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Polycyclic Aromatic Hydrocarbons in Volatile and Particle Phases over the Vicinity of Petrochemical Refinery Areas

(Hidrokarbon Aromatik Polisiklik dalam Fasa Meruap dan Fasa Zarah di Persekitaran Kawasan Penapisan Petrokimia)

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ABSTRACT

The distribution of gaseous and particulate polycyclic aromatic hydrocarbons (PAHs), sources, and human exposure were studied around a petrochemical site located in Melaka, Malaysia from March 2021 - March 2022. Polyurethane (PUF) sampler devices were placed in six different areas to collect gaseous phase PAHs, and a High Volume Air Sampler (HVS) device was placed at a single location to collect $PM_{2.5}$-bound PAHs. PUF samplers utilize porous foam to absorb PAHs passively with zero external power for long-term exposure monitoring. Meanwhile, the HVS device draws air at higher flow rates for more than a day. A total of sixteen PAHs were analyzed in both particulate and gaseous phases. The average concentrations for the gaseous phase (n=48) were 15.90±27.29, 10.41±16.74, 7.47±18.18, 8.19±15.70, 9.39±19.35, and 11.19±28.35 ng/m^3 at Sri Vanathandavar Temple, Masjid Wadhi, Tadika Cahaya, Tadika Pasti, Monforth Youth Centre, and Maha Mariamman Temple, respectively, whereas the average concentration of particulate phase (n=35) was 0.24±0.23 ng/m^3 at SK Sungai Udang. In the gaseous phase, the seasonal variations at sampling sites in Southwest Monsoon (June – September) observed the greatest level at 13.89±4.69 ng/m^3 and the lowest during Intermonsoon 1 (October – November) at 8.22±5.26 ng/m^3. The diagnostic ratio showed that the primary contributors of PAHs in both phases are traffic emissions, petroleum and coal burning. The total Benzo(a)Pyrene equivalent carcinogenic (BaP_{eq}) exposure was 5.27 - 22.02 ng/m^3 in the volatile phase and 1.18 ng/m^3 in the particulate aerosol phase. For carcinogenic risk, the incremental lifetime cancer risk (ILCR) in adults was higher compared to children and adolescents in both gaseous and particulate phases. The Hazard Quotient (HQ) for the adolescent in the gaseous phase (9.86E-03) was relatively higher compared to the particulate aerosol phase (1.01E-03).

Keywords: Carcinogens; passive air sampling; petroleum sites; sources

ABSTRAK

Pengagihan hidrokarbon aromatik polisiklik (PAH) dalam gas dan sumber zarah serta pendedahan manusia kepada PAH telah dikaji di sekitar tapak petrokimia yang terletak di Melaka dari Mac 2021 hingga Mac 2022. Peranti pensampelan poliuretana (PUF) diletakkan di enam lokasi untuk mengumpul PAH fasa gas, manakala satu peranti pengambil sampel udara berkelantangan tinggi (HVS) digunakan di satu lokasi untuk mengumpul PAH terikat $PM_{2.5}$. PUF menyerap PAH secara pasif tanpa kuasa luar untuk pemantauan jangka panjang, sementara HVS menarik udara pada kadar aliran tinggi untuk beberapa hari. Sebanyak enam belas PAH telah dianalisis daripada fasa zarah dan gas. Kepekatan purata untuk fasa gas (n=48) ialah 15.90±27.29, 10.41±16.74, 7.47±18.18, 8.19±15.70, 9.39±19.35 dan 11.19±28.35 ng/m^3 masing-masing di Kuil Sri Vanathandavar, Masjid Wadhi, Tadika Cahaya, Tadika Pasti, Pusat Belia Monforth dan Kuil Maha Mariamman, manakala purata kepekatan zarah di SK Sungai Udang (n=35) ialah 0.24±0.23 ng/m^3. Dalam fasa gas, variasi bermusim di tapak pensampelan pada Monsun Barat Daya (Jun - September) memerhatikan kepekatan purata terbesar pada 13.89±4.69 ng/m^3 dan yang terendah semasa antara monsun 1 (Oktober - November) pada 8.22±5.26 ng/m^3. Nisbah diagnostik mendedahkan bahawa penyumbang utama PAH dalam kedua-dua fasa ialah pelepasan lalu lintas, petroleum dan pembakaran arang batu. Jumlah pendedahan karsinogenik setara Benzo(a)Pirena (BaP_{eq}) ialah 5.27 - 22.02 ng/m^3 untuk fasa gas dan 1.18 ng/m^3 untuk fasa zarah. Untuk risiko karsinogenik, peningkatan risiko kanser seumur hidup (ILCR) pada

