GEOCHEMISTRY OF ALIPHATIC HYDROCARBON COMPOUNDS IN CORE SEDIMENTS FROM SADONG RIVER, MALAYSIA

Supported by tetfund

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Received: February 11, 2023 **Accepted:** April 15, 2023

Abstract

Sadong River is a major river catchment in Sarawak, Malaysia. Sediments are useful tools in understanding the environmental processes and are final sinks of hydrocarbons in rivers. Hydrocarbon biomarkers are organic compounds found in the environment with structures suggesting an unambiguous connection with known contemporary natural products. These specific indicator compounds which are found in extracts of geological and environmental sample such as sediment can be utilized for genetic source correlations. Hydrocarbons derived from biogenic sources appear with low concentrations in river sediments, while high level of concentration in hydrocarbon compounds are attributed to be of anthropogenic source which often lead to petroleum pollution. Several indices were used to distinguish the origin of *n*-alkanes in the river such as Low Molecular Weight to High Molecular weight (LMW/HMW), Carbon Preference Index (CPI) and Average Chain Length (ACL). Assessment of *n*-alkanes in the core sediments showed the hydrocarbons originated from terrestrial sources and anthropogenic sources.

Keywords: hydrocarbon, geological, biogenic, anthropogenic, *n*-alkanes

Introduction

Petroleum hydrocarbons comprised of a wide variety of chemicals with diverse origins (Zaghden *et al*., 2005; Zhang *et al*., 2018). Petroleum pollution has been of great concern for a long time because several components in petroleum that have been proven to have mutagenic and carcinogenic impacts on living organisms (Vaezzadeh *et al*., 2015). Global increase in use of fuel such as petrol and diesel as well as accidental oil spills are the main contributors to the release of petroleum into the environment (Singh *et al*., 2012; Keshavarzifard *et al*., 2018).

Aliphatic hydrocarbons commonly referred to as *n*-alkane consist of a straight chain of carbon and hydrogen with no alkyl branch or substitutes. They are mainly of geologic and anthropogenic sources. Aliphatic hydrocarbons occur naturally as linear, branched, cyclic, saturated and unsaturated. These straight chain hydrocarbons consist of odd and even carbon numbers ranging from C_4 to C_{64} with no alkyl groups (Ibadov & Suleymanov, 2015). The simplest form of alkane is methane, others include branched alkanes, isoprenoids and cycloalkanes. The straight chain molecules are commonly referred to as normal alkanes (*n*-alkanes) while the branched molecules are called iso-alkanes. *n*-Alkanes can be used as molecular markers to assess hydrocarbon contamination and to predict their sources (Duan *et al*., 2010). The aim of this study is to determine the quantity of aliphatic hydrocarbons, to assess the degree of contamination and delineate the relationship between geogenic and anthropogenic sources.

Geology of the Study Area

The geology of the Sadong River basin is not complex, it is made of the basement rocks and sedimentary rocks (Figure 1). There is a mosaic of sedimentary rocks and volcanic rocks together with tertiary alluvial sediments (Hutchison, 2005). Sarawak host sedimentary deposits comprised mainly of siliciclastic rocks such as siltstone, sandstone and shale (Nagarajan *et al*., 2014). The geology of Sarawak is closely associated and linked to the tectonic development of the South China Sea Basin (Liechti *et al*., 1960; Rijks, 1981; Tongkul, 1996; Manoj *et al*., 2014). The basin is underlain by more than 12 km of carbonate and siliciclastic sediments which are Tertiary in age (Madon *et al*., 2013). It is divided into various stratigraphic provinces (Hutchison, 1996; Peng *et al*., 2004; Hutchinson, 2005).

