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SOURCES OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHS) POLLUTION IN MARINE SEDIMENT FROM TUANKU ABDUL RAHMAN NATIONAL PARK, SABAH

(Sumber Pencemaran Hidrokarbon Aromatik Polinuklear (PAH) Dalam Sedimen Marin Dari Taman Negara Tuanku Abdul Rahman, Sabah)

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Abstract

The concentrations of parent and alkyl Polycyclic Aromatic Hydrocarbons (PAHs) in marine sediment samples collected from Tuanku Abdul Rahman National Park, Sabah were determined by using GCMS. The ratio of anthracene to anthracene plus phenanthrane, fluorenthane to fluorenthane plus pyrene, benz[a]anthracene to benz[a]anthracene plus chrysene and indeno[1,2,3-cd]pyrene to indeno[1,2,3-cd]pyrene plus benzo[g,h,i]perylene, compounds were used to identify the sources of PAHs pollution. The total concentration of parent and alkyl PAHs are ranged from 121.7 to 191.5 ng/g dry weight. The concentrations of PAHs pollution in sediments were categorised as a moderate polluted. The ratio values of PAHs compound indicate the origin source of PAHs pollutions in marine sediment sample of Tuanku Abdul Rahman National Park were originated from fossil fuel combustion (pyrolytic).

Keywords: PAHs, national park, pollution source, sediment

Abstrak

Pengukuran kepekatan sebatian hidrokarbon polisiklik aromatik alkil dan induk di dalam sample sedimen marin yang diambil dari Taman Negara Tuanku Abdul Rahman dilakukan dengan menggunakan peralatan GCMS. Penentuan sumber pencemaran PAH ditentukan dengan menggunakan penilaian nisbah sebatian antrasena terhadap antrasena campur fenantrena, fluorantena terhadap fluorantena campur piren, benz[a]antrasena terhadap benz[a]antrasena campur krisena dan indeno[1,2,3-cd]piren terhadap indeno[1,2,3-cd]piren campur benzo[g,h,i]perilena. Jumlah kepekatan sebatian PAH alkil dan induk berada dalam julat 121.7 hingga 191.5 ng/g berat kering. Kepekatan PAH di dalam sedimen dikatogerikan sebagai sederhana tercemar. Nilai nisbah sebatian PAH menunjukan sumber pencemaran PAH di dalam sedimen marin Taman Negara Tuanku Abdul Rahman berasal daripada pembakaran minyak fosil (pirolitik).

Kata kunci: PAH, taman negara, sumber pencemaran, sedimen

Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a class of organic priority pollutants, ubiquitous in the aquatic ecosystems, resistance to biodegradation and show adverse health effects (carcinogenic activity) depending on the molecular weight and structure [Hoffman et al., 1984] [1-5]. PAH can be introduced into the marine environment by various ways such as oil spill, forest fire, combustion of petrol and diesel, coal combustion [3,5-7], urban runoff, domestic and industrial wastewater discharges [4, 6].

Source of PAH pollution can be categorized into two groups; (1) pyrolytic and (2) petrogenic. Pyrolytic PAHs are generated through incomplete combustion of organic matter such as coal, petroleum and wood combustion, industrial operation and power plant using fossil fuels. Petrogenic PAHs are derived from crude oil and petroleum product such as kerosene gasoline, diesel fuel, lubricating oil and asphalt. Petrogenic PAHs are emitted directly to

marine environment through oil spills and routine operation of tankers (e.g. discharge of ballast water) [8]. Source of PAH pollution can be identify by using isomer pair ratios such as anthracene/ (anthracene+phenanthrene); indeno[1,2,3-cd]pyrene/ (indeno[1,2,3-cd]pyrene + benzo[g,h,I]perylene); benz[a]anthracene/ (benz[a]anthracene + chrysene) and fluoranthene/ (fluoranthene + pyrene) [5,9].

Study on PAHs pollution at Tuanku Abdul Rahman national park and others marine park of Sabah are still limited. In this present study, organic contamination (PAHs) was selected because they are important due to carcinogenic and toxicity to the human health and also for baseline data and/or information. The goal of this work was to determine the concentrations of PAH compound in the sediment and to identify the possible source of these compounds whether from anthropogenic or natural processes.