orang dewasa adalah lebih tinggi berbanding kanak-kanak dan remaja dalam kedua-dua fasa gas dan zarah. Darjah bahaya (HQ) untuk kumpulan remaja dalam fasa gas (9.86E-03) adalah lebih tinggi berbanding fasa zarah (1.01E-03).

Kata kunci: Karsinogen; persampelan udara pasif; sumber; tapak petroleum

INTRODUCTION

Persistent organic pollutants (POPs), including polycyclic aromatic hydrocarbons (PAHs), have chemical configurations that include hydrogen and carbon (Abdel-Shafy & Mansour 2016). The United States Environmental Protection Agency (US-EPA) singled out 16 PAHs for their pronounced negative effects, and abundance in the environmental matrix (Ravindra, Wauters & Van Grieken 2008). PAHs can infiltrate different environments, including soil, air, water, and rocks, originating from natural processes and human activities (Biswa et al. 2020). PAHs originate from natural events like wildfires, volcanoes and genesis of fossil fuel but human activities greatly increase their presence in the environment.

PAHs can be categorized into two subgroups based on their respective molecular weight as in low molecular weight (LMW) and high molecular weight (HMW) PAHs. PAHs pose a substantial environmental and public health risk because of their enduring nature, toxicity, and carcinogenicity. Their environmental persistence can lead to their bioaccumulation in living organisms, including humans and subsequent harmful health implications (Sun et al. 2021). PAHs can lead to respiratory and cardiovascular issues, developmental disorders, and cancer in humans, along with oxidative stress, inflammation, and DNA damage. They can also harm aquatic life, causing developmental and reproductive issues (Sun et al. 2021; Vandana et al. 2022).

Numerous investigations in Malaysia have focused on the origins, impact, and amounts of particulate PAHs in various environmental matrices. The seasonal patterns and origins of particulate PAHs in particulate matter (PM_{10} and $PM_{2.5}$) collected from several sites throughout Malaysia were examined by Md Firoz et al. (2015), showing that Kuala Lumpur had the highest concentrations due to combustion processes like coal burning, biomass burning, and vehicle emissions. Nor Azura et al. (2019) studied PAHs' dispersion, origins, and potential health impacts during multiple rainy periods and hazy episodes in Kuala Lumpur, Malaysia. Their methodology included sampling particulate matter during different weather conditions and employing diagnostic ratios to identify PAH sources. The study reported that forest fires were a notable origin of PAHs during the haze episode because their concentration was higher than in the monsoon seasons.

Although extensive research has been conducted on particulate-phase PAHs in Malaysia, there remains a critical gap in understanding the concentrations, sources, and implications of gaseous-phase PAHs. This is particularly concerning because gaseous PAHs, which are present in significant quantities in the atmosphere, have a higher propensity to undergo reactions with oxidizing agents, forming more toxic derivatives (Jia et al. 2021b). Such reactions can exacerbate their health and environmental impacts. Gaseous PAHs are distinct from their particulate counterparts due to their volatility, enabling long-range atmospheric transport and posing unique health risks through inhalation.

Studies conducted in neighbouring regions such as Singapore and Taiwan have reported varying concentrations and behaviours of gaseous PAHs, highlighting the importance of localized investigations to better understand regional air quality and associated health outcomes. However, comparable data for Malaysia remains scarce, leaving a significant gap in the comprehension of air quality dynamics specific to this region.

