SEM SE); **I** – Rim of illite/smectite on detrital grain (D2<sub>1c</sub>, SEM SE). SEI = secondary electron image. Photo: M. Matošević.

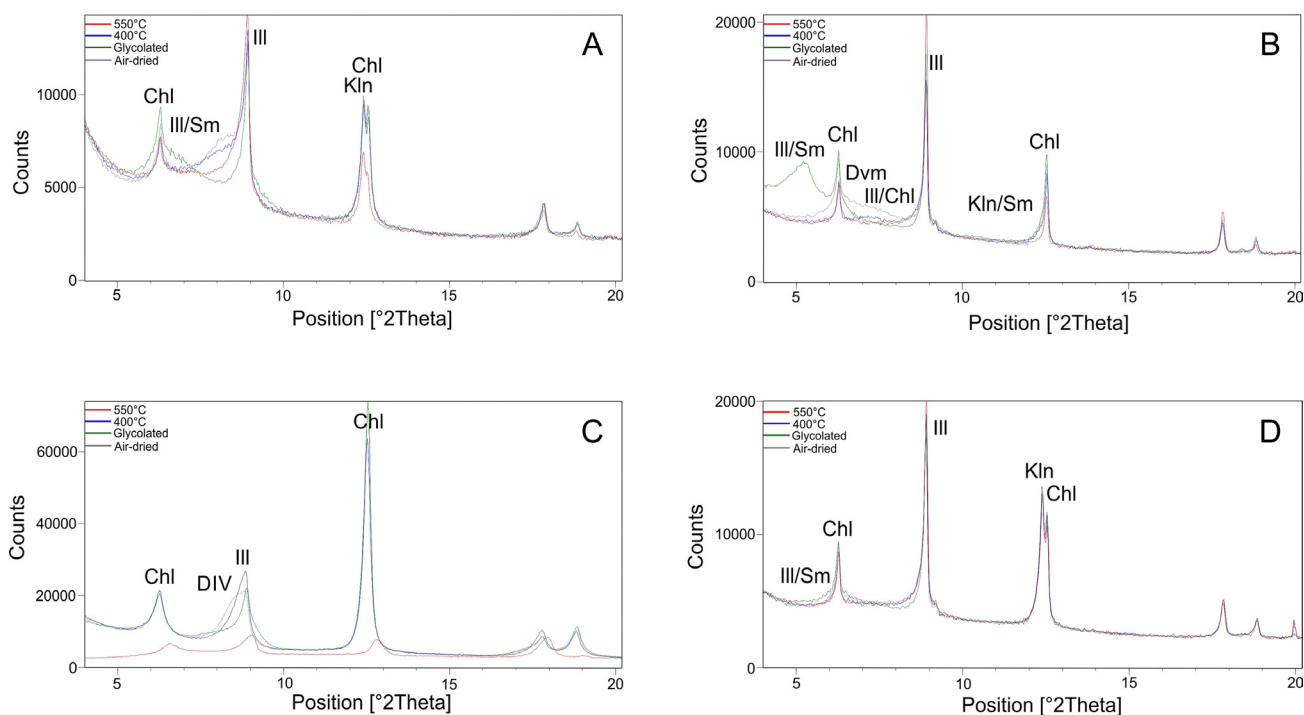
prevalent illite/smectite (**Figure 6 H, I**), illite/chlorite, chlorite/smectite, kaolinite/smectite, dioctahedral vermiculite, and dioctahedral illite/vermiculite are identified based on XRD analyses (**Figure 7**). Questionable occurrences have illite/vermiculite and sepiolite, each in only one sample. For detail clay mineral determination, refer to the results of the XRD analyses (**Supplementary Table 3; Figure 7**).

#### 4.3. Porosity

Porosity measurements conducted in the Sava and Drava depressions reveal distinctive statistical parameters reflecting reservoir characteristics at specific depths. Within the Sava depression, porosity in analyzed samples ranges from a minimum of 3.0% at 3047.55 m depth to a maximum of 34.9% at 827.7 m depth, with a mean value of 19.06% and a median of 19.00%. Correspondingly, within the Drava depression, porosity in analyzed sam-

ples ranges from a minimum of 1.9% at 4141.15 m depth to a maximum of 28.7% at 1398.6 m depth, with a mean value of 14.19% and a median of 13.80%. **Supplementary Table 4** summarizes porosity measurements in both depressions, highlighting the variability across depths and underscoring the heterogeneity of subsurface reservoirs. Relationship of porosity and depth in the Sava and Drava depressions is illustrated in **Figure 8**.

Primary intergranular porosity predominates across all sandstone samples (**Supplementary Table 2; Figure 3 and Figure 4**). As mentioned earlier, the intergranular volume manifests as unfilled, partially filled, or completely filled with carbonate minerals, fine-grained matrix, different types of clay minerals, quartz, and feldspar. Pore diameters typically range from 5-100  $\mu\text{m}$ , with an average size falling within the range of 10-40  $\mu\text{m}$  in both depressions (**Supplementary Table 2; Figure 3 and Figure 4**). While most pores exhibit connectivity or partial connectivity, non-connected pores are discernible as



**Figure 7:** XRD patterns of main clay minerals in the Upper Miocene sandstones of the Sava and Drava depressions: A – Sample S4\_5a; B – Sample S5\_3; C – Sample D4\_5b; D – Sample D6\_8a. Chl = chlorite, Dvm = dioctah. vermiculite, DIV = dioctah. Illite/vermiculite, Ill = illite, Ill/Sm = illite/smectite, Ill/Chl = illite/chlorite, Kln = kaolinite, Kln/Sm = kaolinite/smectite.

well, particularly within samples where carbonate cement establishes grain-to-grain bonds. Secondary porosity, stemming from partial to complete dissolution of detrital grains and cements is chiefly evident in feldspars (both alkali feldspars and plagioclase; **Figure 4 E, F, G**) and phyllosilicates (mica and chlorite; **Figure 4 A, B**), with sporadic occurrences within rock fragments (predominantly carbonates; **Figure 4 D**), and is infrequently also associated with cement (carbonate and feldspar cements). Micropores are present as diminutive pores (less than 2  $\mu\text{m}$ ), primarily linked with matrix within intergranular spaces or differently oriented detrital and authigenic clay minerals (**Figure 4 and Figure 6**), collectively contributing to overall porosity. Fracture porosity, manifested as void spaces within natural fractures, is particularly rare within samples, primarily macroscopically observed in the deepest intervals.