Methodology

Sampling Location

Eleven sampling locations were selected at Tuanku Abdul Rahman National Park of Sabah as shown in Figure 1. The sediment samples were collected using a Ponar grab sampler, and they were transferred into glass bottles with aluminium caps using a stainless steel spatula. The marine sediment samples were stored in glass bottles and stored at below 5^{o}C before analysis. The sampling station coordinates of Tuanku Abdul Rahman National Park were shown in Table 1. 5.0 gram of wet sediment sample was dried in oven at 105^{o}C until constant weight. The percentage of water content in sediment sample and PAHs concentrations in dry weight unit were calculated as described in Reference Method in Marine Pollution Studies No.20 [10].

Table 1: Sampling stations and coordinates of Tuanku Abdul Rahman National Park, Sabah

Station	Latitude	Longitude	Water depth (m)
TAR 1	6^{o} 01.308' N	116^{o} 04.416' E	27.2
TAR 2	6^{o} 02.248' N	116^{o} 02.915' E	33.5
TAR 3	6^{o} 02.634' N	116^{o} 01.371' E	44.5
TAR 4	6^{o} 02.548' N	116^{o} 00.498' E	34.7
TAR 5	5^{o} 58.409' N	116^{o} 02.235' E	31.3
TAR 6	6^{o} 00.015' N	116^{o} 01.834' E	27.0.
TAR 7	5^{o} 58.419' N	116^{o} 02.240' E	21.6
TAR 8	5^{o} 57.648' N	116^{o} 01.969' E	11.2
TAR 9	5^{o} 57.253' N	115^{o} 59.407' E	32.0
TAR 10	5^{o} 58.301' N	115^{o} 59.381' E	36.1
TAR 11	5^{o} 58.039' N	116^{o} 00.401' E	30.0

Soxhlet extraction of marine sediment sample

20 g wet sediment samples were weighted in a glass beaker where about 30 g of Na_2SO_4 was added to the sediment sample, mix together for a homogenous. Sample was added into the soxhlet apparatus and spike with two internal standard for aliphatic fraction (n-octadecene-5µg/l) and PAHs fraction (orto-terphenyl-5µg/l) for recovery assessment. The sample was soxhlet extracted using 250 ml (50:50 v/v) mixture of dichloromethane (DCM) and hexane (analar grade). After 12 hours extraction, the extracted samples were dry up until 1ml by using rotary evaporator. 8.0g of silica gel and 8.0g of alumina were deactivated with 0.16 ml and 0.4 ml deionzed water, respectively. Silica gel and alumina were filling up into silica-alumina column (20mm diameter and 150 mm height). 1ml of extracted sample was fractioned into subfraction using silica-alumina column. Copper granules were used to remove sulphur from the extract, which was then passed through a silica/alumina chromatographic column. Then 30 ml of hexane was eluted pass through into the silica-alumina column to elute aliphatic compounds ($C_{12} - C_{34}$) fraction (F1). Forty (40) ml mixture of DCM and hexane (50:50 v/v) was used to elute the PAHs for fractionation 2 (F2) and followed by 40 ml of hexane-DCM (20:80) for fractionation 3 (F3). The eluent from fractionation 2 and 3 (F2 and F3) was collected and concentrated to a few ml in a rotary evaporator. After that the

concentrated samples were transferred into a vial, and reduced to 1 ml under a gentle nitrogen gas stream. The final solution F2 and F3 were analyzed by GC-MS to determine the aromatic hydrocarbon compounds quantitatively.

