

Organic geochemical assessment of the origin and maturity of crude oils from the Drava Basin, Croatia

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Abstract

Six crude oils (Jagnjedovac, Lepavina, Čepelovac, Letičani, Šandrovac and Pavljani) from the western part of the Drava Basin, in the Croatian part of the Pannonian Basin, were investigated to define origin, depositional environment and thermal maturity. The oil fields are surrounded by the largest gas and gas-condensate fields (Molve, Kalinovac and Stari Gradac), located in the northwestern part of the Drava Basin. The complex structural, lithological, and stratigraphic features of this area resulted in the specific physical characteristics of the hydrocarbon fluids. The study utilized bulk parameters, gas chromatography (GC) of the gasoline fractions and gas chromatography-mass spectrometry (GC-MS) of biomarkers and phenanthrene derivatives of oils and representative samples of gas condensates from the surrounding fields. Although crude oils from the western part of the Drava Basin have different bulk properties (e.g., API gravity, sulphur content, pour point), according to the biomarker "fingerprints", all the analysed oils have a common origin and were generated from siliciclastic source rocks containing a mixture of marine and terrestrial organic matter (OM). Precursor OM was deposited in a deltaic suboxic-oxic environment. Particular differences in the distributions of *n*-alkanes and bulk parameters are attributed to the post-generative alteration (initial stage of biodegradation and evaporative fractionation). Maturity parameters derived from biomarkers and methylphenanthrenes (MP), in combination with the similarity of the gasoline fractions and the MP distributions of oils and surrounding condensates, indicated that five oils represent a mixture of low immature oil and condensate. One sample, Pavljani (geographically the most distant from the gas-condensate fields), is an oil of low maturity generated from source rocks at a vitrinite reflectance of 0.60–0.70 %.

Keywords: Geochemical characteristics, crude oil, Drava Basin, origin, maturation, biomarkers

1. INTRODUCTION

The study area is within the western part of the Drava Basin, in the Croatian part of the Pannonian Basin, covering an area of approximately 2900 km² (MALVIĆ & ĐUREKOVIĆ, 2004). The northwestern part of the Drava Basin contains large gas and gas-condensate accumulations with a series of smaller gas and oil/gas fields extending alongside the western edge of this study. The area is distinguished by sedimentary, hydrodynamic, and tectonic complexity, which accounts for the formation of various types of reservoirs and the specific characteristics of the reservoir fluids (BARIĆ et al., 1991; BARIĆ et al., 1998). This area is also characterised by an irregular pattern of existing fields (MALVIĆ, 2011), where most of the producing oil fields are located in the Bilogora Mt. zone. It is important because field data usually have the most value regarding stratigraphic, tectonic and other analyses (MALVIĆ & ĐUREKOVIĆ, 2004).

Within such a geologically heterogeneous setting, geochemical characterization is essential for understanding hydrocarbon systems. Biomarkers, particularly terpanes and steranes, enable precise crude oil fingerprinting and provide insights

into biological precursors and the depositional environment of the organic matter (PETERS & MOLDOWAN, 1993). Maturity related parameters allow further reconstruction of the thermal evolution of the oils, while gas chromatographic hydrocarbon patterns help to identify migration pathways and potential mixing between crude oils of different origins or maturity levels. These analytical tools are therefore fundamental for resolving questions related to source rocks distribution and hydrocarbon migration in structurally complex basins such as the Drava Basin. Previous studies have addressed some of these issues. BARIĆ et al. (1992) examined sedimentary and geochemical characteristics of the rocks and fluids in the western Drava Basin and identified a distinct group of oils affected by evaporative alteration. In contrast, ALAJBEG et al. (1990), analysing 29 crude oils from the Croatian part of the Pannonian Basin, suggested that all oils have a similar origin and could be classified as one broad group. Despite these contributions, BARIĆ et al. (1992, 2003) emphasized that the precise location of source rocks and the main hydrocarbon migration trends remained insufficiently resolved, largely due to variations in precursor types and maturity levels.

Confirmed source rocks in the northwestern Drava Basin include Lower Miocene to Badenian mudstones, fossil-rich carbonate mudstones, marls and siltstones occurring below 3000 m. These rocks contain predominantly terrigenous organic matter (kerogen type III) and reached high thermal maturity (condensate-gas window), generating hydrocarbons for the major gas-condensate fields of Molve, Kalinovac and Stari Gradac (BARIĆ et al., 1998).

In the Bjelovar Subbasin, organic-rich sediments occur locally (MALVIĆ, 2003a), including Upper Badenian, Sarmatian and Lower Pannonian pelitic deposits with kerogen types II–III, and low to early maturity. However, the overall quantity and quality of these rocks appear insufficient for the hydrocarbons discovered in the area, suggesting that migration from other parts of the Drava Basin played a significant role.

The aim of this study is to determine the origin of crude oils from the western Drava Basin through detailed geochemical characterization, with a specific focus on biomarker parameters used for crude oil fingerprinting and comparison with surrounding gas condensates. This approach provides new insights into the origin, depositional environment and thermal maturity of the oils, while also enhancing the understanding of source rock distribution and hydrocarbon migration pathways within this structurally complex part of the Drava Basin.

2. GEOLOGICAL SETTINGS

The Pannonian Basin is a large extensional basin in a back-arc tectonic setting that formed during the Miocene in Central Europe (TARI et al., 2023; DOLTON, 2006). It is bordered by the Alps to the west and by the Carpathian Mountains to the east and north (Fig. 1a).

The Croatian part of the Pannonian Basin System covers an area approximately 26 000 km² and is divided into the Hrvatsko Zagorje Basin and the North Croatian Basin (Fig. 1b), which are further divided into basins and subbasins traditionally termed depressions and subdepressions: the Mura, Hrvatsko Zagorje, Sava, Drava and Slavonija–Srijem Basin (SEBE et al., 2020; BARIĆ et al., 2000; MALVIĆ, 2003a, 2003b). Each of these basins also has its own local characteristics, similarities and differences in evolution, sedimentary environment, transport mechanisms and tectonics, with the boundaries between basins being mountains (BARIĆ, 2006). All basins, except for the Hrvatsko Zagorje Subbasin, host a significant number of oil and gas production fields.

The Drava Basin is located on the southwestern margin of the Pannonian Basin. Gas and gas condensate fields, Molve, Kalinovac and Stari Gradac (Fig. 1c; marks 1, 2, 3), hosting the largest accumulations of hydrocarbons in this area, are situated in the north-western part of the Drava Basin.

The study area mainly belongs to the western, uplifted part (Bilogora Zone; Fig. 1b) of the Drava Basin, with the largest number of gas and oil/gas fields. From the northwest to the southeast, the oil fields included in this study are the Jagnjedovac, Lepavina, Čepelovac and Šandrovac fields (Fig. 1c; marks a, b, c, e). The only two fields located within the subbasin are the Letičani and Pavljani fields (Fig. 1c; marks d, f), and they are also included here. The Drava Basin contains sediments of Neogene–Quaternary age (Fig. 2). The oldest Miocene sediments are alluvial and are Ottnangian in age (MANDIĆ et al., 2012), they consist of various Palaeozoic and Mesozoic rocks, commonly referred to as the Base Neogene (BNg) surface (RUKAVINA et al., 2023).

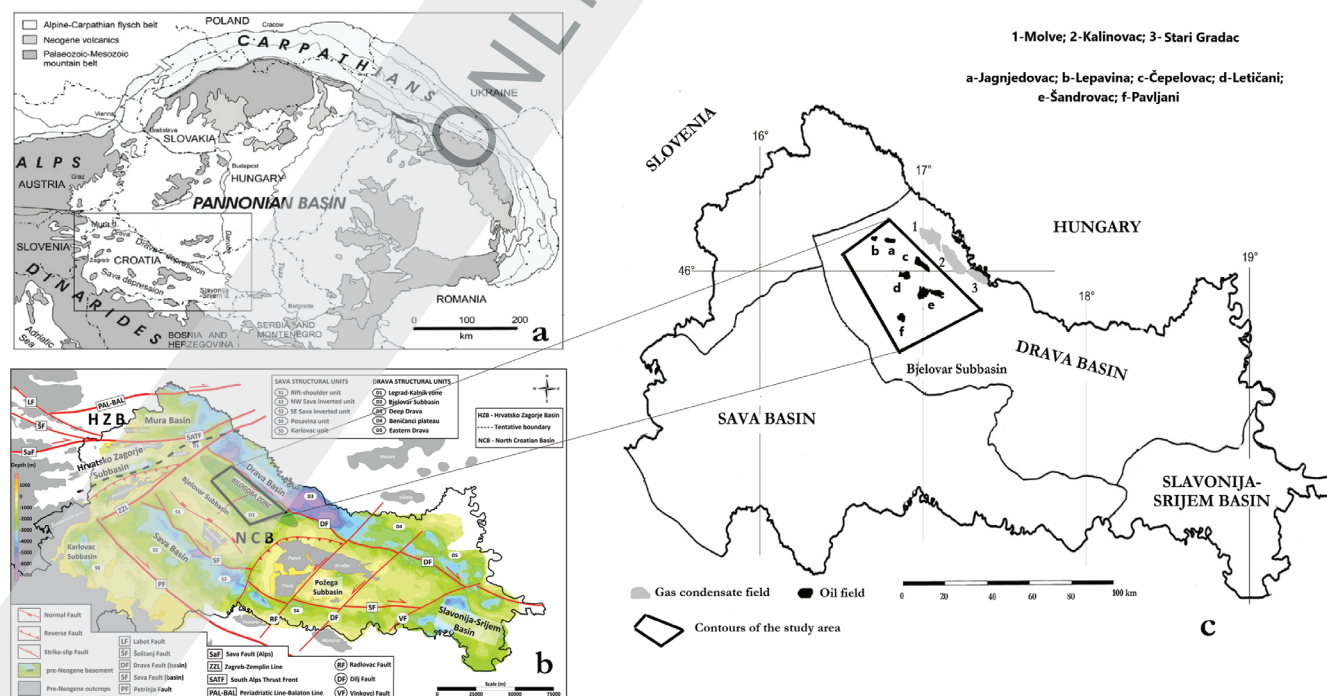


Figure 1. (a) Pannonian Basin; (b) Classification of structures and faults of the Croatian part of the Pannonian Basin (modified from TROSKOT-ČORBIĆ et al., 2026); (c) Locations of samples in the study area, Oils: a – Jagnjedovac; b – Lepavina; c – Čepelovac; d – Letičani; e – Šandrovac; f – Pavljani; Gas-condensates: 1 – Molve; 2 – Kalinovac; 3 – Stari Gradac (modified from BARIĆ et al., 2000).

