

Supplement 1

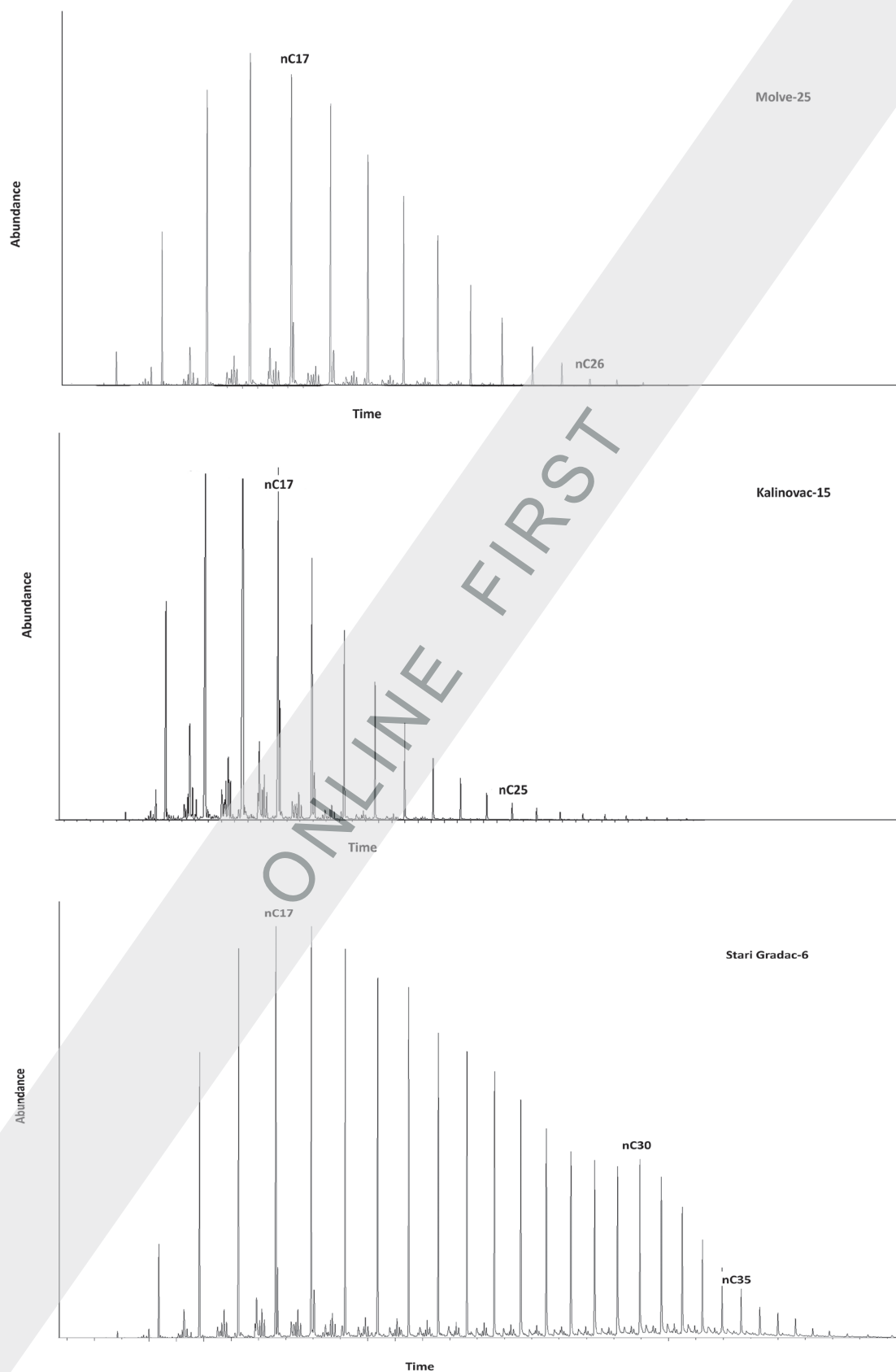


Figure S1. Mass fragmentograms of n -alkanes (m/z 85) of gas-condensates Molve-25, Kalinovac-15 and Stari Gradac-6.

Table S1. Summary of parameters indicative for the post-generative alteration of oils.