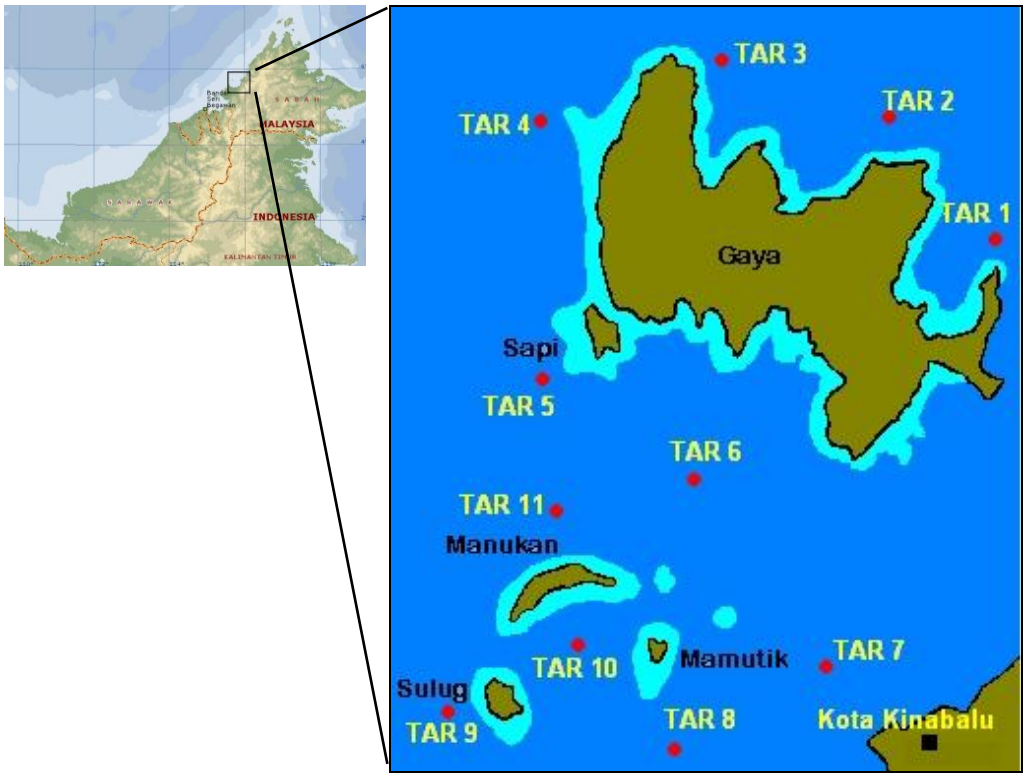

Fig. 1: Sampling stations of Tuanku Abdul Rahman National Park of Sabah

Gas Chromatography–Mass Spectrometry (GC-MS) analysis

The PAHs compounds were analyzed by GC-MS (Shimadzu QP5050A model) using selected ion monitoring (SIM) mode. A 30 m x 0.25 mm x 0.25 µm ZB1 fused silica capillary column (Phenomenex) was used to identify them. Helium was used as carrier gas with a flow-rate of 3 ml/min. The initial injection and interface temperature of GC-MS was setup at 70^{o}C and 270^{o}C respectively. The column temperature was started at 70 ^{o}C, held for 2 min, then increased at 15 ^{o}C/ min up to 300°C and the temperature will be maintained at 300^{o}C for 12 minutes. The PAH analysis took about 1 h to be complete to identification and quantification of the alkyl and parent PAHs base on ion fragmentation and retention time compared to of that the external PAHs standard. The alkyl PAHs such as 1-methyl naphthalene, 1-ethyl naphthalene, 2,3,6-trimethyl naphthalene, 2-methyl phenanthrene , 1-methyl phenanthrene, 3,6-dimethyl phenanthrene, 1-methyl pyrene and 17 parent aromatic hydrocarbons analyzed namely; naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene, perylene, benz[a]anthracene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd)-pyrene, dibenz[a,h]anthracene, and benzo[g,h,i]perylene.

Results and Discussion

Tuanku Abdul Rahman National Park consists of Sapi, Manukan, Mamutik, Sulug and Gaya Island. Twenty-four species of parent and alkyl PAH compounds were measured by using GCMS for source identification of PAHs pollution. The analysed PAHs compound from sediment samples of Tuanku Abdul Rahman National Park were shown in Table 2 and total parent and alkyl PAHs concentrations of each station were shown in Figure 2 (Bar

graph). The total (Σ) concentration of parent and alkyl PAHs compound are ranged from 121.7 to 191.5 ng/g dry weight. The PAH concentrations could be characterized as low, moderate, high and very high ones when the ΣPAHs were 0–100, 100–1000, 1000–5000, and more than 5000 ng/g, respectively [11]. PAHs pollution of Tuanku Abdul Rahman National Park sediments were categorize as a moderate pollution due to their PAH concentration within the range 121.7 to 191.5 ng/g dry weight as shown in Figure 2.

