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Geochemistry of Potential Source Rock and Natural Gas Seepages in the Mangole-Taliabu Archipelago, North Maluku, Indonesia

Taufik Ramli[1], Iwan Sukma Gumilar[2], Ade Yogi[3], Ryan Akbar Fadhillah[4], Rakhmat Fakhruddin[5], and Praptisih[6]

[1] Research Center for Geological Resources, National Research and Innovation Agency, Jl. Cisitu, Sangkuriang Bandung, Jawa Barat 40135, Indonesia. https://orcid.org/0000-0003-0958-0411

[2] Center for Geological Survey, Geological Agency, Ministry of Energy and Mineral Resouces, Jl. Diponegoro No. 57, Bandung, West Java 40122, Indonesia

[3] Center for Geological Survey, Geological Agency, Ministry of Energy and Mineral Resouces, Jl. Diponegoro No. 57, Bandung, West Java 40122, Indonesia

[4] Center for Geological Survey, Geological Agency, Ministry of Energy and Mineral Resouces, Jl. Diponegoro No. 57, Bandung, West Java 40122, Indonesia

[5] Research Center for Geological Resources, National Research and Innovation Agency, Jl. Cisitu, Sangkuriang Bandung, Jawa Barat 40135, Indonesia. https://orcid.org/0000-0001-5448-5654

[6] Research Center for Geological Resources, National Research and Innovation Agency, Jl. Cisitu, Sangkuriang Bandung, Jawa Barat 40135, Indonesia. https://orcid.org/0009-0001-2377-2260

Abstract

The Mangole-Taliabu Archipelago has great potential for natural gas exploration due to its many active natural gas seepages. The limited natural gas exploration activities initiated during the last three decades led to a lack of publications on source rock and natural gas geochemistry. Previous research has postulated that the potential source rocks in this area and its surroundings are the Buya and Salodik formations, and known active natural gas seepages are hypothesised to originate from humic organic matter (type-III kerogen). The research aims to review and re-discuss the geochemical characteristics of potential source rock, the origins and genetics of natural gas seepages, and the correlation between natural gas seepage and source rocks by integrating the existing data with the newly collected data. This research revealed that the potential source rock is the Bobong Formation, aside from the previously confirmed Buya Formation, while the Salodik Formation is not a potential source rock as was suggested in previous publications. The most significant research finding is the type of natural gas seepage which found not only coal-derived gas but also oil-type gas. The calculated Ro of the coal-derived gas source rock is 0.80% – 1.00% and that of oil-type gas source rock ranges between 0.50% – 1.31%. For gas–source rock correlation, the Bobong Formation appears to be the most likely source for RAG/88/012 and RAG/88/014 gas samples, while the Buya Formation is the source for Binono Seep, RA/21/33, RA/21/34, and RA/21/35 gas samples. The new research findings are hoped to have significant implications for increasing the understanding of the petroleum system and encouraging further exploration activities.

Keywords:

source rock; natural gas seepage; organic geochemistry; carbon isotope; Mangole; Taliabu

1. Introduction

The research location is in the Taliabu-Mangole Archipelago, tectonically part of the North Banggai-Sula Microcontinent (see **Figure 1**). This area is underexplored for natural gas, although there are reports regarding active natural gas seepages (**Garrard et al., 1988; Ferdian, 2010**).

The available literature on geochemical studies indicates that the potential source rock can be identified in the Buya Formation's Late Jurassic-Early Cretaceous marine shale and the Miocene marine shales and coals within the Tomori and Matindok formations of Salodik Group (**Garrard et al., 1988; Davies, 1990; Hasanusi et al., 2004; Satyana and Zaitun, 2016**). According to **Garrard et al. (1988)**, the Buya Formation's anoxic shales from the Late Jurassic to Early Cretaceous periods are a promising source rock, with the range of values for total organic carbon (TOC) observed between 1 - 2 wt%. The study acquired hydrogen index (HI) measurements, revealing that most samples are inert (43 samples) and gas-prone (31 samples). Only two samples were shown to be mixed oil and gas-prone, while two others were oil-prone. The maturity of late Jurassic to early Cretaceous sediment from outcrop samples indi-

Corresponding author: Taufik Ramli
e-mail address: taufik.ramli@brin.go.id

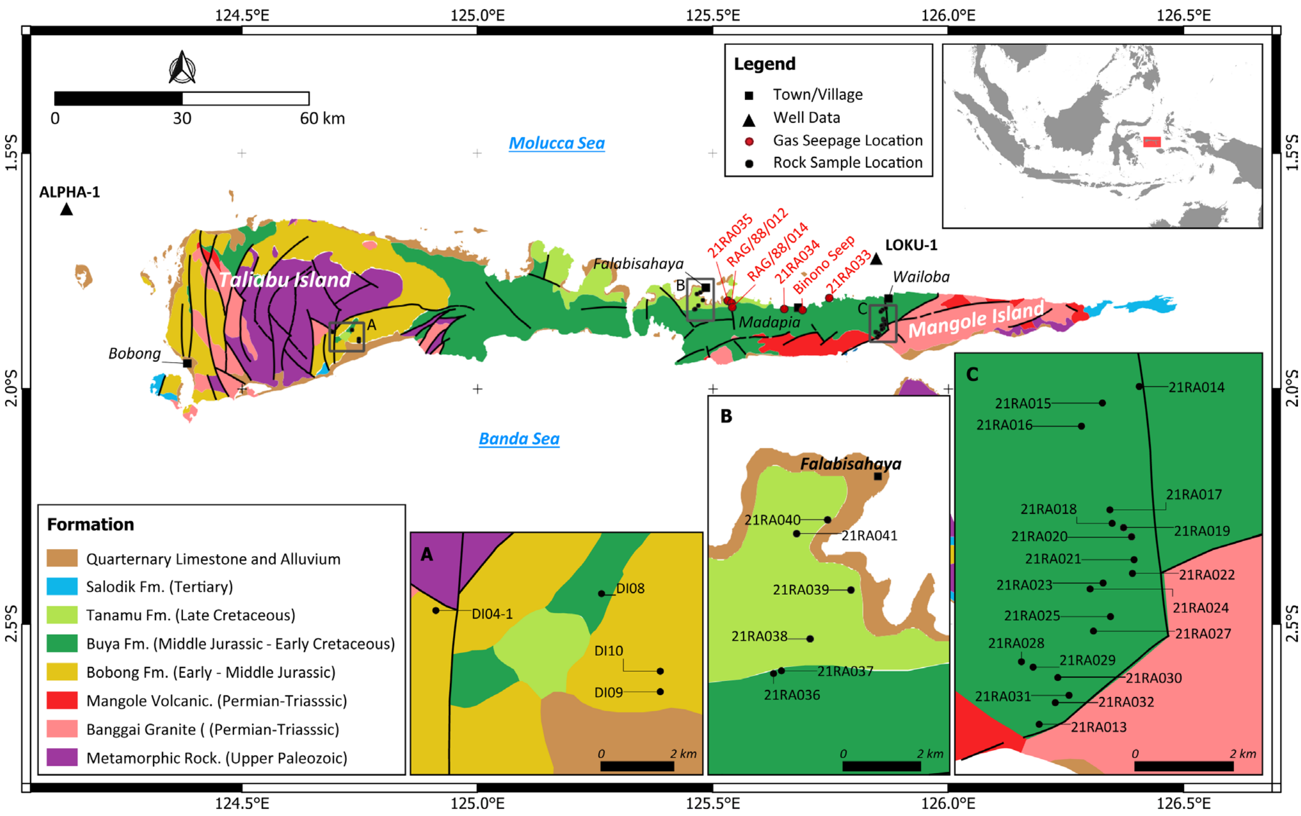

Figure 1: Geological map and data availability (modified from **Garrard et al., 1988; Supandjono and Haryono, 1993; Surono and Sukarna, 1993**)

cates interesting regional variations in the Mangole-Taliabu Archipelago (**Garrard et al., 1988; Satyana and Zaitun, 2016**). The southern part of the archipelago is more mature than its northern counterpart. In the northern part, extending from Taliabu Island to the northern part of Manggole Island, the maturity of potential source rock ranged from immature to fully mature, with the most mature samples in the eastern region. Meanwhile, the same sediments found along the southern part of Mangole Island show post-mature for oil generation (**Garrard et al., 1988**).

Furthermore, Miocene marine shales and coals within the Tomori and Matindok formations of the Salodik Group have also been mentioned as potential source rock by **Davies (1990)** and **Hasanusi et al. (2004)**. According to **Hasanusi et al. (2004)**, interbedded argillaceous limestones, coal, and carbonaceous shales of the Tomori Formation exhibit very good - excellent TOC and S2 (up to 32 wt% and up to 72 mg HC/g rock, respectively), and contained Type II/III kerogen based on HI and kerogen typing data. Meanwhile, the Matindok Formation, which is composed of interbedded shales, sandstone, limestones, and coals, has fair organic richness (TOC up to 0.96 wt%), poor residual hydrocarbon potential (S2 up to 1.48 mg HC/g rock), and primarily composed of Type III kerogen. However, these source rocks are located around 250 km west of this research area. Detailed geochemical analyses of the various potential source rocks from several wells indicate that the hydrocarbons are most likely genetically related to shales within the Tomori Formation of Salodik Group source rocks of Early to Middle Miocene age, with no apparent contribution from Jurassic source rocks (**Hasanusi et al., 2004**).

Garrard et al. (1988) reported that seven active gas seepages had been identified near Falabisahaya, northern Mangole Island. A comprehensive geochemical analysis was conducted on those samples to ascertain their composition, level of maturity, and potential source. The analysis of those gas samples revealed that the gas is thermogenic in origin. The source of the gas is likely humic organic matter and not associated with oil. However, there has been no report of the Jurassic coal presence in the surrounding area of the gas seep site as a potential source rock for the seepage. The closest documented coal outcrop is situated in the western region of Taliabu.

Geochemical characteristics of organic matter of selected rock samples from four rock formations (the Bobong, Buya, Tanamu, and Salodik formations), the origin and genetics of six natural gas seepages, as well as the correlation between natural gas seepage and source rocks are reviewed and re-discussed in this paper, by integrating the existing data with the most recent data available.

2. Geological Setting

The Taliabu-Mangole Archipelago is tectonically included in the Banggai-Sula Microcontinent (**Audley-**