Parameter	Geochemical property of oil	Results
API gravity (°)	≤ 20° severe biodegradation of oil, 20–40° medium oil, 40–45° light oil.	All oils have API > 20°. Lepavina and Letičani oils have API < 30°.
S (%)	High in oils from marine/evaporitic and hypersaline lacustrine reducing environments, elevated in biodegraded oils, low in highly mature condensates, as well as in terrestrial and lacustrine freshwater samples.	Three samples have S content less than 0.5 %. Lepavina and Letičani oils have S content 0.86–0.98 %. Pavljani oil has S content of 0.68 %. Marine/evaporitic reducing environments are ruled out in studied sample set.
PP (°C)	High in oils with high paraffin wax which is indicative for significant contribution of terrestrial plants and/or biodegradation.	PP of oils is in range 9–21 °C (impact of terrestrial OM), only Lepavina and Letičani have PP ≥ 30 °C.
SARA analysis	Lower content of hydrocarbons (≤50 %) is indicative for severe biodegradation.	In all oils content of hydrocarbons exceeds 82 %, indicating no severe biodegradation.
Loss of short-chain n-alkanes (C15–C20)	Initial stages of biodegradation (level 1–2). Evaporation.	Jagnjedovac, Lepavina, Letičani and Šandrovač oils showed loss of short-chain n-alkane homologues.
Toluene/n-heptane ratio in the gasoline fraction	0.2–0.6 in normal oils. >0.6 indicates evaporative fractionation. in biodegraded oils content of gasoline fraction is low.	Toluene/n-heptane >0.60 in Jagnjedovac and Šandrovač oils. Lepavina and Letičani oils have very low content of gasoline fraction.

Interpretation: All samples with the exception of Čepelovac and Pavljani were exposed to post-generative alteration. The Lepavina and Letičani oils were exposed to the initial stage (1–2) of biodegradation, whereas the Jagnjedovac and Šandrovač oils were exposed to evaporative fractionation. However, the observed post-generative alteration cannot influence distributions of steranes, terpanes and phenanthrene derivatives, which are used in the interpretation.

Table S2. Summary of parameters indicative for the origin of the oils.

Parameter	Geochemical property of oil	Results
Paraffin wax and PP	High paraffin wax abundance and high PP are typical for oils derived from terrigenous organic matter (OM).	All oils have paraffin wax ≥6 % and PP ≥9 °C, indicating contribution of terrestrial OM.
Distribution of n-alkanes	Odd long-chain – terrestrial, short-chain – algal and bacterial, short-chain enrichment with maturity, loss of short-chain n-alkanes (C ₁₅ –C ₂₀) at initial stages of biodegradation (1–2), removal the whole series of n-alkanes at biodegradation level 4.	n-Alkanes are the most abundant compounds in saturated fractions. Studied oils contain n-C ₁₄ –n-C ₃₅ alkanes and have high contents of long-chain homologues (> n-C ₂₅).
Distribution of regular C ₂₇ –C ₂₉ steranes	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.	All samples have a characteristic "V" shape, i.e. the highest percentages of C ₂₇ and C ₂₉ steranes, and a slightly smaller proportion of C ₂₈ steranes, indicative for the marine, algal OM with a pronounced influence of terrestrial vegetation.
Sterane/hopane ratio (ST/HOP)	Contribution of algae vs. bacteria. ST/HOP is >1 in marine environments, <1 in lacustrine environments. Also ST/HOP <1 indicates terrigenous microbially reworked OM.	All oils have ST/HOP ratio >1, with exception of Čepelovac, which shows the value of 0.66.
Pr/n-C ₁₇ vs. Ph/n-C ₁₈ diagram	Shows the origin and depositional environment of OM (see Fig. 4). Ratios are very low at biodegradation level higher than 3. Also high maturation reduces values of these parameters.	All oils are plotted in region of type II–III kerogen (mixture of marine and terrestrial OM), deposited under suboxic/oxic conditions. Influence of high maturity or severe biodegradation is ruled out for the studied oils.
Oleanane/C ₃₀ αβ hopane	Indicative for input of angiosperms. Usually high in deltaic environments. Not influenced by maturity and biodegradation. The presence of oleanane is typical for age younger than the Early Cretaceous.	All oils have uniform and high contents of oleanane, with the oleanane/C ₃₀ αβ hopane ratios ≥ 0.13, suggesting high impact of angiosperm plants and deposition in deltaic environment.

Interpretation: All oils originate from the source rocks containing type II–III kerogen (mixture of marine and terrestrial OM). The contribution of terrestrial plants was significant in all samples.

Table S3. Summary of parameters indicative for the depositional environment.