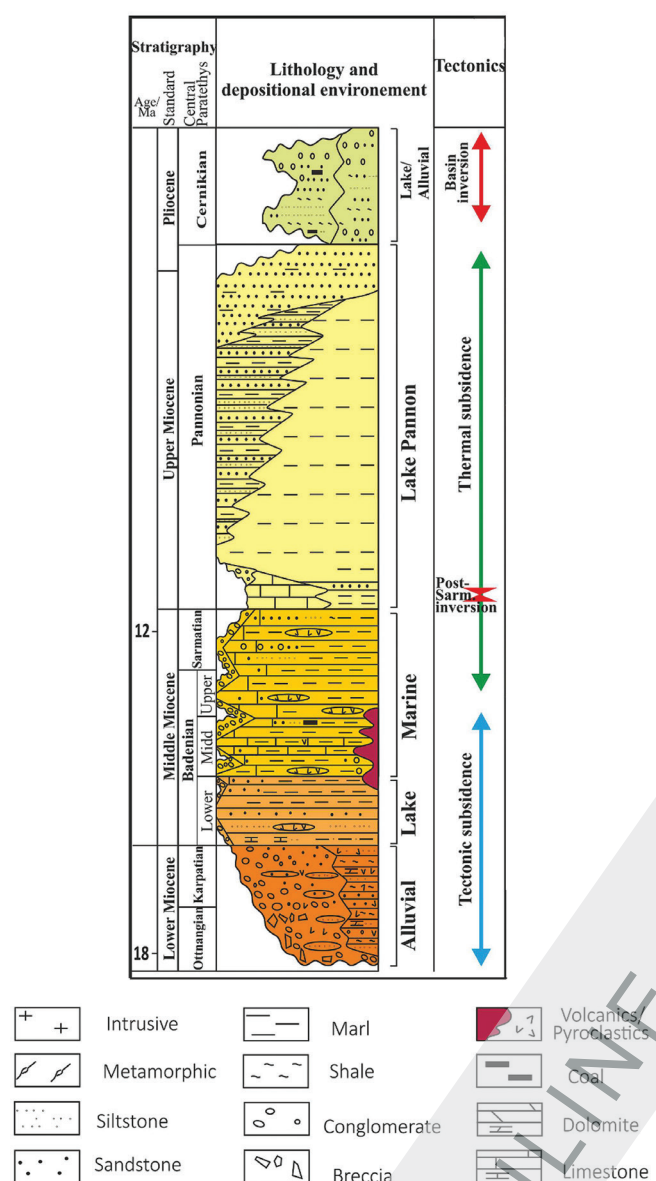


Figure 2. Stratigraphy, lithology, and depositional environment of the Neogene of the North Croatian Basin (modified from RUKAVINA et al., 2023 and PAVELIĆ & KOVAČIĆ, 2018).

Drava Basin development was driven by Neogene rifting, evolving through syn-rift (Early to Middle Miocene) and post-rift (Late Miocene to Quaternary) phases (RUKAVINA et al., 2023; PAVELIĆ & KOVAČIĆ, 2018).

In the syn-rift phase, the Early to Middle Miocene were characterized by the formation of half-grabens and a marine transgression. Initial sediments are Ottnangian–Karpatian transitional deposits. In the Middle Miocene, i.e. in the Badenian phase, the significant marine transgression occurred, leading to deposition of Badenian marine rocks, characterized by clastics (breccias, conglomerates, sandstones), marls and limestones deposited in continental to marine conditions (Fig. 2; RUKAVINA et al., 2023; PAVELIĆ & KOVAČIĆ, 2018). In the Badenian phase, marine environments developed, including protected lagoons and shallow seas, which provided favourable conditions for organic matter (OM) accumulation. The primary source rocks in the Drava Basin are Badenian

marls and calcite-rich marls (BARIĆ, 2006). Late Badenian and Sarmatian ages were characterised by continued marine environments represented by basal conglomerates above the syn-rift/post-rift boundary, algal bioclastic limestones, biocalcarenes, and marls (PAVELIĆ et al., 1998). The Pannonian period was marked by isolation from the open sea and the creation of the Pannonian Lake (Fig. 2; RUKAVINA et al., 2023; SAFTIĆ et al., 2003). Thick turbidite sequences (alternations of sandstone, silt and marl) dominate, transitioning into deltaic and alluvial systems. Upper Miocene sandstones are the most important reservoir rocks in the basin.

The post-rift Phase (Pliocene–Quaternary) was characterized by the shallowing of Lake Pannon, transition to fluvial systems (Fig. 2; RUKAVINA et al., 2023; PAVELIĆ & KOVAČIĆ, 2018) and tectonic inversion (compressive tectonics).

The study area is geologically and geographically separated from the central Drava Basin by the uplift of Mt. Bilogora during the Pliocene and Quaternary. The investigated area has characteristic uplift, which is important as a structural trap for hydrocarbons. As a result of this uplift, the thickness of Neogene–Quaternary sediments rarely exceeds 3000 m, in contrast to more than 7000 m in the central Drava Basin (MALVIĆ, 2011). Reverse faults occur predominantly along the northwestern margins of the structures and define a positive flower structure. Transcurrent dextral displacements on the Periadriatic Drava fault, as well as displacements of the Slayonian Mts. cause the formation of the domino structures on Mt. Bilogora (PRELOGOVIĆ et al., 1998). Both types of uplift are significant because these provide potential structures, as traps for oil and gas.

Reservoirs in the study area consist of Middle to Upper Miocene sandstones occurring at depths of 800 – 1200 m (BARIĆ, 2006). Their permeability ranges from 3 to 216 mD, while porosity varies between 10 % and 29 %, indicating that these sandstones represent relatively good hydrocarbon reservoirs.

Source rocks in the Drava Basin have been identified in both the northwestern and southeastern parts of the basin, as well as at several locations within the Bjelovar Subbasin (BARIĆ, 2006). Variations in source-rock type and maturity reflect the sedimentological and dynamic-thermal complexity of this area (BARIĆ et al., 1992; VELIĆ, 2007).

3. MATERIALS AND METHODS

3.1. Samples

Six crude oil samples were collected from producing fields in the western part of the Drava Basin. Composite samples were taken from storage tanks at the Jagnjedovac, Lepavina, Čepelovac, Letičani, Šandrovac and Pavljani fields. The reservoir depths of the sampled oils range from 404 to 1280 m. All crude oil samples were stored in a refrigerator in clean glass bottles with Teflon stoppers at a temperature of approximately 5 °C until analysis. All laboratory analyses were performed at the INA Exploration and Production Laboratory in Zagreb. The locations of the sampled fields are shown in Figure 1. Due to the closeness of the gas and gas-condensate fields, Molve,

Table 1. General characteristics of investigated oils (a–f) and surrounding gas-condensates (1–3).

Sample	Well depth (m)	API gravity (°)	S (%)	Bulk composition				PP (°C)	Paraffin wax (%)
				Saturates (%)	Aromatics (%)	NSO (%)	Asphaltenes (%)		
Jagnjedovac (a)	757	34.30	0.30	66.82	18.86	11.61	2.71	+9	6.2
Lepavina (b)	404	27.89	0.98	67.60	14.53	13.38	4.04	+33	12.2
Čepelovac (c)	1000	36.76	0.36	71.65	18.72	8.61	1.02	+21	6.1
Letičani (d)	1280	25.95	0.86	57.54	27.60	12.15	2.72	+30	6.4
Šandrovac (e)	910	34.28	0.29	67.68	18.00	12.80	1.52	+3	7.6
Pavljani (f)	1150	33.65	0.68	71.54	16.98	9.80	1.68	+9	6.9
Molve-25 (1)	3338	52.05	0.21	94.40	4.20	1.40	/	-24	0.05
Kalinovac-15 (2)	3442	56.73	0.25	95.20	3.30	1.50	/	-36	0.03
Stari Gradac-6 (3)	3760	52.84	0.14	95.50	2.90	1.60	/	+6	3.5

*PP – pour point, S (%) – content of sulphur.

Kalinovac and Stari Gradac to the studied oil wells, representative gas-condensates from the aforementioned fields (depths range 3338 – 3760 m) are included in this study (Fig 1c; Table 1).

3.2. Bulk composition

The bulk composition of crude oil, specifically the SARA fractions (Saturates, Aromatics, Resins, and Asphaltenes), is commonly determined using Thin-Layer Chromatography with Flame Ionization Detection (TLC-FID) on an Iatroscan MK-6s instrument. The Iatroscan technique is a modification of the thin layer chromatography with a flame ionization detector (TLC-FID). Silica gel is the thin layer on the quartz rods Chromarod S-III (quartz rod: Ø=0,9 mm, particle size 5 µm, d micropore 60 Å). Crude oil samples have been separated on the thin layer with a solvent or mixtures of solvents of different polarity, as follows: *n*-hexane for saturated hydrocarbons (saturates), toluene for aromatic hydrocarbons, (aromatics), dichloromethane/methanol for NSO compounds (polar resins), asphaltene residue remains on the spot where the sample has been applied, on the Chromarod S-III quartz rod (asphaltenes).

The TLC-FID method is preferred over conventional gravimetric SARA methods because it provides much faster analysis, requires less solvent and smaller sample amounts.

3.3. Capillary gas chromatography (GC)

GC analysis of “whole oil” was performed on an Agilent 7890A gas chromatograph fitted with a 50 m x 0.25 mm i. d. DB-petro column with a film thickness of 0.5 µm, using helium as the carrier gas. A constant flow mode and flame ionization detector were employed. The gas chromatography oven temperature was held initially at 35 °C for 10 min, then ramped to 40 °C at 1 °C/min, then increased to 320 °C at 8 °C/min, and maintained at this temperature for 60 min.

3.4. Gas chromatography-mass spectrometry (GC-MS)

Gas chromatography-mass spectrometry (GC-MS) of the saturated and aromatics fractions from crude oils was carried out using a quadrupole mass spectrometer Agilent MS 5975C interfaced to an Agilent 7890A gas chromatograph. GC 7890 A was fitted with 30 m x 0.25 mm i.d. HP-5MS column with

a film thickness of 0.25 µm with helium as the carrier gas. The GC oven temperature was ramped from 60 °C to 145 °C at 15 °C/min, then increased to 315 °C at 2 °C/min, and maintained at this temperature for 15 min. The mass spectrometer was operated in electron impact (EI) mode at 70 eV. Saturates and the aromatics fractions from crude oils were dissolved in *iso*-octane and analysed in full-scan mode (50–560 scan range). Terpanes and steranes were identified using *m/z* 191 and 217 fragments, respectively. Identification of phenanthrene and methylphenanthrenes was based on multiple ion fragmentogram *m/z* 178 and 192, respectively.

4. RESULTS AND DISCUSSION

4.1. General characteristics of the investigated oils

The well depths and bulk parameters of the investigated oils are shown in Table 1. Crude oils are classified as heavy, medium, and light on the basis of their API gravity value. API gravity is ≤20, 20–40°, and 40–45°, respectively, while condensates have an API gravity value >45° (DEMAISON, 1977). The samples analysed in this study have API values in the range 20–40° (Table 1) and are classified as medium API gravity crude oils. API gravity is a bulk physical property that can be used as a crude indicator of thermal maturity and biodegradation (JARVIE & LUNDELL, 2001). Some of the investigated crude oils have relatively high API values of 34–36° (Table 1) and accordingly, these samples are expected to be more mature or less biodegraded than the samples with lower API gravity values. However, API gravity is not a highly diagnostic parameter; although higher API gravity (lighter oil) generally indicates higher maturity, it cannot distinguish between oils of similar density generated from different sources or affected by biodegradation and evaporation. Only, API gravities below 20° usually indicate severe biodegradation, due to the removal of lighter hydrocarbons from oils, primarily *n*-alkanes and then naphthalenes, phenanthrenes and isoprenoids (HEAD et al., 2003). Gas condensates have apparently higher API gravities 52.0–56.7° consistent with high maturity.

Sulphur compounds are undesirable in crude oil because of the cost incurred for sulphur removal and environmental problems associated with these compounds. Crude oil is classified as sweet or sour depending on its sulphur content, with sweet crudes containing less than 0.5 % sulphur and sour

crudes containing more than 1 %. (AWADH, 2024; DEMAISON, 1977). The sulphur content of the analysed samples is different, some of them (Jagnjedovac, Čepelovac and Šandrovac) have a very low content of sulphur (about 0.3 %). These samples also exhibited higher API gravities, which may be attributed to their higher maturity, since maturation processes are usually associated with the defunctionalisation and cracking reactions resulting in enrichment of light hydrocarbons (PRINZLER & PAPE, 1964). On the other hand, oil samples Pavljani, Letičani and Lepavina, have higher sulphur contents of 0.68 %, 0.86 % and 0.98 %, respectively (Table 1). The elevated sulphur content associated with lower API, particularly in Letičani and Lepavina, may imply lower maturity or possible post-generative alteration. Finally, higher sulphur contents have been reported in oils from the marine/evaporitic reducing environments (PRINZLER & PAPE, 1964). Therefore, the observed relationship between sulphur and API gravity values are not sufficient to make any firm conclusion and should be supported by other analysis. Gas condensates have low sulphur contents ≤ 0.25 % (Table 1) in accordance with a significant contribution of terrestrial OM and high maturity.