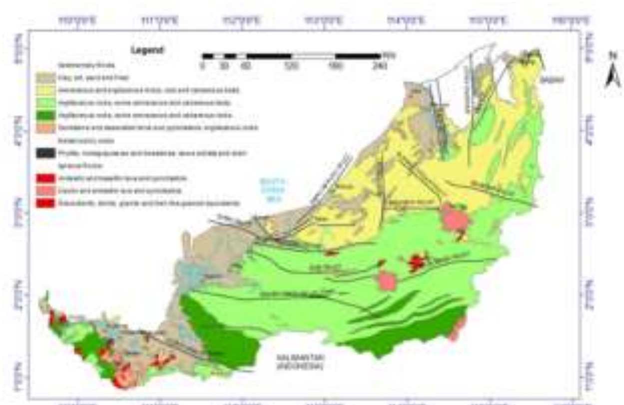

Figure 1: Simplified geological map of the Sarawak Basin (Modified after Mineral and Geoscience Department of Malaysia 2012; Adepehin *et al*., 2019)

The environment of the Sadong River has several economic activities such as farming, fishing, peat swamps, deserted coal mining site, boat docks and tourist spots. About 20 km upstream of this river, it bifurcates towards the east at Simunjan where coal has been mined in the 19th to 20th century around an isolated mountain (Bryant, 2003). In recent times, coastal and estuary pollution have become a global crisis in the natural ecosystem.

Materials and Methods

A total of three core sediments were also collected from three stations of Sadong River, Malaysia using a gravity corer. The corer tube length was about 100 cm and core tube size (diameter) was 5 cm. The core sediment was sliced into 2.5 cm interval, this interval was to complement visual, textural description and deposition of sediments. The river sediment samples were later air dried and sieved prior to extraction. The targeted hydrocarbon components were extracted using Soxhlet extractor, followed by column chromatography fractionation thereafter analyzed on capillary GC-MS. The Soxhlet extraction technique is capable to separate and isolate the geolipid from the mixture compounds and impurities. The geolipid was then fractionated on silica gel column chromatography to aliphatic hydrocarbon. Extraction of sediment was carried out following the procedure as outlined by Vaezzadeh *et al*. (2015). The geolipid was separated on a glass column chromatography (1.1 cm × 25 cm) packed with 4 g of silica gel purchased from Merck with mesh size of 0.040-0.063 mm. The geolipid extract was placed on the top of silica gel and eluted with 20 mL of *n*-hexane. Concentrations of *n*-alkanes in sediments were determined using a Shimadzu Gas Chromatography-Mass Spectrometer (GC-MS) model QP 2010.

Results and Discussion

Concentration of *n*-alkanes in the core sediments from Sadong River varied from upper layer to lower layer in Sebangan (BS1), Sadong Jaya (BS2) and Sebemban (BS6) core sediments as shown in Figures 2- 4. The *n*-alkanes in the three sediment cores comprised of *n*-alkanes ranged between C_9-C_{33} with low concentration of C_9-C_{12}. The core sediments of Sebangan (BS1) was characterized with very low concentration of C_9-C_{12} at the upper layer to lower layer in all core sediments with the exception of layer 42.5-45.0 cm which had a concentration of C_9 at 0.84 µg/g, C_{10} was 1.08 ug/g, C_{11} was 0.27 µg/g C_{12} was 0.15 ug/g and C_{13} was 0.98 µg/g. All the layers of core sediments had a low concentration of *n*-alkane C_9 with the exception of upper layer with concentration 1.19 µg/g, lower layer 40.0-42.5 cm with concentration 0.61 µg/g and 42.5-45.0 cm layer with concentration 0.84 µg/g. The middle layer of core sediments of Sadong River of Sebangan (BS1) at layers of 15.0-17.5 cm, 20.0-22.5 cm, 27.5-30.0 cm and 30.0-32.5 cm have a high concentration of *n*-alkanes C_{21}-C_{29}. The highest concentration of HMW *n*-alkane C_{26} was at layer 42.5-45.0 cm with concentration 103.66 µg/g.

The layers of the core sediments of Sadong Jaya (BS2) have low concentration of *n*-alkanes of C_9 to C_{13} except at layer 15.0-17.5 cm which had concentration of 3.93 µg/g, 7.28 µg/g and 0.48 µg/g for C_9, C_{10} and C_{11}, respectively. The highest concentration of *n*-alkanes was detected at layer 22.5-25.0 cm of C_{29}. There was an increase in the concentration of *n*- alkanes C_{21} to C_{25} in the layers of the core sediments. Whereas in the core sediments of Sebemban (BS6), the middle layer 20.0-22.5 cm had high concentration of *n*-alkanes C_{22} to C_{29} with concentration 47.17 µg/g and 55.53 µg/g. The increase of *n*-alkanes concentration of C_{20} to C_{24} with concentration 15.46 µg/g to 19.43 µg/g. The concentration was consistent at the lowest layer 40.0-42.5 and 42.5-45.0 cm.