Parameter	Geochemical property of oil	Results
Pr/Ph ratio	Generally redox indicator Pr/Ph >1 in oxidative environments; Pr/Ph <1 in reducing environments; Pr/Ph >3 often in coal. Also influenced by source and maturity (increases at high maturity).	The Pr/Ph ratios for the studied oils range from 0.9 to 1.4, which is characteristic of suboxic/oxic environment. Higher values of this parameter (≈ 1.4) are detected in Čepelovac and Pavljani oils.
Distribution of C_{31} – C_{35} $\alpha\beta$ homohopanes and C_{35}/C_{34} $\alpha\beta$ homohopane ratio	There is no exponential decrease in abundance from C_{31} to C_{35} $\alpha\beta$ hopanes in marine reducing environments. The C_{35}/C_{34} $\alpha\beta$ hopane ratio >0.8 in suboxic, and >1 in marine reducing environments. Notably declining trend in abundance from C_{31} to C_{35} $\alpha\beta$ homohopanes and the C_{35}/C_{34} $\alpha\beta$ hopane ratio ≤ 0.6 in terrigenous and lacustrine environments (with exception of saline and hypersaline lakes which show similar patterns as marine environments).	The distributions of C_{31} – C_{35} $\alpha\beta$ homohopanes of investigated oils show loss of content of homohopanes with increasing number of carbon atoms. The C_{35}/C_{34} $\alpha\beta$ hopane ratio of studied oils in range 0.89–1.10 indicates suboxic environments, with exception of Čepelovac (C_{35}/C_{34} $\alpha\beta$ hopane ratio = 0.62), which suggests oxidizing conditions.
Sterane/hopane ratio (ST/HOP)	ST/HOP is >1 in marine suboxic-reducing environments. ST/HOP <1 in lacustrine environments and in terrigenous microbially reworked OM.	All oils have a ST/HOP ratio >1, with the exception of Čepelovac, which shows the value of 0.66.
Pr/ n - C_{17} vs. Ph/ n - C_{18} diagram	See Table S2.	See Table S2.
Oleanane/ C_{30} $\alpha\beta$ hopane	See Table S2.	See Table S2.
Gammacerane/ C_{30} $\alpha\beta$ hopane	Elevated concentrations of gammacerane and high gammacerane/ C_{30} $\alpha\beta$ hopane ratio are typical for the OM of sediments deposited, below the stratified water column in marine and non-marine environments, which may be due to hypersaline conditions or temperature gradients in the water body.	The gammacerane/ C_{30} $\alpha\beta$ hopane ratios ≤ 0.06 in all samples denote deposition of precursor OM in the shallow water column, without significant stratification.
C_{29} $\alpha\beta$ norhopane/ C_{30} $\alpha\beta$ hopane (nH/H)	Usually higher than 0.6 in marine carbonates, in combination with C_{35}/C_{34} $\alpha\beta$ homohopane ratio >1.0 and Pr/Ph ratio <0.80.	C_{29} $\alpha\beta$ norhopane/ C_{30} $\alpha\beta$ hopane < 0.55 in all samples in combination with the Pr/Ph ratio > 0.90 indicate shales.
Ts/(Ts+Tm)	Usually low in carbonate environments and elevated in shales (attention – the ratio increased with maturity and pH).	All studied oils have uniform and relatively high Ts/(Ts+Tm) ratio in range 0.37–0.42. Since the influence of maturity can be ruled out (see Table S4), the obtained results indicate marine shales.

Interpretation: Parameters indicative for the depositional environment show that the oils originated from marine shales deposited in a deltaic environment. Precursor organic matter of all oils was deposited in a shallow water column under suboxic conditions, with the exception of the Čepelovac oil. The latter oil also originates from marine shales, but precursor organic matter was deposited under oxidizing conditions.

Table S4. Summary of parameters indicative for maturation.