Pour point (PP) is generally associated with the waxy nature of crude oils (CHINENYEZE & EKENE, 2017). High paraffin abundance is usually reflected by high concentration of C_{20+} *n*-alkanes in crude oil. Crude oil derived from terrigenous OM generally has high paraffinic content and high PP (SHAGUFTA & TAHIRA, 2013). All analysed samples have a high content of paraffin wax (>6 %), and accordingly a high PP. Some samples including Lepavina and Letičani, have a pour point exceeding values of $+30$ °C (Table 1), which may be indicative for the significant input of land plant OM or resulted from the post-generative alteration, as suggested by a lower API and higher sulphur content. In contrast to crude oils, gas-condensates have a very low PP (-36 – $+6$ °C, Table 1), as expected at high maturity level.

The bulk compositions (SARA) of the investigated oils from the western part of Drava Basin are given in Table 1. The bulk compositions are very similar, with saturates and aromatics exceeding 82 %, which suggests that the oils were probably not exposed to severe biodegradation. However, the apparently lower content of a saturated fraction in the Letičani oil (57.54 %) and the highest content of asphaltenes in the Lepavina oil (4.04 %; Table 1) are an additional indication that these two crude oils probably have undergone a post-generative process, which is reflected through all the bulk properties discussed. Gas condensates consist almost of hydrocarbons (≥ 98.4 %) with the predominance of saturated hydrocarbons (≥ 94.4 %; Table 1).

4.2. Origin and depositional environment of organic matter

The prevalence of *n*-alkanes in the saturated fractions of all the studied samples (Fig. 3) indicates that oils were not exposed to biodegradation or experienced the maximum biodegradation level of 1–2 (HEAD et al., 2003). This result implies that distributions of acyclic isoprenoids ($>C_{15}$), phenanthrenes, steranes and terpanes are not altered by microbial activity and

can be used for interpretation of sources, depositional environment and maturity. The mass fragmentograms (m/z 85) of saturated fractions (Fig. 3) are represented by n - C_{14} – n - C_{35} alkanes, exhibiting high contents of long-chain homologues ($>n$ - C_{25}). This indicates a high contribution of terrestrial OM (BOURBONNIERE & MEYERS, 1996), consistent with the high content of paraffin waxes and pour points (Table 1). Furthermore, high contents of long-chain *n*-alkanes are an indicator of low maturity, since at the higher maturity level cracking occurs, resulting in the prevalence of homologues with the shorter chain. The *n*-alkane patterns are very similar in samples a – Jagnjedovac, b – Lepavina and d – Letičani showing unimodal distributions with maxima in range n - C_{26} – n - C_{28} and apparent loss of short-chain homologues. This implies evaporation or a low level of biodegradation, because light hydrocarbons are more prone to microbial degradation (HEAD et al., 2003). Furthermore, the lower input of algal OM and/or lower maturity should be also considered. Samples c – Čepelovac and f – Pavljani have bimodal *n*-alkane distributions with maxima at n - C_{17} and n - C_{29} – n - C_{30} , which is typical for mixed aquatic/terrestrial OM. The results suggest lower evaporation/biodegradation and/or a higher contribution of aquatic precursor material/higher maturity of samples c and f compared to a, b, d. However, the prevalence of long-chain *n*-alkanes, particularly in c – Čepelovac oil is evident (Fig. 3c). This oil also exhibits a certain similarity with the previously discussed oils (a, b, d) that is reflected through a reduced abundance of short-chain homologues. Finally, sample e – Šandrovac shows an obviously different *n*-alkanes pattern, with maxima at mid-chain homologues n - C_{23} – n - C_{25} (Fig. 3). The precursors of these *n*-alkanes are aquatic macrophytes, microalga and vascular plants (FICKEN et al., 2000). This result suggests a higher contribution of aquatic precursor OM to the Šandrovac oil. However, the evident cut-off in the distribution of short-chain *n*-alkanes (C_{15} – C_{20}) in the Šandrovac oil (Fig. 3e) also suggests evaporation or a low level of biodegradation. Therefore, distributions of *n*-alkanes indicate that samples a – Jagnjedovac, b – Lepavina, d – Letičani and e – Šandrovac were exposed to certain post-depositional alteration, in contrast to c – Čepelovac and f – Pavljani. This has already been postulated for the b – Lepavina and d – Letičani oils based on bulk parameters. Despite the observed differences in *n*-alkane distributions, the CPI values (BRAY & EVANS, 1961) are quite uniform 1.01–1.06 (Table 2).

The Pr/n - C_{17} versus Ph/n - C_{18} plot proposed by SHANMUGAM (1985) provides useful information regarding kerogen source and depositional conditions of the OM. All the analysed oils are plotted in the region of a type II–III kerogen mixture (a mixture of marine and terrestrial OM), deposited under suboxic/oxic conditions (Fig. 4). However, positions of the Čepelovac and Pavljani oils in this plot suggests deposition of OM in a more oxidizing environment and/or with a greater contribution from terrestrial sources in comparison to the other studied samples.

In the gas-condensates, *n*-alkane distributions are dominated by low-chain homologues (C_{15} – C_{18}), resulting from high maturity, which is associated with intense cracking of long-chain *n*-alkane homologues (Fig. S1). However, the

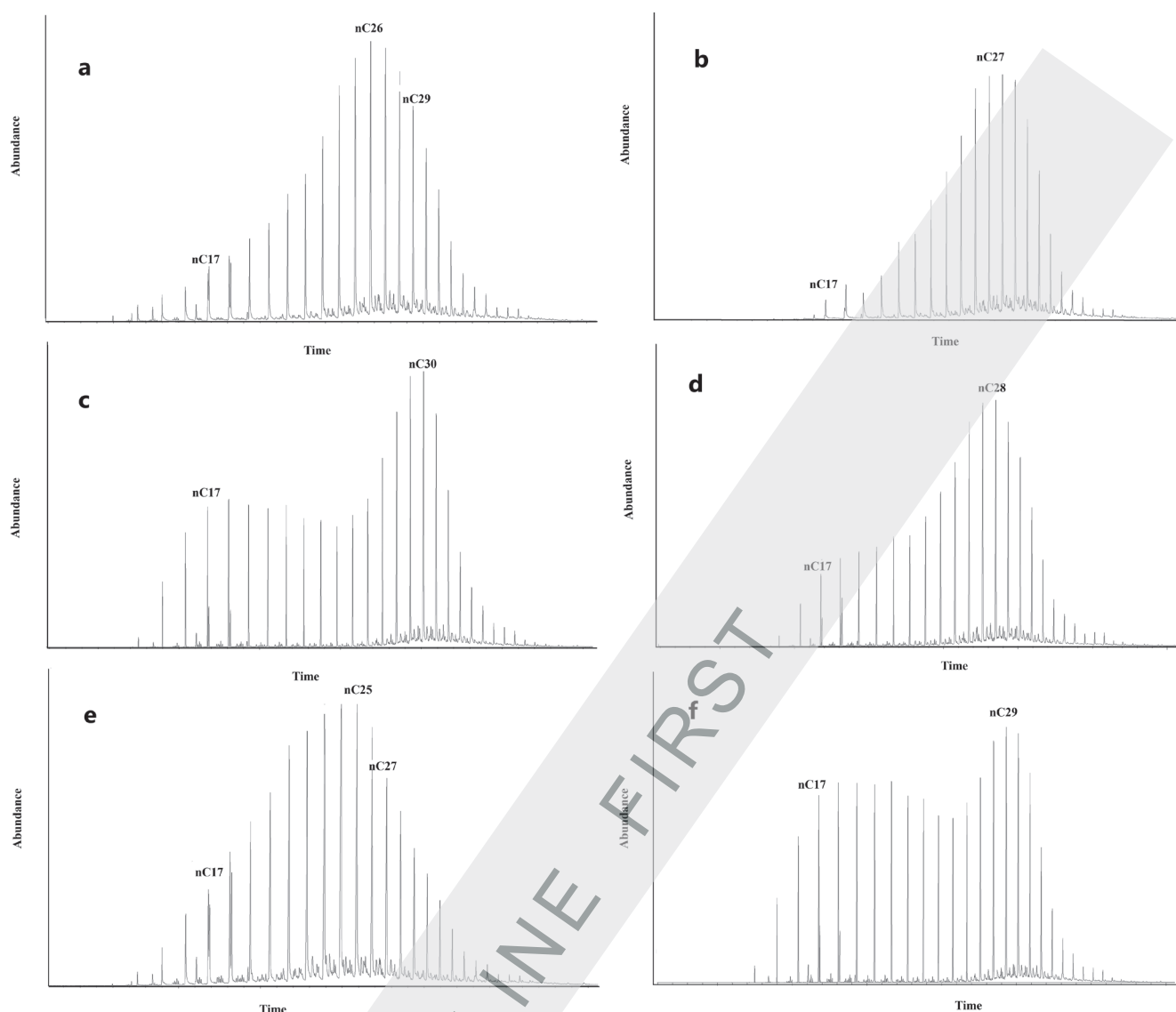


Figure 3. Mass fragmentograms of *n*-alkanes ($m/z = 85$) of investigated oils: **a** – Jagnjedovac; **b** – Lepavina; **c** – Čepelovac; **d** – Letičani; **e** – Šandrovac; **f** – Pavljani.

presence of considerable amounts of long chain *n*-alkanes along with CPI values of about 1.2 (Table 2) in the Molve and Kalinovac samples denotes a significant contribution from terrestrial OM.

In the $Pr/n-C_{17}$ versus $Ph/n-C_{18}$ diagram (Fig. 4), gas-condensates also plotted in the region of a type II–III kerogen mixture deposited under suboxic/oxic conditions. However, due to high maturity, the $Pr/n-C_{17}$ and $Ph/n-C_{18}$ ratios are low

Table 2. Gas chromatography properties of investigated crude oils (a–f) and surrounding gas-condensates (1–3) (API is given for comparison).