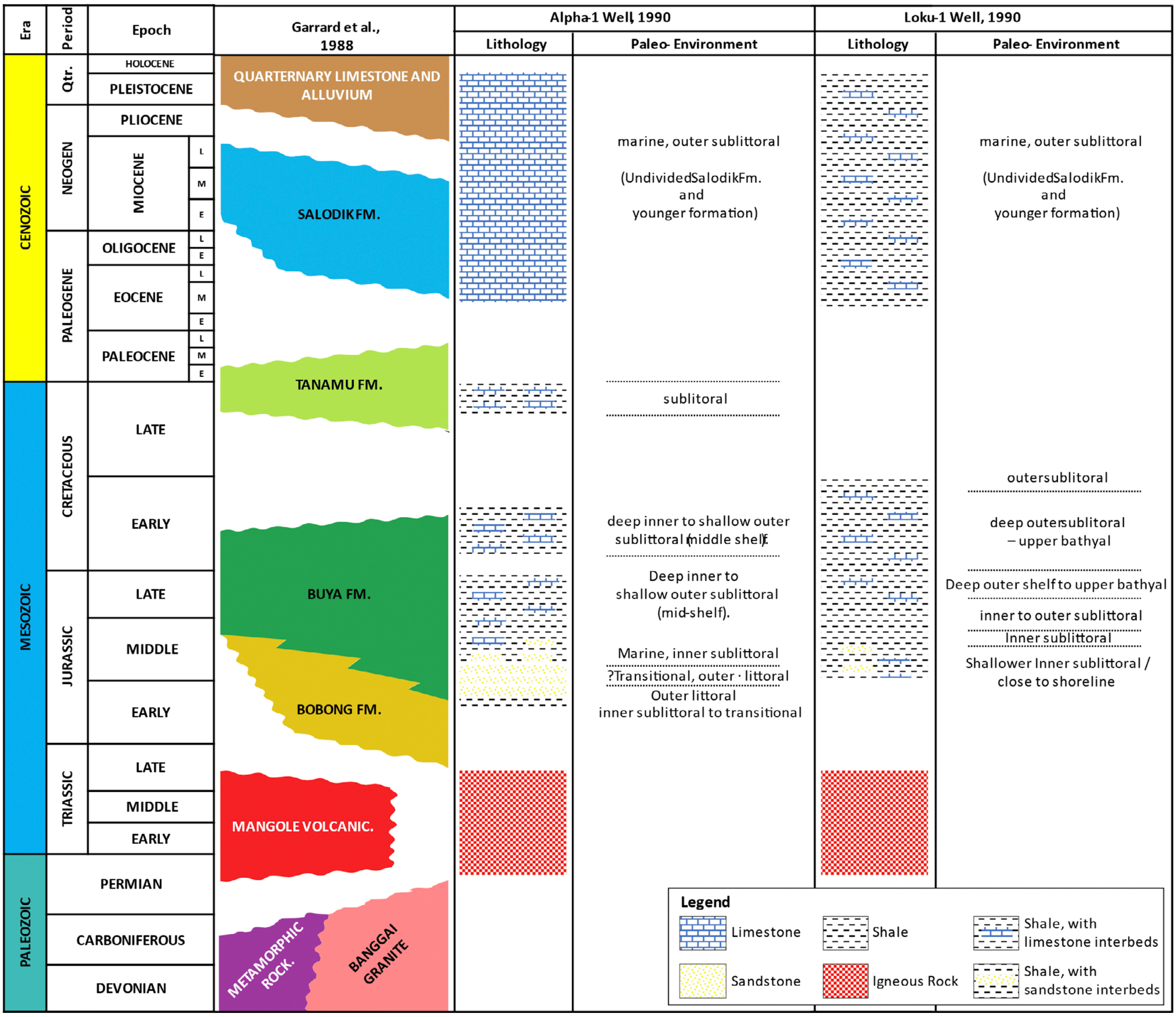

Figure 2: Regional chronostratigraphy of Taliabu-Mangole Archipelago (**Garrard et al., 1988**), including the well summary of Alpha-1 and Loku-1 (**Brown et al., 1990a, 1990b**)

Charles et al., 1972; Metcalfe, 1990; Simandjuntak and Barber, 1996). This island is considered to come from the northern boundary of the Australian Continent (**Klompé, 1954**). This separation is thought to have occurred during the Mesozoic or up to the Paleogene, and the island subsequently experienced displacement through the Great Sorong Fault due to the Philippine Sea Plate's motion (**McCaffrey et al., 1980, 1981; McCaffrey, 1982**).

According to **Garrard et al. (1988)** findings and the results obtained from the drilling of two wells, namely Alpha-1 and Loku-1, situated off the north and west coasts of the Mangole-Taliabu Archipelago (as depicted in **Figure 2**), it can be inferred that the Banggai-Sula microcontinent is covered by metamorphic, granite, and volcanic rocks. The Metamorphic Basement is comprised of lithologies such as slate, schist, and gneiss. It is postulated that the Metamorphic Basement deformation occurred in the Upper Paleozoic, and then Granite Banggai intruded this metamorphic rock over the Late Permian to the Early Triassic (**Pigram et al., 1985; Garrard et al., 1988; Supandjono and Haryono, 1993**). The Granite Banggai is comprised of a heterogeneous assemblage of rock varieties, encompassing red orthoclase-dominant granite, granodiorite, quartzdiorite, microdiorite, syenite porphyry, aplite, and pegmatite (**Garrard et al., 1988**). According to **Pigram et al. (1985)**, the absolute age of that intrusive rock ranges from the Late Permian to Triassic. The Mangole Volcanics Formation spans a vertical range of 1000 m and comprises various rock types, including rhyolite, dacite, ignimbrite, lithic tuff, and breccia. This formation is predominantly situated above water level, exhibiting well-defined layering, ranging in colour from reddish-brown to greyish-green, and often displaying signs of modification. **Sukamto and Simandjuntak (1983)** reported that the acid extrusives had yielded ages of 210 ± 25 Ma and 330 ± 90 Ma. These findings suggest that the extrusives were likely formed around the same time as the Banggai Granite, as noted by **Garrard et al. (1988)**. The early Jurassic Bobong Formation unconformably overlies the volcanic rock. The Bobong Formation comprises a combination of conglomerate and sandstone interlayered with shale and coal and locally contains pyrite nodules. The upper part of the Bobong Formation is interfingering with the lower Buya Formation, consisting of shale, mudstone, and calcareous claystone intercalated with fine quartz sandstone and calcarenite. Ammonite and Belemnite fossils indicate geological periods ranging from the Middle Jurassic to the Late Jurassic. In the Jurassic, a magmatic event formed the diabas dike, which intruded the Bobong and Buya formations. The Buya Formation is reported to be unconformably overlain by marly fossiliferous limestones of the Tanamu Formation (**Pigram et al., 1985; Garrard et al., 1988; Supandjono and Haryono, 1993**). Moreover, the Tanamu Formation is observed to be unconformably superimposed by the Salodik Formation of the Middle Eocene-Miocene era, characterised by the presence of alternating limestones and marls. Limestones and Alluvium sediment of the Quaternary age overlap unconformably across all pre-existing formation units (**Garrard et al., 1988**).

3. Data and Method

A total of 46 rock samples was collected from the outcrop of the Mesozoic Bobong, Buya, Tanamu, and Salodik formations in Mangole-Taliabu Archipelago of North Maluku, as well as six natural gas seepage samples (sampling locations are shown in **Figure 1**). It should be mentioned that even though the geological map (see **Figure 1**) suggest sample DI-04-1, DI-10, and DI-09 are taken from the Bobong Formation, based on lithological characteristics and paleontological data, the author classified them as representing the Buya Formation. All of these outcrop data were combined with the existing subsurface data from Alpha-1 well (**Brown et al., 1990a**) and Loku-1 well (**Brown et al., 1990**). The subsurface data consists of 29 sidewall core samples from the Alpha-1 well, as well as 86 sidewall cores and 20 ditch-cutting samples from the Loku-1 well. Total organic carbon (TOC) measurements and pyrolysis analysis were performed on each collected rock sample. Moreover, from all collected rock samples, 74 samples were measured for vitrinite reflectance (Ro) (see **Table 1**), and ten samples were selected for Gas Chromatography (GC) and Gas Chromatography-Mass Spectrometry (GC-MS) analysis (see **Table 2**). The dataset of natural gas seepage comprises six samples, three of which were gathered from BP Research's unpublished reports (RAG/88/012, RAG/88/014, and Binono Seep) (**Dee, 1989**), while the remaining three were recently collected by the Taliabu Oil and Gas Prospecting 2021 Team (TOGP 2021 Team), Center for Geological Survey. The six natural gas seepage samples were subjected to stable carbon isotope analysis (see **Table 4**), but only three samples were subjected to molecular compositions (see **Table 3**).

Potential source rock characterisation is generally shown by histograms of TOC, residual hydrocarbon potential (S2), Hydrogen Index (HI), and Tmax, where each parameter is grouped according to the categories of **Peters and Cassa (1994)**. A cross-plot of S2 versus TOC was utilised to identify the organic richness and generating potential (**Espitalie et al., 1985**). The S2 against TOC cross-plot was also used to identify the quality or organic matter type (**Langford and Blanc-Valleron, 1990**). Moreover, the cross-plot of HI against Tmax was applied to identify the type of organic matter and determine the maturity level. The determination of maturity level can also be achieved by utilising plots of Tmax and Ro data against its depth. Furthermore, GC and GC-MS data were used to identify the source of organic matter, namely the cross-plot of Pr/nC_{18} against Ph/nC_{17} (after **Shanmugam, 1985**), the cross-plot of C_{29}/C_{27} regular steranes against Pristane/Phytane ratio (Pr/Ph) (**Adegoke et al., 2014**), and the C_{27}-C_{29} steranes ternary diagram (**Huang and Meinschein, 1979**). Several cross-plots were used for the origin and genetics of natural gas seeps that several scholars have published, namely Schoell's Diagram (**Schoell, 1983**), the cross-plots for differentiating coal-derived gas and oil-associated gas (**Dai, 1992**), and the modified Bernard's Diagram (**Whiticar, 1994**). **Equation 1** for coal-derived gas and **Equation 2** for oil-type gas, obtained by **Schoell (1983),** was applied to estimate the calculated Ro based on the isotopic composition of methane. By comparing the calculated Ro with the measured Ro of the potential source rock, it is possible to infer that the correlation between natural gas and its source rock can be carried out.

$$\text{Coal-derived gas} \quad : \delta^{13}C_1 = 14\lg Ro - 28 \qquad (1)$$

$$\text{Oil-associated gas} \quad : \delta^{13}C_1 = 17\lg Ro - 42 \qquad (2)$$

Where:

$\delta^{13}C_1$ – the value of methane isotopic composition (‰),

Ro – the value of vitrinite reflectance (%).

4. Results and Discussion

This section presents the findings of this research. It includes the geochemical characteristics of potential source rocks, the origins and genetics of natural gas seepages, and the correlation between natural gas seepage and its source rock.

4.1. Source Rock

The description and interpretation of the Geochemical characteristics of each formation and their hydrocarbon potential are summarised in **Table 1**. They consist of

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Table 1: The geochemical characteristics of each formation in the research area and their hydrocarbon potential. Raw data can be seen in **Supplementary Material**.

Formation	**Bobong Formation**	**Buya Formation**	**Tanamu Formation**	**Salodik Formation**
Age	**E. Jura**	**Mid. Jura - E. Creta**	**Late Cretaceous**	**Tertiary**
Total Organic Carbon / TOC (wt%)	0.13 to 3.62 (13)	0.01 to 2.61 (161)	0.12 to 0.21 (5)	0.55 to 0.67 (2)
	0.84	0.73	0.17	0.61
Residual Hydrocarbon Potential / S2 (mg HC/g rock)	0.16 to 6.96 (13)	0.01 to 3.64 (157)	0.07 to 0.12 (2)	0.65 to 0.68 (2)
	1.16	0.90	0.10	0.67
Production index (S1/(S1+S2))	0.02 to 0.18 (13)	0.02 to 0.83 (157)	0.08 to 0.22 (2)	0.02 to 0.03 (2)
	0.08	0.12	0.15	0.03
Hydrogen index / HI (mg HC/g TOC)	53 to 203 (13)	7 to 375 (157)	39 to 57 (2)	97 to 124 (2)
	124.38	122.51	48.00	110.50
Kerogen type	III Gas Prone	III, II/III, IV Gas & Oil-Prone	III and IV None	III Gas Prone
Rock-Eval Tmax	433 to 443 (13)	303 to 559 (153)	423 (1)	426 to 426 (2)
	438.46	445.50		426.00
Vitrinite Reflectance	0.4 to 0.43 (2)	0.3 to 2.34 (71)	n/a	0.32 (1)
	0.42	1.01		
Maturity	Immature	Immature – Post Mature	Immature	Immature
Hydrocarbon potential	Fair - Good	Fair	Poor	Poor

Note: n.a. = not applicable. The numerator represents the geochemical parameters range. The denominator represents the average values. In the numerator, the number in parentheses represents the total number of outcrops and wells samples.

Mesozoic sediments of the Bobong, Buya, and Tanamu formations and the Tertiary Salodik Formation.

4.1.1. The Bobong Formation.

The Early Jurassic of the Bobong Formation (only from Alpha-1 well) was geochemically characterised from 13 samples (see **Table 1**). This formation exhibited a range of 0.13 to 3.62 wt% in TOC content, with an average amount of 0.84 wt% (see **Table 1**). It demonstrates poor to very good organic carbon content (see **Table 3**) (**Peters and Cassa, 1994**). The very good TOC content was collected from Alpha-1 well at 689 m and 720 m depths. The residual hydrocarbon potential (S2) of 13 rock samples was measured, yielding a range of 0.16 to 6.96 mg HC/g and an average value of 1.16 mg HC/g rock (see **Table 1**), indicating poor to good residual generation potential. The fair to good residual generation potential was also recorded from Alpha-1 well at depths 689 m and 720 m. Although Total Organic Carbon (TOC) is an index of the organic content present in source rock, it may not be an adequate indicator for accurately evaluating the quantity of organic matter (**Peters and Cassa, 1994; Dembicki, 2009, 2017**) because sediments contain a variety of organic matter. Particular organic matter can generate hydrocarbons, whereas others exhibit inertness (**Tissot et al., 1974**). Carbon must associate with hydrogen to generate hydrocarbon. The indicator of hydrogen content in organic matter is needed to assess organic matter richness. Hydrogen content can be estimated indirectly using Rock-Eval pyrolysis, namely using the value of residual hydrocarbon potential / S2 (**Dembicki, 2009**). As a result, the TOC value against S2 cross-plot is frequently used to assess organic matter richness. This cross-plot shows that from 13 samples, only two samples from depths of 689 m and 720 m indicate fair - good hydrocarbon-generating potential (see **Figure 4**).