Parameter	Geochemical property of oil	Results
Distribution of <i>n</i> -alkanes	Odd long-chain – terrestrial, short-chain – algal and bacterial, short-chain enrichment with maturity, loss of short-chain <i>n</i> -alkanes (C ₁₅ –C ₂₀) at initial stages of biodegradation (1–2), removal the whole series of <i>n</i> -alkanes at biodegradation level 4.	High contents of long-chain <i>n</i> -alkanes in all samples are indicator of low maturity.
C ₂₉ $\alpha\alpha$ 20(S)/ $\alpha\alpha$ 20(S+R) sterane i.e., 20S/(20S+20R)	Increases with maturity and reach approximate equilibrium value of 0.52–0.55 at vitrinite reflectance 0.80–0.85 %, which corresponds to the peak (maximum) of the "oil window".	All samples have 20S/(20S+20R) sterane ratio < 0.36 indicating low maturity (corresponding to vitrinite reflectance less than 0.80 %).
C ₂₉ $\beta\beta$ 20(R)/($\alpha\alpha$ 20(R) + $\beta\beta$ 20(R)) sterane	Increases with maturity. Reach value of 0.25 at the beginning (initial stage) of oil window (vitrinite reflectance $\approx$ 0.60 %) and reach approximate equilibrium value of 0.67–0.71 at vitrinite reflectance 0.90–0.95 %, which corresponds to the beginning of the late stage of the "oil window".	All samples have C ₂₉ $\beta\beta$ 20(R)/($\alpha\alpha$ 20(R) + $\beta\beta$ 20(R)) sterane ratio higher than 0.30, but less than 0.67. This indicates that corresponding source rocks at least reached initial stage of oil window (vitrinite reflectance 0.60 %).
The ratio of diasterane to sterane (Dia/Reg)	The diasterane to sterane ratio continuously increases up to vitrinite reflectance $\geq$ 1 %. However, elevated values are also observed in samples from source rocks enriched in clay-type minerals, as well as in samples whose organic matter was deposited in more oxic environments with lower pH (increased acidity). In addition, it may have elevated values at high level of biodegradation.	All studied oils have a diasterane/sterane ratio $\leq$ 0.17. Since other sterane and hopane parameters, as well as <i>n</i> -alkane distributions suggested a maturity level corresponding to the vitrinite reflectance of 0.60–0.70 %, the low values of this parameter are logical. However, apparent presence of diasteranes at low maturity level is consistent with marine shales as the source rocks.
C ₃₁ $\alpha\beta$ 22(S) hopane/C ₃₁ $\alpha\beta$ (22S+22R) hopane i.e., 22(S)/22(R + S)	Increases with maturity. Reach approximate equilibrium value of 0.57–0.62 at the beginning (initial stage) of oil window (vitrinite reflectance $\approx$ 0.60 %).	Oil samples have the 22(S)/22(R + S) ratio in the range of approximate equilibrium values, confirming that corresponding source rocks reached the initial stage of oil window (vitrinite reflectance 0.60 %).
C ₃₀ $\beta\alpha$ hopane (moretane)/ $\alpha\beta$ hopane i.e. M/H	Decreases with maturity. Approximate equilibrium value is in range 0.15–0.05. 0.05 is the equilibrium value in samples older than the Tertiary, while for Tertiary and younger samples value of 0.15 is taken as equilibrium. It is assumed that equilibrium value corresponds to the vitrinite reflectance 0.70–0.75 %, which roughly corresponds to the first third of the "oil window".	Oil samples have a M/H ratio in the range 0.12–0.15 suggesting that corresponding source rocks reached the first third of the "oil window" (vitrinite reflectance 0.60–0.70 %).
Methylphenanthrene index-1, MPI-1, Calculated vitrinite reflectance Rc = 0.6 * MPI-1 + 0.37	MPI-1 approximately continuously increases from 0.4 to 1.6 in the vitrinite reflectance range from 0.6–1.3 % and then decreases to 0.5 at the end of stage of gas condensate formation (vitrinite reflectance $\geq$ 2 %).	MPI-1 values and corresponding calculated vitrinite reflectance (Rc) values indicate low maturity of Pavljani oil (MPI-1 = 0.48, Rc = 0.66 %), moderate maturity of Čepelovac and Letičani oils (MPI-1 = 0.61–0.75; Rc = 0.74–0.82 %) and maturity corresponding to the peak to beginning of late stage of oil window (MPI-1 = 0.88–0.89; Rc = 0.90 %) for Jagnjedovac, Lepavina and Šandrovac oils.

Interpretation: 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 of 0.60 – 0.70 %. Other studied oils Jagnjedovac, Lepavina Šandrovac, Čepelovac and Letičani are mixtures of low mature oil (which contain steranes and hopanes) and gas condensates from the surrounding gas and gas-condensate fields (Molve, Kalinovac and Stari Gradac), which do not contain steranes and hopanes (due to the high maturity). Gas condensates have higher concentrations of phenanthrene (P) and methylphenanthrenes (MP), since the contents of these aromatic hydrocarbons increase with maturity. Therefore, in the mixtures, sterane and hopane distributions represent low mature oil, while P and MP distributions mirror those in gas-condensates. Based on the obtained results (differences in maturity according to the biomarker ratios and MP-indices/MP distributions), Jagnjedovac, Lepavina and Šandrovac oils have a higher contribution of gas condensate in the mixtures than the Čepelovac and Letičani oils, the latter having been located in the central part of study area. Comparison of chromatograms of gasoline fractions (Section 4.3) and methylphenanthrene distributions in 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). On the other hand, the Šandrovac oil (P similar to 2-MP) was most probably mixed with both Kalinovac (P slightly dominates over 2-MP) and Stari Gradac (2-MP dominates over P) gas condensates. These results are consistent with the locations of the oils and gas condensates (Fig. 1).