Potential of Sawahlunto and Sangatta Coal as Methane Producers based on Aspects of Organic Geochemistry of Aromatic Fractions

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**Abstract.** Identification of aromatic hydrocarbon fraction biomarkers in Sawahlunto and Sangatta coal samples has been carried out to determine their organic geochemical characteristics. Biomarker analysis using Gas Chromatography-Mass Spectrometry (GC-MS) instruments shows the distribution of triterpenoid naphthalene, phenanthrene, and pentacyclic polyaromatic compounds. The presence of 1,2,5-TMN compounds in the SL-02 coal sample which was not found in the KD-04 sample indicates that the input of organic matter comes from the reduction of β amyrin in higher plants in Angiospermae which is deposited in an oxic environment. The distribution of alkylphenanthrenes in the SL-02 sample such as methyl phenanthrene, dimethyl phenanthrene and trimethyl phenanthrene indicates coal with medium maturity. In the KD-04 sample, only dimethyl phenanthrene was found, indicating that the coal has a low maturity rating. Compounds 2 MP and 1 MP in the SL-02 coal sample have a high abundance and a Methyl Phenanthrene Index (MPI) value = 0.55, while in the KD-04 coal sample no methyl phenanthrene compound was found and the MPI value = 0. This indicates low coal maturity and indicates that the SL-02 coal sample is more mature than the KD-04 coal sample. The SL-02 coal sample is dominated by the abundance of the compound 1,2,9-trimethyl-1,2,3,4-tetrahydropicene, while the KD-04 coal sample is dominated by the abundance of the compound 2,2,9-trimethyl-1,2,3,4-tetrahydropicene. The difference in abundance indicates that the aromatization process in the KD-04 coal sample occurs in a more acidic environment than the SL-02 coal sample. On the other hand, both samples tend to produce prone gas which is classified as type III kerogen and has the potential to produce coalbed methane (CBM).

Keywords: Organic geochemistry, coalbed methane, biomarkers, aromatic hydrocarbons, Sawahlunto, Sangatta.

# INTRODUCTION

The islands of Kalimantan and Sumatra are the largest coal-producing islands in Indonesia [1]. Coal production in Indonesia in 2023 will reach 775 million tons, which is the second largest in the world [2]. This large amount of production is only used in the steam power plant (PLTU), the cement industry, metallurgy, fertilizer, textiles, paper, briquettes and other industries [3]. The use of coal for PLTU produces exhaust gas that can pollute the environment [4]. So it is necessary to develop the potential of coal which produces cleaner exhaust gas. One of the developments in coal potential is through diversification such as coalbed methane (CBM). CBM has great potential to be developed because it has more environmentally friendly exhaust gas compared to direct use of coal [5]. In recent years, CBM has been in the spotlight because this energy source forms natural gas naturally during the burial process and is considered one of the renewable alternative energy commodities in Indonesia. The gas trapped in the Sawahlunto and Sangatta coal layers is predicted to have potential as CBM [6], [7].

CBM is coal that contains natural gas (methane gas) in its pores [8] and is classified as a sweet gas producer. This gas is formed by coalification which does not contain hydrogen sulfide and can interfere during the process of using it as fuel. The formation of CBM can occur biogenically with the help of microbes and thermogenically with the help of geothermal energy. Orem & Finkelman, (2013) reported that biogenic methane gas is formed from low-rank coal, while Fallgren et al., (2013) said that more methane gas is produced by bituminous coal than lignite and subbituminous. In addition, subbituminous and highly volatile bituminous coals have a higher percentage of methane gas than anthracite [9].

The coal resources used in this study came from the Sangatta Mine, East Kalimantan and the Sawahlunto Mine, West Sumatra. The Sangatta Mine is included in the Kutai Basin, which has medium-quality coal reserves

(calorific value between 5,800-7,100 kcal/kg) [10]. Meanwhile, the Sawahlunto coal mine is included in the Ombilin Basin, which has a sub-bituminous coal rank, namely coal with medium maturity with a calorific value

> 7,200 kcal/kg) [7]. The coal needs to be analyzed to determine its potential as a CBM producer. This is because each coal has its own uniqueness and potential, so several related studies need to be carried out before being used, one of which is through an organic geochemical approach [11]. Through the organic geochemical aspect approach based on the analysis of sediment biomarkers, a coal sample's potential can be determined [12]. The presence of biomarkers in geological sediment samples can be linked to the depositional environment, the source of organic matter, and thermal maturity [7], [13].