The HI value of the Bobong Formation ranges from 53 to 203 mg HC/g TOC (see **Table 1, Figure 3**), which suggests type-III kerogen (gas-prone) dominates in this formation (**Peters and Cassa, 1994**). A Plot of TOC against residual hydrocarbon potential (see **Figure 5**) and Hydrogen index against Tmax temperature (**Figure 6 a**) also confirmed that the Bobong Formation is dominated by type-III kerogen (gas-prone). The values of the pristane/phytane ratio of samples from the Alpha-1 well at a depth of 720m (see **Table 2**) is greater than 1, which is indicative that the source rock was deposited in suboxic to oxic environment (**Didyk et al., 1978; Peters et al., 2005**). The cross-plot of the ratio pristane /nC_{17} against phytane /nC_{18} of source rock extract suggests that kerogen Type III terrestrial organic matter is the source input under oxidising depositional conditions (see **Figure 7a**). The cross-plot of C_{29}/C_{27} regular steranes against Pr/Ph (Adegoke et al., 2014) confirms the dominance of land plants as the primary organic matter of source rock samples in the suboxic to oxic environment (see **Figure 7b**). The ternary diagram of $C_{27\text{-}29}$ ster-

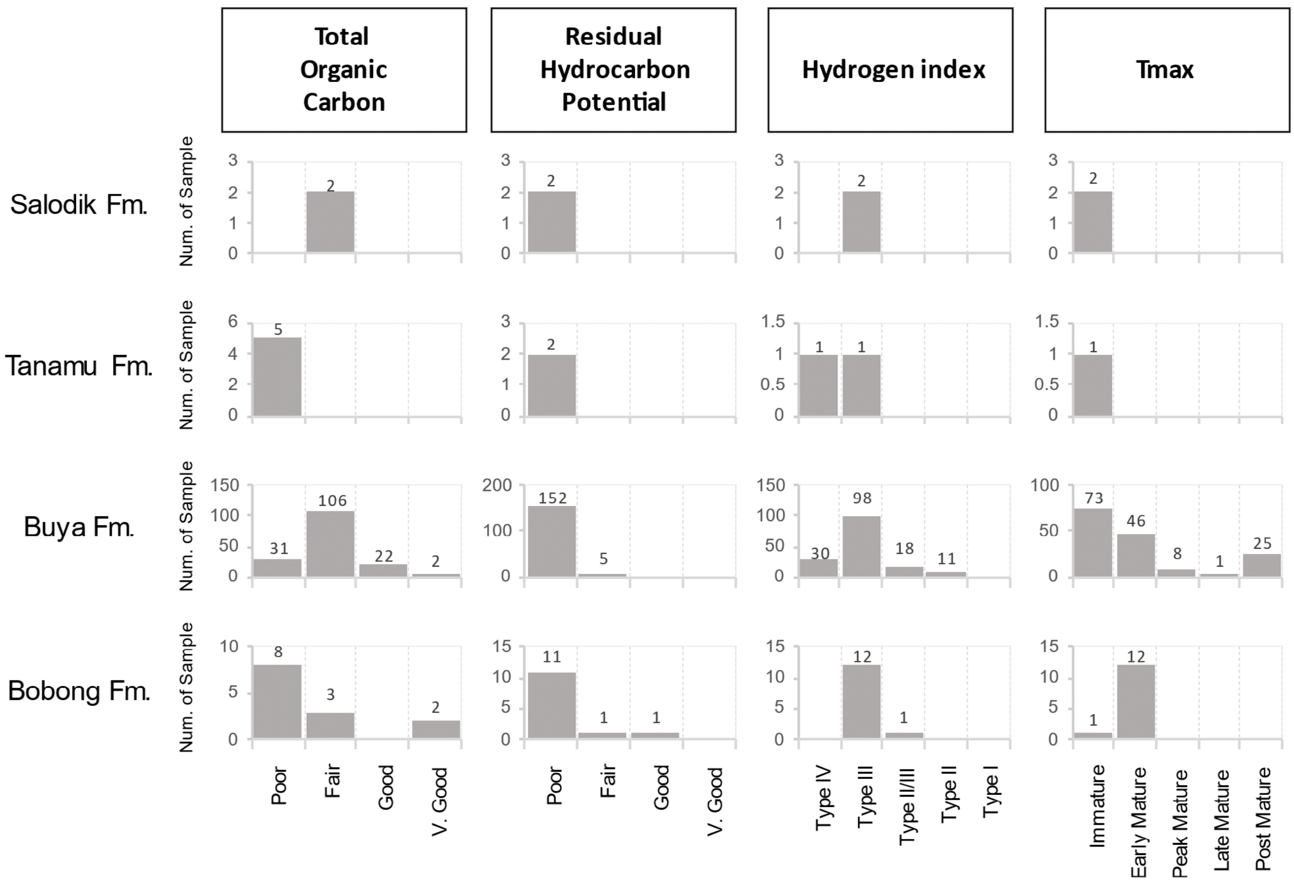

Figure 3: The histograms depicting the Total Organic Carbon, residual petroleum potential, Hydrogen Index, and Tmax of each formation were analysed. The categorisation of each parameter has been carried out following the classification system proposed by **Peters and Cassa (1994)**.

anes also indicates that organic matter originates from source facies deposited in a transition zone (estuarine or bay) (**Huang and Meinschein, 1979**).

The thermal maturation interpretation is based on Ro values of between 0.40 and 0.43% plus corresponding pyrolysis Tmax values of 433° to 443°C. Thermal Maturity parameters recorded through the Bobong Formation of the Alpha-I well suggest the sediments are immature. Although, based on Tmax data, the value > 430°C (see **Figure 6b**) is regarded as entering the maturity window (**Espitalie et al., 1985; Mukhopadhyay et al., 1995**), but higher Tmax values can occur due to chemical weathering/oxidation and kerogen reworking (**Copard et al., 2002; Yang and Horsfield, 2020**).

4.1.2. The Buya Formation.

The Middle Jurassic to Early Cretaceous, sediment rich in organic matter of the Buya Formation, was sampled and geochemically analysed from Alpha-1 well, Loku-1 well, and outcrop. This formation was geochemically characterised based on analyses of 161 samples, i.e. 14 samples from Alpha-1 wells, 104 samples from Loku-1 wells, and 43 samples from outcrop. Their capability as source rock potential is slightly under the Bobong Formation. The highest TOC content equals 2.61 wt%, and the average reaches 0.73 wt% (see **Table 1**). From the histogram of total organic carbon content (see **Figure 3**), most samples are fair source rock quality. The residual hydrocarbon potential (S2) exhibits a range of values between 0.01 to 3.64 mg HC/g rock, with an average of 0.9 mg HC/g rock (see **Table 1, Figure 5**). The TOC value against S2 cross-plot shows that almost all samples showed poor residual petroleum potential and only five samples showed fair residual petroleum potential (see **Figure 4**), namely from Alpha-1 well (406 m, 452 m, and 504 m) and Loku-1 well (2285.5 and 2386 m).

The Buya Formation has an HI value between 7 to 375 mg HC/g TOC, which means the results are wide-ranged, encompassing type-IV to type II/III of kerogen types (see **Table 1, Figure 3**). The cross-plot of TOC against S2 (see **Figure 5**) and HI against Tmax temperature (see **Figure 6a**) indicates that most of the Buya Formation, except the sample from outcrop, is characterised by an admixture of type-II and type-III kerogen. The findings of this research are consistent with prior observations (**Garrard et al., 1988**) that proved the presence of oil-prone kerogens in several outcrop samples, as confirmed by microscopic analysis. These kerogens are believed to generate significant quantities of oil when

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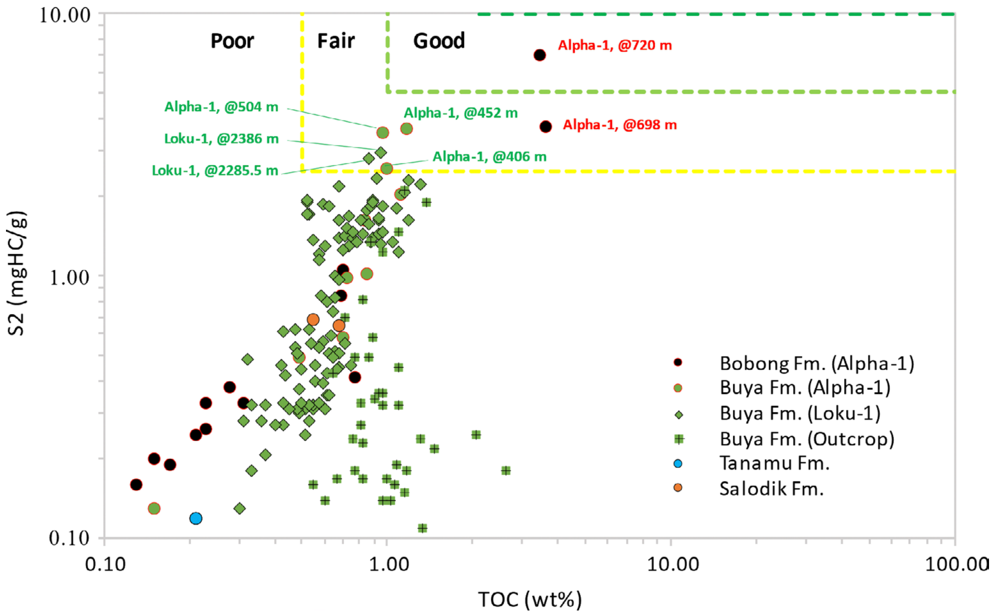

Figure 4: The cross-plot of S2 against TOC for assessing the quantity of organic matter (after **Espitalie et al., 1985**)

subjected to optimal maturation conditions. Furthermore, the pristane/phytane ratio values of the Buya Formation (as presented in **Table 2**) vary from 1.87 to 9.97, indicating that the deposition of the source rock was in a suboxic to oxic environment (**Didyk et al., 1978; Peters et al., 2005**), although samples of the high level of their maturity may impact DI-08-C and DI-04-1A's high values of this ratio (**ten Haven et al., 1987; Kotarba et al., 2014**). The cross-plot of the ratio pristane /nC_{17} against phytane /nC_{18} of source rock extract from all of the Buya Formation sample suggests that kerogen Type III derived from terrestrial organic matter is the source input under oxidising depositional conditions (see **Figure 7a**). The cross-plot of C_{29}/C_{27} regular steranes against Pr/Ph (**Adegoke et al., 2014**) also indicates that all samples of the Buya Formation were deposited in a suboxic to oxic environment with input of organic matter from land plants and algae-water bacteria (see **Figure 7b**). The land plant matter is the primary organic matter input of the Bobong Formation in the Alpha-1; meanwhile, algae-water bacteria matter is the primary organic matter input in Loku-1 well, with land plant matter as minor input. The veracity of this claim is corroborated by the ternary cross-plot diagram of the $C_{27\text{-}29}$ steranes (see **Figure 7c**) (**Huang and Meinschein, 1979**).