Based on the PAH isomer pair ratio measurement compiled by Yunker et al., [9]; Anth/(Anth+Phen) ratio less than 0.1 indicates source of PAH dominance of petroleum and ratio higher than 0.1 indicates dominance of combustion; Fl/(Fl+Py) ratio less then 0.4 shown petroleum source, 0.4 to 0.5 petroleum combustion (pyrolytic) and more than 0.5 combustion of coal, grasses and wood; B[a]A/B[a]A+Chry ratio less than 0.2 petroleum source, 0.2 to 0.35 petroleum and combustion and more than 0.35 combustion source; and IP/IP+BghiP ratio less than 0.2 petroleum, 0.2 to 0.5 petroleum combustion and more than 0.5 combustion of coal, grasses and wood .

Isomer pair ratio of Anth/(Anth+Phen) of Tuanku Abdul Rahman National Park sediment ranged from 0.66 to 0.73 as shown in Table 2. These indicated the source of PAHs pollution is originated from the combustion. The ratio of Benz[a]Anthracene to Benz[a]Anthracene + chrycene (B[a]A/B[a]A+Chry) shown four of the stations (TAR 01, TAR 04, TAR05 and TAR11) was not calculated due to not detected of the Benz[a]Anthracene compound of that stations. However, the ratio of B[a]A/(B[a]A+Chry) for other stations are ranged from 0.54 to 0.60, which indicated the source of PAHs pollution from combustion. Most isomer pair ratio values of IP/(IP+BghiP) in all stations are 0.41 except for TAR 04, TAR 05 and TAR 10. TAR 04 and TAR 05 stations were not calculated due to not detected of Indeno(1,2,3-cd)pyrene and the ratio for TAR 10 is 1.0 due to not detected of Benzo[g,h,i]perylene compound. According to Yunker et al., [9] the IP/(IP+BghiP) values of 0.20 to 0.50 indicate the source of PAH pollution from petroleum combustion (pyrolytic). The Fl/(Fl+py) ratios in all station ranged from 0.45 to 0.47, these clearly indicated the source of PAH pollution in Tuanku Abdul Rahman National Park was originated from petroleum combustion (pyrolytic).

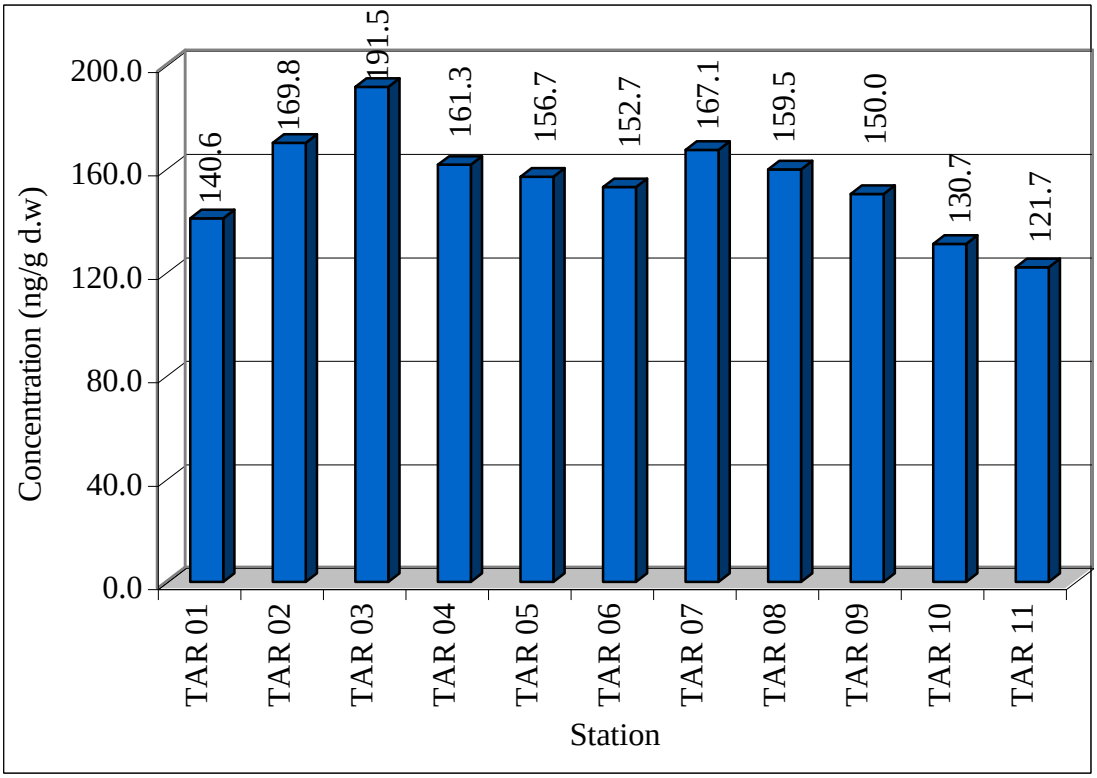

Fig 2: Total parent and alkyl PAHs concentration of Tuanku Abdul Rahman National.Park, Sabah.