Polyaromatic biomarker groups such as phenanthrene in sediments can be used to determine the level of maturity, type of kerogen, and source of organic matter [14], [15]. The maturity indicator of a sediment can also be determined based on the methyl phenanthrene index (MPI) value, which is the ratio of the abundance of β isomers (2-methylphenanthrene and 3-methylphenanthrene) to α isomers (1-methylphenanthrene and 9-methylphenanthrene) [16], [17]. In addition, the high abundance of 2-methylphenanthrene, accompanied by high vitrinite levels, indicates type III kerogen and tends to produce gas (gas prone) [17], [18]. Through the analysis of aromatic hydrocarbon fraction biomarkers, it can be seen that the characteristics of Sawahlunto and Sangatta coal tend to be gas-prone or oil-prone. Based on this information, coal can be explored and converted into CBM.

# EXPERIMENTAL

## Sample

Coal samples were obtained from the Sawahlunto mine, West Sumatra and the Sangatta mine, East Kalimantan. Coal samples were washed with running water and dried in direct sunlight. After drying, each Sawahlunto and Sangatta coal sample was finely ground to a size of 200 mesh [7].

## Extraction

A total of 60 grams of each coal sample SL-02, Sawahlunto and KD-04, Sangatta were extracted using Soxhlet for 120 hours in 250 ml of the azeotropic solvent dichloromethane:methanol (9:1 v/v) [19]. A total of 50 mg of extract from each sample was fractionated using the preparative thin layer chromatography (TLC) method on silica glass plates (GF254) with n-hexane eluent. After elution, it was then dried, and the spots were observed using UV at a wavelength of 254 nm to obtain the aromatic hydrocarbon fraction [13].

## GC-MS test

The aromatic hydrocarbon fraction obtained previously was dissolved in an n-hexane solvent. Then the sample was injected in splitless mode and analyzed using an Agilent Gas Chromatograph (6890N) coupled to an Agilent mass selective detector (D5975C) equipped with an HP-5MS fused silica capillary column (60 m × 0.25 mm, 0.33 μm film thickness). Helium was used as a carrier gas at a rate of 1 mL/min (constant flow), and a mass spectrometer with an electron ionization source operated at an energy of 70 eV. The initial temperature in the GC-SM analysis was set to 70 °C, then held for 1 minute. The program instrument was run at minute 5. The operating temperature used was an isothermal temperature of 70 °C maintained for 1 minute, increased to 180

°C at a rate of 10 °C/minute. Then the temperature was raised again to 315 °C with a speed set at 4 °C/min. The final isothermal temperature was set at 315 °C and maintained for 30 minutes [20].