Sample	Well depth (m)	API gravity (°)	Pr/nC_{17}	Ph/nC_{18}	Pr/Ph	CPI	Benzene/ nC_6	Toluene/ nC_7
Jagnjedovac (a)	757	34.30	0.66	0.60	1.19	1.06	0.44	1.22
Lepavina (b)	404	27.89	0.71	0.68	0.91	1.01	0.64*	0.13*
Čepelovac (c)	900	36.76	0.56	0.45	1.38	1.03	0.32	0.34
Letičani (d)	1280	25.95	0.77	0.79	1.02	1.03	0.41*	0.61*
Šandrovac (e)	910	34.28	0.67	0.67	1.07	1.02	0.48	0.88
Pavljani (f)	1150	33.65	0.56	0.40	1.40	1.05	0.84	0.43
Molve-25 (1)	3338	52.05	0.26	0.20	1.70	1.21	1.42	1.83
Kalinovac-15 (2)	3442	56.73	0.26	0.21	1.91	1.20	0.54	0.85
Stari Gradac-6 (3)	3760	52.84	0.25	0.19	1.48	1.02	0.41	0.70

*Pr – pristane; Ph – phytane; CPI – Carbon Preference Index (BRAY & EVANS, 1961)

**CPI = $\frac{1}{2}[(C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{26}+C_{28}+C_{30}+C_{32}+C_{34}) + (C_{25}+C_{27}+C_{29}+C_{31}+C_{33})/(C_{24}+C_{26}+C_{28}+C_{30}+C_{32})]$

***Note: Values of Benzene/ nC_6 and Toluene/ nC_7 for Lepavina and Letičani oils should be considered with caution due to the very low content of gasoline fraction

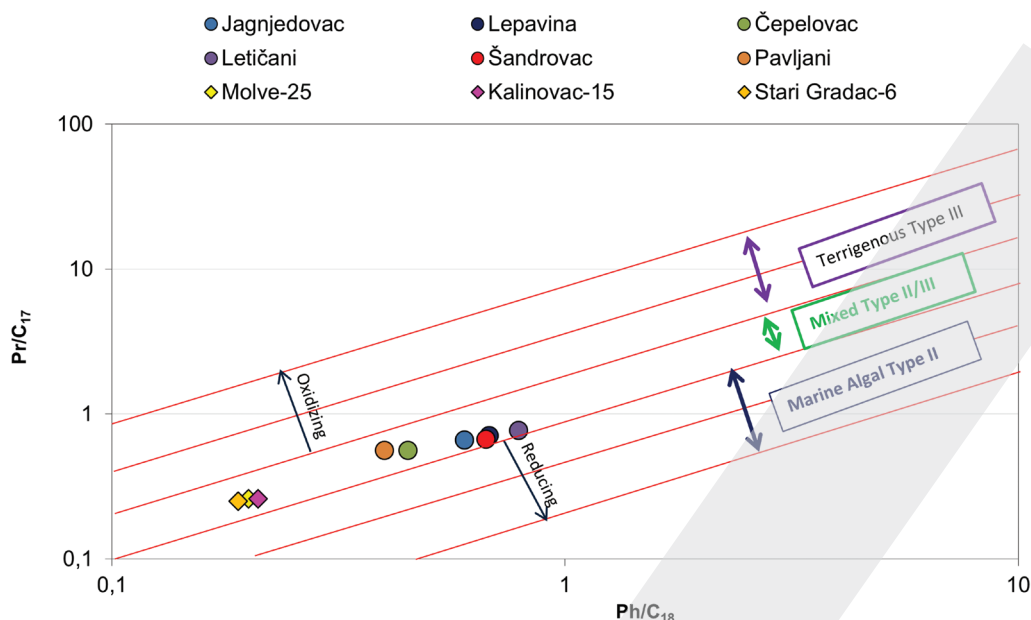


Figure 4. Pr/nC_{17} versus Ph/nC_{18} plot of investigated oils and gas-condensates (SHANMUGAM, 1985). Pr – pristine, Ph – phytane.

(≤ 0.26 and ≤ 0.21 , respectively; Table 2), signifying rather higher maturity than usual for the source/depositional conditions, resulting in the underestimation of terrestrial input and oxidation state of the depositional environment. High Pr/Ph ratios (≥ 1.48 ; Table 2) in gas-condensate samples are mostly caused by high maturity, since this parameter gradually increases with maturation (PETERS et al., 2005), and also indicates oxic conditions and a significant impact of terrestrial OM.

The origin of the investigated crude oils was further assessed using the abundances and distributions of sterane and terpane biomarkers. These biomarkers are absent in gas-condensates due to the high maturity.

Despite the differences in the general characteristics of the investigated oils (Table 1) and the distributions of *n*-alkanes (Fig. 3), biomarker fingerprints indicate similarities between the crude oils (Figs. 5, 6; Table 3).

All the studied oils showed similar distributions of C_{27} – C_{29} $\alpha\alpha\alpha$ R steranes (Fig. 5; Table 4). The use of the distribution of regular steranes in the evaluation of the sources and depositional environment of OM is based on evidence that C_{27} sterane originates dominantly from marine plankton, C_{28} sterane from yeast, fungi, plankton and algae, while C_{29} sterane originates from higher plants, and from brown and green algae (VOLKMAN, 2003).

The relative abundances of C_{27} – C_{29} steranes (Table 4) imply an origin of the oils from source rocks of mixed kerogen types (terrestrial and marine). The pattern of C_{27} – C_{28} – C_{29} regular steranes for all samples (Fig. 5) has a characteristic "V" shape, i.e. the highest percentages are for the C_{27} and C_{29} steranes, and a slightly smaller proportion of the C_{28} steranes is indicative for the marine, algal OM with a pronounced influence of terrestrial components (VOLKMAN, 2003). In all the investigated oil samples (except Čepelovac crude oil), steranes prevail over hopanes (ST/HOP ratio >1 ; Table 4), indicating a greater contribution of algae and terrestrial plants than bacteria in the precursor OM (MOLDOWAN et al., 1985;

TISSOT & WELTE, 1984). The result coincides with the distributions of *n*-alkanes and regular steranes, as well as data from the Pr/nC_{17} vs. Ph/nC_{18} plot (Fig. 4).

Table 3. Peak identification of terpanes and steranes in *m/z* 191 and 217 mass chromatograms of the investigated oils.

Peak	Compound
$C_{23}TT$	C_{23} tricyclic terpane
$C_{24}TT$	C_{24} tricyclic terpane
Ts	18 α (H)-22,29,30-trisnorhopane
Tm	17 α (H)-22,29,30-trisnorhopane
$C_{29}\beta\alpha$	C_{29} 17 β (H)-21 α (H)-moretane
$C_{29}\alpha\beta$	17 α (H)-21 β (H)-30-norhopane
O	18 α (H)-oleanane
$C_{30}\alpha\beta$	17 α (H)-21 β (H)-hopane
M	C_{30} 17 β (H)-21 α (H)-moretane
31S, 31R	17 α (H)-21 β (H)-30-homohopane (22S, 22R)
32S, 32R	17 α (H)-21 β (H)-30-bishomohopane (22S, 22R)
33S, 33R	17 α (H)-21 β (H)-30-trishomohopane (22S, 22R)
34S, 34R	17 α (H)-21 β (H)-30-tetrakishomohopane (22S, 22R)
35S, 35R	17 α (H)-21 β (H)-30-pentakishomohopane (22S, 22R)
27 $\beta\alpha$ d S	13 β (H)-17 α (H)-diacholestane (20S)
27 $\beta\alpha$ d R	13 β (H)-17 α (H)-diacholestane (20R)
27 $\alpha\alpha$ S	5 α (H), 14 α (H), 17 α (H)-cholestane (20S)
27 $\beta\beta$ R	5 α (H), 14 β (H), 17 β (H)-cholestane (20R)
27 $\beta\beta$ S	5 α (H), 14 β (H), 17 β (H)-cholestane (20S)
27 $\alpha\alpha$ R	5 α (H), 14 α (H), 17 α (H)-cholestane (20R)
28 $\alpha\alpha$ S	5 α (H), 14 α (H), 17 α (H), 24-methyl-cholestane (20S)
28 $\beta\beta$ R	5 α (H), 14 β (H), 17 β (H), 24-methyl-cholestane (20R)
28 $\beta\beta$ S	5 α (H), 14 β (H), 17 β (H), 24-methyl-cholestane (20S)
28 $\alpha\alpha$ R	5 α (H), 14 α (H), 17 α (H), 24-methyl-cholestane (20R)
29 $\alpha\alpha$ S	5 α (H), 14 α (H), 17 α (H), 24-ethyl-cholestane (20S)
29 $\beta\beta$ R	5 α (H), 14 β (H), 17 β (H), 24-ethyl-cholestane (20R)
29 $\beta\beta$ S	5 α (H), 14 β (H), 17 β (H), 24-ethyl-cholestane (20S)
29 $\alpha\alpha$ R	5 α (H), 14 α (H), 17 α (H), 24-ethyl-cholestane (20R)

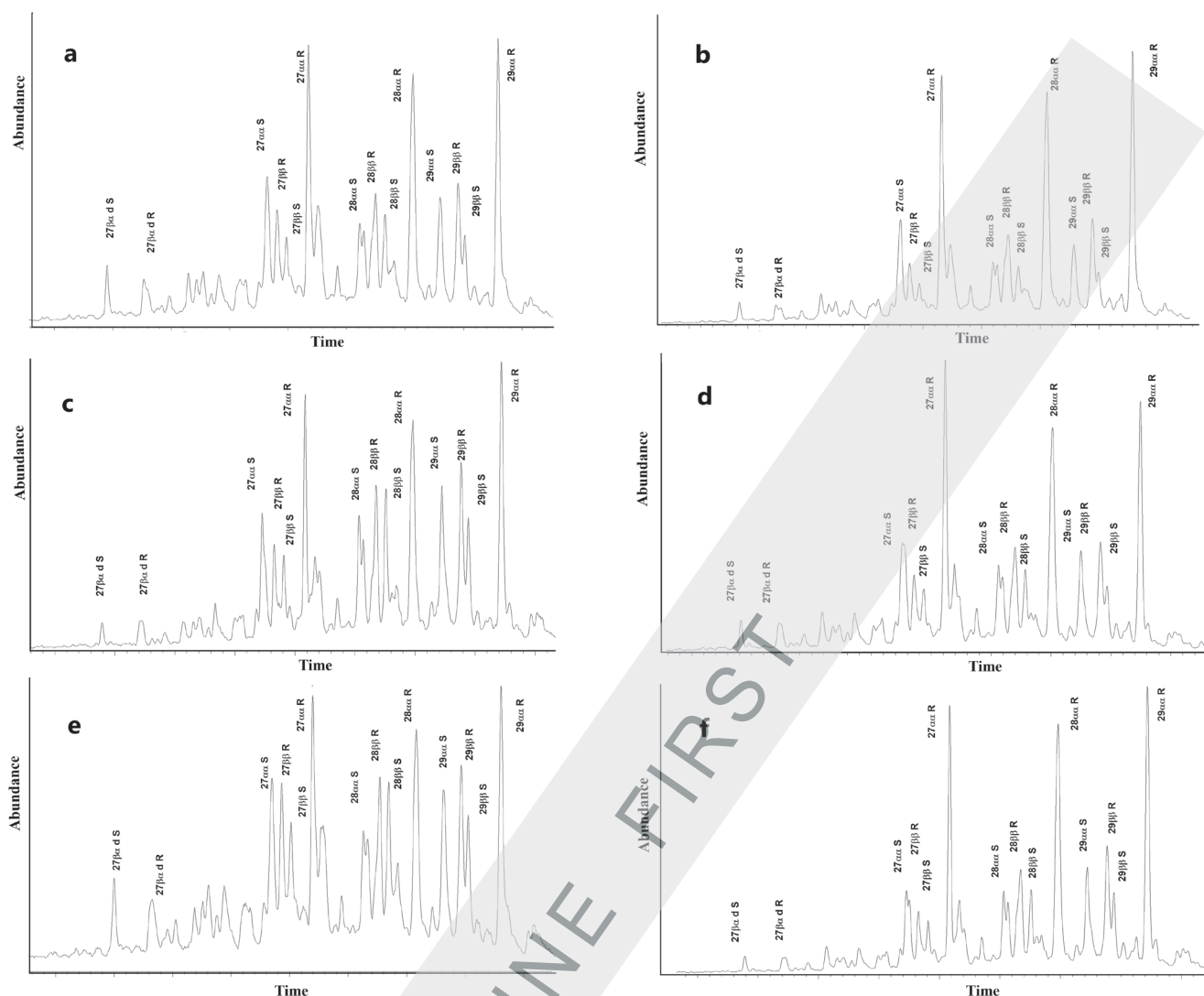


Figure 5. Mass chromatograms of steranes ($m/z = 217$) of the investigated oils: **a** – Jagnjedovac; **b** – Lepavina; **c** – Čepelovac; **d** – Letičani; **e** – Šandrovac; **f** – Pavljani. Identification is given in Table 3.