This study addresses this pressing research gap by providing the first comprehensive dataset on gaseous-phase PAHs in Malaysia. By focusing on their concentrations, sources, and potential health implications, this research offers novel insights into an underexplored aspect of air pollution in the region, thereby contributing to informed policy-making and public health interventions. Therefore, the aims of this research were to (a) determine the level of PAHs in gaseous and particulate phases; (b) evaluate the risk factor of PAHs among multiple age groups near the petrochemical site, and (c) identify the plausible origins of PAHs near the petrochemical site.

METHODOLOGY

DESCRIPTION OF THE RESEARCH AREA AND SAMPLE COLLECTION

The sampling area comprised a total of seven sites near the Petrochemical Refinery Site which is located at Sungai Udang, Melaka (Figure 1). Within the Tangga Batu parliamentary constituency, the coordinates (2°16'34.14"N, 102°7'58.17"E) denote a busy area, primarily due to the presence of several nearby schools. Sampling sites were chosen in public places frequented by the targeted age group in this study.

PASSIVE AIR SAMPLING FOR GASEOUS SAMPLES

Six out of the seven sites, including Sri Vanathandavar Temple, Masjid Wadhi, Tadika Cahaya Sufi Caw, Tadika Pasti Al-Huda, Monforth Youth Centre, and Sri Maha Mariamman Temple were chosen to collect gaseous phase PAHs. These sites ranged from 0.9 km to 9.47 km away from the Petrochemical Refinery Site. Table 1 summarizes the

FIGURE 1. Six passive sampling sites for gaseous PAHs (white-colored box), one active sampling site for $PM_{2.5}$(orange- colored box) and a red-colored box representing a petrochemical site at Sungai Udang, Melaka

TABLE 1. Latitude and longitude of each passive sampling locations

Passive sampling site	Sampling Site	Latitude	Longitude	Distance from petrochemical site
Sri Vanathandavar Temple	Industrial	2°17'1.78"N	102° 8'7.47"E	0.9 km
Masjid Wadhi	Industrial	2°16'13.19"N	102°8'58.60"E	1.97 km
Tadika Cahaya Sufi Caw	Rural	2°15'34.89"N	102° 9'9.41"E	2.86 km
Tadika Pasti Al-Huda	Rural	2°15'15.58"N	102°10'54.55"E	5.96 km
Monforth Youth Centre	Roadside	2°16'19.51"N	102°10'7.47"E	4 km
Dewi Sri Mariamman Temple	Residential	2°16'47.77"N	102°13'4.47"E	9.47 km

latitude and longitude of each sampling site for collecting gaseous phase PAHs. Passive air samplers, positioned 15-20 meters above ground, were used for monthly sampling between March 2021 and March 2022.

Polyurethane foam (PUF) (TISCH TE-1014, ½ × 5½) was used to collect gaseous samples. The PUFs were positioned horizontally in between two stainless steel bowls at the middle axis. The stainless bowls were used to prevent interferences from climatic conditions like rain or direct sunlight. The bowls were wiped and cleaned with n-hexane before fixing them. PUF was used due to its cost-efficiency, ease of extraction, and high collection efficiency (Guo et al. 2011).

HIGH VOLUME SAMPLING (HVS) FOR $PM_{2.5}$ SAMPLE

In this study, airborne particulates were collected using a high volume sampler (HVS) with a quartz fiber filter (203 mm × 254 mm, Whatman™ UK) at a flow rate of 1.13 m^3 min^{-1}. HVS is a device used to collect large volumes of ambient air samples. By passing air through a pre-weighed filter, suspended particles are captured, enabling further assessment of the particulate matter collected. Particles exceeding 10 µ are retained on the impaction plate of the sampling head, whereas smaller particles effectively pass through to be captured on the filter. Particulate samples were collected continuously for 24 h at 12.00 pm from January 2022 to March 2022 at Sekolah Kebangsaan (SK) Sungai Udang, Melaka. SK Sungai Udang was located 2 km from the petrochemical refinery site.