Table 2: Parent and alkyl PAHs concentration (ng/g d.w) in marine sediment samples collected from Tuanku Abdul Rahman National Park, Sabah

Compound	Station			
	TAR 01	TAR 02	TAR 03	TAR 04
Naphthalene	7.85 ± 0.86	6.47 ± 0.72	8.23 ± 0.90	9.35 ± 1.02
1-Methyl Naphthalene	6.18 ± 0.08	5.76 ± 0.08	7.23 ± 0.09	7.99 ± 0.10
1-Ethyl Naphthalene	2.85 ± 0.08	2.82 ± 0.08	3.74 ± 0.10	3.66 ± 0.10
2,3,6 -Trimethyl Naphthalene	2.50 ± 0.02	2.47 ± 0.02	3.12 ± 0.02	3.12 ± 0.02
Acenaphthylene	n.d	4.35 ± 0.12	5.61 ± 0.16	5.69 ± 0.16
Fluorene	7.02 ± 0.13	7.05 ± 0.13	8.98 ± 0.17	8.81 ± 0.17
Acenaphthene	2.26 ± 0.10	2.23 ± 0.09	3.49 ± 0.15	3.12 ± 0.13
Phenanthrene	5.71 ± 0.34	5.17 ± 0.31	6.98 ± 0.42	6.78 ± 0.41
Anthracene	12.25 ± 2.07	12.23 ± 2.07	13.72 ± 2.33	14.23 ± 2.41
2-Methyl Phenanthrene	7.37 ± 1.16	6.94 ± 1.08	8.23 ± 1.29	8.40 ± 1.32
1-Methyl Phenanthrene	1.66 ± 0.32	1.53 ± 0.28	1.87 ± 0.36	2.03 ± 0.37
3,6-Dimethyl Phenanthrene	5.35 ± 1.74	5.29 ± 1.71	6.98 ± 2.26	6.50 ± 2.10
Fluoranthene	5.59 ± 0.16	5.99 ± 0.17	8.35 ± 0.24	7.32 ± 0.20
Pyrene	6.66 ± 0.37	6.82 ± 0.37	9.73 ± 0.53	8.54 ± 0.47
1-Methyl Pyrene	4.99 ± 0.60	5.05 ± 0.61	5.74 ± 0.68	n.d
Perylene	14.03 ± 5.46	14.34 ± 5.58	14.84 ± 5.77	15.99 ± 6.24
Chrysene	13.32 ± 2.53	16.81 ± 3.19	15.34 ± 2.91	15.31 ± 2.91
Benz[a]anthracene	n.d	19.51 ± 2.93	20.70 ± 3.11	n.d
Benzo[b]fluoranthene	5.02 ± 0.80	4.96 ± 0.08	5.26 ± 0.85	5.72 ± 0.91
Benzo[k]fluoranthene	3.04 ± 0.33	3.01 ± 0.33	3.19 ± 0.35	3.47 ± 0.39
Benzo[a]pyrene	4.64 ± 0.69	4.58 ± 0.69	4.86 ± 0.74	5.28 ± 0.80
Indeno [1,2,3-cd)pyrene	4.64 ± 1.43	4.58 ± 1.43	4.86 ± 1.52	n.d
Dibenz[a,h]anthracene	4.16 ± 2.10	4.11 ± 2.05	4.36 ± 2.20	4.74 ± 2.35
Benzo[g,h,i]perylene	2.71 ± 0.14	2.68 ± 0.14	2.84 ± 0.14	3.09 ± 0.16
∑Parent and Alkyl PAH	140.6	169.8	191.5	161.3
Anth/(Anth+Phen) ratio	0.68	0.70	0.66	0.68
Fl/(Fl+Py) ratio	0.46	0.47	0.46	0.46
B[a]A/(B[a]A +Chry) ratio	n.c	0.54	0.57	n.c
IP/(IP+BghiP) ratio	0.41	0.41	0.41	n.c

n.d – below detection limit; n.c – not calculated. Anth = Anthracene; Phen = Phenanthrene; Fl = Fluoranthene; Py = Pyrene; B[a]A=Benz[a]anthracene; Chry =Chrysene; IP=Indeno[1,2,3-cd]pyrene; BghiP=Benzo[g,h,I]perylene.