# RESULTS AND DISCUSSION

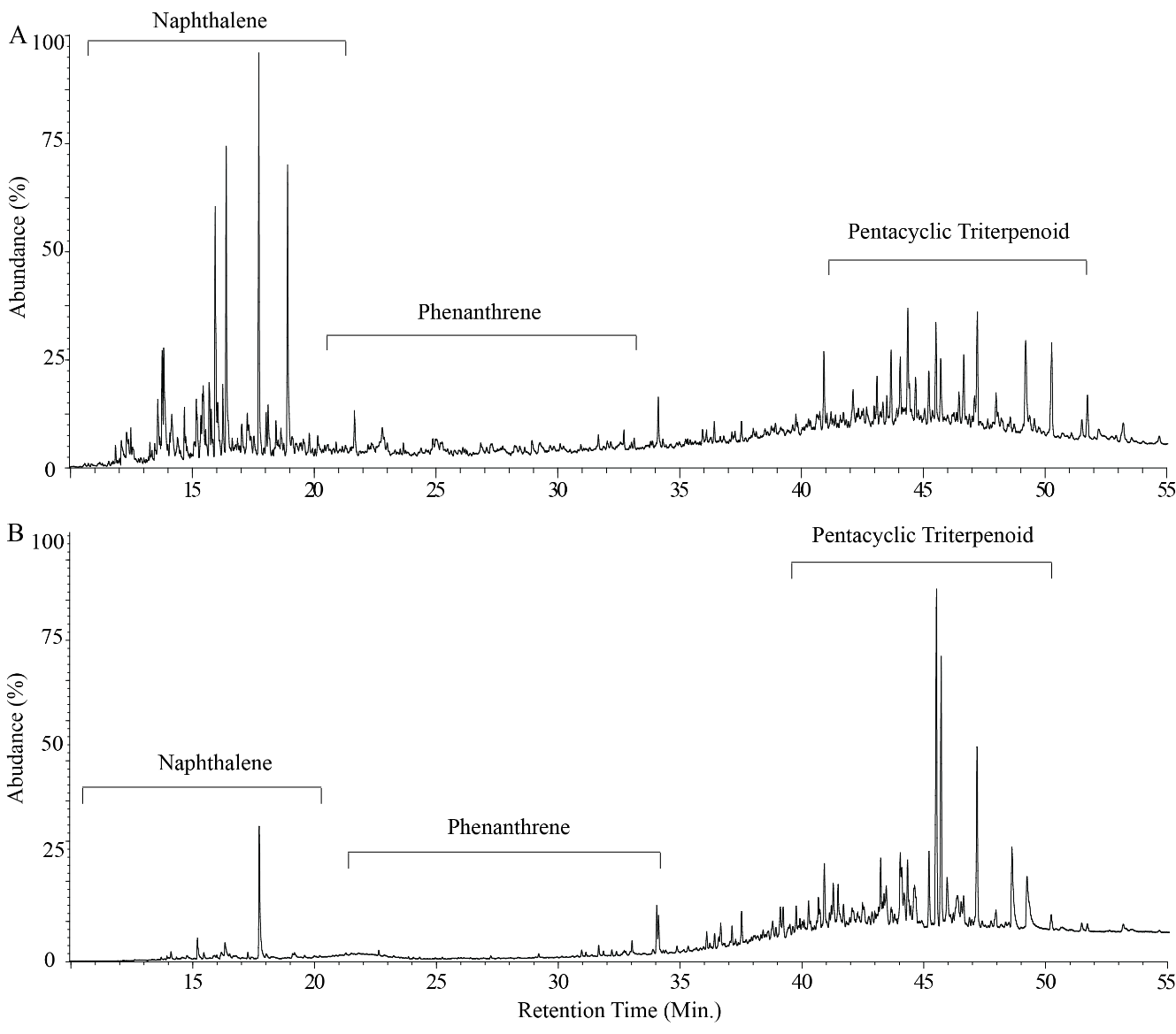
## Extraction and Fractionation of Coal

Extraction was carried out on two coal samples (SL-02 and KD-04) and the total organic extracts were 1.325 g (2.21%) and 2.124 g (3.69%) respectively. Fractionation was carried out using the Thin Layer Chromatography (TLC) method on EOT to obtain the aromatic hydrocarbon fraction. The aromatic hydrocarbon fraction obtained was then desulfurized with copper (Cu) powder. The results of each fractionation of the SL-02 and KD-04 samples obtained were 18.6 mg and 8.8 mg, respectively. The aromatic fractions of both samples were identified for their biomarker structure using GC-MS.

## Identification of Aromatic Hydrocarbon Biomarkers

The total ion chromatogram (TIC) in **Fig. 1** (A and B) was produced in KG-SM analysis on the aromatic hydrocarbon fraction of coal SL-02, Sawahlunto and KD-04, Sangatta. Identification of the structure of each component was carried out based on the specific fragmentogram provided and comparing the mass spectrum fragmentation data obtained with the mass spectrum published by previous researchers [13], [21], [30],