Terpane distributions in oils are characterized by a very low proportion of tricyclic terpanes and the dominance of C_{30} $\alpha\beta$ hopane (Fig. 6). Very low abundance of tricyclic terpanes is indicative of low maturity and a significant contribution of terrestrial OM. The Šandrovac sample (Fig. 6e) has a slightly higher content of tricyclic terpanes, which coincides with the

distribution of n -alkanes (Fig. 3e) and argues for a slightly larger contribution of algal OM.

In all the investigated oils, oleanane is present as an indicator of an angiosperm terrigenous influence (PHILP & GILBERT, 1986), as well as an indicator that the OM of the source rocks is younger than the Early Cretaceous (MOLDOWAN et al.,

Table 4. Biomarker parameters of investigated oils based on sterane (m/z 217) distributions.

Sample	$\beta\beta$	20S/(20S+20R)	S27 (%)	S28 (%)	S29 (%)	Dia/Reg	ST/HOP
Jagnjedovac (a)	0.34	0.28	33.86	30.17	35.97	0.11	1.49
Lepavina (b)	0.30	0.21	33.16	30.72	36.12	0.05	2.44
Čepelovac (c)	0.41	0.35	33.15	29.05	37.80	0.07	0.66
Letičani (d)	0.32	0.27	38.47	28.85	32.67	0.07	2.27
Šandrovac (e)	0.41	0.36	33.86	29.17	36.98	0.17	1.23
Pavljani (f)	0.33	0.26	33.65	30.95	35.40	0.04	1.08
E.V.	0.67–0.71	0.52–0.55	/	/	/	/	/

* $\beta\beta = C_{29}\beta\beta$ (S+R) steranes/ ΣC_{29} ($\alpha\alpha + \beta\beta$) steranes (including S and R isomers); 20S/(20S+20R) = $C_{29\alpha\alpha}$ (S) sterane/ $C_{29\alpha\alpha}$ (S+R) sterane; S27 (%) = $100 * C_{27\alpha\alpha}$ (R) sterane / $\Sigma C_{27} - C_{29\alpha\alpha}$ (R) steranes; S28 (%) = $100 * C_{28\alpha\alpha}$ (R) sterane / $\Sigma C_{27} - C_{29\alpha\alpha}$ (R) steranes; S29 (%) = $100 * C_{29\alpha\alpha}$ (R) sterane / $\Sigma C_{27} - C_{29\alpha\alpha}$ (R) steranes; Dia/Reg = $C_{27\beta\alpha}$ (S) diasterane/ $C_{27\alpha\alpha}$ (R) sterane; ST/HOP = Σ steranes/ Σ hopanes; E.V. – Equilibrium value

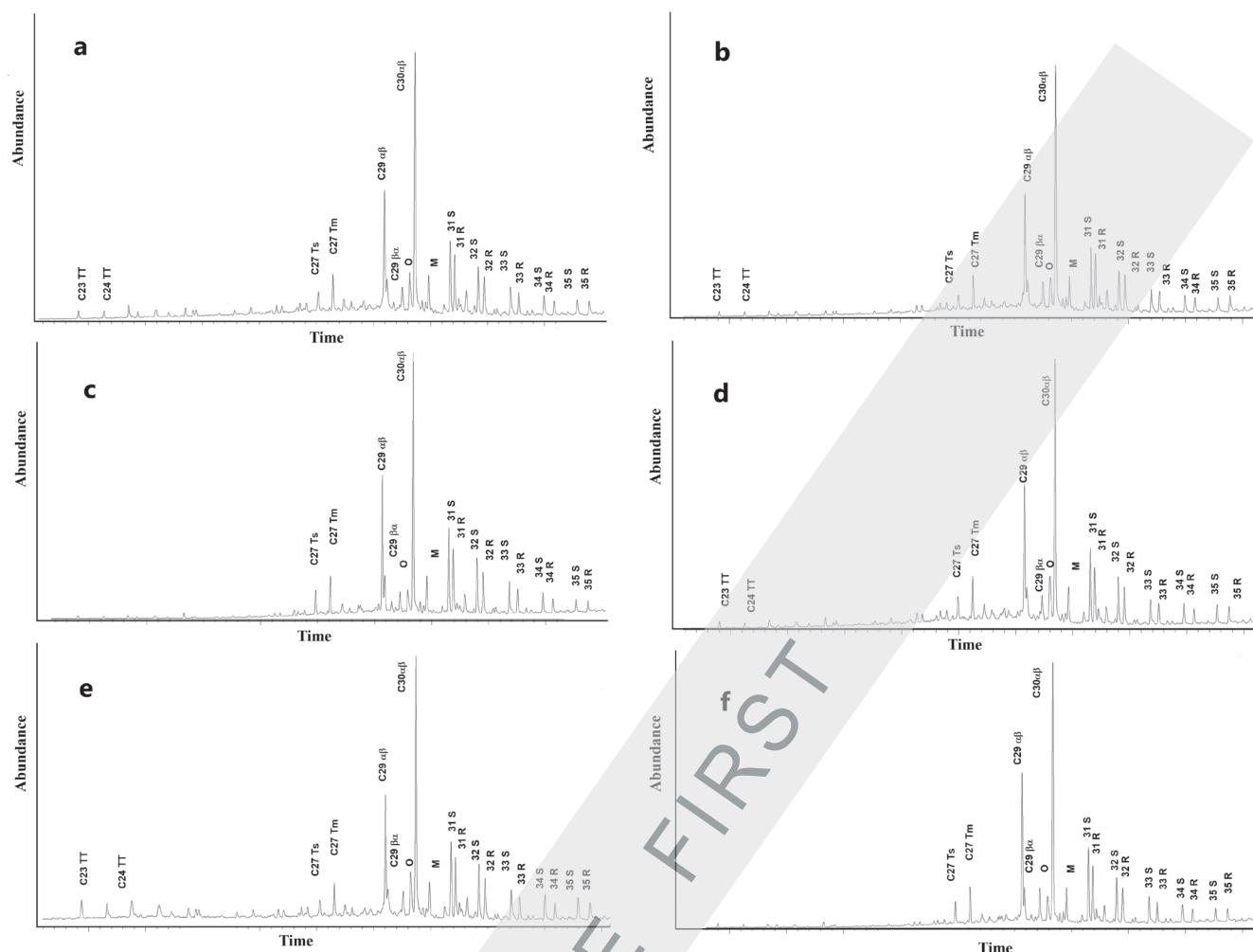


Figure 6. Mass chromatograms of terpanes ($m/z = 191$) of the investigated oils: **a** – Jagnjedovac; **b** – Lepavina; **c** – Čepelovac; **d** – Letičani; **e** – Šandrovac; **f** – Pavljani. Identification is given in Table 3.

1994). The relatively high oleanane/ C_{30} $\alpha\beta$ hopane ratios ≈ 0.20 (O/H; Table 5) are indicative of deltaic environments (MURRAY et al., 1997). Earlier investigations revealed that the diagenetic contact of seawater with higher-plant organic matter in shales dramatically enhances hydrogenation of amyrine precursors and the preservation of oleanane (MURRAY et al., 1997; PETERS et al., 2000). A mixed marine/terrestrial precursor OM of the oils, deduced from the distributions of n -alkanes (Fig. 3), steranes (Fig. 5), steranes/hopanes ratio (Table 4) and Pr/ n - C_{17} vs. Ph/ n - C_{18} plot (Fig. 4)

fits well with the deltaic environment. The C_{31} 22 (R) hopane/ C_{30} hopane ratio < 0.25 may be evidence of marine depositional environments (Table 5).

Despite numerous limitations (e.g. maturity or source material dependence; ALBRECHT et al., 1976; GOOSSENS et al., 1984), the pristane/phytane (Pr/Ph) ratio is used as a redox parameter (DIDYK et al., 1978). The Pr/Ph ratios for the studied oils range from 0.9 to 1.4 (Table 2), which is characteristic of a suboxic/oxic environment (DIDYK et al., 1978). However, evidently higher values of this parameter

Table 5. Biomarker parameters of investigated oils based on terpane (m/z 191) distributions.

Sample	O/H	G/H	C_{35}/C_{34} $\alpha\beta$ hopane	nH/H	M/H	Ts/(Ts+Tm)	22S/(22S+22R)	(C_{24}/C_{23}) TT	$C_{31}R/H$
Jagnjedovac (a)	0.20	0.05	0.89	0.48	0.12	0.41	0.53	0.95	0.21
Lepavina (b)	0.19	0.05	1.10	0.44	0.14	0.37	0.51	0.88	0.22
Čepelovac (c)	0.13	0.03	0.62	0.49	0.15	0.42	0.57	N.C.	0.23
Letičani (d)	0.24	0.04	1.06	0.51	0.13	0.40	0.57	N.C.	0.19
Šandrovac (e)	0.24	0.06	0.90	0.46	0.13	0.37	0.54	0.84	0.25
Pavljani (f)	0.14	0.04	0.91	0.54	0.14	0.37	0.57	N.C.	0.20
E.V.	/	/	/	/	0.15	/	0.57–0.62	/	/

*O/H = Oleanane/ C_{30} $\alpha\beta$ hopane; G/H = Gammacerane/ C_{30} $\alpha\beta$ hopane; nH/H = C_{29} $\alpha\beta$ hopane/ C_{30} $\alpha\beta$ hopane; M/H = C_{30} $\beta\alpha$ moretane/ C_{30} $\alpha\beta$ hopane; 22S/(22S+22R) = C_{31} $\alpha\beta$ (S) hopane/ C_{31} $\alpha\beta$ (S+R) hopane; $C_{31}R/H$ = C_{31} $\alpha\beta$ (R) hopane/ C_{30} $\alpha\beta$ hopane; N.C. – not calculated due to the absence of tricyclic terpanes; E.V. – Equilibrium value; for other abbreviations see Table 3

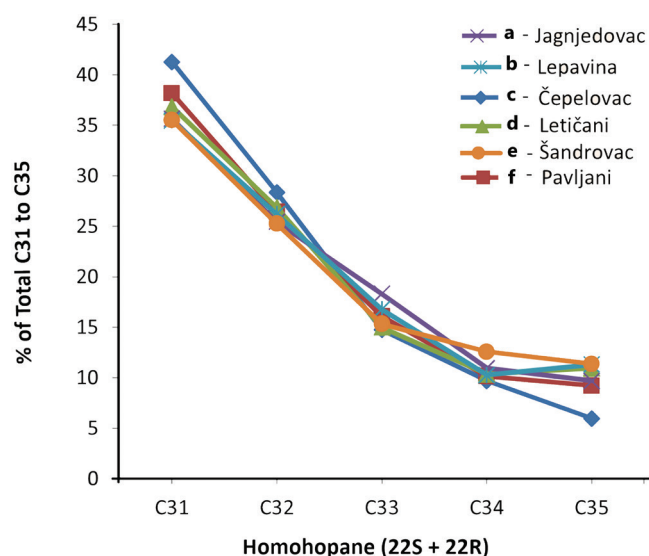


Figure 7. Distributions of C_{31} – C_{35} $\alpha\beta$ homohopanes of the investigated oils: a – Jagnjedovac; b – Lepavina; c – Čepelovac; d – Letičani; e – Šandrovac; f – Pavljani.

(≈ 1.4) in c – Čepelovac and f – Pavljani oils suggest deposition in a more oxidizing environment, consistent with the position of these samples in the plot shown in Fig. 4.