Quartz fiber filters were utilized to collect particulate PAHs which were pretreated before sampling by baking at 500 °C for 5 h to get rid of any probable impurities (Kim et al. 2012). The filters were heated and then kept in a desiccator for a whole day (24 h). The filters were measured using a five-digit high-resolution electronic

microbalance (Denver, Model TB-2150, USA) before and post-sampling to identify the mass of particulates. The filters were then sampled, secured in aluminum foil, tied up in plastic bags with zipper and refrigerated at 4 °C until further investigations were conducted (Watson et al. 2017).

EXTRACTION OF GASEOUS SAMPLES

The PUFs were pretreated by cleaning with soxhlet-extracted using n-hexane and acetone (1:1) (Kim et al. 2012) for 24 h, then dried in a pre-cleaned bell jar chamber and kept in a sealed aluminum foil until collection. After 30 days of sampling, the PUFs were cautiously pulled out from the stainless-steel bowl and stored in a pre-cleaned zipped aluminum foil. These samples were maintained at -20 °C before continuing with extraction. After sampling, the collected PUFs were soxhlet-extracted by following the procedure implied by Srimurali et al. (2015). Extract from PUF disks was collected for 6 h at 40 °C in 200 mL of acetone and n-hexane (1:1). A rotary vacuum evaporator was utilized to decrease the extract to 1 mL, while pre-concentration was completed by solid phase extraction (SPE) cartridges. A volume of 25 mL mixture consisting of n-hexane: dichloromethane in a ratio of 1:1 was utilized to elute the PAHs from the cartridge that had been conditioned by 10 mL of n-hexane. The eluents were reduced to 500 µL with the aid of nitrogen gas (N_2) and then fed into gas chromatography-flame ionization detection (GC-FID).

The concentration of targeted gaseous PAHs in the atmosphere was obtained using the equation herewith:

$$\text{PAH in ambient air (ng/m}^3\text{)} = C / (3.5 \times 30) \qquad (1)$$

where C represents the concentration of PAHs in PUF, 3.5 reflects the PAS linear sampling rate and 30 represents the sampling period in days.

EXTRACTION FOR PARTICULATE SAMPLES

For particulate phase analysis, the filters were divided up into tiny pieces and put inside a small beaker. The particulate phase extraction procedure was conducted according to a prior study by Md Firoz et al. (2015). Initially, 20 mL of dichloromethane (DCM) solvent was added to the beaker containing the filter sample. The filter samples were extracted by ultrasonic agitation for 20 min and repeated two more times. After filtering the extracts with glass microfibre filters (Whatman™, UK), the resulting solution was lessened to a volume of 200 µL while passing through a mild stream of N_2 gas. The extraction process was continued with the pre-concentration of extracts using SPE cartridges. Upon conditioning the cartridge with 10 mL of n-hexane under a light suction, the cartridges were filled with the extract solutions and the elution of DCM: n-hexane (1:9) was then carried out. The collected elutes were then reduced to 500 µL using N_2 gas and further tested by GC- FID.

ANALYSIS USING GC-FID

GC-FID (Agilent Technologies 7890A, GC System, USA) was employed to assess the samples. The capillary column used was (DB-5MS) with an internal diameter of 30 m × 0.25 mm, length of 30 m and 0.25 µm film thickness. The GC column temperature was configured to start at 60 °C, increase to 150 °C at a rate of 20 °C min^{-1}, increase to 300 °C at a rate of 4 °C min^{-1}, and then reach the isothermal mode for 6 min at 300 °C. The carrier gas, helium (He), was kept flowing at a rate of 1.0 mL per min.