In general, based on Tmax (303 to 559°C) and Ro (0.3 to 2.34%Ro) data as well as production index (0.02 to 0.83), samples of the Buya Formation from Alpha-1 well, Loku-1 well, and from outcrop data, indicate a wide-ranged thermal maturity level, encompassing immature to post mature (see **Table 1, Figure 6**). Most of the samples obtained from the outcrop exhibit post-mature characteristics (see **Figure 6a**). The observed phenomenon may be attributed to potential intrusion or the possibility that said samples had been buried at a significant depth but have since been cropped out (**Dembicki, 2017; Dow, 1977**). At the Alpha-1 well, the range of Tmax and Ro data is 425 to 431°C and 0.3 to 0.43 %, respectively, indicating this formation is thermally immature for hydrocarbon generation. This is corroborated by a low PI value (<0.1), indicating a poor kerogen conversion to liquid hydrocarbons. However, several samples from the Alpha-1 well at depth 655.5 m, 660 m, 675 m, 677 m, and 679 m, where the PI value range between 0.28 to 0.83, is considered anomalously high. This is due to the poor recovery of pyrolysate (S2) relative to the yield of free bitumen (S1) (see **Table 1**). Maturity parameters recorded for the Buya Formation in Loku-1 suggest the Middle Jurassic-Early Cretaceous sediments are thermally immature to marginally mature. The Tmax values span between 427 to 448°C, indicating that they have not yet reached the level of significant hydrocarbon generation or expulsion. Measured Ro data (0.32 to 0.74%) displays a progressive increase with depth through the interval (see **Figure 6b**). The range of values recorded generally agrees with the interpreted level of maturation, although some reworking of the vitrinite particles is identified. Higher Tmax values are also identified at the upper part of the Buya Formation in the Loku-1 well, which can be assigned to chemical weathering/oxidation and kerogen reworking (**Copard et al., 2002; Yang and Horsfield, 2020**).

4.1.3. The Tanamu Formation.

The geochemical characteristics of the Tanamu Formation were solely determined through pyrolysis analysis data, as presented in **Table 1**, **Figure 3**, and **Figure 5**. The Tanamu Formation exhibits a predominantly low total organic carbon (TOC) value, varying from 0.12 to 0.21 wt%. The residual hydrocarbon potential (S2) is low, with the highest observed measurement being 0.12

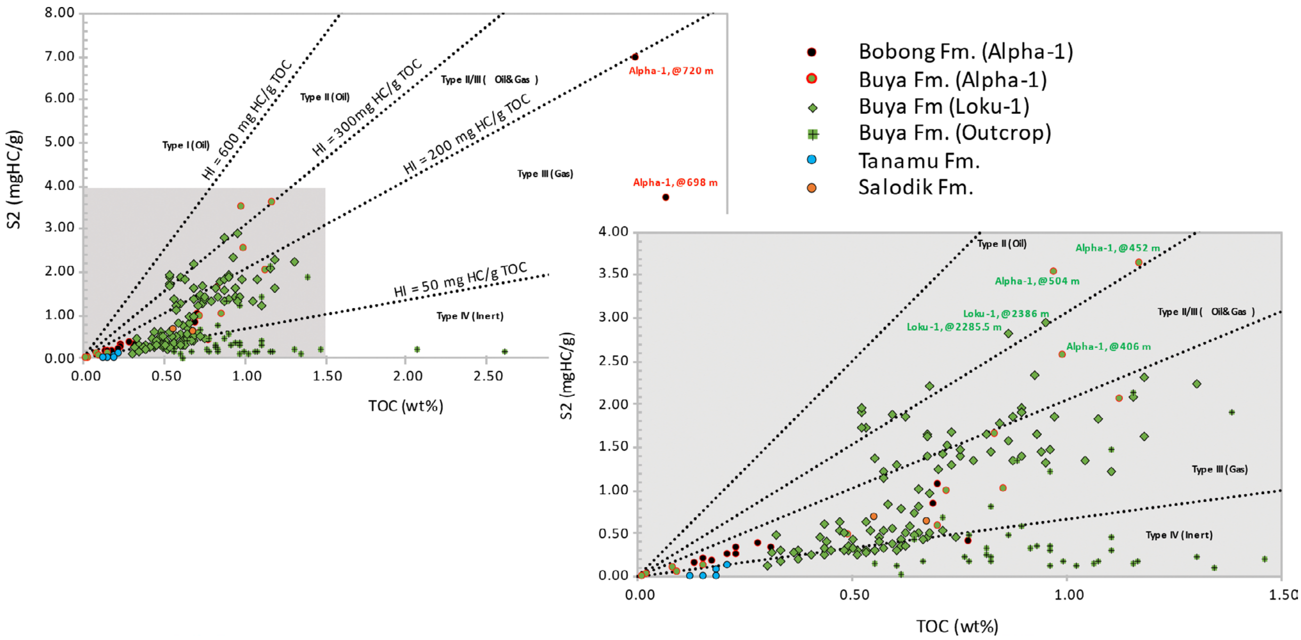

Figure 5: The cross-plot of S2 versus TOC to determine the kerogen type of organic matter, according to **Langford and Blanc-Valleron (1990)**

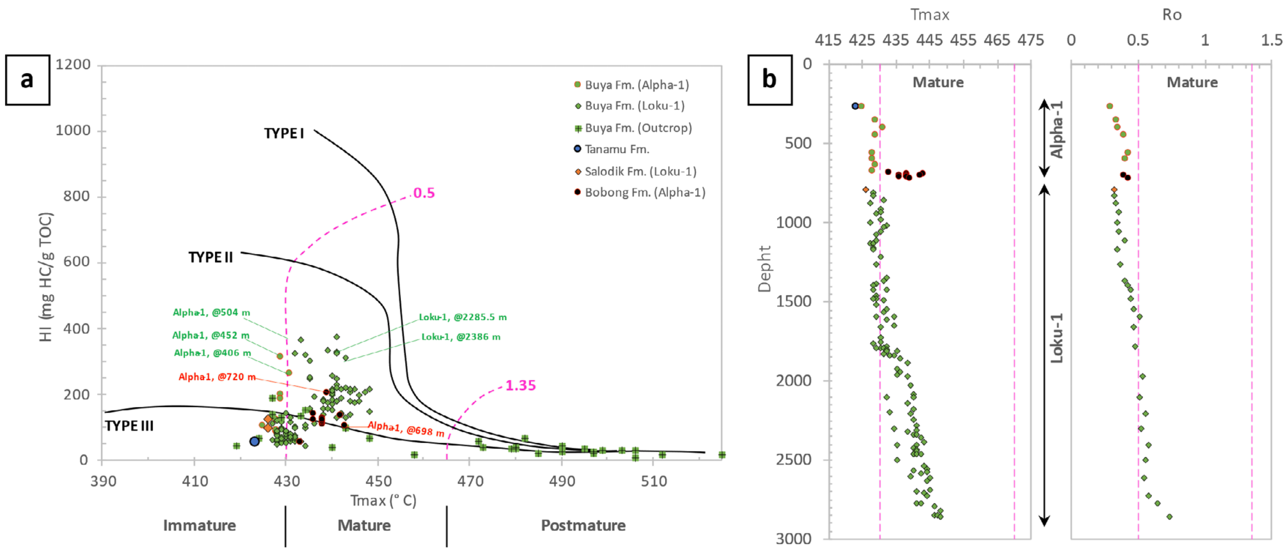

Figure 6: a) The cross-plot of HI against Tmax reflects the source rock thermal maturity (**Mukhopadhyay et al., 1995**), b) the Plot of Tmax and %Ro against depth.

mg HC/g rock. The hydrogen index of two samples from the Alpha-1 well with a range between 39 to 57 mg HC/g TOC (see **Table 1, Figure 3**) suggests that samples contain type IV kerogen (no hydrocarbon-generating capacity). Moreover, a cross-plot S2 against TOC also confirms that inference. Based on the pyrolysis Tmax and production index (below 435°C and 0.1), the thermal maturity of the Tanamu Formation penetrated by the Alpha well is interpreted as being thermally immature. Although there is a production index value reaching 0.22 in well Alpha-1 at a depth of 262.5 m, this is due to the poor recovery of pyrolysate (S2) relative to the yield of free bitumen (S1), which is not significant (see **Table 1**), which was identified in the Buya Formation in the Alpha-1 well. Thus, the Tanamu Formation does not have the capability as a source rock in the research area. The findings presented herein agree with the conclusions provided by **Garrard et al. (1988)**, who revealed that the Tanamu Formation displays a low organic matter content and lacks the capacity to generate hydrocarbon.

4.1.4. The Salodik Formation and Younger.

The geochemical characteristics of organic matter in the Salodik Formation were conducted solely through the pyrolysis analysis data, similar to the approach taken

Table 2: Gas Chromatography (GC) and Gas Chromatography-Mass Spectrometry (GC-MS) data for the Bobong and Buya formations

Well/Outcrop	Depth (m) / No Sample	Formation	N-alkanes & isoprenoids			Steranes			
			Pr/Ph	Pr/nC_{17}	Ph/nC_{18}	C_{27}	C_{28}	C_{29}	C_{29}/C_{27}
Alpha-1	406	Buya Fm.	1.87	0.8	0.33	38.11	17.68	44.21	1.16
Alpha-1	452	Buya Fm.	2.05	1.01	0.51	19.35	24.19	56.45	2.92
Alpha-1	504	Buya Fm.	1. 93	0.84	0.35	41.23	18.42	40.35	0.98
Loku-1	2099.5	Buya Fm.	2.45	0.79	0.33	30.72	21.5	47.78	1.56
Loku-2	2386	Buya Fm.	3.82	0.83	0.22	48.90	14.29	36.81	0.75
Loku-3	2421.5	Buya Fm.	3.56	0.7	0.21	49.73	15.3	34.97	0.70
Loku-4	2858	Buya Fm.	3.33	0.43	0.13	53.59	15.69	30.72	0.57
Outcrop	DI-08-C	Buya Fm.	9.97	3.53	0.55	44.06	15.38	40.56	0.92
Outcrop	DI-04-1A	Buya Fm.	6.05	7.64	0.84	51.31	14.12	34.57	0.67
Alpha-1	720	Bobong Fm.	2.16	0.93	0.34	35.54	21.9	42.56	1.20

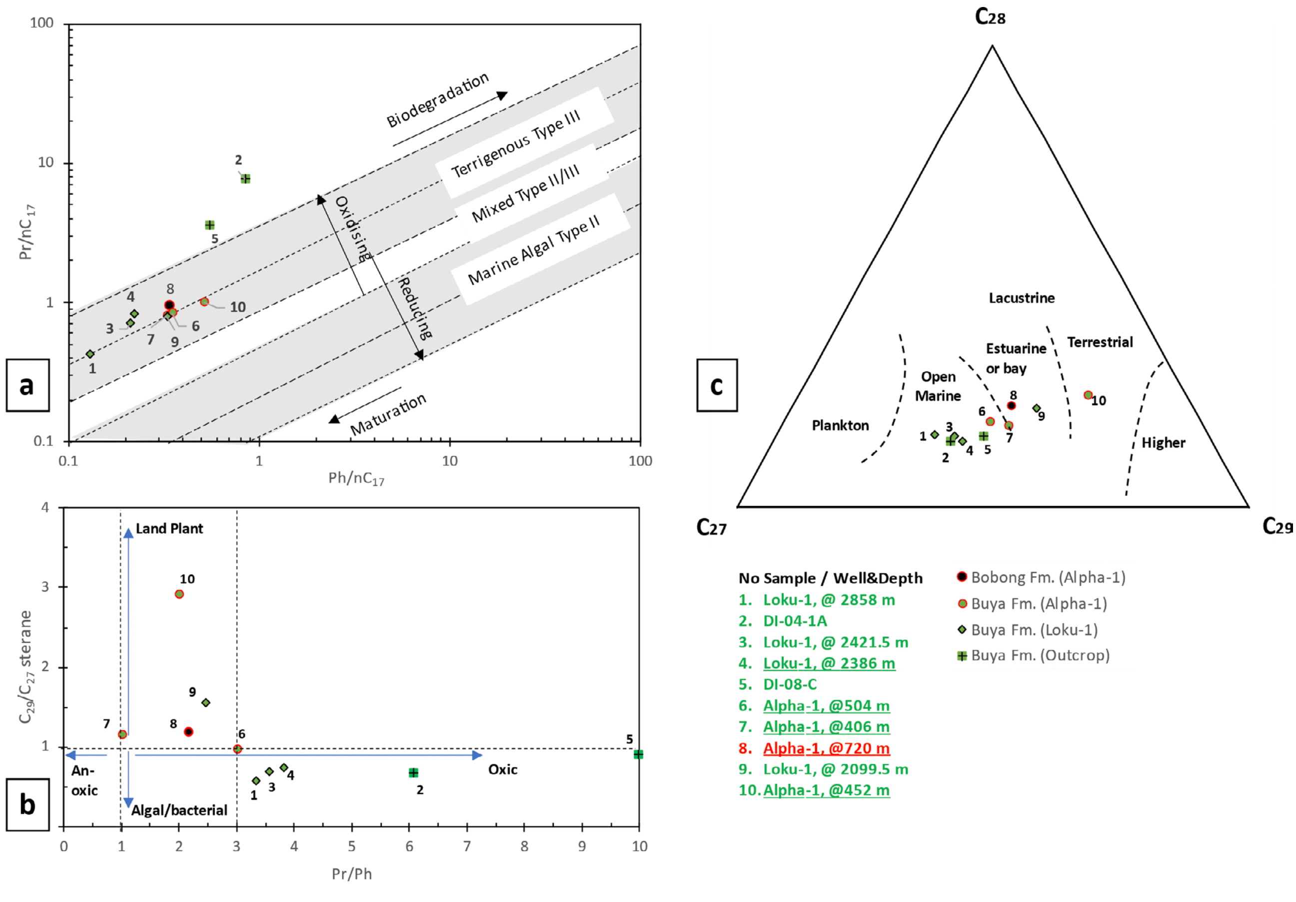

Figure 7: a) Cross-plot of Pr/nC_{18} against Ph/nC_{17} (after **Shanmugam,1985**), b) Cross-plot of C_{29}/C_{27} regular steranes against Pr/Ph (**Adegoke et al., 2014**), c) Ternary diagram of C_{27}-C_{29} steranes (**Huang and Meinschein, 1979**).