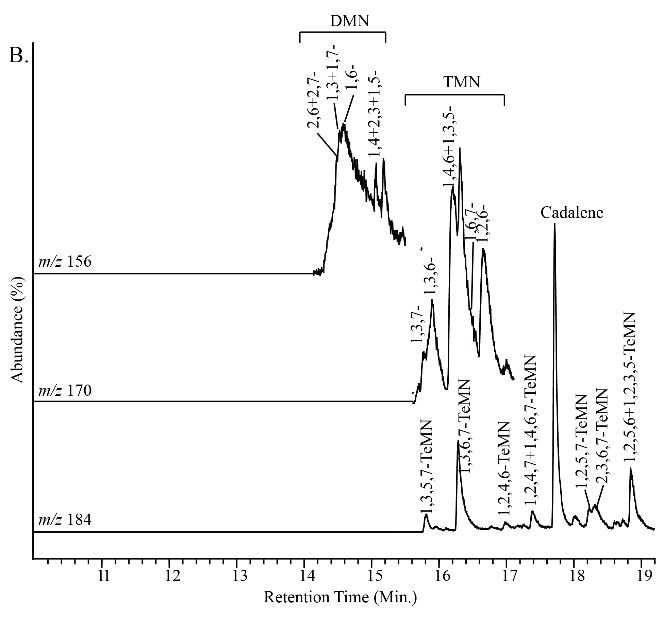
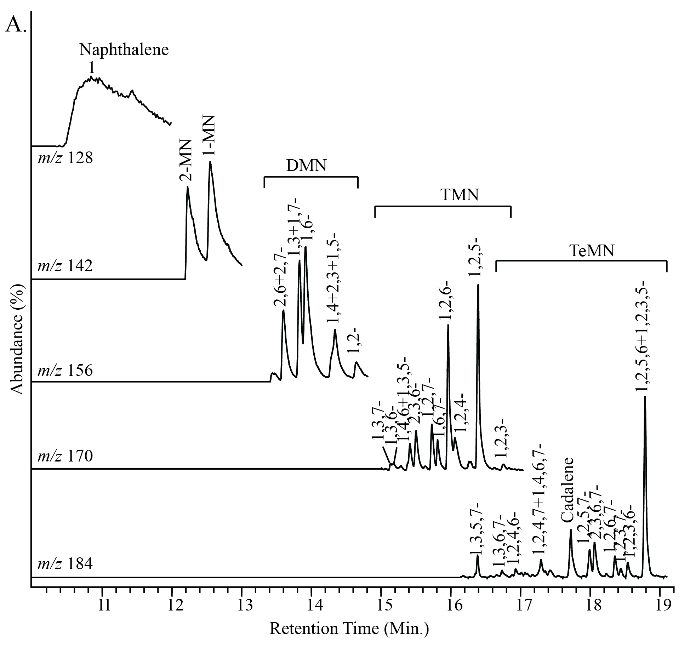
[22]–[29]. Biomarker analysis of the aromatic hydrocarbon fraction in coal samples SL-02 and KD-04 showed the presence of naphthalene, phenanthrene, and polyaromatic pentacyclic triterpenoid compound groups.



**FIGURE 1.** Total ion chromatograms (TIC) of Aromatic Hydrocarbon fractions (a) sample SL-02, (b) sample KD-04

Naphthalene Biomarker Group

The presence of the naphthalene compound group from coal samples SL-02 and KD-04 can be seen in **Fig. 2** (A and B) and identified based on specific fragmentograms of naphthalene (N, m/z 128), methylnaphthalene (MN, m/z 142), dimethylnaphthalene (DMN, m/z 156), trimethylnaphthalene (TMN, m/z 170), and tetramethylnaphthalene (TeMN, m/z 184).



**FIGURE 2.** Abundance of naphthalene derivative compounds (a) Sample SL-02, (b) Sample KD-04

Alkylated aromatic hydrocarbons are constituents widely used as molecular indicators of thermal maturity. This is due to the isomerization of substituted naphthalene and phenanthrene containing alkyl groups in the α-position into more stable forms with substituents in the β-position. Generally, α-substituted isomers of alkyl groups in the α-position become more stable structures with substituents in the β-position because α-substituted isomers of aromatic hydrocarbons are sterically hindered and thus exhibit less thermodynamic stability [25].

In the KD-04 coal sample, no naphthalene and dimethylnaphthalene compounds were found, while in the SL-02 coal sample, naphthalene and dimethylnaphthalene compounds were found. Samples with low maturity are indicated by the dominance of 1-MN over 2-MN [31]. The presence of 2-MN and 1-MN in the SL-02 coal sample indicates medium maturity. However, the maturity of the SL-02 coal sample is higher than that of the KD-04 coal sample because no 2-MN and 1-MN compounds were found. The SL-02 coal sample, which is a coal in the process of maturing, has a higher abundance of 1-MN compared to 2-MN, possibly because there is input from other sources at the 1-MN concentration.