The relative distribution of C_{31} – C_{35} homohopanes (extended hopanes) in crude oils is also used as an indicator of redox potential during deposition of the source rocks (PETERS et al., 1995; PETERS et al., 2005). The distributions of C_{31} – C_{35} $\alpha\beta$ homohopanes of the investigated oils (Fig. 7) are uniform and indicative for suboxic/oxic environments. This coincides with low values of the gammacerane/ C_{30} $\alpha\beta$ hopane ratios

≤ 0.06 (G/H; Table 5), denoting a rather shallow water column, without significant stratification (SINNINGHE DAMSTÉ et al., 1995).

The most rapid loss of content of homohopanes with increasing number of carbon atoms in the Čepelovac sample (Fig. 7) suggests oxic conditions during OM deposition. This coincides with the previously discussed steranes/hopanes ratio < 1 for this oil (Table 4), because activity of aerobic bacteria, which represent the main sources of hopanes (OURISSON et al., 1979), is favoured under oxic conditions. However, the flatter profile of the contents of C_{33} – C_{35} homohopanes in the Šandrovac oil (Fig. 7) fits with a higher contribution of algal OM (less oxic settings) as deduced from the distribution of n -alkanes (Fig. 3) and slightly higher abundance of tricyclic terpanes (Fig. 6). Furthermore, the C_{35}/C_{34} hopane ratios slightly above 1 in samples Lepavina and Letičani (Fig. 7; Table 5) are in accordance with the lowest Pr/Ph ratios (Table 2) and the highest steranes/hopanes ratio (Table 4) in the studied sample set, suggesting suboxic depositional conditions.

Values of the C_{29} norhopane/ C_{30} hopane ratio < 0.60 (nH/H; Table 5) associated with the aforementioned distributions of homohopanes (Fig. 7) are indicative for marine shales as source rocks (FAN et al., 1987). Furthermore, the relatively high values of the Ts/(Ts+Tm) ratio (0.37–0.42; Table 5) and the presence of diasteranes, associated with the low values of typical biomarker maturity parameters (Tables 4, 5; see Section 4.4), argues for an abundance of clay minerals in the source rocks.

However, biomarker similarities between the studied crude oils are the primary indicators of a common source facies (depositional environment and OM input); such similarities do

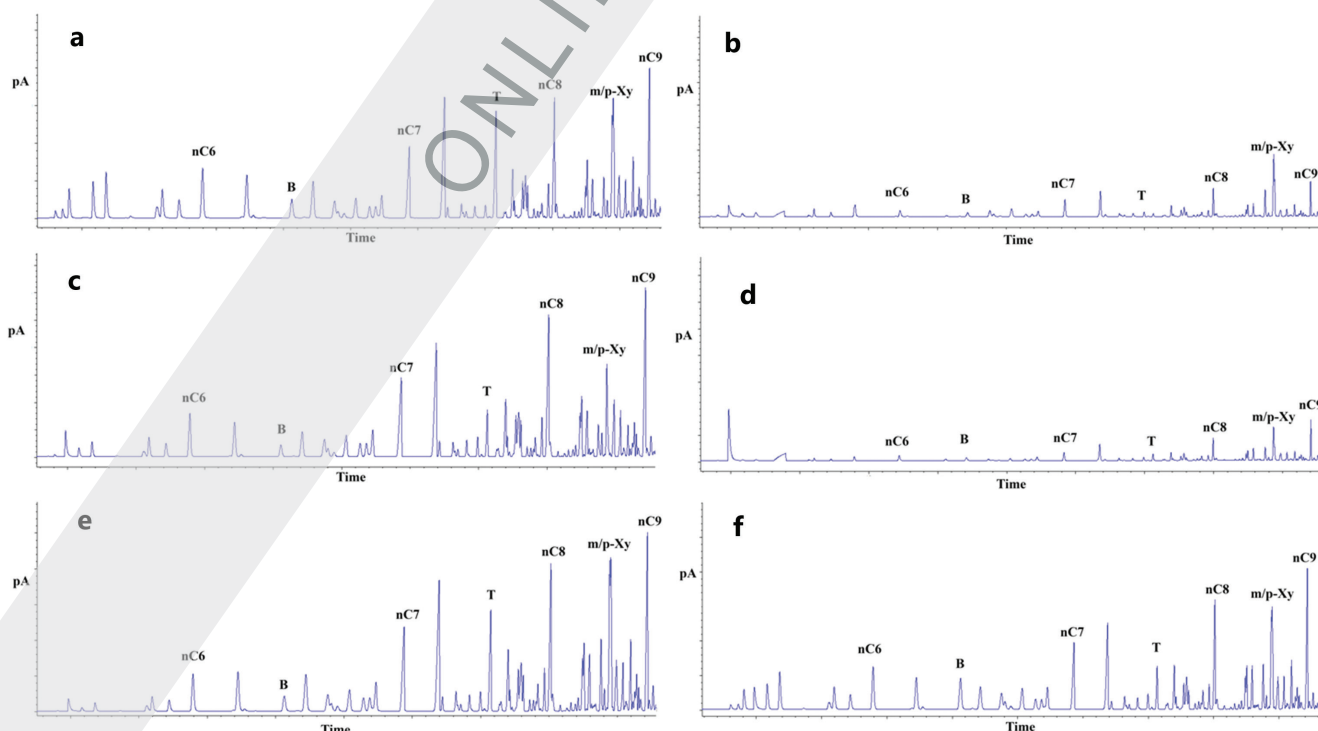


Figure 8. Gasoline fractions of the investigated oils: a – Jagnjedovac; b – Lepavina; c – Čepelovac; d – Letičani; e – Šandrovac; f – Pavljani. nC6 – n -hexane; B – benzene; nC7 – n -heptane; T – toluene; nC8 – n -octane; m/p-Xy – meta/para-xylene; nC9 – n -nonane.

not necessarily imply identical maturity or migration history, because specific biomarker parameters can be influenced differently by thermal evolution, migration, or post-generative alterations.

4.3. Gasoline fractions and post-generative alteration

Parts of gas chromatograms of whole oils representing the distributions of hydrocarbons in the gasoline fraction are shown in Figure 8. All oils contain light aromatics (benzene – B, toluene – T, and xylene – Xy; THOMPSON, 1983)

consistent with the contribution of terrigenous OM. The toluene/*n*-heptane ratio (THOMPSON, 1983) is <1 in all samples (confirming marine algal input), except for the Jagnjedovac oil in which it exceeds 1 (1.22; Table 2). It was proposed that “normal oils” which were not exposed to post-generative alterations have a toluene/*n*-heptane ratio in the range of 0.2–0.6 (THOMPSON, 1983).

Because the sterane and hopane distributions are generally similar in all oils (Figs. 5, 6), the increased toluene/*n*-heptane ratio for the Šandrovac oil and particularly the Jagnjedovac oil

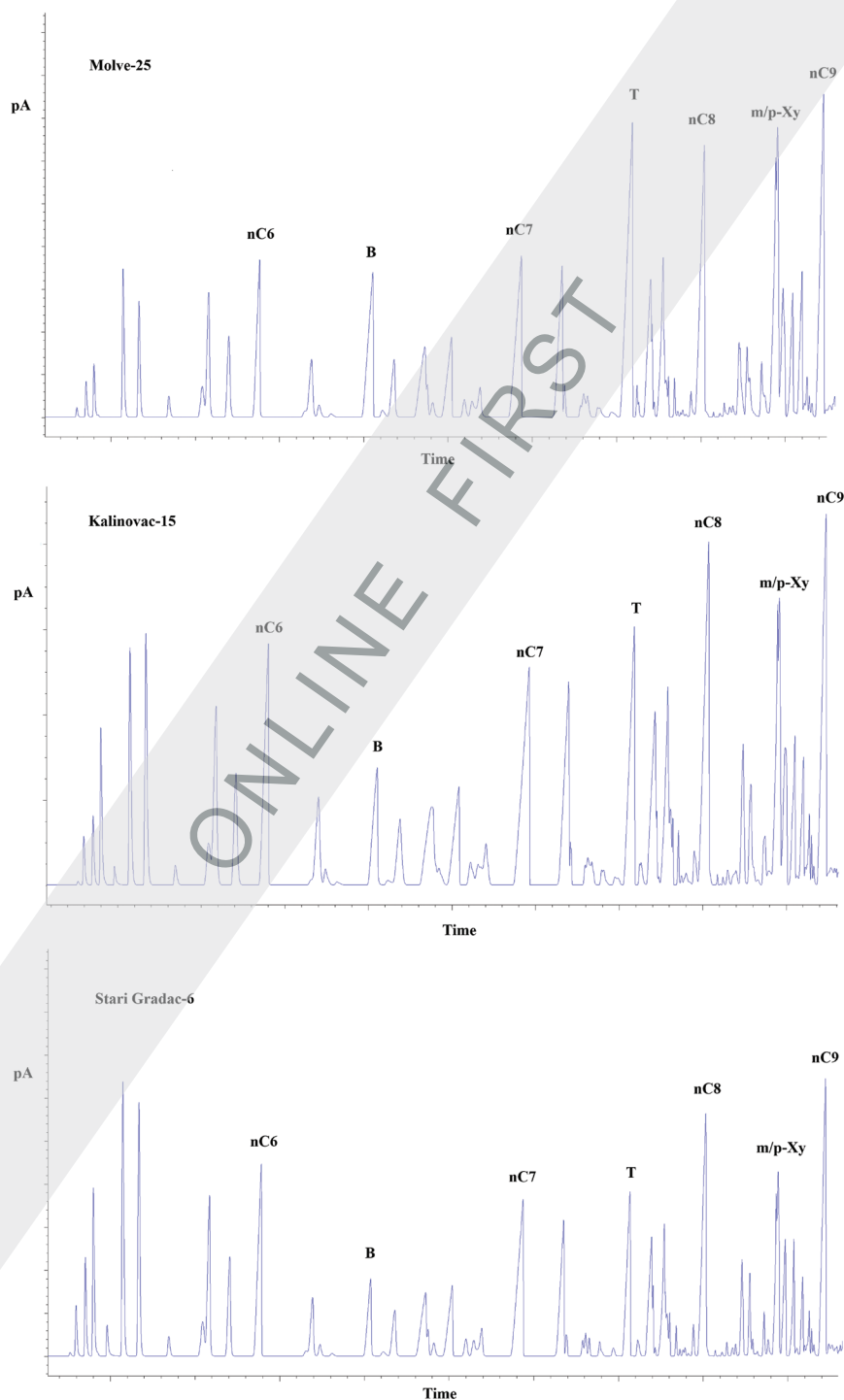


Figure 9. Gasoline fractions of gas-condensates Molve-25, Kalinovac-15 and Stari Gradac-6; nC6 – *n*-hexane; B – benzene; nC7 – *n*-heptane; T – toluene; nC8 – *n*-octane; m/p-Xy – *meta/para*-xylene; nC9 – *n*-nonane.

(Table 2), indicates an evaporative fractionation process, rather than differences in the precursor material (THOMPSON, 1987). However, the Lepavina and Letičani crude oils differ from other samples primarily due to the lack of hydrocarbons in the gasoline fraction (Fig. 8b, d). This argues for post-generative alterations (including a 1–2 level of biodegradation) and is also suggested by bulk parameters (Table 1).

The studied oil fields are situated in the area surrounding the large gas and gas-condensate fields Molve, Kalinovac and Stari Gradac in the northwestern part of the Drava Basin (Fig. 1). As has already been mentioned, gas condensates do not contain hopanes and steranes. Consequently, these biomarkers cannot be used for correlating crude oils and gas-condensates (PETERS & MOLDOWAN, 1993; MARINOVIĆ & BIONDIĆ, 2023).