## 5. Discussion

### 5.1. Uniformity of sandstones and diagenetic alterations

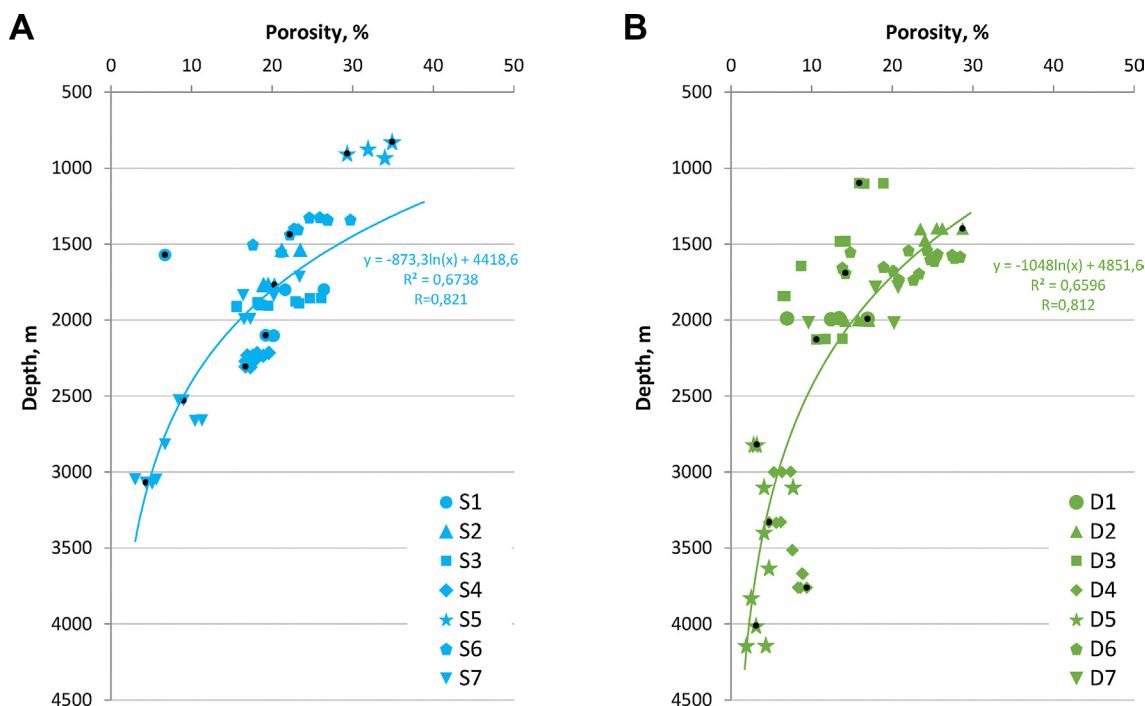
The sandstones within the Sava and Drava depressions exhibit remarkable similarities in grain size distribution, mineralogical composition, and diagenetic alterations (**Supplementary Tables 1-3; Figures 3-7**). This uniformity in grain shape and modal composition suggests the shared provenance for the detrital material previously investigated by **Matošević et al. (2023a,**

**2024)**. This study further elucidates comparable diagenetic processes within the depressions, particularly highlighting the compaction progressions and cementation that significantly influence sandstone porosity. As sandstone layers compact over time under the pressure of overlying sediments, intergranular spaces diminish, leading to a reduction in porosity with depth (**Figure 3 and Figure 8**). Various grains undergo rearrangement during shallow and deep burial, with ductile grains (e.g., volcanic rock fragments, mudrock intraclasts, metasediments, and phyllosilicates; **Figure 3 and Figure 4**) and matrix/pseudomatrix (**Figure 4 C**) plastically deforming, while others partially dissolve (e.g., feldspar and carbonate rock fragments; **Figure 4**) or may even fracture (e.g., quartz). These processes contribute to overall compaction of sandstones (**Waugh, 1971; Pittman and Larese, 1991; Bjørlykke and Egeberg, 1993; Worden et al. 1997; Worden and Burley, 2003**). Additionally, chemical compaction, including pressure dissolution of silicate minerals present in the sandstones, also contributes, but less significantly, to porosity reduction with increasing burial depth (**Robin, 1978**).

### 5.2. Grain contacts, compaction dynamics, and cementation

Examination of grain contacts reveals a spectrum of contact types evolving through compaction and associated pressure dissolution processes, ranging from point-contacts to interpenetrative (even sutured) contacts (**Fig-**





**Figure 8:** Depth-wise distribution of porosity values in the Upper Miocene sandstones of the Sava (A) and Drava (B) depressions, showing reductions in porosity with depth. Black dots indicate samples that underwent detailed petrographic, SEM-EDS and XRD analyses. For detailed measurement values per depth per sample, refer to the **Supplementary Table 4**.

ure 3). The dynamic nature of compaction highlights the significance of comprehending the evolution of grain contacts, which promote the development of more extensive contacts over time and depth (Taylor, 1950; Robin, 1978). Moreover, water expulsion during compaction facilitates cementation, with quartz and carbonate cements emerging as prominent contributors to porosity reduction in the sandstones within both depressions, manifesting in various forms (Figure 3 and Figure 5). Quartz overgrowths and microcrystalline quartz cement contribute to pore space reduction (Figure 5), particularly at burial diagenesis temperatures surpassing 70°C (Bjørlykke and Egeberg, 1993). Quartz cement predominantly occurs in sandstones deeper than 1000 m, with the expected temperatures in the subsurface resulting from elevated geothermal gradient within the North Croatian Basin (e.g., Cvetković et al., 2019; Macenić et al., 2020; Tuschl et al., 2022). Carbonate cement, primarily composed of calcite and Fe-dolomite/ankerite, significantly influences porosity reduction during both eogenesis and mesogenesis in the sandstones from both depressions (Figure 3 C, D and Figure 5 A, B, C). Calcite typically represents the early eogenetic grain-to-grain pore-filling cement fabrics, precipitating from pore waters post-deposition, irrespective of the primary mineralogy of the sandstones (Figure 3 D and Figure 5 A). Late-stage calcite cement and Fe-dolomite/ankerite are typically associated with dissolution and recrystallization of pre-existing carbonate minerals, including carbonate rock fragments, and cements, and precipitation from pore fluids in deeper intervals (Worden and Burley, 2003, and references there-

in). Fe-dolomite/ankerite, and infrequently Fe-calcite, dominate among carbonate cements during mesogenesis, crystallizing at temperatures approximately 100°C (Morad, 1998; Worden and Burley, 2003), and locally fill pores and pore throats in the form of rhombohedral crystals, thereby contributing to the reduction of porosity and local permeability of the sandstones (Figure 5 B). Some burial calcite and Fe-dolomite/ankerite cement may also originate from crystallization due to mass fluid and solute transfer, migrated from organic mudrocks intercalated with sandstones in the basin (Jackson and Beales, 1967; Baines et al., 1991). Additionally, it is known that oil can also influence carbonate cementation, particularly during biodegradation and oxidation processes (Ehrenberg and Jakobsen, 2001). Calcite and Fe-dolomite/ankerite cements emerge as the predominant factors responsible for porosity reduction in the Upper Miocene sandstones of the Sava and Drava depressions.