Dimethylnaphthalene (DMN) compounds in coal samples SL-02 and KD-04 were identified based on fragmentogram m/z 156 as shown in **Fig. 2** (A and B). There are 10 possible isomers in the DMN structure, namely 1,2; 1,3; 1,4; 1,5; 1,6; 1,7; 1,8; 2,3; 2,6; and 2,7. Methyl groups substituted at the β position (C-2, C-3, C-6, or C-7) are more stable than those at the α position (C-1, C-4, C-5, or C-8). The maturity of organic matter is indicated by the dominance of more stable isomers. Coal samples SL-02 and KD-04 have lower abundances of stable DMN isomers (2,6- and 2,7-DMN) compared to other less stable DMN isomers, indicating immature coal. The DMN compound in both samples showed almost the same distribution. There was a difference in the abundance of 1,6-DMN against 1,3- and 1,7-DMN, in the SL-02 coal sample it had a higher abundance of 1,6-DMN against 1,3- and 1,7-DMN compared to the KD-04 coal sample. The high peak intensity of 1,6-DMN against 1,3- and 1,7-DMN and the low abundance of 2,6-; 2,7-; and 1,2-DMN indicated that the SL-02 coal sample was more mature than the KD-04 coal sample, but both samples were in the process of reaching maturity. The high abundance of 1,6-DMN compounds found in the SL-02 coal sample came from terrestrial plants and marine algae [32], [33].

Trimethylnaphthalene (TMN) compound was identified based on fragmentogram m/z 170. The presence of TMN determines the source of organic matter and the environmental conditions of deposition (Peters et al., 2005). In the SL-02 coal sample, the isomer 1,2,5-TMN has the highest abundance, while in the KD-04 coal sample, the isomer with the highest abundance is 1,4,6+1,3,5-TMN. The presence of 1,2,5-TMN compounds in high abundance in SL-02 coal is an indicator of organic matter input originating from the degradation of oleanane-type triterpenoids in higher plants, especially angiosperms [34]–[36]. On the other hand, oleanane-type triterpenoids (β-amyrin) can be converted into aromatic hydrocarbons during diagenesis to produce 1,2,5-TMN and 1,2,7-TMN through aromatization of rings A, B, D, E and cleavage of ring C [35], [36].

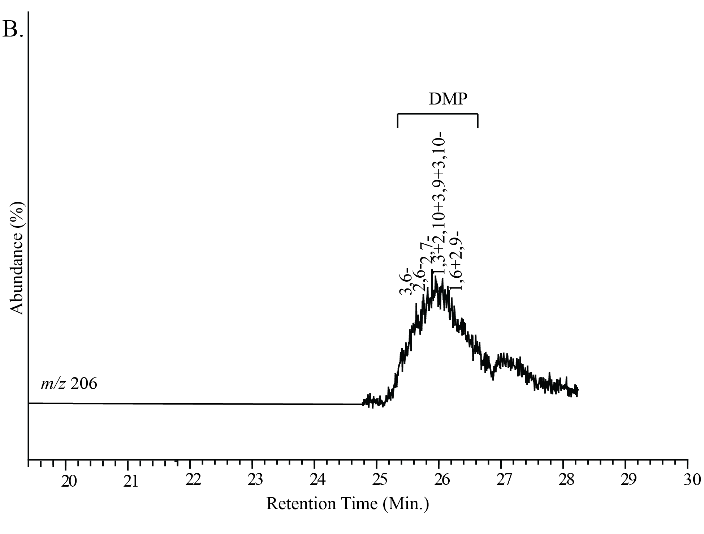
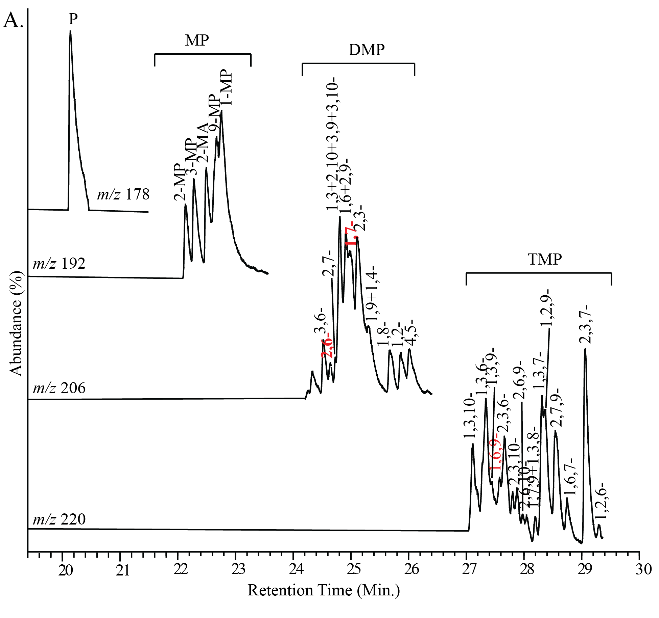
The intensity of tetramethylnaphthalene (TeMN) in coal samples was identified based on the fragmentogram m/z 184. In the SL-02 coal sample, the highest isomer was 1,2,5,6+1,2,3,5-TeMN, while in the KD-04 coal sample, 1,3,6,7-TeMN was the highest isomer. The biomarker 1,2,5,6-TeMN is an α substitution isomer, while 1,3,6,7-TeMN is a β substitution isomer. The methyl group at the β position is more stable at high temperatures than the α position, so the presence of 1,3,6,7-TeMN, which is slightly lower than 1,2,5,6-TeMN, indicates coal maturity [36], [37]. It is suspected that 1,2,5,6-TeMN is formed from the aromatization process of β-amyrin compounds originating from angiospermae and bacterial input [38].