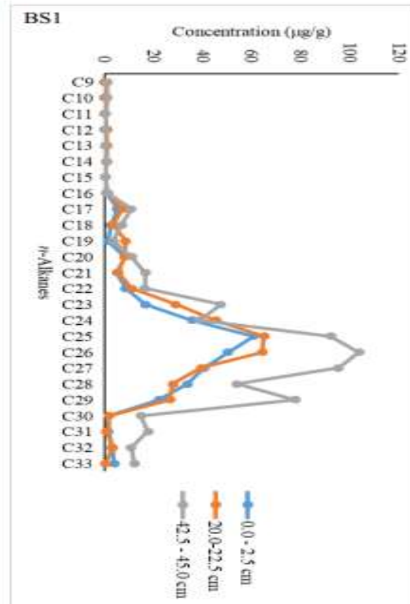

Figure 2: Variation of *n*-alkanes at upper layer, middle layer and lower layer in core sediments from BS1 of Sadong River

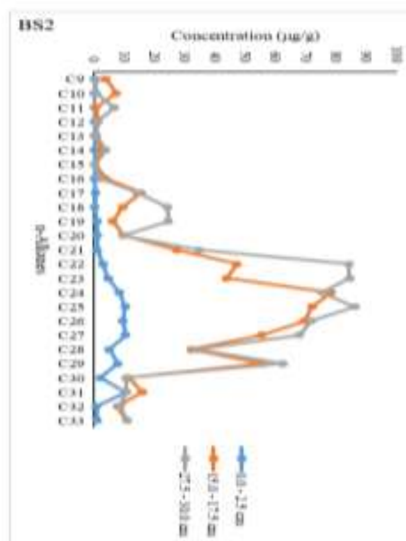

Figure 3: Variation of *n*-alkanes at upper layer, middle layer and lower layer in core sediments from BS2 of Sadong River

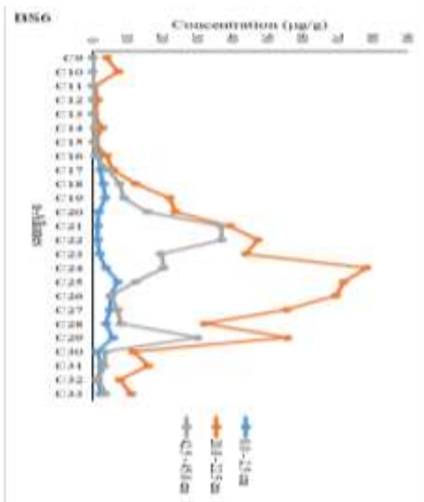

Figure 4: Variation of *n*-alkanes at upper layer, middle layer and lower layer in core sediments from BS6 of Sadong River

Several diagnostic indices were used to determine the aliphatic hydrocarbons pollution in the core sediments from Sadong River. The diagnostic indices are the total *n*-alkanes (TNA), Carbon Preference Index (CPI), Average Chain Length (ACL), Low Molecular Weight to High Molecular Weight (LMW/HMW) and Major Hydrocarbon (MH) (Sakari *et al*., 2008a; Omorinoye *et al*., 2019a). The ratio of LMW/HMW *n*-alkanes is an indicator of freshness of the released hydrocarbons in the river and its sources (Duan *et al*., 2019).

The total *n*-alkanes (TNA) was calculated in order to differentiate between natural and petrogenic aliphatic hydrocarbons (Sakari *et al*., 2008a and 2008b). The TNA is the sum of the concentration of *n*-alkanes from C_9 to C_{33}. Figure 5 shows the vertical profile of TNA in the core sediments from Sadong River. The total *n*-alkanes recorded in core sediments from Sadong River was > 100 µg/g in all layers. The TNA in core sediments from Sadong River varied from upper layer to lower layers of core. The TNA in the layer of core sediments of Sebangan (BS1) at 0.0-2.5 cm, 20.0-22.5 cm and 42.5- 45.0 cm was 624 µg/g, 715 µg/g and 1274 µg/g respectively. This indicated the lowest layer at 42.5- 45.0 cm had a high TNA, TNA of the layer of core sediment of Sadong Jaya (BS2) from Sadong River at 2.5-5.0 cm, 15.0-17.5 cm and 27.5-30.0 cm was 569 µg/g, 646 µg/g and 799 µg/g respectively. The upper layer of core sediment at 0.0-2.5 cm was 88 µg/g with an increase deeper depth. High levels of *n*-alkanes in some layers of the core sediment of Sebangan (BS1) may be indicative of anthropogenic inputs of hydrocarbons due to the activities in the vicinity of Sadong River.