Continued Table 2:

Compound	Station			
	TAR 05	TAR 06	TAR 07	TAR 08
Naphthalene	6.88 ± 0.76	7.07 ± 0.78	5.95 ± 0.65	5.79 ± 0.64
1-Methyl Naphthalene	6.00 ± 0.08	5.31 ± 0.07	4.05 ± 0.05	3.98 ± 0.05
1-Ethyl Naphthalene	3.50 ± 0.09	2.78 ± 0.08	2.53 ± 0.07	2.53 ± 0.07
2,3,6 -Trimethyl Naphthalene	3.25 ± 0.02	2.53 ± 0.02	2.53 ± 0.02	2.29 ± 0.01
Acenaphthylene	5.38 ± 0.15	4.67 ± 0.13	4.68 ± 0.13	4.46 ± 0.13
Fluorene	8.88 ± 0.17	7.58 ± 0.14	7.59 ± 0.14	7.24 ± 0.14
Acenaphthene	3.50 ± 0.15	2.40 ± 0.10	n.d	n.d
Phenanthrene	6.75 ± 0.41	5.43 ± 0.32	5.57 ± 0.34	5.19 ± 0.31
Anthracene	13.63 ± 2.31	13.14 ± 2.23	13.03 ± 2.21	12.42 ± 2.11
2-Methyl Phenanthrene	8.13 ± 1.27	7.58 ± 1.19	7.97 ± 1.26	7.36 ± 1.16
1-Methyl Phenanthrene	1.88 ± 0.36	1.77 ± 0.34	1.77 ± 0.34	1.81 ± 0.34
3,6-Dimethyl Phenanthrene	6.75 ± 2.20	5.94 ± 1.91	5.82 ± 1.87	5.43 ± 1.74
Fluoranthene	7.38 ± 0.21	6.44 ± 0.18	6.33 ± 0.18	5.91 ± 0.17
Pyrene	9.13 ± 0.50	7.45 ± 0.41	7.09 ± 0.39	6.87 ± 0.38
1-Methyl Pyrene	5.50 ± 0.66	5.31 ± 0.64	5.44 ± 0.65	5.19 ± 0.62
Perylene	14.76 ± 5.77	n.d	14.93 ± 5.81	14.47 ± 5.66
Chrysene	13.76 ± 2.62	13.90 ± 2.64	13.92 ± 2.64	13.27 ± 2.53
Benz[a]anthracene	n.d	20.97 ± 3.15	21.00 ± 3.15	20.02 ± 3.00
Benzo[b]fluoranthene	5.28 ± 0.85	5.33 ± 0.85	5.34 ± 0.85	5.09 ± 0.81
Benzo[k]fluoranthene	3.20 ± 0.35	3.23 ± 0.35	3.24 ± 0.35	3.09 ± 0.34
Benzo[a]pyrene	4.88 ± 0.74	4.93 ± 0.74	4.93 ± 0.74	4.70 ± 0.71
Indeno [1,2,3-cd)pyrene	n.d	4.93 ± 1.52	4.93 ± 1.52	4.70 ± 1.46
Dibenz[a,h]anthracene	4.38 ± 2.20	n.d	4.43 ± 2.20	4.22 ± 2.10
Benzo[g,h,i]perylene	2.85 ± 0.15	2.88 ± 0.15	2.88 ± 0.15	2.75 ± 0.14
∑Parent and Alkyl PAH	156.7	152.7	167.1	159.5
Anth/(Anth+Phen) ratio	0.67	0.71	0.70	0.71
Fl/(Fl+Py) ratio	0.45	0.46	0.47	0.46
B[a]A/(B[a]A +Chry) ratio	n.c	0.60	0.60	0.60
IP/(IP+BghiP) ratio	n.c	0.41	0.41	0.41

n.d – below detection limit; n.c – not calculated. Anth = Anthracene; Phen = Phenanthrene; Fl = Fluoranthene; Py = Pyrene; B[a]A=Benz[a]anthracene; Chry =Chrysene; IP=Indeno[1,2,3-cd]pyrene; BghiP=Benzo[g,h,I]perylene.