Other naphthalene derivatives were identified based on the fragmentogram m/z 183 in both samples indicating the presence of cadalene. This compound is commonly found in petroleum and coal samples as an indicator of organic matter sources and thermal maturity [39], [40]. The presence of cadalene and isocadene compounds indicates that the organic matter input comes from higher plants. Cadalene is a compound formed from the polymerization of polycadynene, which is found abundantly in the resin of Dipterocarpaceae Angiospermae plants [33], [41]. The presence of the cadalene compound in both samples indicates that the coal sample comes from higher plants and has medium thermal maturity.

Phenanthrene Biomarker Group

Identification of phenanthrene derivatives in SL-02 and KD-04 coals using fragmentogram data of phenanthrene (P, m/z 178), methylphenanthrene (MP, m/z 192), dimethylphenanthrene (DMP, m/z 206) and trimethylphenanthrene (TMN, m/z 220). The identification was carried out by comparing specific fragmentograms, retention times, and mass spectra in previously published studies [22], [34], [42], [43].

The distribution of phenanthrene derivative compounds is depicted in **Fig. 3A** for sample SL-02 and **Fig. 3B** for sample KD-04. The phenanthrene compound group generally originates from diterpenoid compounds found in higher plant resins, generally Gymnospermae [44].



**FIGURE 3.** Abundance of Phenanthrene derivative compounds (a) Sample SL-02, (b) Sample KD-04

The distribution of alkylphenanthrene is useful for determining the maturity level of coal. The increase in coal maturity is indicated by the isomerization of alkylphenanthrene (α-isomer to β-isomer) and dealkylation of alkylphenanthrene (α-isomer to phenanthrene or β-isomer to phenanthrene) in the distribution of its compounds [17]. In general, the increase in coal thermal maturity is indicated by the change of alkylphenanthrene compounds substituted from the less stable α-position to the more stable β-isomer. The distribution of methylphenanthrene compounds in SL-02 coal shows the highest abundance of 1-MP compounds, which are associated with high terrestrial plant input. The 1-MP compound comes from the precursor of pimaric acid in higher plant resins, so it is generally found in type III kerogen, while the 9-MP compound is dominated by marine organic matter and is found in type I and II kerogen [39], [45], [46]. The distribution of MP in mature organic material is indicated by an increase in the abundance of 2- and 3-MP over 9- and 1-MP [17]. The SL-02 coal sample has a distribution of 9- and 1-MP with a more dominant intensity than 3- and 2-MP as seen in **Table**

**1**. This condition indicates that the coal is still heading towards the maturation process. Meanwhile, the KD-04 coal sample does not have a distribution of methylphenanthrene compounds. This indicates that the KD-04 coal sample has a lower thermal maturity than the SL-02 coal sample.