Upper layer at 0.0-2.5 cm of core sediments of Sebemban (BS6) was 137 µg/g, the middle layer at 20.0-22.5 cm was 1249 µg/g and lowest layer at 42.5-45.0 cm was 477 µg/g. It is suggested that some distinctive attributes of the ecosystem such as occurrence of vegetal organic matter and detritus incorporated into the sediment which caused high deposition and accumulation of hydrocarbons (Vaezzadeh *et al*., 2015).

The ratios of LMW hydrocarbons ($<C_{24}$) relative to HMW *n*-alkane ($>C_{24}$) have been used to evaluate the relative contribution of the LMW and HMW hydrocarbons in Sadong River as shown in Figure 6. The odd number carbons of *n*-alkanes are derived from biogenic sources, while the even carbon number *n*-alkane are usually derived from anthropogenic sources (Aly Salem *et al*., 2014).

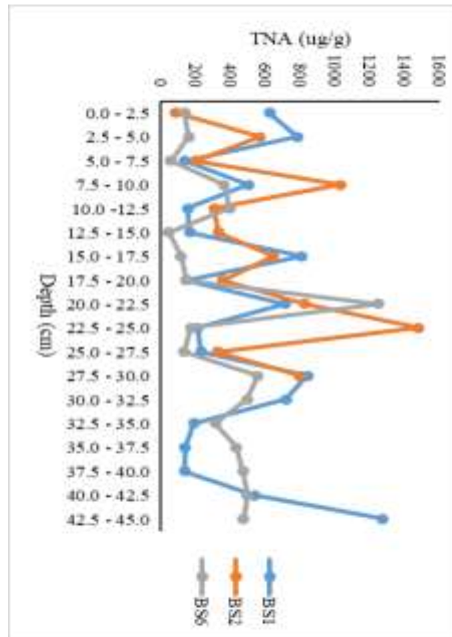

Figure 5: The vertical profile of TNA in core sediments from Sadong River

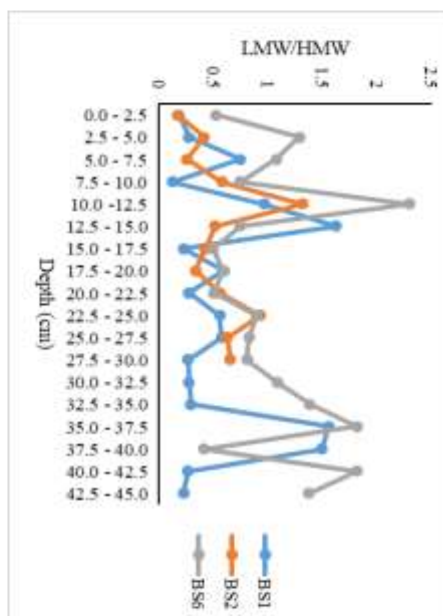

Figure 6: The vertical profile of LMW/HMW in core sediments from Sadong River

Elevated concentration of TNA in all core sediments may be due to the input from natural and anthropogenic activities. Concentrations of TNA below 50 µg/g is usually considered as uncontaminated (Zegouagh *et al*., 1998), while values above 100 µg/g are considered as contaminated and indicated the presence of petroleum biomarkers (Readman *et al*., 2002). The *n*-alkanes concentration in all layers were less than 100 µg/g except in core sediments at layer of 42.5-45.0 cm of Sebangan (BS1), layer 7.5-10.0 cm, 20.0-22.5 cm and 22.5-25.0 cm of Sadong Jaya (BS2). Thus, the value of TNA in all sediment cores indicate that they are slightly polluted with *n*-alkanes (Vaezzadeh *et al*., 2015).