Gas-condensates and low maturity oils (see Section 4.4) generally have different compositions of light hydrocarbons due to the significant difference in maturity. Therefore, for comparison of the gasoline fractions C_7 – C_9 hydrocarbons are considered, as they are the most abundant in gasoline fractions of crude oils (Fig. 8) and less volatile compared to C_5 – C_6 hydrocarbons. The gasoline fraction of the Jagnjedovac oil (Fig. 8a) shows a composition closely comparable to that of the Molve condensate (Fig. 9). Both fluids are characterized by the predominance of *n*-nonane, elevated concentrations of light aromatic hydrocarbons (T and Xy; BARIĆ, 2006) and showed the pattern: T > *n*-heptane; T slightly lower than *n*-octane. In contrast, the Šandrovac oil (Fig. 8e) resembled the distribution of T, *n*-heptane, *n*-octane and *n*-nonane in the Kalinovac and Stari Gradac gas condensates (*n*-nonane dominates; T slightly prevails over *n*-heptane; *n*-octane is much higher than T; Fig. 9). The observed similarities in the gasoline fractions indicate that the Jagnjedovac and Šandrovac oils may contain condensate components, implying possible mixing between locally generated oils and the Molve, Kalinovac and Stari Gradac gas condensates.

As mentioned before, in the Bjelovar Subbasin, source rocks occur only locally (MALVIĆ, 2003a) and consist of mixed marine and terrestrial organic matter (kerogen types II–III), with low to early maturity. Generally, sufficient quantities of source rocks and the quality of the organic matter are not proven. Almost all the hydrocarbon reserves discovered in the Bilogora zone migrated from the northwestern Drava Basin, where gas-prone Type III kerogen, source rocks for the Molve, Kalinovac and Stari Gradac gas condensates—played a significant role in charging the reservoirs in the study area. Furthermore, the observed similarity between the gasoline fractions of the studied oils and the gas condensates (Figs. 8, 9) supports the interpretation that locally generated oils in the Bjelovar Subbasin were mixed with condensate from the northwestern Drava Basin, located to the north and northeast of the Bilogora zone.

4.4. Maturity assessment

From the perspective of exploration studies, crude oil thermal maturity may be considered as one of the most important geochemical characteristics. The maturation process involves cracking, isomerisation and aromatization reactions, as well

as alkylation and dealkylation of aromatics rings (STOJANOVIĆ et al., 2007; MRKIĆ et al., 2011). Hopanes and steranes represent the biomarkers which are commonly used for maturity assessment (MACKENZIE et al., 1985; PETERS & MOLDOWAN, 1993). The ratio of thermodynamically more stable 22S- and the thermodynamically less stable 22R-epimer of C_{31} – C_{35} $\alpha\beta$ hopanes is used as a maturity parameter. Calculating the value of the ratio 22 (S)/22 (S+R) for the C_{31} – C_{35} series of 17 α (H) 21 β (H) hopanes in a large number of samples resulted in almost identical mean values (0.55, 0.58, 0.60, 0.62, and 0.59 for C_{31} – C_{35} hopane, respectively) being obtained. However, most studies use the C_{31} - or C_{32} homologue ratios, because they are usually present in high concentrations in samples, while C_{34} and especially C_{35} $\alpha\beta$ hopane is present in a significant amount only in samples from reducing environments (PETERS et al., 1995; PETERS et al., 2005). From the analysis of a large number of samples (SEIFERT & MOLDOWAN, 1980), it was found that equilibrium values of the C_{31} 22 (S)/22 (S+R) hopane ratio are in the range of 0.57–0.62 which corresponds to an initial stage of oil generation (vitrinite reflectance, ≈ 0.6 %). The studied oils have the C_{31} $\alpha\beta$ (22S)/ C_{31} $\alpha\beta$ (22S+22R) hopane ratio in the equilibrium range or slightly lower (0.51–0.57; Table 5). However, values of the C_{30} moretane/ C_{30} hopane ratio (M/H, 0.12–0.14; Table 5) are in the equilibrium range for Tertiary oils (0.10–0.15), which corresponds to a vitrinite reflectance of 0.70–0.75 % (the first third of the “oil window”; SEIFERT & MOLDOWAN, 1980; PETERS et al., 2005). Therefore, slightly lower values than the equilibrium ones of the C_{31} $\alpha\beta$ (22S)/ C_{31} $\alpha\beta$ (22S+R) hopane ratio, can be attributed to the Tertiary age and insufficient time for epimerization, at C-22 in hopanes to reach equilibrium (GEORGE et al., 2001).

Ratios used to evaluate the maturity of crude oil from sterane biomarkers are based on thermodynamically more stable 5 α (H), 14 α (H), 17 α (H) 20 (S) and 5 α (H), 14 β (H), 17 β (H) 20 (R and S) and less stable 5 α (H), 14 α (H), 17 α (H) 20 (R)-isomers of C_{27} – C_{29} -steranes (SEIFERT & MOLDOWAN, 1986). Most often, C_{29} sterane concentrations are applied to the ratios, because the abundance of these steranes does not interfere with other peaks using routine GC-MS analysis (PHILP, 1985).

The sterane isomerisation indices $C_{29}20S / (20S + 20R)$ and $C_{29}20S / (20S + 20R)$ indicate low maturity for the studied oil samples (Table 4). The equilibrium values for the parameter $C_{29}20S / (20S + 20R)$ are 0.52–0.55 and corresponds to the peak of the “oil window” (vitrinite reflectance ≈ 0.80 – 0.85 %). The equilibrium values for the $C_{29}20S / (20S + 20R)$ sterane ratio are 0.67–0.71, which corresponds to the beginning of the late stage of the oil window (vitrinite reflectance ≈ 0.90 – 0.95 %) (SEIFERT & MOLDOWAN, 1986). For the Jagnjedovac, Lepavina, Pavljani and Letičani oils, very low values of sterane maturation parameters were obtained, whereas the Šandrovac and Čepelovac oil showed slightly higher values, which are also apparently lower than the equilibrium ones (Table 5). Therefore, based on biomarker parameters, the maturity of the oils corresponds to the early stage of oil generation (low maturity, vitrinite reflectance, 0.60–0.70 %).

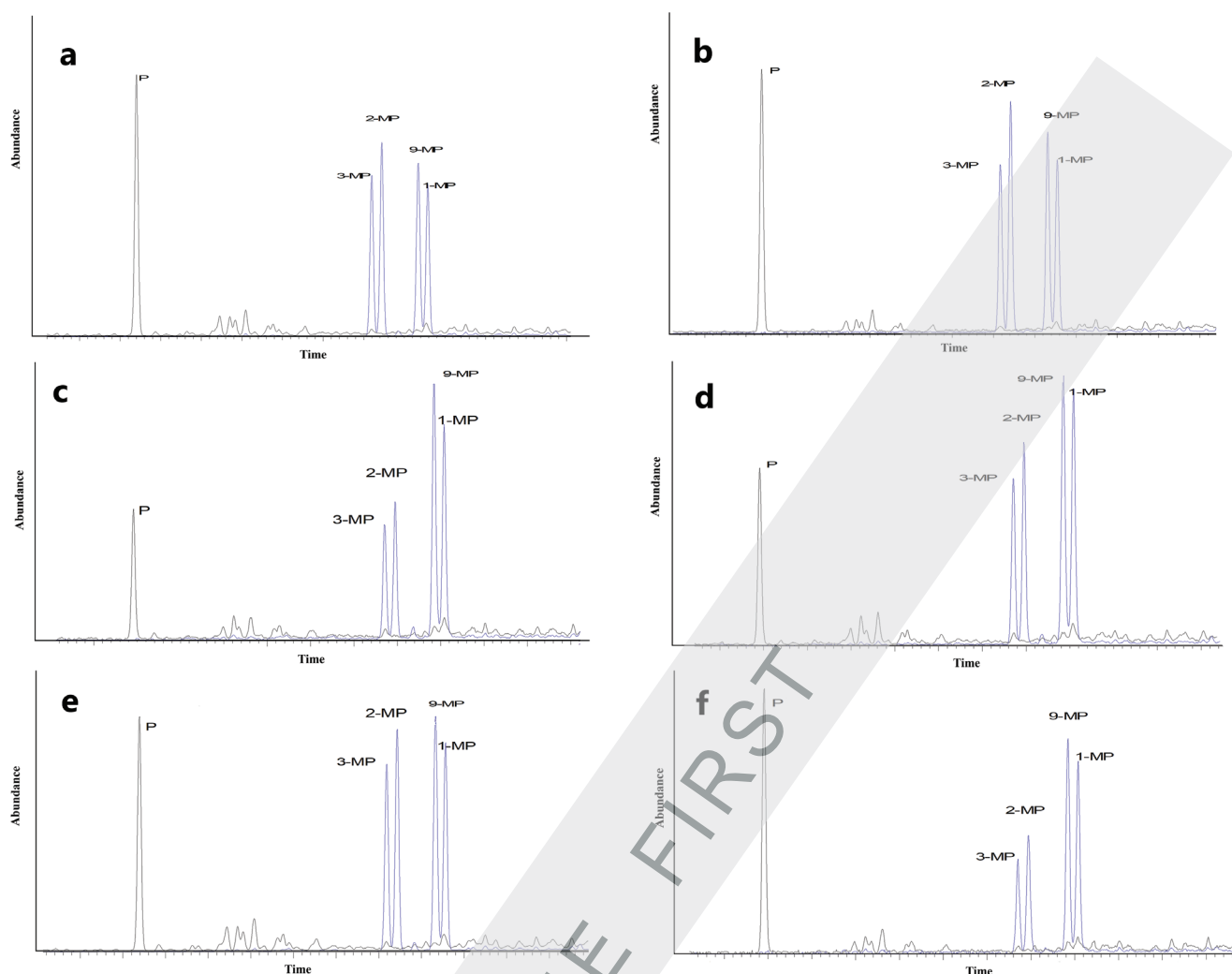


Figure 10. Mass chromatograms of phenanthrene and methylphenanthrenes ($m/z = 178 + 192$) of the investigated oils: **a** – Jagnjedovac; **b** – Lepavina; **c** – Čepelovac; **d** – Letičani; **e** – Šandrovac; **f** – Pavljani. P – phenanthrene; MP – methylphenanthrene.

In contrast to the biomarker parameters, the maturity ratios derived from phenanthrene and its methyl-derivatives can be used at higher maturities (vitrinite reflectance $\geq 1.00\%$) (SZCZERBA & ROSPONDEK, 2010). Distributions of methylphenanthrene (MP) isomers differ in the studied oils. Samples Čepelovac, Letičani and Pavljani show a prevalence of thermodynamically less stable α -isomers (1-MP and 9-MP), consistent with low maturity. In the Jagnjedovac, Lepavina and Šandrovac oils, the prevalence of thermodynamically more stable β -isomers (2-MP and 3-MP) is observed (Fig. 10). The maturation parameters based on the aromatic hydrocarbons (RADKE et al., 1982; RADKE, 1987), such as the methylphenanthrene indices (MPI-1, MPI-2 and MPI-3) are shown in Table 6. The obtained results indicate that the Jagnjedovac, Lepavina and Šandrovac oils are mature petroleum samples (MPI-3 ≈ 1), whereas the other oils exhibit lower maturity (MPI-3, 0.52–0.67). The vitrinite reflectance calculated from MPI-1 (R_c ; RADKE & WELTE, 1983) varies over a wide range, $R_c = 0.66$ – 0.90% (Table 6), showing the high maturity (peak to beginning of late stage of oil window) of the Jagnjedovac, Lepavina and Šandrovac oils ($R_c = 0.90\%$), moderate maturity for the Čepelovac and Letičani oils ($R_c = 0.74$ – 0.82%), and low maturity of the Pavljani oil ($R_c = 0.66\%$). Therefore, based on

both biomarker and phenanthrene maturity ratios the Pavljani oil has low maturity. On the other hand, there is an inconsistency between the sterane and hopane, and methylphenanthrene ratios for the other oils. Biomarker maturation parameters

Table 6. Maturity parameters of investigated oils (a–f) and surrounding gas-condensates (1–3) based on phenanthrene and methylphenanthrene ($m/z 178+192$) distributions.