Continued Table 2:

Compound	Station		
	TAR 09	TAR 10	TAR 11
Naphthalene	5.72 ± 0.63	5.30 ± 0.58	5.19 ± 0.57
1-Methyl Naphthalene	3.93 ± 0.05	3.89 ± 0.05	3.77 ± 0.05
1-Ethyl Naphthalene	2.50 ± 0.07	2.36 ± 0.06	2.36 ± 0.06
2,3,6 -Trimethyl Naphthalene	2.26 ± 0.01	2.24 ± 0.01	2.24 ± 0.01
Acenaphthylene	4.41 ± 0.12	4.36 ± 0.12	4.36 ± 0.12
Fluorene	n.d	n.d	n.d
Acenaphthene	n.d	2.12 ± 0.09	n.d
Phenanthrene	5.12 ± 0.31	4.71 ± 0.28	4.60 ± 0.28
Anthracene	12.38 ± 2.11	12.25 ± 2.09	12.27 ± 2.09
2-Methyl Phenanthrene	7.38 ± 1.16	6.95 ± 1.10	6.84 ± 1.07
1-Methyl Phenanthrene	1.67 ± 0.32	1.53 ± 0.28	1.53 ± 0.28
3,6-Dimethyl Phenanthrene	5.36 ± 1.74	5.30 ± 1.71	5.31 ± 1.71
Fluoranthene	5.83 ± 0.16	5.54 ± 0.15	5.54 ± 0.15
Pyrene	6.79 ± 0.37	6.48 ± 0.36	6.49 ± 0.36
1-Methyl Pyrene	5.00 ± 0.60	n.d	n.d
Perylene	14.05 ± 5.50	13.90 ± 5.42	13.92 ± 5.42
Chrysene	13.10 ± 2.49	12.96 ± 2.47	12.97 ± 2.47
Benz[a]anthracene	19.77 ± 2.97	19.56 ± 2.94	n.d
Benzo[b]fluoranthene	5.02 ± 0.80	4.97 ± 0.80	4.98 ± 0.80
Benzo[k]fluoranthene	3.05 ± 0.33	3.02 ± 0.33	3.02 ± 0.33
Benzo[a]pyrene	4.64 ± 0.69	4.59 ± 0.69	4.60 ± 0.69
Indeno [1,2,3-cd)pyrene	4.64 ± 1.43	4.59 ± 1.43	4.60 ± 1.43
Dibenz[a,h]anthracene	4.17 ± 2.10	4.12 ± 2.05	4.13 ± 2.05
Benzo[g,h,i]perylene	2.71 ± 0.14	n.d	2.69 ± 0.14
∑Parent and Alkyl PAH	150.0	130.7	121.7
Anth/(Anth+Phen) ratio	0.71	0.72	0.73
Fl/(Fl+Py) ratio	0.46	0.46	0.46
B[a]A/(B[a]A +Chry) ratio	0.60	0.60	n.c
IP/(IP+BghiP) ratio	0.41	1.00	0.41

n.d – below detection limit; n.c – not calculated. Anth = Anthracene; Phen = Phenanthrene; Fl = Fluoranthene; Py = Pyrene; B[a]A=Benz[a]anthracene; Chry =Chrysene; IP=Indeno[1,2,3-cd]pyrene; BghiP=Benzo[g,h,I]perylene.

Conclusion

The Σ concentration of parent and alkyl PAHs compound in marine sediment samples collected from Tuanku Abdul Rahman National Park are ranged from 121.7 to 191.5 ng/g dry weight. The PAHs pollution of Tuanku Abdul Rahman National Park can be categorized as moderately polluted with total parent and alkyl PAHs from 100 – 1000 ng/g dry weight.

Based on isomer pairs ratios of PAHs compounds, with the ratio value of Anth/Anth+Phen, Fl/Fl+Py, IP/(IP+BghiP) and B[a]A/B[a]A +Chry in all station clearly indicate, the source of PAHs pollution in marine sediment sample of Tunku Abdul Rahman National Park areas were dominated by petroleum combustion (pyrolytic). The source of PAHs pollution from petroleum combustion probably originated from combustion of diesel and gasoline (petrol) from vehicle engine.

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