The CPI of *n*-alkanes has been used to differentiate between natural and petrogenic aliphatic hydrocarbons (Omorinoye *et al*., 2019b). The CPI of *n*-alkanes in the core sediments of Sadong River are presented in Figure 7. The CPI at Sebangan (BS1) for layers of 0.0-2.5 cm, 20.0-22.5 cm and 42.5-45.0 cm were 1.24, 1.14 and 1.47, respectively. The CPI values between 1 to 3 are related to marine micro-organisms and recycled organic matter (Tahir *et al*., 2015; Commendatore *et a*l., 2012). The CPI for core sediments of Sadong Jaya (BS2) at layers 0-2.5 cm, 15.0-17.5 cm and 27.5-30.0 cm (BS2) were 1.93, 1.39 and 1.55, respectively. The CPI of core sediments at Sebemban (BS6) at layers 0.0-2.5 cm, 20.0-22.5 cm and 42.5-45.0 cm were 1.76, 1.39 and 2.29, respectively.

The vertical trend of CPI in core sediments of Sebangan (BS1), Sadong Jaya (BS2) and Sebemban (BS3) was zig-zag from upper layer to lower layer. Core sediments of Sebangan (BS1) and Sadong Jaya (BS2) showed a similar vertical trend for CPI as shown in Figure 7 as the CPI values increased gradually from uppermost surface and decreased in middle of core. In BS1, the CPI increased form middle towards the bottom of core with a sharp decrease in deepest having lowest CPI value of 0.34 in the 37.5-40.0 cm layer and the highest CPI of 5.53 in the middle core 25.0-27.5 cm layer. The relative high value is an indication of biogenic contributions. However, a significant decrease of CPI to 0.34 at layer 37.5-40.0 cm could be an indication of petrogenic contribution due to the fact that Sebangan which is the estuary and may be subjected to some of the anthropogenic activities in the area. The average CPI in Sadong Jaya (BS2) is greater than 1 with the exception of 7.5-10.0 cm layer and 12.5-15.0 cm layer having CPI values 0.95 and 0.97 respectively which indicated petrogenetic input from petroleum products.

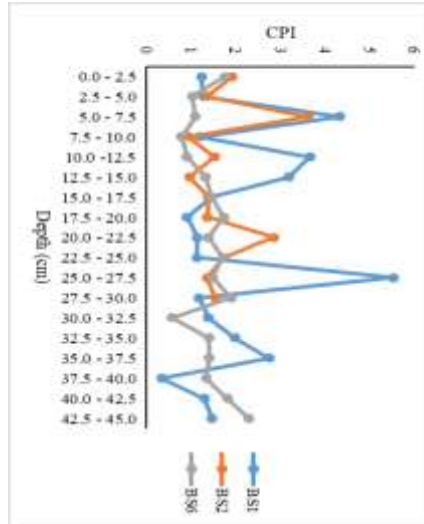

Figure 7: The vertical profile of CPI in core sediments from Sadong River

In Sebemban (BS6), the average CPI is between 1-2 which is related to marine micro-organisms and recycled organic matter. CPI value in 10.0-12.5 cm was 0.91 and 30.0-32.5 cm was 0.58, this attributed to petrogenetic input from petroleum products. High CPI value in BS6 was found in lowest layer 42.5-45.0 cm. Among all the core sediments from Sadong River, there is predominance of biogenic sources. Hence, a mixture of hydrocarbon sources is suggested which include petrogenic inputs, terrigenous sources, aquatic microorganisms and/or recycles organic matter (Duan *et al*., 2019).

Figure 8 shows the variation of ACL in core sediments from Sadong River. The trend of ACL in core sediments was zig zag from the upper layer to the lower layer of Sebangan (BS1), Sadong Jaya (BS2) and Sungai Buloh (BS3). The ACL of *n*-alkanes in core sediments layers 0.0-2.5 cm, 20.0-22.5 cm and 42.5-45.0 cm respectively, of Sebangan (BS1) was 26.63, 26.45 and 27.39. The ACL values was observed to increase in the middle core and fluctuated towards the middle core with an increase at lower layer of core. Sadong Jaya (BS2) was characterised by an increase in the upper layer at 28.10 at 0.0-2.5 cm, a consistent pattern in the middle layer at 27.46 and an increase in the lower layer of core 22.5-25.0 cm and decreased to 27.25 at 27.5-30.0 cm. Sebemban (BS6) was characterised by a fairly consistent pattern form upper layer to bottom of core with upper layer 0.0-2.5 cm was 27.91, 20.0-22.5 cm was 27.46 and 42.5-45.0 cm was 28.24.