### 5.3. The role of clay minerals

The clay mineral assemblage in both the Sava and Drava depressions exhibits similar major components, including illite, chlorite, kaolinite, and other identified species (Supplementary Table 2 and Supplementary Table 3; Figure 6 and Figure 7), independent of sample depth. However, SEM observations indicate a shift from detrital to authigenic clays with increasing depth. Authigenic clay minerals play a crucial role in altering pore space and fluid flow within the sandstones (Worden and Burley, 2003). They primarily act as pore-filling cement or coatings on the main grains.



Diagenetic illite, especially fibrous types formed at temperatures above 70°C (**Figure 6 A, B**), is associated with potassium-bearing formation waters, significantly reducing permeability by blocking pore throats and reducing pore networks without notably affecting porosity (**Neasham, 1977; Warren and Curtis, 1989**). Authigenic chlorite typically formed below 2000 m depth (with temperatures exceeding 90–100°C; **Ehrenberg, 1993; Aagaard et al., 2000**). As coatings on mineral grains, chlorite decreases porosity and permeability by clogging pore throats and reducing pore network connectivity (**Figure 6 C**). Conversely, in deeper intervals, it may locally inhibit quartz cementation by preventing quartz overgrowths (**Figure 6 D**). Kaolinite is commonly clustered within pores or adhering to primary grains from shallower intervals (**Figure 6 E, F, G**), resulting from the decomposition of primary grains like feldspars. It has a dual effect on porosity and permeability – reducing primary intergranular, but locally increasing secondary intragranular porosity. Kaolinite is more abundant in samples with higher porosity, i.e., in samples having less carbonate cement, and is usually absent in the deepest horizons, especially in the Drava depression (**Supplementary Table 3**). Additionally, the reaction of kaolinite with K-feldspar to produce illite and quartz is significant, particularly at temperatures higher than about 70°C but becomes pervasive at temperatures exceeding 130°C (**Worden and Burley, 2003**). Moreover, kaolinite, typical of weathering profiles and early diagenesis, forms vermiform masses that occupy and locally fill pores, with possible replacement by chlorite in deeper potassium-deficient systems. Highly saline water and common aqueous metals, such as Na, K, Ca, and Mg, can locally affect clay, carbonate, and feldspar mineral stability in the sandstones, causing various diagenetic changes, such as additional albitization of K-feldspar and illitization of smectite and kaolinite (**de Caritat and Barker, 1992; Worden et al., 1999**). These results to some extent coincide with those obtained from the earlier study on the pelitic sediments from the Sava depression (c.f., **Grizelj et al., 2011**), particularly concerning the composition and alteration of clay minerals and carbonates with increasing depth – e.g., smectite, kaolinite, and calcite are gradually replaced by illite-smectite, illite, chlorite, Cadolomite/ankerite, and albite in deeper intervals of exploration wells.

Variations in mixed-layered clay species occur, with illite/smectite being the most frequent, followed by illite/chlorite. The presence of illite/smectite, though widespread, is generally absent at depth intervals around 2500–3000 meters. Although the replacement of smectite by illite in sandstones accompanies early stages of oil generation, illite content in illite/smectite remains relatively consistent depthwise in the sandstones (**Supplementary Table 3**), likely due to conflicting influences of detrital and authigenic illite. Illite crystallinity follows a more regular pattern throughout the drilled sections, with

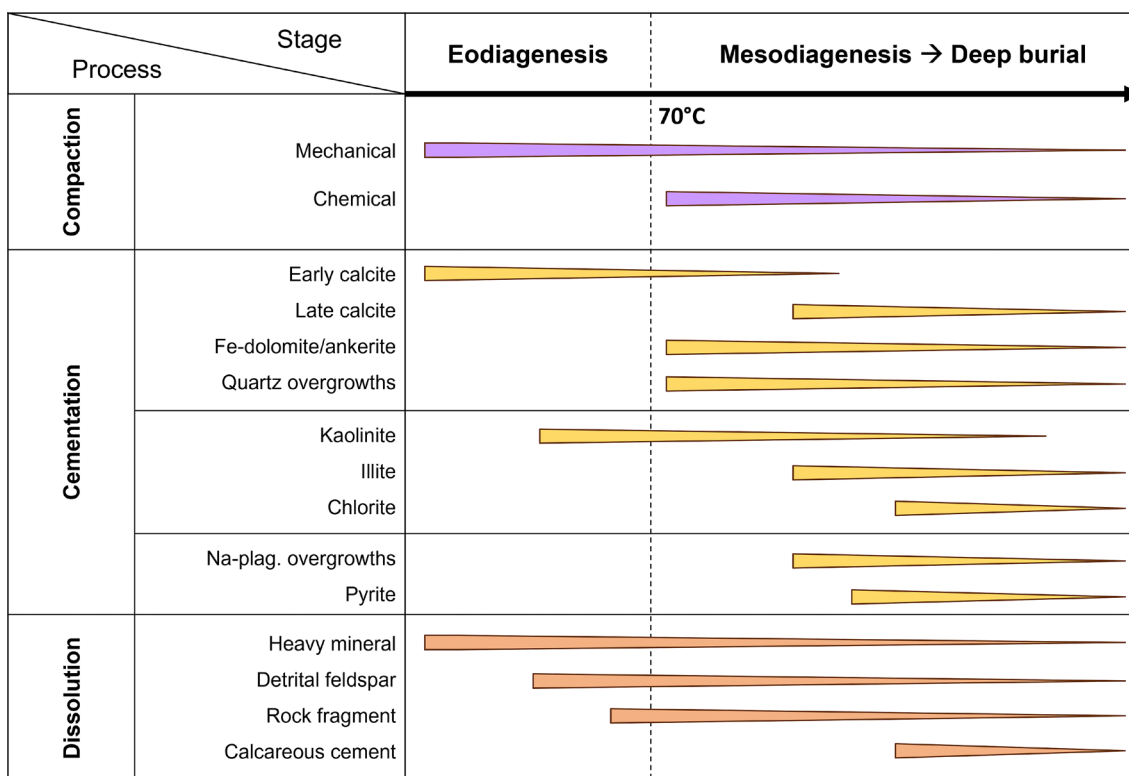
higher crystallinity observed in shallower horizons, possibly inherited from detrital illite, while crystallinity decreases in deeper horizons due to diagenetic changes. Illite/chlorite occurrence was recorded only in the Sava depression sample set, less prominent in the intervals 1500–2300 m in depth (**Supplementary Table 3**). Other clays are sparse: kaolinite/smectite and dioctahedral vermiculite were observed only in two samples from the shallowest horizons of the Sava depression, likely detrital and indicating transitional stages in clay mineral formation during weathering; dioctahedral illite/vermiculite seems more widespread among the samples of both depressions but generally occurs at deeper horizons, suggesting fine changes in mixed layered phases caused by diagenesis (**Supplementary Table 3**).