with the Tanamu Formation. Two claystones analysed from Loku-1 well (789m and 792m) show fair organic richness with a TOC range of 0.55 to 0.67 wt% and low value of residual petroleum potential (S2< 2.5 mg HC/g rock), indicating poor source potential (see **Table 1, Figure 3, Figure 5**). Hydrogen Index (HI) of 97 mg HC/g TOC and 124 mg HC/g TOC indicate the samples are type III kerogen (gas-prone) (see **Table 1, Figure 3**). Moreover, the cross-plot of S2 against TOC confirms that inference (see **Figure 5, Figure 6**). According to the Tmax and Ro values, the maturity level of this formation can be considered immature (see **Figure 6**). Based on the findings of the geochemical analysis carried out on the Salodik Formation, it can be inferred that the forma-

Table 3: The molecular composition of natural gas seepages from the Taliabu-Mangole Archipelago

Sample code	Component (Mol%)											Dryness
	Air	CO_2	N_2	CH_4	C_2H_6	C_3H_8	iC_4H_{10}	nC_4H_{10}	iC_5H_{12}	nC_5H_{12}	C_6H_{14}	C_1/C_{1-5}
RAG/88/012	8.68	1.03	3.06	82.08	3.20	1.22	0.19	0.30	0.10	0.05	0.04	0.94
RAG/88/014	1.86	3.05	0.96	89.45	2.89	1.12	0.17	0.23	0.10	0.04	0.05	0.95
Binono Seep	-	2.83	35.43	59.72	1.35	0.52	0.08	0.05	-	-	-	0.97
RA/21/33	-	-	-	-	-	-	-	-	-	-	-	-
RA/21/34	-	-	-	-	-	-	-	-	-	-	-	-
RA/21/35	-	-	-	-	-	-	-	-	-	-	-	-

Table 4: Isotopic composition and value of calculated Ro for each natural gas seepage from the Taliabu-Mangole Archipelago

Sample code	Stable isotopes (‰)				Calculated %Ro (Schoell., 1983)
	$\delta^{13}C(CH_4)$	$\delta^{13}C\ (C_2H_6)$	$\delta^{13}C\ (C_3H_8)$	$\delta^{13}C\ (CO_2)$	
RAG/88/012	-27.98	-26.55	-	-40.85	1.00
RAG/88/014	-29.32	-23.98	-	-39.41	0.80
Binono Seep	-40.00	-29.40	-31.30	-34.70	1.31
RA/21/33	-49.10	-45.70	-13.80	-28.30	0.50
RA/21/34	-45.70	-46.60	-9.00	-26.80	0.61
RA/21/35	-46.40	-	-	-34.60	0.55

tion does not possess the characteristics of a potential source rock within the research area. The present findings contradict the results proclaimed by **Garrard et al. (1988)**, **Davies (1990)**, and **Hasanusi et al. (2004)**, who reported that the Miocene marine shales and coals present in the Tomori and Matindok formations of the Salodik Group exhibit excellent potential as source rocks due to their substantial organic matter content.

4.2. Natural Gas Seepage

The following section provides the geochemical characteristics, origins, and genetics of natural gas seepages. This evaluation is based on molecular and isotopic composition data.

4.2.1. Geochemical characteristics

The molecular composition of natural gas seepage in the research area is shown in **Table 3**. Only three of six gas seepage samples were subjected to gas composition analysis: RAG/88/012, RAG/88/014, and Binono Seep. The gaseous samples comprise alkane (C_{1-n}) and nonhydrocarbon gas, as presented in **Table 3**. The alkane gases comprise CH_4 and C_{2-n} components, with a range of 59.72% - 89.45% and 2% - 5.15%, respectively. The category of nonhydrocarbon gases encompasses various gases such as air, which ranges from 1.86% to 8.68%; CO_2, which ranges from 1.03% to 3.05%; and N_2, which ranges from 0.96% to 35.43%. According to **Schoell (1980)**, the gas dryness coefficient (C_1/C_{1-n}) range suggests that the gases are predominantly wet (see **Table 3**).

The carbon isotope composition of natural gas seepage is presented in **Table 4**. The range of $\delta^{13}C(CH_4)$ values observed is between -49.10‰ and -27.98‰, whereas the range $\delta^{13}C(C_2H_6)$ values is between -46.60‰ and -23.98‰. Furthermore, the range of $\delta^{13}C(C_3H_8)$ values is from -31.30‰ to -9.00‰, while the range of $\delta^{13}C(CO_2)$ values is from -40.85‰ to -26.80‰. However, it was noted that there was a partial reversal trend in carbon isotope values in two samples, namely Binono Seep and RA/21/34. Specifically, in Binono Seep, the $\delta^{13}C(C_2H_6)$ value was found to be greater than the $\delta^{13}C(C_3H_8)$ value, while in RA/21/34, the $\delta^{13}C(CH_4)$ value was greater than the $\delta^{13}C(C_2H_6)$ value (refer to **Table 4** for further details).

4.2.2. Origins and Genetic

Several studies have demonstrated that the utilisation of gas chemical composition and stable carbon isotopes analysis can be a valuable method for figuring out the origin and genetics of natural gas seepage (**Schoell, 1980, 1983, 1984; Whiticar, 1994; Dai, 1992; Dai et al., 2004, 2014, 2022; Kotarba et al., 2009, 2014; Liu et al., 2018, 2019; Milkov and Etiope, 2018; Zhang et al., 2018; Z. Xu et al., 2019; W. Xu et al., 2022**). Thermogenic, microbial, and abiogenic (inorganic) gas are the three main types of natural gas (**Whiticar, 1994; Sherwood et al., 2013**). The distinguishing characteris-

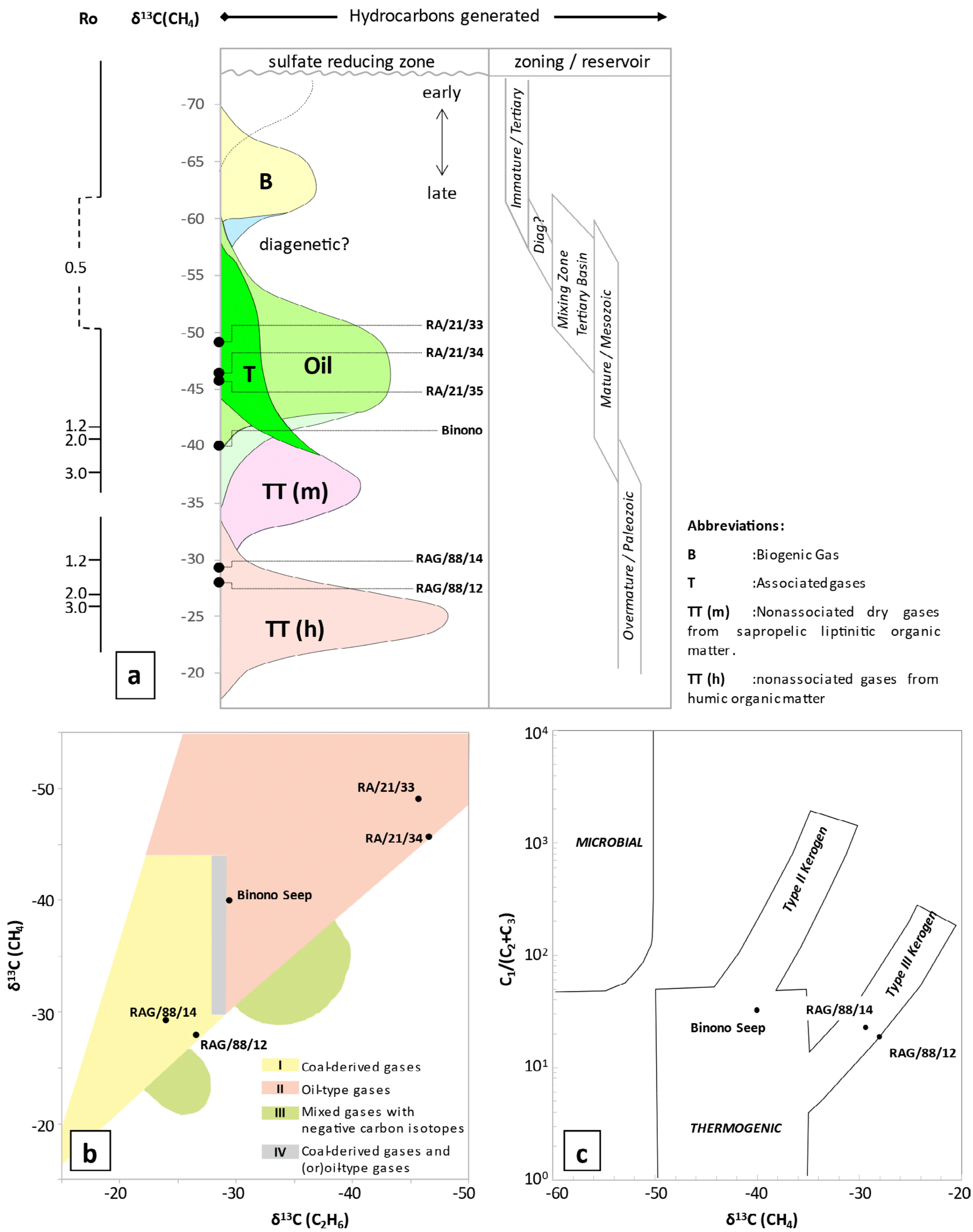

Figure 8: Origin and genetic characterisation of natural gas seepages using several cross-plots, a) Schoell's Diagram (**Schoell, 1983**), b) the cross-plots for distinguishing coal-derived gas and oil-associated gas (**Dai, 1992**), and c) the modified Bernard's Diagram (**Whiticar, 1994**).

tic of thermogenic gas is its normal carbon isotopic distribution pattern (following the order of $\delta^{13}C(CH_4) < \delta^{13}C(C_2H_6) < \delta^{13}C(C_3H_8)$) and the value of $\delta^{13}C(CH_4)$ exhibits a range of ~ -55‰ to -20‰ (**Dai et al., 2004; Galimov, 2006; Sherwood et al., 2013**). Whereas microbial gas shows a significant decline in $\delta^{13}C(CH_4)$ values (below -60‰), and the composition of C_{2+} alkane molecules is less than 1% (**Barker & Fritz, 1981; Sherwood et al., 2013; Whiticar et al., 1986**). The identification of the inorganic type of natural gas can be inferred by a reversal carbon isotopic distribution pattern ($\delta^{13}C(CH_4) > \delta^{13}C(C_2H_6) > \delta^{13}C(C_3H_8)$), as previously proposed by **Dai et al. (2004)** and **Galimov (2006)**. Furthermore, as **Dai et al. (1996)** proposed, natural gas's origin can also be identified by examining CO_2 concentration and $\delta^{13}C(CO_2)$ values. The content of CO_2 in organic natural gas is lower than 15%, and its $\delta^{13}C(CO_2)$ value is lower than -10‰. Conversely, inorganic natural gas exhibits a higher CO_2 content and $\delta^{13}C(CO_2)$ value greater than 60% and -8‰, respectively.