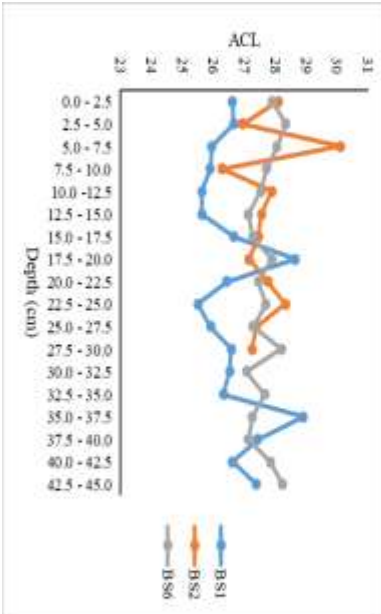

Figure 8: The vertical profile of ACL in core sediments from Sadong River

Table 1 shows the MH for hydrocarbons in core sediments from Sadong River. The MH of hydrocarbons in sediments core of Sebangan (BS1) was C_{25} which represents terrestial sources with the exception of middle layer at depth 17.5-20.0 cm with C_{24} and layer 37.5-40.0 cm with C_{22} indicating marine sources. This showed that *n*-alkanes in core sediments of Sebangan (BS1) which is located at the estuary were contributed by a mixture of terrestial vascular plant, phytoplankton, algae and petroleum. Guo & Fang (2012) reported that *n*-alkanes originated from microorganisms such as algae, plankton and bacteria have short hydrocarbon chain with major hydrocarbons at C_{15}, C_{17} and C_{19}. Domination of C_{20} and C_{21} alkanes indicated *n*-alkanes derived from oil, automobile exhaust and fossil fuel combustion. The long chains *n*-alkanes between C_{25}-C_{33} are derived from terrestial higher plants.

Table 1: Major hydrocarbon (MH) in core sediments from Sadong River

	MH		
Depth	BS1	BS2	BS6
0.0-2.5	C_{25}	C_{31}	C_{25}
2.5-5.0	C_{25}	C_{22}	C_{14}
5.0-7.5	C_{25}	C_{33}	C_{20}
7.5-10.0	C_{25}	C_{26}	C_{24}
10.0-12.5	C_{25}	C_{22}	C_{21}
12.5-15.0	C_{25}	C_{26}	C_{9}
15.0-17.5	C_{25}	C_{24}	C_{25}
17.5-20.0	C_{24}	C_{25}	C_{25}
20.0-22.5	C_{25}	C_{27}	C_{24}
22.5-25.0	C_{25}	C_{20}	C_{20}
25.0-27.5	C_{25}	C_{24}	C_{20}
27.5-30.0	C_{25}	C_{25}	C_{29}
30.0-32.5	C_{25}	-	C_{28}
32.5-35.0	C_{25}	-	C_{21}
35.0-37.5	C_{29}	-	C_{21}
37.5-40.0	C_{22}	-	C_{26}
40.0-42.5	C_{25}	-	C_{22}
42.5-45.0	C_{26}	-	C_{22}

'-' - data not available

Conclusion

Anthropogenic sources of *n*-alkanes in this river are land-based sources particularly from the activities around the banks of the river such as use of fertilizers and pesticides on oil palm plantation and rice paddies, exhaust fumes from boat docking as well as shipping. Domination of *n*-alkanes originating from vascular plants were observed with a significance of HMW *n*-alkanes with odd carbon numbers in the surface sediments. The hydrocarbons are mainly derived from natural sources such as terrestial vascular plants, algae and micro-organisms with minimal impact of anthropogenic activities as shown by major hydrocarbons in the river core sediments.

Acknowledgment

Authors are grateful to Universiti Malaysia Sarawak

Conflict of Interest

The authors declare that there is not any conflict of interests regarding the publication of this manuscript.

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