Smectite clay minerals and their interstratified types, known for reducing permeability due to their small grain size and swelling properties (**Hermanns Stengele and Plötze, 2000**), are prevalent in shallower horizons (**Supplementary Table 3**). However, aforementioned illite and chlorite abundance can also significantly reduce permeability through the drilled sections.

Furthermore, common feldspar mineral cements and overgrowths on main grains in the Upper Miocene sandstones from both the Sava and Drava depressions, although much less abundant than quartz and carbonates, also contribute to pore-filling cements (**Figure 5 H**), with burial diagenesis promoting their stability (**Morad et al., 1989**). The rarely present pyrite cement (**Figure 5 I**), usually formed under the microbial reduction of ferric iron and sulphates due to influx of saline water and reduction of hematite in the presence of hydrocarbons during both eogenesis and mesogenesis (**Love, 1967; Elmore et al., 1987**), has an insignificant impact on porosity reduction in the sandstones from both depressions.

#### 5.4. Porosity evolution and reservoir quality

Understanding porosity in sandstones involves a complex interplay of depositional, compaction, and cementation processes (**Worden et al., 2000**). The changes in porosity observed with depth in both depressions underscore the cumulative effects of grain rearrangement, compaction, and cementation, emphasizing the dynamic nature of diagenetic processes shaping these reservoirs' properties. Initially, primary intergranular porosity dominates in both depressions (**Figure 3 and Figure 4**), with observable reduction in intergranular pore sizes with depth. Furthermore, it is observed that secondary porosity, resulting from dissolution and redistribution processes, plays a significant role in the overall porosity evolution of the studied sandstones, which conforms with the observations reported by **Schmidt and McDonald, 1979** and **Giles and de Boer, 1990**. This secondary porosity is primarily associated with feldspar, pylosilicates, and carbonate rock fragments (**Figure 4 B, D, E, F, G**). Additionally, microporosity, primarily at-



**Figure 9:** Generalized paragenetic sequence of diagenetic processes in the Upper Miocene sandstones of the North Croatian Basin based on analyzed samples from the Sava and Drava depressions. The same diagenetic processes (compaction, cementation, and dissolution) were observed in both Sava and Drava depressions.

tributed to matrix within intergranular spaces or variously oriented detrital and authigenic clay minerals (**Figure 6**), contributes collectively to overall porosity. Notably, the dissolution of heavy minerals which was documented at deeper sections of studied sandstones (**Matošević et al., 2023a, 2024**) does not significantly affect the overall proportion of porosity (**Figure 4 I**).

Established relationship between porosity and depth of the studied sandstones, suggests a heterogeneous nature of the Upper Miocene reservoirs' quality within the study area (**Supplementary Table 4; Figure 8**). Relatively strong relationship between porosity and depth in both depressions implies that compaction plays a significant role in porosity reduction of studied sandstones (**Figure 8**), but also that influence of other diagenetic processes generally intensifies with depth. The close alignment between mean and median values indicates a relatively even distribution of data around the central value, with no significant outliers disproportionately influencing the mean.

Moreover, the identified diagenetic paragenetic sequences unveil the intricate interplay between mineralogical transformations and evolution of petrophysical properties (**Figure 9**), ultimately defining reservoir quality and potential production (**Worden and Morad, 2000**). These findings underscore the importance of considering diagenetic processes in understanding reservoir

quality and the potential implications for production strategies in the broader context of the Pannonian Basin System and similar geological settings. Further research in this field is warranted to refine our understanding of these processes and their implications for reservoir management and exploration efforts.

## 6. Conclusions

This study sheds light on the diagenetic evolution of the Upper Miocene lacustrine sandstones in the North Croatian Basin, providing insights into their reservoir characteristics relevant for geoenery potential. Uniform diagenetic processes were found in both the Sava and Drava depressions, with carbonate cementation, especially calcite and Fe-dolomite/ankerite, being the main contributors to porosity reduction, along with other cements like clay minerals, quartz, feldspar, and pyrite. Compaction-induced porosity reduction, evidenced by evolving grain contacts and pressure dissolution, is widespread, leading to decreased porosity with depth. Porosity measurements confirm this trend, with values decreasing from 34.9% to 3.0% in the Sava depression and from 28.7% to 1.9% in the Drava depression, respectively. Primary intergranular porosity predominates, with secondary porosity, mainly intragranular from dissolution processes, also significant. Pore diameters typi-

cally range from 5-100  $\mu\text{m}$ . Microporosity is particularly attributed to matrix, some detrital grains, and differently oriented clay minerals. The observed paragenetic sequences elucidate the intricate relationship between mineralogical transformations and evolution of petrophysical properties of the reservoirs. Clay minerals, both detrital and authigenic, interact in a complex manner with other diagenetic processes, further influencing porosity and permeability. The main clay minerals identified include illite, chlorite, kaolinite, and mixed-layered illite/smectite. Authigenic clay minerals appear at specific depths, acting as pore-filling cement or coatings on mineral grains, and additionally inhibit fluid flow. Variations in mixed-layered clay species reflect the dynamic nature of diagenetic processes. These findings highlight the complexity of diagenetic interactions and their implications for reservoir quality and potential productivity. Understanding these processes is crucial for predicting reservoir quality and fluid migration pathways. This study also highlights the need for future exploration, particularly concerning permeability measurements and the interaction of diagenetic changes and permeability of the sandstones at various depths. A more extensive sampling and exploration wells network should be included in further studies to gain a broader understanding of the Upper Miocene sandstone reservoirs' quality and their differences between depressions in the North Croatian Basin and wider Pannonian Basin System.

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## APPENDIX

### SUPPORTING INFORMATION

Supporting Information is hosted online at <https://hr-cak.srce.hr/ojs/index.php/rgn/article/view/30841>

## SAŽETAK

### Procjena kvalitete ležišta: Otkrivanje dijagenetskih promjena mineraloškim i petrofizičkim analizama gornjomiocenskih jezerskih pješčenjaka u Panonskome bazenskom sustavu, Hrvatska