This research reveals that the $\delta^{13}C(CH_4)$ value of natural gas seepage ranges from -49.10‰ to -27.98‰, with a C_{2+} alkane content exceeding 1%. Subsequently, the CO_2 content and the carbon isotope ratio of the CO_2 exhibit values below 15% and -10‰, respectively. Furthermore, the distribution pattern of carbon isotope exhibited by the natural gas seepage indicates typical normal patterns, confirming that gas is primarily thermogenic gas derived from organic sources. The partially reversed alkane carbon isotope trend shown by the Binono Seep and RA/21/34 samples can be associated with mixing gases from various sources and maturation, secondary alteration, bacterial oxidation, and thermal sulfate reduction/TSR (**Dai et al., 2004; Hao et al., 2008; Liu et al. 2018; Quan et al., 2019**). Moreover, this trend reversal, particularly in the case of RA/21/34, where $\delta^{13}C(CH_4)$ is slightly greater than $\delta^{13}C\ (C_2H_6)$, can be associated with the increased pore pressure in the n-alkane system, as discussed by **Galimov (2006)**. This increase has the potential to affect the chemical equilibrium towards the higher analogues and possibly could lead to the partial transformation of C_1, resulting in an increase ^{13}C isotope in methane and a decrease in ethane ($\delta^{13}C(CH_4) > \delta^{13}C(C_2H_6)$). Additionally, it might be worthwhile considering the suggestion of **Sandvik et al. (1992)** that a delay of expulsion due to lower Hydrogen Index might lead to an abnormal increase in pore pressure and subsequent impact on the isotope distribution.

According to the data plotting shown on Schoell's diagram (see **Figure 8a**), it has been observed that the genetics of natural gas seepage can be classified into two distinct types. These categories include associated gases (Binono Seep, RA/21/33, RA/21/34, and RA/21/35), as well as non-associated gases (RAG/88/012 and RAG/88/014), which are derived from humic organic matter. According to **Schoell (1983)**, "associated gas" refers to the gas obtained during the cracking of petroleum, whereas "non-associated gas" is obtained mainly during the cracking of kerogen. However, if kerogen and petroleum undergo significant burial, the thermal alteration within a post-mature setting could result in dry non-associated gases forming from both (**Schoell, 1983**).

Furthermore, based on the $\delta^{13}C(CH_4)$ - $\delta^{13}C(C_2H_6)$ cross-plots (see **Figure 8b**), which are based on the extensive empirical research by Dai (**Dai, 1992; Dai et al., 2014**), it was revealed that the gases in the research area can be classified into two distinct types. It was found that the RAG/88/012 and RAG/88/014 gas samples appeared in a zone of coal-derived gases that were generated from type III kerogen. Meanwhile, the Binono Seep, RA/ 21/33, and RA/21/34 gas samples belong to oil-type gas zones.

The dryness coefficients ($C_1/(C_2 + C_3)$) against $\delta^{13}C(CH_4)$ cross-plot, Bernard's Diagram (**Whiticar, 1994**), were also employed to determine the genetics of natural gas. This diagram can distinguish kerogen type and thermal maturity (**Xu et al., 2019**). Due to not all samples having chemical composition data, only RAG/88/012, RAG/88/014, and Binono Seep data are plotted in the Bernard diagram. As depicted in **Figure 8c**, all samples belong to thermogenic gas. The Binono Seep source mixes type II and type III kerogens, whereas the sources of RAG/88/012 and RAG/88/014 seepage are predominantly composed of type III kerogens (see **Figure 8c**).

Consequently, natural gas seepage in the Mangole-Taliabu Archipelago is of organic and thermogenic origin. RAG/88/012 and RAG/88/014 gases are coal-derived from humic organic matter (type III kerogen). In contrast, Binono Seep, RA/21/33, RA/21/34, and RA/21/35 are oil-type gases derived from source rocks containing a mixture of type II and III kerogen (see **Figure 8b**). Thus, in addition to corroborating the previously reported genetic gas type documented in **Garrard et al. (1988)** and **Satyana and Zaitun (2016)**, this research has also unveiled another genetic gas type.

4.3. Gas–Source Rock Correlation

To comprehensively understand the relationship between potential source rocks and active natural gas seepages within the research area, an approach was performed by comparing the genetic characteristics of gas seepages with the kerogen type and maturation of potential source rocks. This approach was performed because the natural gas source is tightly related to its source rock (**Zhou et al., 2016**).

According to the organic matter type, the Bobong Formation sediments from the Alpha-1 well, having organic matter dominated by type III kerogen, are the most likely source for coal-derived gases derived from humic organic matter (RAG/88/012 and RAG/88/014). Meanwhile, the organic matter of the Buya Formation from the Loku-1 well is mainly composed of an admixture of type-II and type-III kerogen, which tend to generate oil and gas, is most likely the source rock for oil-type gases derived from a mixture of type II and III kerogen.

Isotope ^{13}C of methane ($\delta^{13}C(CH_4)$) is susceptible to variations with maturity (**Stahl, 1974, 1977; Stahl and Carey, 1975**). Therefore, the $\delta^{13}C(CH_4)$ value in natural gas allows the identification of their source rocks by determining the maturity of the organic matter (**Stahl & Carey, 1975**). This approach has been extensively employed in correlating gas to sources rock (**Stahl & Carey, 1975; Chen et al., 2021; Jian et al., 2021; Tian et al., 2021; Feng et al., 2023**). In order to estimate the calculated Ro, this research employed **Equation 1** and **Equation 2**, which were established by **Schoell (1983)** based on the published data from **Stahl and Carey**

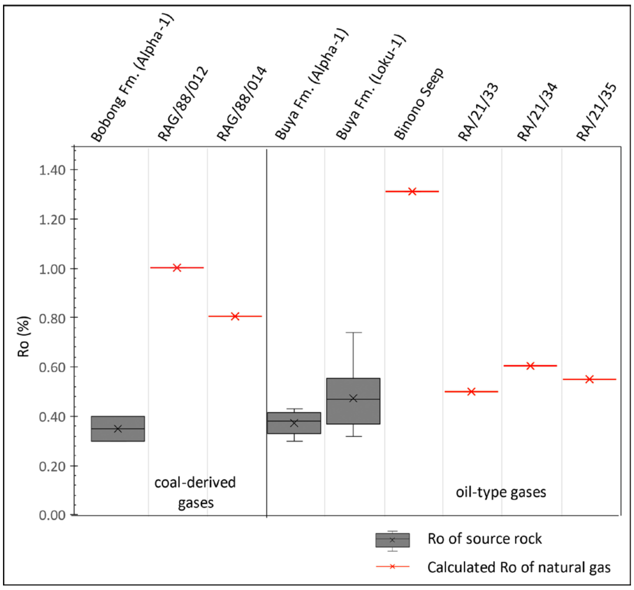

Figure 9: The box plot shows the comparison between the measured Ro of potential source rock with the calculated Ro of natural gas seepage based on isotope ^{13}C of methane.

(1975) and **Stahl et al. (1977)**. The decision to use these equations was based on their relevance to the scenario of hydrocarbon generation from reburied source rock that followed extensive uplift and erosion (**Chen et al., 2021**). According to **Garrard et al. (1988)**, there is indeed evidence of regional unconformity events in the Paleogene due to the uplift and erosion of Mesozoic sediment.

Equation 1 was applied to determine the calculated Ro value of coal-type gas samples. From the results of this calculation, it is known that the Ro values for RAG/88/012 and RAG/88/014 are 1 % and 0.80 %, respectively (see **Table 4**). In contrast, the maturation of potential source rocks from the Bobong Formation only ranges from 0.40% Ro to 0.43% Ro, indicating that the Bobong Formation is insufficiently mature to produce gas. Consequently, it can be concluded that the gas seepages found at RAG/88/012 and RAG/88/014 were not derived from the Bobong Formation in the Alpha-1 well (see **Figure 9**). The coal-derived gases may have been generated from the Bobong Formation at greater depth and maturation levels near the seepage locations. Previously, **Watkinson (2011)** and **Surjono et al. (2019)** suggested that the Bobong Formation extends into the offshore area north of the Mangolle-Taliabu Archipelago.

Furthermore, **Equation 2** was employed to calculate the Ro value of gas from oil-generating source rock. The calculated Ro of gas sample from the Binono Seep, RA/21/33, RA/21/34, and RA/21/35 are 1.31 %Ro, 0.50 %Ro, 0.61 %Ro, and 0.55 %Ro, respectively (see **Table 4**). As mentioned, the Buya Formation maturity value in the Loku-1 well ranges between 0.32 %Ro and 0.74 %Ro. Thus, the calculated Ro of the oil-generating source rock gas, except for the Binono Seep, coincides with the measured Ro of the Buya Formation in the Loku-1 well (see **Figure 9**), suggesting that the gas may originate from the Buya Formation in the Loku-1 well. Meanwhile, the source for the Binono Seep is also believed to derive from the Buya Formation, but not from the part penetrated by the Loku-1 well, yet from the parts of the Buya Formation at a deeper depth and higher maturity level. Namely, the calculated Ro based on isotope ^{13}C of methane from the Binono Seep is greater than the Ro range of the Buya Formation in Loku-1 well (see **Figure 9**). Nevertheless, according to the interpretation of **Watkinson (2011)** and **Surjono et al. (2019)**, the Buya Formation deepens toward the north from the Binono Seep, where it possibly reaches an adequate maturity level.

5. Conclusions

The results obtained from conducting organic geochemical analyses on potential source rocks, along with the examination of the molecular and isotopic composition of natural gas seepages in the Taliabu-Mangole Islands area, show that:

1. The potential of the Bobong Formation as source rock is considered to be fair to good, as evidenced by its total organic carbon content (TOC) of up to 3.62 wt%, the range of residual hydrocarbon potential between 0.16 to 6.96 mg HC/g rock, and type-III kerogen composition, which is known to be gas-prone. The available data from Alpha-1 well suggests that the formation is still an immature source rock. However, to the east of the Alpha-1 well, near the gas seepage, the Bobong Formation is at greater depth and maturity.
2. The Buya Formation has variable hydrocarbon potential, from poor to fair. The maximum value of the total organic carbon content (TOC) is 2.61 wt%, whereas the residual hydrocarbon potential (S2) spans from 0.01 to 3.64 mg HC/g rock. Admixture of type II and type III kerogen dominates in the Buya Formation. The organic matter maturity of the Buya Formation varies with depth, namely immature – post-mature conditions.
3. The Tanamu Formation and the Salodik Formation have no potential as source rocks in the research area.
4. The origin of all natural gas seepages is organic and thermogenic.
5. Two distinct types of natural gas seepage were found in the research area, one composed of coal-derived gas that derived from humic organic matter of type-III kerogen (RAG/88/012 and RAG/88/014) and the other composed of oil-type gases that derived from mixed-sources of type II and type III kerogen (Binono Seep, RA/21/33, RA/21/34 and RA/21/35).

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6. The Bobong Formation, which has a greater maturity level than that in the Alpha-1 well, looks to be the most likely source for the gas samples RAG/88/012 and RAG/88/014.
7. The Buya Formation, which was penetrated by the Loku-1 well, is the source for RA/21/33, RA/21/34, and RA/21/35. Meanwhile, the source for Binono Seep is also from the Buya Formation but at a deeper depth and higher maturity level.

As an additional point, it is necessary to acknowledge that certain limitations may exist in the present research and require further research to address them. The first shortcoming of the research is that there is no gas composition measurement for samples RA/21/33, RA/21/34, and RA/21/35, which was attributed to insufficient sample volume. Therefore, in interpreting the origin and genetic migration of gas, it is assumed that these samples exhibit a gas composition that is relatively similar to that of the Binono Seep sample. Conducting the molecular composition measurement of those unmeasured natural gas seepage samples is needed to address that. Another shortcoming is that the gas-source rock correlation mostly based on indications or indirect evidence. It is essential to conduct 3D basin modelling to comprehensively depict the distribution and maturity model of potential source rocks, as well as to simulate the generation, migration, and accumulation of hydrocarbons in the research area. The identified faults should also be investigated as possible pathways for natural gas to migrate from the kitchen area to either accumulate into closures or seep onto the surface. So, this further research is expected to validate the preliminary results of gas-source rock correlation in this present research.