**TABLE 1.** The presence of methyl phenanthrene (MP) compounds in coal samples SL-02 and KD-04

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| **No.** | **Compound** | ***m/z*** | **% Abundance** | |
| **SL-02** | **KD-04** |
| 1 | 2 Methylphenanthrene | 192 | 8.59 | - |
| 2 | 3 Methylphenanthrene | 14.73 | - |
| 3 | 2 Methylphenanthrene | 13.70 | - |
| 4 | 9 Methylphenanthrene | 18.52 | - |
| 5 | 1 Methylphenanthrene | 44.46 | - |
| MPI (1.5 × ([2−MP] + [3 − MP])/ (P+[1−MP]+[9−MP])) | | | 0,55 | 0 |
| Log 1 MP/9-MP | | | 0,38 | - |

Maturity indicators based on aromatic hydrocarbons can be reviewed based on the number of isomers. The number of more stable isomers increases with the increasing thermal maturity of a sediment sample. The MPI parameter is based on the shift from the less stable 1-MP and 9-MP to the more stable 2-MP and 3-MP, so that the increase in the MPI parameter value is directly proportional to the increase in thermal maturity [47]. The MPI parameter value in the SL-02 coal sample is 0.55, indicating medium maturity, while the KD-04 coal sample has an MPI parameter value of 0, indicating low maturity.

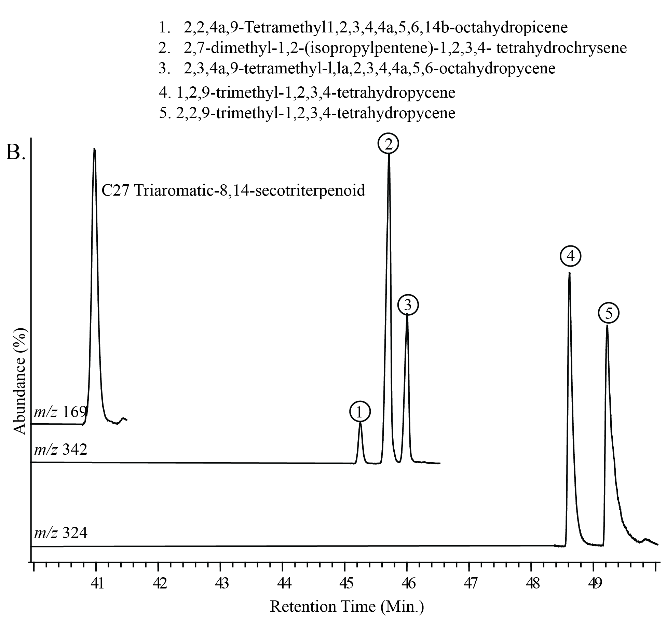
The increase in thermal maturity is also based on the distribution of DMP isomers. The most stable DMP isomers include 2,6-; 2,7-; 3,6-; 2,3-; 2,10-; and 2,9- DMP experiences an increase in abundance with increasing maturity. Conversely, less stable isomers, such as 1,7-; 1,6-; 1,2-; 1,8-; and 1,9-DMP, will decrease in abundance with increasing maturity [48]. The presence of 2,3-DMP with a fairly high intensity also indicates the origin of organic compounds derived from diterpenoid acids, such as pimaric acid, which is a component of conifers. The abundance of 2,3-DMP compounds in the SL-02 coal sample indicates that Gymnospermae plants also provide input on the availability of organic material in the SL-02 coal apart from the source of Angiospermae plants. Sample SL-02 showed that the organic matter input came from pimaric acid precursors in higher plant resins [39], [45], [46].

The high abundance of less stable DMP isomers indicates that the SL-02 and KD-04 coal samples have not reached the maturity level [46], [49]. However, there is a fairly high abundance of 2,3-; 1,7-; 1,3+2,10+3,9+3,10-; 1,6+2,9-DMP compounds in the SL-02 coal sample, which indicates that the coal sample continues to move towards the medium-high thermal maturity process. While in the KD-04 coal sample, the abundance of 3,6-; 2,6-; 2,7-; 1,3+2,10+3,9+3,10-; 1,6+2,9-DMP compounds is quite high, indicating that the coal sample is moving towards the medium thermal maturity process. This indicates that the SL-02 coal sample is more mature than the KD-04 coal sample.

The dominance of 1,2,6-TMP isomers indicates terrestrial organic matter input [50]. The presence of 2,3,6- and 2,3,7-TMP compounds is used as an indicator of coal maturity because its isomeric structure is stable so that it remains available in organic material that experiences a high temperature increase process [48]. The stability of TMP will be high if the methyl group is substituted at the βββ position, such as 2,3,6-TMP and at the αββ position, such as 2,6,9-TMP; 2,6,10-TMP; 2,7,9-TMP; 1,3,6-TMP and 1,3,7-TMP. Therefore, the isomers 1,3,6- and 2,7,9-TMP with quite high abundance with αββ substituents indicate the SL-02 coal sample at medium to high maturity [21], [22]. The distribution of TMP isomers in the SL-02 coal sample still shows a high abundance of TMP with less stable isomers. In the KD-04 coal sample, no TMP compounds were found. So it can be concluded that the thermal maturity of the KD-04 coal sample is lower than that of the SL-02 coal sample. The lower input of Gymnospermae plants in the KD-04 coal sample compared to the SL-02 coal sample is associated with the absence of MP and TMP compounds.