Gornjomiocenski jezerski pješčenjaci Sjevernohrvatskoga bazena, smještenoga u jugozapadnome dijelu Panonskoga bazenskog sustava, čine važna ležišta ugljikovodika, no razvoj njihovih dijagenetskih procesa još uvijek nije dovoljno istražen. Ovo istraživanje nudi sveobuhvatan pregled dijageneze tih pješčenjaka analizirajući uzorke iz istražnih bušotina u Savskoj i Dravskoj depresiji. Koristeći se petrografskim analizama, skenirajućom elektronskom mikroskopijom s energijski disperzivnom rendgenskom spektroskopijom (SEM-EDS), rendgenskom difrakcijom (XRD) i petrofizičkim mjerenjima, cilj istraživanja bio je objasniti dijagenetske procese koji utječu na kvalitetu ležišta nafte i plina i njihovu produktivnost. Rezultati otkrivaju konzistentnu distribuciju veličine zrna, modalnoga sastava i dijagenetskih promjena pješčenjaka u objema depresijama. Kompakcija, koja se očituje kroz razvoj zrnskih kontakata i tlačno otapanje, dovodi do smanjenja poroznosti s dubinom. Karbonatni cementi, osobito kalcit i Fe-dolomit/ankerit, glavni su čimbenici smanjenja primarne međuzrnske poroznosti, uz minerale glina, kvarc, feldspate itd. Sekundarna poroznost rezultat je otapanja minerala i procesa redistribucije koji također znatno utječu na cjelokupan razvoj poroznosti. Minerali glina, detritalni i autigeni, pokazuju kompleksno međudjelovanje s drugim dijagenetskim procesima, dodatno utječući na smanjenje poroznosti i propusnosti. Autigeni minerali glina, uključujući ilit, klorit i kaolinit, djeluju kao cementi koji ispunjavaju pore ili se javljaju u vidu prevlaka na zrnima, sprečavajući dodatno protok fluida. Paragenetski procesi ocrtavaju složeni odnos između mineraloških transformacija i petrofizičkih svojstava definirajući kvalitetu ležišta. Razumijevanje dinamike dijagenetskih procesa ključno je za predviđanje kvalitete ležišta, putove migracija fluida i ugljikovodičnu produktivnost. Ovo istraživanje popunjava bitnu prazninu u znanju o dijagenezi gornjomiocenskih jezerskih pješčenjaka u jugozapadnome dijelu Panonskoga bazena, pružajući uvide važne za energetski sektor i podržavajući održivi razvoj resursa u regiji.

#### Ključne riječi:

pješčenjački rezervoari, dijageneza, poroznost, Sjevernohrvatski bazen, kasni miocen

#### Author's contribution

**Mario Matošević (1)** (PhD student at the Faculty of Mining, Geology and Petroleum Engineering, University of Zagreb, and Geology Expert at INA – Industrija nafte d.d.) provided sample collection, sample preparation, petrographic, mineralogical, and SEM-EDS analyses, data interpretation, interpretation of petrophysical results, conceptualization and drafting of the article, and graphical representations. **Nenad Tomašić, PhD (2)** (Full Professor at the Faculty of Science, University of Zagreb) provided sample preparation, XRD analyses, data interpretation, interpretation of XRD results and comparison with petrophysical data. **Adaleta Perković (3)** (PhD and Core Analyses Expert at INA – Industrija nafte d.d.) provided sample preparation, petrophysical (porosity) measurements, data interpretation, interpretation of petrophysical results and comparison with mineralogical data, and graphical representations. **Štefica Kampić (4)** (Expert at the Faculty of Science, University of Zagreb) provided sample preparation and assistance with XRD analyses. **Marijan Kovačić, PhD (5)** (Full Professor at the Faculty of Science, University of Zagreb) provided conceptualization and drafting of the article, data interpretation, interpretation of mineralogical results, and contributing with geology of the Neogene North Croatian Basin. **Davor Pavelić, PhD (6)** (Full Professor at the Faculty of Mining, Geology and Petroleum Engineering, University of Zagreb) provided conceptualization and drafting of the article, data interpretation, and contributing with geology of the Neogene North Croatian Basin.

### 3. DISCUSSION

#### 3.1. Provenance analysis

The Upper Miocene sandstones from the Sava and Drava depressions share similar compositions, primarily classified as feldspatho-litho-quartzose carbonaticlastic sandstones (**Garzanti, 2016, 2019**). They predominantly consist of carbonate sedimentary rock fragments such as dolostones and limestones, alongside fragments of various metamorphic and igneous rock fragments (including mica schists, quartzites, gneisses, slates, phyllites, and granitoid rocks). The composition suggests their origin from cover strata with contributions from both metamorphic and igneous sources. Detected small-scale originals of source rocks indicate recycled carbonate-clastic, axial belt metamorphic complex, and continental-block orogenic provenance (**Garzanti, 2016**). Examining the modal composition using the QFL or QtFL classification schemes proposed by **Dickinson (1985)** and **Weltje (2006)**, these sandstones align with a tectonic setting indicative of a recycled orogen, such as subduction complex or fold-thrust belt. This tectonic setting reflects the collisional processes within the Alpine-Carpathian-Dinaridic orogenic system, which amalgamate various tectonic units.

The heavy mineral composition in the Upper Miocene sandstones of the North Croatian Basin has been observed to bear similarities both on surface and in deep exploration wells (**Šćavničar, 1979; Kovačić & Grizelj, 2006**), suggesting a consistent association. This similarity, despite potential diagenetic modifications, indicates a reflection of the source area's composition. The heavy minerals present in the sandstones likely originate from two primary groups of parent rocks: low- to medium-grade dynamo-thermal metamorphic rocks and felsic to intermediate igneous rocks, either as primary minerals or their alteration products. Certain heavy minerals, such as rutile, tourmaline, zircon, and barite, exhibit resistance to weathering and transport, possibly suggesting a recycled sedimentary origin as well. Rock fragments in the sandstones, excluding limestones and dolostones, correspond to potential sources of the heavy mineral assemblage. Therefore, it is reasonable to infer that the sandstones from the Sava and Drava depressions predominantly derive their mineral content from carbonate rocks, followed by low to medium-grade metamorphic rocks, and to a lesser extent, granitoid rocks.

Results suggest that the Upper Miocene sandstones likely originate from Mesozoic carbonates, Proterozoic to Palaeocene low- to medium-grade metamorphic rocks, and



Palaeozoic granitoids found in the Eastern Alps and Southern Alps. There may have been some minor contributions from detritus sourced from the Western Carpathians. However, significant input from the Dacia tectonic mega-unit of the Eastern and Southern Carpathians, as well as from the Dinarides, is excluded, despite their closer geographic proximity to the North Croatian Basin than the Eastern Alps. The absence of significant amounts of mafic and ultramafic rocks, radiolarites, and heavy minerals like chrome spinel in the Upper Miocene sandstones of the Sava and Drava depressions further supports the exclusion of the Dinarides, Southern Carpathians, and Eastern Carpathians as primary source areas (cf. **Lužar-Oberiter et al. 2009; Ustaszewski et al. 2009, 2010; Lužar-Oberiter et al. 2012**). A sole provenance from the Western Carpathians would have led to a higher content of volcanic or volcanoclastic material (e.g., **Seghedi & Downes 2011**). Therefore, based on the lithic fragments identified in the sandstones, the Eastern Alps are proposed as the most likely major source area.