Sample	MPI-1	R_c	MPI-2	MPI-3
Jagnjedovac (a)	0.88	0.90	0.95	1.05
Lepavina (b)	0.88	0.90	0.95	1.05
Čepelovac (c)	0.61	0.74	0.64	0.52
Letičani (d)	0.75	0.82	0.81	0.67
Šandrovac (e)	0.89	0.90	0.96	0.90
Pavljani (f)	0.48	0.66	0.56	0.53
Molve-25 (1)	1.04	1.68	1.02	1.17
Kalinovac-15 (2)	1.50	1.40	1.30	1.73
Stari Gradac-6 (3)	2.61	0.73	1.97	3.05

*MPI-1 = $1.5 \cdot (3\text{-MP}+2\text{-MP})/(P+9\text{-MP}+1\text{-MP})$; $R_c = 0.6 \cdot (\text{MPI-1}) + 0.37$ for oils, $R_c = 2.30 - 0.60 \cdot \text{MPI-1}$ for gas-condensates; MPI-2 = $3 \cdot 2\text{-MP}/(P+1\text{-MP}+9\text{-MP})$; MPI-3 = $(3\text{-MP}+2\text{-MP})/(9\text{-MP}+1\text{-MP})$

**P – phenanthrene; MP – methylphenanthrene

indicated low maturity, whereas the methylphenanthrene indices supported the mature stages of hydrocarbon generation. A possible reason for this disagreement could be that the oils represent a mixture of light mature oil (similar to condensate, which occur in the studied area; Fig. 1) and a low mature oil that has dissolved in a mature one. Namely, the condensates have no steranes and terpanes, and therefore their distributions in mixtures correspond to the low mature oil. On the other hand, mature oils/condensates have higher contents of phenanthrene derivatives than low mature oils, and therefore the mature patterns of methylphenanthrenes predominate in the oil mixture. This explanation fits well with the geographical locations of oils (Fig. 1). Namely, oils a–e are close to the gas/gas-condensate fields (1–3), while the location of oil f (Pavljani) is more distant, which probably disabled mixing. Based on the obtained results (differences in maturity according to biomarker ratios and MP-indices and MP distributions), the Jagnjedovac, Lepavina and Šandrovac oils have higher amounts of gas condensate in their mixtures than the Čepelovac and Letičani oils, the latter being located in the central part of the study area (Fig. 1). Comparison of chromatograms of the gasoline fractions (Section 4.3) and methylphenanthrene distributions in the oils and gas condensates (Figs. 10, 11), reveals that the Jagnjedovac and Lepavina oils had a contribution from the Molve gas condensate (P dominates over 2-MP). In contrast, the Šandrovac oil (P similar to 2-MP) was most probably mixed with both the Kalinovac (P slightly dominates over 2-MP) and Stari Gradac (2-MP dominates over P) gas condensates. These results are consistent with findings from comparison of the gasoline fractions (Figs. 8, 9) and the locations of the oils and gas condensates (Fig. 1).

4.5. Characteristics of source rocks of crude oils

Maturity parameters derived from biomarkers and methylphenanthrenes, together with the similar compositions of the gasoline fractions (Figs. 8, 9) and similar MP distributions (Figs. 10, 11) in the analysed oils and surrounding condensates, indicate that five crude oils represent mixtures of low mature crude oil and gas condensate. A possible migration pathway is the Drava Fault, which, due to its regional extent and tectonic activity, may have acted as a structural corridor enabling hydrocarbons generated in the northwestern Drava Basin to migrate toward the Bjelovar Subbasin and mix with locally derived low mature oils. Despite the limited source potential in the Bjelovar Subbasin, the low mature Pavljani oil was probably derived from hydrocarbon sources containing Type II–III kerogen, detected in the triangle between Rovišće, Letičani and Galovac–Pavljani. Furthermore, as suggested by MALVIĆ (2003b), these sources probably had particular contributions to the oil fields Letičani and Šandrovac. Since the Lake Pannon successions are diachronous, multiple potential source intervals probably exist, showing laterally variations in maturity.

Indeed, source rocks of Badenian, Sarmatian and lower Pannonian age, which have different lithological characteristics (mudstones, shales and silts) and maturity (vitrinite reflectance 0.3–2.2 %), are detected in the Bjelovar depression at depths between 1100 and 3450 m (BARIĆ, 2006). Considering the vitrinite reflectance of 0.6–0.7 % (indicated from biomarker

ratios), the temperature of hydrocarbon generation (T) and the relative depths of source rocks (D) of low mature oil have been estimated using the formulas created for prolific hydrocarbons production areas in the Pannonian Basin: $T = \ln(Rc/0.2096)/0.0085$; $D = 3.67 \times T^{1.33}$ (KOSTIĆ, 2010). The results obtained showed that the temperature of hydrocarbons generation ranges from 124 to 142 °C, which corresponds to the relative depths of source rocks at 2230–2670 m. The elevated heating rates in the study area probably caused rapid maturation of OM in the source rocks, associated with the generation and expulsion of hydrocarbons, as well as the establishment of specific thermodynamic conditions, while also changing the composition of the fluids. Since the high

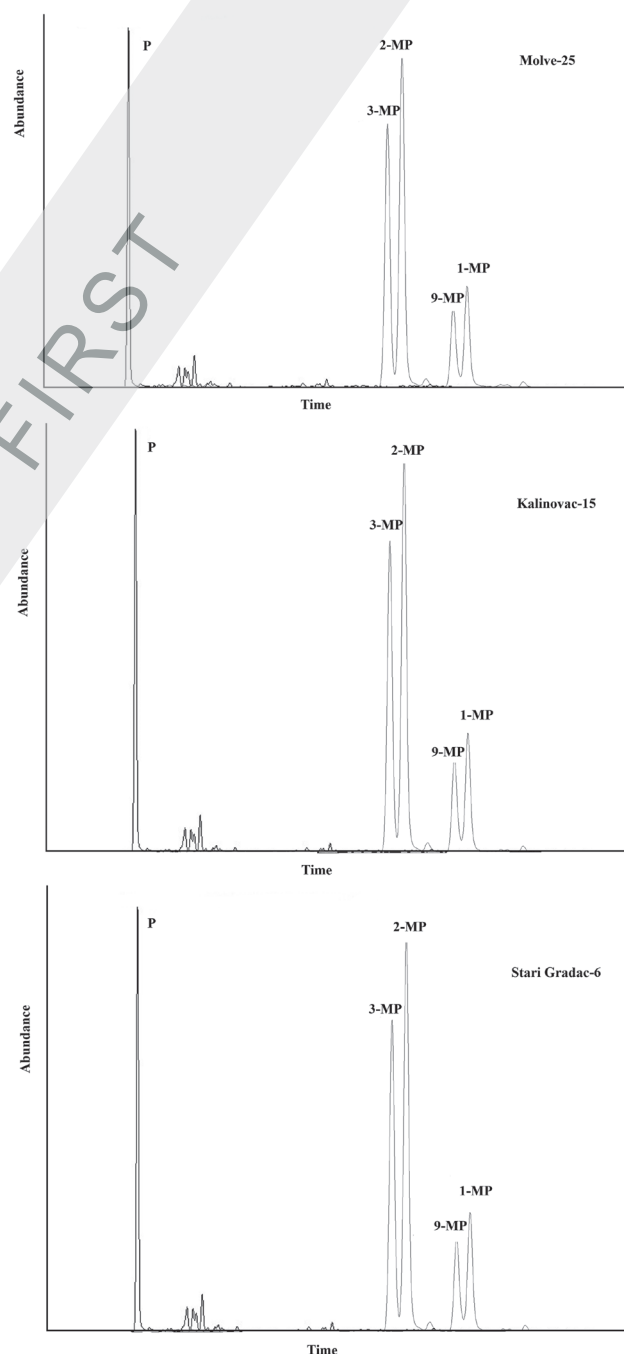


Figure 11. Mass chromatograms of phenanthrene and methylphenanthrenes ($m/z = 178 + 192$) of the gas-condensates Molve-25, Kalinovac-15 and Stari Gradac-6. P – phenanthrene; MP – methylphenanthrene.

heating rates narrow the oil window that corresponds to the lower vitrinite reflectance (0.5–0.9%; WELTE, 1989; SWEENEY & BURNHAM, 1990), it can be suggested that source rocks of low mature oil ($R_c = 0.6$ –0.7 %) at least in part, are in the main stage of the oil window, rather than in its initial stage. However, insufficient time for the isomerisation of steranes, which are more energy demanding and a slower process than isomerisation of hopanes (PETERS et al., 2005), resulted in the reduced sterane maturity ratio. This is in accordance with the results of BARIĆ et al. (1992), related to the presence of source rocks, containing a mixture of Type II and III kerogen in the Bjelovar Basin, which are within the main stage of the oil window.

From all the above it is obvious that the petroleum fields located on Bilogora Mt. have a very complicated structural-tectonic history which has influenced the chemical composition and maturity of the discovered hydrocarbons.

5. CONCLUSION

Six crude oils (Jagnjedovac, Lepavina, Čepelovac, Letičani, Šandrovac and Pavljani) from the western part of the Drava Basin, which are surrounded by the largest gas and gas-condensate fields (Molve, Kalinovac and Stari Gradac) in the Croatian part of the Pannonian Basin, were investigated to determine origin, depositional environment and thermal maturity.

The API gravities of the studied samples ranged from 25.95 to 36.76° classifying them as medium crude oils. According to the sulphur content, most of the studied oils are sweet, but it should be noted that this parameter varied over a relatively wide range of 0.30–0.98 %. The oils have a high content of paraffins (>6 %), while the Pour point varies between +9 and +33 °C (Table S1).

Despite particular differences in the bulk parameters, the sterane and terpane “fingerprints” suggest a common origin for all samples. Oils were generated from siliciclastic source rocks containing a mixture of marine and terrestrial organic matter (Table S2). Precursor organic matter was deposited in a deltaic suboxic-oxic environment (Table S3). Particular differences in the distributions of *n*-alkanes and bulk parameters are attributed to the post-generative alterations (initial stage of biodegradation and evaporative fractionation), which are indicated based on *n*-alkane patterns and compositions of the gasoline fractions (Table S1). Maturity parameters derived from biomarkers and methylphenanthrenes (Table S4) accompanied with similar compositions of the gasoline fractions of oils and surrounding condensates, indicate that the five oils (Jagnjedovac, Lepavina, Čepelovac, Letičani, Šandrovac), represent a mixture of low mature oil and surrounding condensate. The obtained geochemical results are in accordance with the geological interpretation that source rocks from the northwestern Drava Basin played a significant role in charging the hydrocarbons’ reservoirs in the Bjelovar Subbasin. One sample, Pavljani (geographically the most distant from the gas-condensate fields) is a low mature oil which was generated at maturity corresponding to the vitrinite reflectance 0.60–0.70 %. The estimated temperature of hydrocarbon generation of the source rocks in the Bjelovar Subbasin ranges from 124 to 142 °C,

which corresponds to their relative depths of 2230–2670 m. The obtained results provide particular implications for regional petroleum system modelling.

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