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References

Adegoke, A.K., Abdullah, W.H., Hakimi, M.H. and Yandoka, B.M.S. (2014): Geochemical characterisation of Fika Formation in the Chad (Bornu) Basin, northeastern Nigeria: Implications for depositional environment and tectonic setting. Applied Geochemistry, 43, 1–12. https://doi.org/10.1016/j.apgeochem.2014.01.008

Audley-Charles, M.G., Carter, D.J. and Milsom, JS (1972): Tectonic Development of Eastern Indonesia in Relation to Gondwanaland Dispersal. Nature Physical Science, 239 (90), 35–39. https://doi.org/10.1038/physci239035a0

Barker, J. F. and Fritz, P. (1981): The occurrence and origin of methane in some groundwater flow systems. Canadian Journal of Earth Sciences, 18(12), 1802–1816. https://doi.org/10.1139/e81-168

Brown, S., Frame, P., Quidayan, V., Rumsari, W. and Harrington, J. (1990a): Biostratigraphy and Geochemistry of The Alpha-1A Well, Offshore Sula Island, Indonesia, Inc. Volume I. Core Laboratories, File No.: GSI-90148.

Brown, S., Frame, P., Quidayan, V., Rumsari, W. and Harrington, J. (1990b): Biostratigraphy and Geochemistry of The Loku-1 Well, Offshore Sula Island, Indonesia, Inc. Volume I and II. Core Laboratories, File No.: GSI-90059.

Chen, J., Wang, X., Chen, J., Ni, Y., Xiang, B., Liao, F., He, W., Yao, L. and Li, E. (2021): New equation to decipher the relationship between carbon isotopic composition of methane and maturity of gas source rocks. Science China Earth Sciences, 64(3), 470–493. https://doi.org/10.1007/s11430-020-9692-1

Copard, Y., Disnar, J.R. and Becq-Giraudon, J.F. (2002): Erroneous maturity assessment given by Tmax and HI Rock-Eval parameters on highly mature weathered coals. International Journal of Coal Geology, 49(1), 57–65. https://doi.org/10.1016/S0166-5162(01)00065-9

Dai, J. (1992): Identification and distinction of various alkane gases. Science in China (Scientia Sinica) Series B, 35(10), 1246–1257.

Dai, J., Gong, D., Ni, Y. and Wu, W. (2014): Genetic types of the alkane gases in giant gas fields with proven reserves over 1000×108m 3 in China. Exploration & Exploitation, 32(1), 1–18. https://doi.org/10.2307/90006067

Dai, J., Ni, Y., Liu, Q., Wu, X., Yu, C., Gong, D., Hong, F., Zhang, Y. and Yan, Z. (2022): Carbon Dioxide and its Carbon Isotopic Composition of Natural Gas in the Sichuan Basin, SW China. Frontiers in Earth Science, 10. https://doi.org/10.3389/feart.2022.857876

Dai, J., Song, Y. and Da, C. (1996): Geochemistry and Accumulation of Carbon Dioxide Gases in China. Am. Assoc. Petrol. Geol. Bull. 80, 1615–1626. https://doi.org/10.1306/64EDA0D2-1724-11D7-8645000102C1865D

Dai, J., Xia, X., Qin, S. and Zhao, J. (2004): Origins of partially reversed alkane $\delta^{13}C$ values for biogenic gases in China. Organic Geochemistry. 35(4), 405–411. https://doi.org/10.1016/j.orggeochem.2004.01.006

Davies, I. C. (1990): Geological and Exploration Review of The Tomori PSC, Eastern Indonesia. In: Proceedings Indonesian Petroleum Association, 19th Annual Convention, IPA90-223, 41-67.

Dee, M. P. (1989): Isotopic Analysis of The Binono Gas Seep and Sula Field Samples from Taliabu and Mangole Island, Indonesia Geochemistry Branch of BP Research, Report No.: GCB/43/89

Dembicki, Jr.H. (2009): Three common source rock evaluation errors made by geologists during prospect or play appraisals. Am. Assoc. Petrol. Geol. Bull. 93(3), 341–356. https://doi.org/10.1306/10230808076

Dembicki, Jr.H. (2017): Source Rock Evaluation. In Practical Petroleum Geochemistry for Exploration and Production. – Elsevier, 61–133. https://doi.org/10.1016/B978-0-12-803350-0.00003-9

Didyk, B.M., Simoneit, B.R.T., Brassell, S.C. and Eglinton, G. (1978): Organic geochemical indicators of palaeoenvironmental conditions of sedimentation. Nature, 272(5650), 216–222. https://doi.org/10.1038/272216a0

Dow, W.G. (1977): Kerogen studies and geological interpretations. Journal of Geochemical Exploration, 7, 79–99. https://doi.org/10.1016/0375-6742(77)90078-4

Espitalie, J., Deroo, G. and Marquis, F. (1985): La pyrolyse Rock-Eval et ses applications. Deuxième partie (*Rock-Eval pyrolysis and its applications. Second part*). Revue de l'Institut Français Du Pétrole, 40(6), 755–784. https://doi.org/10.2516/ogst:1985045

Feng, D., Liu, C., Tian, J., Ran, Y., Awan, R. S., Zeng, X., Zhang, J. and Zang, Q. (2023): Natural Gas Genesis, Source and Accumulation Processes in Northwestern Qaidam Basin, China, Revealed by Integrated 3D Basin Modeling and Geochemical Research. Natural Resources Research, 32(1), 391–412. https://doi.org/10.1007/s11053-022-10127-7

Ferdian, F. (2010): Evolution and Hydrocarbon Prospect of The North Banggai-Sula Area: An Application of Sea Seep[tm] Technology for Hydrocarbon Exploration in Under-explored Areas. In: Proceedings Indonesian Petroleum Association, 34th Annual Convention, IPA10-G-014, 1–14.

Galimov, E.M. (2006): Isotope organic geochemistry. Organic Geochemistry, 37(10), 1200–1262. https://doi.org/10.1016/j.orggeochem.2006.04.009

Garrard, RA, Supandjono, J.B. and Surono. (1988): The Geology of The Banggai-Sula Microcontinent, Eastern Indonesia. In: Proceedings Indonesian Petroleum Association, 17th Annual Convention, IPA88-11.01, 23–52.

Hao, F., Guo, T., Zhu, Y., Cai, X., Zou, H. and Li, P. (2008): Evidence for multiple stages of oil cracking and thermochemical sulfate reduction in the Puguang gas field, Sichuan Basin, China. Am. Assoc. Petrol. Geol. Bull. 92(5), 611–637. https://doi.org/10.1306/01210807090

Hasanusi, D., Abimanyu, R., Artono, E. and Baasir, A. (2004): Prominent Senoro Gas Field Discovery in Central Sulawesi. In: Proceedings IPA & AAPG, Deep-water and Frontier Exploration in Asia & Australasia Symposium, DFE04-OR-007, 177–197.

Huang, W.Y. and Meinschein, W.G. (1979): Sterols as ecological indicators. Geochimica et Cosmochimica Acta, 43(5), 739–745. https://doi.org/10.1016/0016-7037(79)90257-6

Jian, L., Jixian, T., Chenglin, L., Dehao, F., Xu, Z., Mai, Z., Zeqing, G., Hua, K., Awan, R. S., Zhijie, L. and Sai, Y. (2021): Geochemical characteristics and source analysis of natural gas in the saline lacustrine basin in the Western Qaidam basin. Journal of Petroleum Science and Engineering, 201, 108363. https://doi.org/10.1016/j.petrol.2021.108363

Klompé, Th.H.F. (1954): The structural importance of the Sula Spur (Indonesia). Journal for Natural Science, 110, 21–44.

Kotarba, M.J., Curtis, J.B. and Lewan, M.D. (2009): Comparison of natural gases accumulated in Oligocene strata with hydrous pyrolysis gases from Menilite Shales of the Polish Outer Carpathians. Organic Geochemistry, 40(7), 769–783. https://doi.org/10.1016/j.orggeochem.2009.04.007

Kotarba, M.J., Więcław, D., Dziadzio, P., Kowalski, A., Kosakowski, P. and Bilkiewicz, E. (2014): Organic geochemical study of source rocks and natural gas and their genetic correlation in the eastern part of the Polish Outer Carpathians and Palaeozoic-Mesozoic basement. Mar. Pet. Geo. 56, 97–122. https://doi.org/10.1016/j.marpetgeo.2014.03.014

Langford, F.F. and Blanc-Valleron, M. (1990): Interpreting Rock-Eval Pyrolysis Data Using Graphs of Pyrolizable Hydrocarbons vs. Total Organic Carbon (1): Am. Assoc. Petrol. Geol. Bull. 74. https://doi.org/10.1306/0C9B238F-1710-11D7-8645000102C1865D

Liu, Q., Jin, Z., Li, H., Wu, X., Tao, X., Zhu, D. and Meng, Q. (2018): Geochemistry characteristics and genetic types of natural gas in central part of the Tarim Basin, NW. China. Mar. Pet. Geo. 89, 91–105. https://doi.org/10.1016/j.marpetgeo.2017.05.002

Liu, Q., Wu, X., Wang, X., Jin, Z., Zhu, D., Meng, Q., Huang, S., Liu, J. and Fu, Q. (2019): Carbon and hydrogen isotopes of methane, ethane, and propane: A review of genetic identification of natural gas. Earth-Science Reviews, 190, 247–272. https://doi.org/10.1016/j.earscirev.2018.11.017

McCaffrey, R. (1982): Lithospheric deformation within the Molucca Sea arc-arc collision: Evidence from shallow and intermediate earthquake activity. Journal of Geophysical Research, 87(B5), 3663. https://doi.org/10.1029/JB087iB05p03663

McCaffrey, R., Silver, E.A. and Raitt, R.W. (1980): Crustal structure of the Molucca Sea collision zone, Indonesia. In: Hayes, D.E. (ed.): The Tectonic and Geologic Evolution of Southeast Asian Seas and Islands. – American Geophysical Union, 23, 161–177. https://doi.org/10.1029/GM023p0161

McCaffrey, R., Silver, E.A. and Raitt, R.W. (1981): Seismic refraction studies in the East Arm, Sulawesi–Banggai islands region of eastern Indonesia. In: Barber, AJ and Wiryosujono, S. (eds.): The Geology and Tectonics of Eastern Indonesia. Geological Research and Development Centre, 2, 321–325.

Metcalfe, I. (1990): Allochthonous terrane processes in Southeast Asia. Philosophical Transactions of the Royal Society of London. Series A, Mathematical and Physical Sciences, 331(1620), 625–640. https://doi.org/10.1098/rsta.1990.0094

Milkov, A.V. and Etiope, G. (2018): Revised genetic diagrams for natural gases based on a global dataset of >20,000 samples. Organic Geochemistry, 125, 109–120. https://doi.org/10.1016/j.orggeochem.2018.09.002

Mukhopadhyay, P.K., Wade, J.A. and Kruge, M.A. (1995): Organic facies and maturation of Jurassic/Cretaceous rocks, and possible oil-source rock correlation based on pyrolysis of asphaltenes, Scotian Basin, Canada. Organic Geochemistry, 22(1), 85–104. https://doi.org/10.1016/0146-6380(95)90010-1

Peters, K. E. and Cassa, M. R. (1994): Applied Source Rock Geochemistry. In: Magoon, L.B. and Dow, Q.G. (eds.): The Petroleum System-From Source to Trap. – American Association of Petroleum Geologists, 93–120. https://doi.org/10.1306/M60585C5

Peters, K.E., Walters, C.C. and Moldowan, J.M. (2005): The Biomarker Guide (Second Edition, Vol. 2). Cambridge

University Press. New York, 499 p. https://doi.org/10.1017/CBO9781107326040

Pigram, C. J., Surono and Supandjono, J. B. (1985): Origin of the Sula Platform, eastern Indonesia. Geology, 13(4), 246–248. https://doi.org/10.1130/0091-7613(1985)13%3C246:OOTSPE%3E2.0.CO;2

Quan, Y., Liu, J., Hao, F., Bao, X., Xu, S., Teng, C. and Wang, Z. (2019): Geochemical characteristics and origins of natural gas in the Zhu III sub-basin, Pearl River Mouth Basin, China. Mar. Pet. Geo. 101, 117–131. https://doi.org/10.1016/j.marpetgeo.2018.12.007

Sandvik, E. I., Young, W. A. and Curry, D. J. (1992): Expulsion from hydrocarbon sources: the role of organic absorption. Organic Geochemistry, 19(1–3), 77–87. https://doi.org/10.1016/0146-6380(92)90028-V

Satyana, A. H. and Zaitun, S. (2016, May): Origins of Oils and Gases at Banggai – Sula Microcontinent, Eastern Sulawesi – North Moluccas: Constraints from Biomarkers and Isotope Geochemistry – Implications for Further Exploration of Cenozoic and Pre-Cenozoic Objectives. In: Proceedings Indonesian Petroleum Association, 40th Annual Convention, IPA16-575-G, 1–27.