Provenance from metamorphic units of the Eastern Alps is supported by the transparent heavy mineral suites in the sandstones. The sandstones predominantly contain almandine-rich garnet along with chloritoid, biotite, kyanite, epidote, staurolite, and zoisite. Similar mineral assemblages are found in modern river sediments originating from metamorphic rocks of the Eastern Alps, dominated by the ALCAPA (Adria-derived) tectonic mega-unit, including the Austroalpine and Peninic nappes (e.g., Drava and Mura; **Schmid et al., 2008, 2020; Garzanti et al., 2010; Arató et al., 2021**). The main sources of these minerals are 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**). While the moderately high ZTR index ( $15 \pm 8$ ) might indicate significant recycling from older sandstones (**Hubert, 1962; Garzanti, 2017**), in this case, it is primarily due to diagenetic dissolution of unstable heavy minerals underground. The selective dissolution of ferromagnesian minerals indicates that the heavy mineral assemblages have been altered, particularly in the Drava depression, and thus do not represent the original mineral suite. Therefore, the focus was on garnet, a mineral relatively resistant to burial diagenesis (**Morton & Hallsworth, 2007; Garzanti et al., 2018**). Garnet grains mainly belong to Type B, typical of amphibolite-facies metasedimentary rocks and granitoids (**Mange & Morton, 2007**), with some Type C, found in high-grade metamafic rocks and quartz-biotite gneisses, and Type A minerals, found in granulite-facies metasediments. This classification aligns with the garnet variation diagram by **Aubrecht et al., (2009)** revealing most garnets align within amphibolite-facies conditions as well (Group 4, 5, 6, and 7).

This provenance inference of the Eastern Alps is supported by predominant paleoflow directions from W/NW and N/NW, as observed in outcrop measurements and interpreted from seismic profiles (progradation of the Upper Miocene sandstones' clinofolds) in both depressions (Ivković et al., 2000; Saftić et al., 2003; Kovačić et al., 2004; Kovačić & Grizelj, 2006; Magyar et al., 2013; Sebe et al., 2020; Špelić et al., 2023). In some parts of the Drava depression, sedimentary systems locally exhibit clinofold formations with shelf-margin slope progradation from N/NE towards S/SW, as evidenced by seismic data. These findings collectively indicate the Eastern Alps (potentially including the Southern Alps) as significant sources of the Upper Miocene sandstones, with a possible minor contribution from the Carpathians (Western Carpathians and potentially Eastern Carpathians; Šćavničar, 1979; Kovačić & Grizelj, 2006; Kovačić et al., 2011; Magyar et al., 2013; Sztanó et al., 2013). Furthermore, no significant progradation of sandstone clinofolds from seismic profiles suggests a primary material inflow from S, SW, or SE directions into the Sava and Drava depressions.

The similar composition of the sandstones in the Sava and Drava depressions suggests they were part of the same sedimentary system within the lake, with a common dispersal path from source to sink, possibly connected by the Paleo-Sava and/or Paleo-Drava rivers. Uncertainty persists regarding their connection with the Paleo-Danube (Magyar et al., 2013; Sebe et al., 2020), necessitating further provenance studies from the Hungarian part of the Pannonian Basin System. The substantial influx of detritus into Lake Pannon during the Late Miocene (Bérczi et al., 1988; Juhász, 1994; Magyar et al., 2013; Pavelić & Kovačić, 2018; Andelković & Radivojević, 2021) resulted in significant sediment thicknesses in the Sava and Drava depressions (Ivković et al., 2000; Saftić et al., 2003; Kovačić et al., 2004; Kovačić & Grizelj, 2006; Malvić & Velić, 2011; Pavelić & Kovačić, 2018). Local parts of the North Croatian Basin basement are unlikely to have been significant sources of the material due to their relatively small volumes. Islands within the lake and surrounding peninsulas might have contributed smaller amount of detritus (Bérczi, 1988; Kovačić & Grizelj, 2006; Sztanó et al., 2015). However, their contribution was likely limited as the lake reached its largest extent around 10.5 Ma ago (Magyar et al., 1999), submerging all islands due to high subsidence rates and a wet climate (Pogácsás, 1984; Sztanó et al., 2013; Balázs et al., 2018). Moreover, Upper Miocene sediments currently cover parts of the North Croatian Basin's mountains, indicating significant uplift after the Late Miocene, during the later compressional phase of the basin evolution in the Pliocene and Quaternary (e.g., Pavelić, 2001; Tomljenović & Csontos, 2001; Márton et al., 2002; Pavelić & Kovačić, 2018;

**Kurečić et al., 2021**). During the Late Miocene, the Eastern Alps experienced notable uplift, coinciding with rapid subsidence of the Pannonian Basin System, and facilitating substantial fluvial sediment transfer from the mountains to the basin. Previous studies in the North Croatian Basin support 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 geochemical composition of the studied sandstones reflects the presence of both detrital and authigenic minerals. This is evident in the enrichment of CaO and MgO, attributed to the prevalence of calcite, dolomite, and ankerite, both detrital and diagenetic in origin. The abundant carbonate cement contributes to the enrichment of Ca and Mg, while Na, K, Al, and Fe are depleted compared to the Upper Continental Crust standard (UCC; **Taylor & McLennan, 1985**). Calcite cement is predominant in deeper-water sediments, particularly in the Drava depression, although fully cemented sandstones are also found in shallower deposits, making it challenging to ascertain provenance information solely from the geochemical data. The lack of correlation between Ba and K<sub>2</sub>O, alongside the presence of high Ba content in certain samples, indicates the presence of barite as confirmed by SEM-EDS analyses.

Comparative analyses of the geochemical signatures of the Upper Miocene sandstones in the North Croatian Basin with sediments from adjacent basins in the Po-Adriatic region, focusing on elements like Ni, Cr, or V indicative of mafic-ultramafic source rocks (e.g., **McLennan et al., 1993; Amorosi et al., 2022**), suggest a closer resemblance to river sediments originating from the Eastern Alps rather than the Dinarides or the Carpathians. Consistent CIX and alpha values imply minor weathering effects, with very low alpha indices for Ca and Mg and low values for Sr reflecting carbonate abundance. The narrow range between 1 for Rb and 1.5 for Na indicates minimal weathering intensity. Additionally, analyses of the Th/U ratio dismiss significant weathering processes, aligning with the inconsequential impact of sedimentary sorting or recycling as indicated by the Zr/Sc ratio (**McLennan et al., 1993**). These findings corroborate petrographic analyses suggesting early-stage weathering from high mountain relief area.