Aromatic pentacyclic triterpenoid group

The pentacyclic aromatic triterpenoid groups identified in samples SL-02 and KD-04 include monoaromatic pentacyclic triterpenoids, triaromatic pentacyclic triterpenoids, tetraaromatic pentacyclic triterpenoids and pentaaromatics. The abundance of the pentacyclic aromatic triterpenoid group can be seen in **Fig. 4**.



**FIGURE 4.** Abundance of triaromatic pentacyclic triterpenoid compounds in the m/z 342 fragmentogram, tetraaromatic pentacyclic triterpenoids based on the m/z 324 fragmentogram (a) SL-02 coal sample, (b) KD-04 coal sample

Biomarkers as indicators of Angiospermae plants are supported by a high-intensity fragmentogram m/z 342 that reveals the presence of 2,2,4a,9-tetramethyl-1,2,3,4,4a,5,6, 14b-octahydropysene molecules and 2,7-dimethyl-1,2-(isopropylpenteno)-1,2,3,4-tetrahydrochrysene compounds with a triaromatic ursane framework (partial m/z 257) [30], [49], [51]. The high abundance of 2,7-dimethyl-1,2-(isopropylpenteno)-1,2,3,4-tetrahydrochrysene compounds with a triaromatic ursane framework is an indicator of immature coal [49]. Therefore, considering that the analyzed coal samples are not yet mature and are supported by type III kerogen, the coal has the potential to be a CBM that tends to be gas-prone. However, the abundance of 2,7-dimethyl-1,2-(isopropylpenteno)-1,2,3,4-tetrahydropicene is relatively high compared to 1,2,4a,9-tetramethyl-1,2,3,4,4a,5,6,14b-octahydropicene, so SL-02 and KD-04 coals are estimated to have gas-prone potential [52]. This is also supported by the presence of triaromatic C27-8,14-secotriterpenoid compounds in SL-02 and KD-04 coals, which are formed by the cleavage of bonds in the C ring and are considered intermediates for the formation of trimethylnaphthalene from β-amyrin [53], [54].

The compound 1,2,9-trimethyl-1,2,3,4-tetrahydropycene is thought to originate from α-amyrin (ursane-type pentacyclic triterpenoid), while the compound 2,2,9-trimethyl-1,2,3,4-tetrahydropycene is a transformation product of β-amyrin with an oleanan framework [27], [43]. Both compounds are isomeric pairs consisting of geminal-dimethyl groups from each precursor with a fully hydrogenated E-ring [55]. The presence of both isomers indicates the input of organic matter from higher Angiospermae plants in coal samples SL-02 and KD-04. The SL-02 coal sample is dominated by the abundance of 1,2,9-trimethyl-1,2,3,4-tetrahydropicene,

while the KD-04 coal sample is dominated by the abundance of 2,2,9-trimethyl-1,2,3,4-tetrahydropicene. The difference in abundance indicates that the aromatization process in the KD-04 coal sample occurs in a more acidic environment than the SL-02 coal sample.

# CONCLUSION

Aromatic fraction biomarker analysis of coal samples SL-02 and KD-04 provides information on the composition of the compounds contained. The aromatic compounds found in both samples consist of three main groups, namely the group of naphthalene derivative compounds with a sesquiterpenoid framework, the group of phenanthrene derivative compounds with a diterpenoid framework, and the group of aromatic pentacyclic triterpenoid derivative compounds. The study of these compounds characterizes organic material originating from precursors of terrestrial higher plants, especially Angiospermae, which are prominent vegetation on the islands of Sumatra and Kalimantan, as well as the presence of bacterial input. The distribution of biomarkers in coal samples SL-02 and KD-04 indicates that both samples are approaching thermal maturity, where the SL-02 coal sample has medium maturity while the KD-04 coal sample has low maturity. However, both samples have a gas-prone (III) rating, which has the potential to produce CBM.

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