Schoell, M. (1980): The hydrogen and carbon isotopic composition of methane from natural gases of various origins. Geochimica et Cosmochimica Acta, 44(5), 649–661. https://doi.org/10.1016/0016-7037(80)90155-6

Schoell, M. (1983): Genetic Characterisation of Natural Gases. Am. Assoc. Petrol. Geol. Bull. 67(12), 2225–2238. https://doi.org/10.1306/AD46094A-16F7-11D7-8645000102C1865D

Schoell, M. (1984): Stable Isotopes in Petroleum Research. In: Brooks, J. and Welte, D. (eds.): Advances in Petroleum Geochemistry. – Elsevier, 1, 215–245. https://doi.org/10.1016/B978-0-12-032001-1.50009-2

Shanmugam, G. (1985): Significance of Coniferous Rain Forests and Related Organic Matter in Generating Commercial Quantities of Oil, Gippsland Basin, Australia. Am. Assoc. Petrol. Geol. Bull. 69(8), 1241–1254. https://doi.org/10.1306/AD462BC3-16F7-11D7-8645000102C1865D

Sherwood, O. A., Travers, P. D. and Dolan, M. P. (2013): Compound-Specific Stable Isotope Analysis of Natural and Produced Hydrocarbon Gases Surrounding Oil and Gas Operations. In: Ferrer, I. and Thurman, E. M. (Eds.): Advanced Techniques in Gas Chromatography–Mass Spectrometry (GC–MS–MS and GC–TOF–MS) for Environmental Chemistry. – Elsevier, 61, 347–372. https://doi.org/10.1016/B978-0-444-62623-3.00015-0Simandjuntak, TO and Barber, AJ (1996): Contrasting tectonic styles in the Neogene orogenic belts of Indonesia. In: Hall, R. and Blundell, D. (eds.): Tectonic Evolution of Southeast Asia, – Geological Society Special Publications, 106, 185–201. https://doi.org/10.1144/GSL.SP.1996.106.01.12

Stahl, W. J. (1974): Carbon isotope fractionations in natural gases. Nature, 251(5471), 134–135. https://doi.org/10.1038/251134a0

Stahl, W. J. (1977): Carbon and nitrogen isotopes in hydrocarbon research and exploration. Chemical Geology, 20, 121–149. https://doi.org/10.1016/0009-2541(77)90041-9

Stahl, W. J. and Carey, B. D. (1975): Source-rock identification by isotope analyses of natural gases from fields in the Val Verde and Delaware basins, west Texas. Chemical Geology, 16(4), 257–267. https://doi.org/10.1016/0009-2541(75)90065-0

Stahl, W. J., Wollanke, G. and Boigk, H. (1977): Carbon and nitrogen isotope data of Upper Carboniferous and Rotliegend natural gases from north Germany and their relationship to the maturity of the organic source material. In: Campos, R. & Gonji, J. (eds.): Advances in organic geochemistry 1975. – Enadisma., 539–559.

Sukamto, R. and Simandjuntak, T. O. (1983): Tectonic relationship between geologic provinces of western Sulawesi, eastern Sulawesi and Banggai-Sula in the light of sedimentological aspects. Bulletin Geological Research and Development Centre, 7, 1–12.

Supandjono, J. B. and Haryono, E. (1993): Geology of the Banggai Sheet, Sulawesi, Maluku 1:250,000. Geological Survey of Indonesia, Directorate of Mineral Resources, Geological Research and Development Centre.

Surjono, S. S., Asy'ari, M. R. and Gunawan, A. (2019): Application of Seabed Core Geochemistry for Further Exploration in Banggai-Sula Area. In: Rossetti, F., Blanc, A. C., Riguzzi, F., Leroux, E., Pavlopoulos, K., Bellier, O. and Kapsimalis V. (Eds.): The Structural Geology Contribution to the Africa-Eurasia Geology: Basement and Reservoir Structure, Ore Mineralisation and Tectonic Modelling. – Springer, 171–173. https://doi.org/10.1007/978-3-030-01455-1_36

Surono and Sukarna, D. (1993): Geology of the Sanana Sheet, Maluku 1:250,000. Geological Survey of Indonesia, Directorate of Mineral Resources. Geological Research and Development Centre.

ten Haven, H.L., de Leeuw, J.W., Rullkötter, J. and Damsté, J.S.S. (1987): Restricted utility of the pristane/phytane ratio as a palaeoenvironmental indicator. Nature, 330(6149), 641–643. https://doi.org/10.1038/330641a0

Tian, J., Li, J., Kong, H., Zeng, X., Wang, X. and Guo, Z. (2021): Genesis and accumulation process of deep natural gas in the Altun foreland on the northern margin of the Qaidam Basin. Journal of Petroleum Science and Engineering, 200, 108147. https://doi.org/10.1016/j.petrol.2020.108147

Tissot, B., Durand, B., Espitalié, J. and Combaz, A. (1974): Influence of Nature and Diagenesis of Organic Matter in Formation of Petroleum. Am. Assoc. Petrol. Geol. Bull. 58(3), 499–506. https://doi.org/10.1306/83D91425-16C7-11D7-8645000102C1865D

Watkinson, I. M., Hall, R. and Ferdian, F. (2011): Tectonic re-interpretation of the Banggai-Sula–Molucca Sea margin, Indonesia. In: Hall, R., Cottam, M.A. and Wilson, M.E.J. (eds.): The SE Asian Gateway: History and Tectonics of the Australia-Asia Collision. – Geological Society of London, 355(1), 203–224. https://doi.org/10.1144/SP355.10

Whiticar, M.J. (1994): Correlation of Natural Gases with Their Sources. In: Magoon, L.B. and Dow, Q.G. (eds.): The Petroleum System-From Source to Trap. – American Association of Petroleum Geologists, 261–284. https://doi.org/10.1306/M60585C16

Whiticar, M. J., Faber, E. and Schoell, M. (1986): Biogenic methane formation in marine and freshwater environments: CO2 reduction vs. acetate fermentation—Isotope evidence. Geochimica et Cosmochimica Acta, 50(5), 693–709. https://doi.org/10.1016/0016-7037(86)90346-7

Xu, W., Zheng, G., Ma, X., Fortin, D., Fu, C. C., Li, Q., Chelnokov, G. A. and Ershov, V. (2022): Chemical and isotopic features of seepage gas from mud volcanoes in southern margin of the Junggar Basin, NW. China. Applied Geochemistry, 136. https://doi.org/10.1016/j.apgeochem.2021.105145

Xu, Z., Peng, B., Feng, Y., Liu, L., Fang, C., Shao, M., Jia, K., Wu, K. and Chen, Y. (2019): Characteristics and origins of the natural gas and implications for gas-source correlation in deep formations of the Songliao Basin, NE China. Energies, 12(24): https://doi.org/10.3390/en12244641

Yang, S. and Horsfield, B. (2020): Critical review of the uncertainty of Tmax in revealing the thermal maturity of organic matter in sedimentary rocks. International Journal of Coal Geology, 225, 103500. https://doi.org/10.1016/j.coal.2020.103500

Zhang, S., He, K., Hu, G., Mi, J., Ma, Q., Liu, K. and Tang, Y. (2018): Unique chemical and isotopic characteristics and origins of natural gases in the Paleozoic marine formations in the Sichuan Basin, SW China: Isotope fractionation of deep and high mature carbonate reservoir gases. Mar. Pet. Geo. 89, 68–82. https://doi.org/10.1016/j.marpetgeo.2017.02.010

Zhou, F., Zhang, Y., Liu, Z., Sui, G., Li, G., Wang, C., Cui, S., Zhang, Y., Wang, J. and Zhu, J. (2016): Geochemical characteristics and origin of natural gas in the Dongping–Niudong areas, Qaidam Basin, China. Journal of Natural Gas Geoscience, 1(6), 489–499. https://doi.org/10.1016/j.jnggs.2016.11.008

SAŽETAK

Geokemija potencijalnih matičnih stijena i izdanaka prirodnoga plina u arhipelagu Mangole-Taliabu, Sjeverni Maluku, Indonezija

Arhipelag Mangole-Taliabu ima velik potencijal za istraživanje prirodnoga plina zbog brojnih aktivnih izdanaka prirodnoga plina. Ograničenost istraživačkih aktivnosti započetih tijekom posljednjih triju desetljeća dovele su do nedostatka publikacija o geokemijskim značajkama matičnih stijena i prirodnoga plina. Prethodna istraživanja pretpostavila su da su potencijalne matične stijene u istraživanome području i njegovoj okolici vezane uz formacije Buya i Salodik, a pretpostavlja se i da poznati aktivni izdanci prirodnoga plina potječu od humusne organske tvari (kerogen tipa III). U okviru ovoga istraživanja cilj je bio dati pregled i korištenjem postojećih i novoprikupljenih podataka ponovo raspraviti o geokemijskim karakteristikama potencijalnih matičnih stijena, podrijetlu i genetici prirodnoga plina te korelaciji između prirodnoga plina koji se pojavljuje na izdancima matičnih stijena. Ovo istraživanje otkrilo je da, osim prethodno potvrđene formacije Buya, formacija Bobong predstavlja formaciju s potencijalnim matičnim stijenama, dok u formaciji Salodik nisu zapažene potencijalne matične stijene, na što su upućivali rezultati prijašnjih istraživanja. Najvažniji rezultat istraživanja vezan je uz definiranje tipa prirodnoga plina na izdancima u kojemu nije pronađen samo plin dobiven iz ugljena kao matične stijene, već i plin iz nafte. Izračunani Ro za ugljenu matičnu stijenu kreće se između 0,80 % i 1,00 %, dok za naftno-matičnu stijenu varira između 0,50 % i 1,31 %. Korelacija između uzoraka prirodnoga plina s izdanaka i matične stijene upućuje na to da je formacija Bobong vjerojatno izvor za uzorke plina RAG/88/012 i RAG/88/014, dok je formacija Buya izvor za uzorke plina Binono Seep, RA/21/33, RA/21/34 i RA/21/35. Novi rezultati ovih istraživanja trebali bi imati znatan utjecaj na povećanje razumijevanja naftnoga sustava i poticanje daljnjih istraživačkih aktivnosti.

Ključne riječi:
matična stijena, izdanci prirodnoga plina, organska geokemija, izotop ugljika, Mangole, Taliabu

Authors' contribution

Taufik Ramli (M.Eng, Researcher) conducted fieldwork to collect rock and natural gas seep samples, proposed concepts and methodologies, interpreted source rock and gas geochemistry data, prepared visualisation of tables and figures, and wrote the original drafts. **Iwan Sukma Gumilar** (M.Eng, Researcher) conducted fieldwork as a team leader to collect rock and natural gas seep samples and proposed concepts and methodologies. **Ade Yogi** (M.Eng, Researcher) conducted fieldwork to collect rock and natural gas seep samples. **Ryan Akbar Fadhillah** (B.Sc, Researcher) conducted fieldwork to collect rock and natural gas seep samples. **Rakhmat Fakhruddin** (Dr. candidate, Researcher) supervised the geochemistry interpretation result and the original draft. **Praptisih** (Dr. candidate, Researcher) supervised the geochemistry interpretation result and the original draft.

Supplementary materials

Supplementary Material can be downloaded at the link: https://hrcak.srce.hr/ojs/index.php/rgn/article/view/26645/14242