### **3.2. Diagenetic alterations**

The Upper Miocene sandstones from the Sava and Drava depressions share similar diagenetic changes. This study explores these processes, focusing on compaction and cementation, which significantly affect the porosity of the sandstones. As sedimentary layers compact



under pressure, the spaces between grains in sandstones decrease, reducing porosity with depth. Different grains undergo rearrangement during burial, with some deforming plastically (e.g., volcanic rock fragments, mudrock intraclasts, metasediments, and phyllosilicates) while others dissolve (e.g., feldspar and carbonate rock fragments) or fracture (e.g., quartz). These processes, observed in both shallow and deep burial conditions, lead to overall compaction of the sandstones (**Waugh, 1971; Pittman & Larese, 1991; Bjørlykke & Egeberg, 1993; Worden et al., 1997; Worden & Burley, 2003**). Chemical compaction, particularly pressure dissolution of silicate minerals, also contributes to porosity reduction with increasing burial depth, although to a lesser extent (**Robin, 1978**).

Analysis of grain contacts unveils a variety of contact types evolving during compaction and pressure dissolution, ranging from point-contacts to interpenetrative (even sutured) contacts. Understanding the evolution of grain contacts is crucial due to the dynamic nature of compaction, which leads to the development of more extensive contacts over time and depth (**Taylor, 1950; Robin, 1978**). Compaction-induced water expulsion promotes cementation, with quartz and carbonate cements being prominent contributors to porosity reduction in the sandstones within both depressions, appearing in various forms.

Quartz overgrowths and microcrystalline quartz cement contribute to pore space reduction, particularly at burial diagenesis temperatures exceeding 70° C (**Bjørlykke & Egeberg, 1993**). Quartz cement is prevalent in sandstones deeper than 1000 m, aligning with the average geothermal temperatures within the North Croatian Basin (e.g., **Macenić et al., 2020; Tuschl et al., 2022**).

Carbonate cement, mainly composed of calcite and Fe-dolomite/ankerite, plays a significant role in porosity reduction during both eogenesis and mesogenesis in sandstones from both depressions. Calcite typically represents early eogenetic grain-to-grain pore-filling cement fabrics, precipitating from pore waters post-deposition, regardless of the primary mineralogy of the sandstones. Late-stage calcite cement and Fe-dolomite/ankerite are typically associated with dissolution and recrystallization of pre-existing carbonate minerals, including carbonate rock fragments and cements, and precipitation from pore fluids in deeper intervals. Fe-dolomite/ankerite, and occasionally Fe-calcite, dominate among carbonate cements during mesogenesis, crystallizing at temperatures around 100° C (**Morad, 1998; Worden & Burley, 2003**), filling pores and pore throats in the form of rhombohedral crystals, thereby reducing porosity and local permeability of the sandstones. Some burial calcite and Fe-dolomite/ankerite cement may also originate from crystallization due to mass fluid and solute transfer, migrated from organic mudrocks intercalated with sandstones in the basin

(**Jackson & Beales, 1967; Baines et al., 1991**). Furthermore, oil can influence carbonate cementation, especially during biodegradation and oxidation processes (**Ehrenberg & Jakobsen, 2001**). Calcite and Fe-dolomite/ankerite cements emerge as the predominant factors responsible for porosity reduction in the Upper Miocene sandstones of the Sava and Drava depressions.

The clay mineral composition in both the Sava and Drava depressions shows similar major components, such as illite, chlorite, kaolinite, and other identified species, regardless of sample depth. However, SEM observations reveal a transition from detrital to authigenic clays with increasing depth. Authigenic clay minerals play a crucial role in altering pore space and fluid flow within the sandstones (**Worden & Burley, 2003**), primarily by acting as pore-filling cement or coatings on the main grains.

Diagenetic illite, particularly fibrous types formed at temperatures above 70° C, is associated with potassium-bearing formation waters, significantly reducing permeability by blocking pore throats and reducing pore networks without notably affecting porosity (**Neasham, 1977; Warren & Curtis, 1989**). Authigenic chlorite typically forms below 2000 m depth (with temperatures exceeding 90–100° C; **Ehrenberg, 1993; Aagaard et al., 2000**), reducing porosity and permeability by clogging pore throats and decreasing pore network connectivity. Conversely, in deeper intervals, it may locally inhibit quartz cementation by preventing quartz overgrowths. Kaolinite, commonly found clustered within pores or adhering to primary grains from shallower intervals, results from the decomposition of primary grains like feldspars. It has a dual effect on porosity and permeability, reducing primary intergranular but locally increasing secondary intragranular porosity. Kaolinite is more abundant in samples with higher porosity, usually absent in the deepest horizons, especially in the Drava depression. Additionally, the reaction of kaolinite with K-feldspar to produce illite and quartz is significant, particularly at temperatures greater than about 70° C but becomes pervasive at temperatures exceeding 130° C (**Worden & Burley, 2003**). Moreover, kaolinite, typical of weathering profiles and early diagenesis, forms vermiform masses that occupy and locally fill pores, with possible replacement by chlorite in deeper potassium-deficient systems. Highly saline water and common aqueous metals, such as Na, K, Ca, and Mg, can locally affect clay, carbonate, and feldspar mineral stability in the sandstones, causing various diagenetic changes, such as additional albitization of K-feldspar and illitization of smectite and kaolinite (**de Caritat & Barker, 1992; Worden et al., 1999**).

Variations in mixed-layered clay species occur, with illite/smectite being the most frequent, followed by illite/chlorite. The presence of illite/smectite, though widespread, is

generally absent at depth intervals around 2500-3000 m. Although the replacement of smectite by illite in sandstones accompanies early stages of oil generation, illite content in illite/smectite remains relatively consistent depthwise in the sandstones, likely due to conflicting influences of detrital and authigenic illite. Illite crystallinity follows a more regular pattern throughout the drilling profile, with higher crystallinity observed in shallower horizons, possibly inherited from detrital illite, while crystallinity decreases in deeper horizons due to diagenetic changes. Illite/chlorite occurrence was recorded only in the Sava depression sample set, less prominent in the intervals 1500-2300 m in depth. Other clays are sparse: kaolinite/smectite and dioctahedral vermiculite were observed only in two samples from the shallowest horizons of the Sava depression, likely detrital and indicating transitional stages in clay mineral formation during weathering; dioctahedral illite/vermiculite seems more widespread among the samples of both depressions but generally occurs at deeper horizons, suggesting fine changes in mixed layered phases caused by diagenesis.

Smectite clay minerals and their interstratified types, known for reducing permeability due to their small grain size and swelling properties (**Hermanns Stengele & Plötze, 2000**), are prevalent in shallower horizons. However, illite and chlorite abundance can also significantly reduce permeability through the drilling sections.

Furthermore, common feldspar mineral cements and overgrowths on main grains in the sandstones, although much less abundant than quartz and carbonates, also contribute to pore-filling cements, with burial diagenesis promoting their stability (**Morad et al., 1989**). The rarely present pyrite cement, usually formed under the microbial reduction of ferric iron and sulphates due to influx of saline water and reduction of hematite in the presence of hydrocarbons during both eogenesis and mesogenesis (**Love, 1967; Elmore et al., 1987**), has an insignificant impact on porosity reduction in the sandstones.

Understanding the porosity dynamics in sandstones involves intricate interactions among deposition, compaction, and cementation processes (**Worden et al., 2000**). The variations in porosity with depth in depressions highlight the cumulative impacts of grain rearrangement, compaction, and cementation, emphasizing the dynamic nature of diagenetic processes shaping underground reservoir properties. Initially, primary intergranular porosity predominates in both depressions, gradually diminishing with depth. Additionally, secondary porosity resulting from dissolution and redistribution processes, primarily associated with feldspar, phyllosilicates, and carbonate rock fragments, significantly influences porosity evolution (**Schmidt & MacDonald, 1979; Giles & de Boer, 1990**). Microporosity, attributed to matrix within intergranular spaces or clay minerals, also contributes to overall porosity.



The dissolution of heavy minerals with depth has minimal impact on overall porosity proportions.

Examining the variability in porosity values across depths suggests a heterogeneous reservoir quality within the Upper Miocene formations in the study area. The strong depth-porosity relationship indicates a general decrease in porosity with increasing depth, with very similar correlation coefficients in both depressions ( $R=0.821$  for the Sava depression,  $R=0.812$  for the Drava depression). The alignment between mean and median values suggests an even distribution of data without significant outliers disproportionately affecting the mean.

Moreover, diagenetic paragenetic sequences reveal the complex relationship between mineralogical transformations and petrophysical properties, ultimately defining reservoir quality and production potential (**Worden & Morad, 2000**). These findings underscore the significance of considering diagenetic processes in understanding reservoir quality and their implications for production strategies, particularly in the broader context of the Pannonian Basin System and similar geological settings. Further research is necessary to enhance our understanding of these processes and their implications for reservoir management and exploration efforts.

## 4. CONCLUSION

The Upper Miocene lacustrine sandstones from the Sava and Drava depressions in the North Croatian Basin (SW part of the Pannonian Basin System) exhibit similar characteristics in terms of composition, provenance, and diagenetic evolution.

The sandstones from both depressions are predominantly feldspatho-litho-quartzose carbonaticlastic and are interpreted to have been derived from an area of a recycled orogen. Modal composition and the composition of rock fragments and heavy minerals, along with results from geochemical analyses, suggest an equivalent external source terrain without major local influences. Parent rocks include carbonates (dolostones and limestones), metamorphic rocks (mica schist, quartzites, gneisses, phyllites), and plutonic rocks (granitoids) mainly from the Eastern Alps orogenic belt (ALCAPA tectonic mega-unit), including Austroalpine and Penninic nappes. Poorly 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, indicates the significant effects of selective diagenetic dissolution in the basin and of the possible addition of detritus recycled from older siliciclastic deposits. Late Miocene uplift of the Alps and subsidence of the Pannonian Basin System created the conditions that led to massive fluvial sediment transfer from the orogen into Lake Pannon. The virtually uniform 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 possibly associated with Paleo-Sava and/or Paleo-Drava rivers, following mainly the NW to SE transport direction of detritus, which is also characteristic of the progradation system of the Paleo-Danube but opposite to the system in the SE part of the Pannonian Basin System.

The Upper Miocene sandstones from the North Croatian Basin differ in terms of their texture and composition from the Lower and Middle Miocene sandstones in this area, which are of more local provenance. The results of the study show that no major morphological barriers were present between the Sava and Drava depressions that could have obstructed the inflow of detrital material via rivers and deltas into Lake Pannon during the Late Miocene. These insights, crucial for a better understanding of sedimentary processes and the geological evolution of the Pannonian Basin System, offer a novel perspective to distinguish the Upper

Miocene sandstones from other sedimentary units within the basin, highlighting the delineation of mineralogical and geochemical signatures, which enables improved reservoir characterization, facilitating chemo-stratigraphic assessment and prognosticative modelling in the subsurface.

The Upper Miocene sandstones from the Sava and Drava depressions are the largest oil and gas reservoir rocks in Croatia and are significant for the energy sector, for further hydrocarbon exploration and production activities, as well as initiatives related to energy transition, such as geothermal energy and geological carbon capture and storage technologies. This study offered a comprehensive investigation into the diagenetic processes affecting reservoir quality and hydrocarbon productivity.

Uniform diagenetic processes were found in both the Sava and Drava depression, with consistent grain size distribution, and carbonate cementation (calcite and Fe-dolomite/ankerite) being the main contributor to porosity reduction, along with other cements like clay minerals, quartz, and feldspar. Compaction-induced porosity reduction is evidenced by evolving grain contacts and pressure dissolution. Porosity measurements confirmed decreasing porosity with depth. Primary intergranular porosity predominates, with secondary porosity, mainly intragranular from dissolution processes, also being significant. Microporosity is particularly attributed to matrix, some detrital grains, and differently oriented clay minerals. The observed paragenetic sequences elucidate the intricate relationship between mineralogical transformations and evolution of petrophysical properties of the reservoirs. Clay minerals, including identified illite, chlorite, kaolinite, and mixed-layered types, interact in a complex manner with other diagenetic processes further influencing porosity and permeability. Authigenic clay minerals appear at specific depths, acting as pore-filling cement or coatings on mineral grains, and additionally inhibit fluid flow.

Understanding diagenetic processes is crucial for predicting reservoir quality and fluid migration pathways. The study highlights the need for future exploration, particularly concerning permeability measurements and the interaction of diagenetic changes and permeability of the sandstones at various depths to gain a broader understanding of the Upper Miocene sandstone reservoirs' quality and their differences between depressions in the North Croatian Basin and wider Pannonian Basin System.



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## 6. BIOGRAPHY OF THE AUTHOR

Mario Matošević was born on August 2, 1982, in Zagreb, Croatia. After completing his primary education, he attended the Classical Gymnasium in Zagreb. In 2002, he commenced his studies at the Department of Geology, Faculty of Science, University of Zagreb, where he graduated in 2009, earning the title master of geology on the thesis topic concerning chemical and mineral composition of the Danube stream sediments. During his studies, he worked as a demonstrator for General Paleontology and Paleontology of Invertebrata at the Division of Geology and Paleontology. Since 2012, he has been employed in INA – Industrija nafte d.d. as a geology expert in the Exploration & Production Laboratory. In this role, he has actively participated in both internal and external professional and scientific projects. His work includes conducting mineralogical and petrographic analyses, as well as interpreting data obtained through light polarizing and scanning electron microscopes. His research focuses on the characterization of source and reservoir rocks within the Pannonian Basin System and the Adriatic Basin. In 2016, he was awarded a scholarship by the Faculty of Mining, Geology, and Petroleum Engineering at the University of Zagreb for postgraduate PhD studies in Applied Geosciences, Mining, and Petroleum Engineering. He has authored eight scientific papers published in journals indexed by Web of Science and Scopus.

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