QUALITY ASSURANCE AND QUALITY CONTROL (QA/QC)

In this analysis, five calibration mixtures were prepared from a standard solution of PAHs (Sigma-Aldrich's Certified Standard Reference Material (CRM47543, Polycyclic Aromatic Hydrocarbons Mix; 1 × 1 mL; 2000 µg mL^{-1}). The mixture was diluted to 0.1 ppm, 0.5 ppm, 1.0 ppm, 2.0 ppm, and 3.0 ppm and treated as a reference solution which was analyzed using GC-FID with a similar method as a sample. The mixture containing 16 PAH standards, namely, naphthalene (Nap), acenaphthene (Ace), acenaphthylene, (Acy), anthracene (Ant), fluorene (Flr), phenanthrene, (Phe), fluoranthene (Flt), pyrene (Pyr), benzo(a)anthracene (BaA), chrysene (Chr), benzo(b)fluoranthene (BbF), benzo(k)fluoranthene (BkF), benzo(a)pyrene (BaP), indeno(1,2,3-cd)pyrene (IcP), dibenzo(h)anthracene (DhA), and benzo(g,h,i)perylene (BgP). Each compound detected was quantified using a standard calibration curve and its retention time served as an indicator for the sample. With correlation values of $R^2 \geq 0.99$, the calibration curves of the targeted 16 PAHs indicated fairly good linearity. The average recoveries of naphthalene-d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12, and perelyne-d12 were in the range of 66.1% - 106.9% for gaseous and 83% - 137.2% for particulate PAHs, respectively.

Field and laboratory blanks were used to examine quality control and quality assurance. Laboratory method blank is to check on any potential contamination in the lab where the unused filter/PUF is extracted using the same method as the sample filter/PUF. Additionally, during each sampling period, at least one PUF and filter were transported to the field and back without air passing through the sampler, acting as field blanks (Wang et al. 2019). To volatilize and eliminate any organic contaminants, all apparatus used in this method was washed, soaked in solvent (hexane), and cleaned with purified water before being baked (5 h at 200 °C) (Anas et al. 2014). A solvent blank was injected before and after each calibration set. The targeted compounds were not detected in field blanks or laboratory blanks.

SOURCE APPORTIONMENT USING DIAGNOSTIC RATIO (DR)

The source of PAHs in the surrounding atmosphere can be identified through qualitative analysis. DR is commonly

used as a standard strategy to recognize the potential origins of PAH congeners. Implementing this approach has the notable benefit of being more detailed and simpler than other techniques. The most prevalent diagnostic ratios in the present study were Ant/Ant+Phe, Flt/Flt + Pyr, BaA/BaA + Chr, IcP/IcP + BgP, BaP/BgP, and BaA/Chr (Nur Ain Nazirah et al. 2023). Selected PAHs have well-documented environmental behavior and detection limitations, allowing accurate quantification across various studies. Selective PAHs for DRs are chosen due to their stability and resistance to environmental degradation, making them reliable long-term monitoring markers. Furthermore, several PAHs are classified as priority pollutants due to their toxicological characteristics, emphasizing their significance in environmental assessments. In short, the use of certain PAHs for diagnostic ratios is justified by their distinct production patterns, stability, relative abundance, suitability for accurate analysis, and regulatory compliance (Mali et al. 2022).

HEALTH RISK ASSESSMENT (HRA) OF PAHS

Human exposure to both carcinogenic and non-carcinogenic PAHs may lead to carcinogenesis and adverse health outcomes. Thus, it is critical to ascertain whether the PAH levels at the sampling sites pose significant health risks. PAH exposure in humans occurs through inhalation, ingestion or dermal exposure (Sousa et al. 2022). This study examines the effects of inhaling PAHs on children, adolescents, and adults by calculating the HRA.

BaP, a potent carcinogenic PAH, serves as a standard for comparing the toxicity of other PAHs. The combined toxicity of various PAHs can be quantified as a single value relative to BaP through the calculation of benzo(a) pyrene toxicity equivalent concentration (BaP_{eq}). This method is commonly used in published journals to assess the health risks of PAHs (Md Firoz et al. 2015; Pu et al. 2022). Equation (2) can be applied to calculate the BaP_{eq} to estimate the toxicity and health risk of PAHs (Yu et al. 2008):

$$BaP_{eq} = C_i \times TEF \quad (2)$$

where C_i stands for the concentration of targeted PAH and TEF denotes the compound's toxic equivalency factor.

Before evaluating exposure to carcinogenic and non-carcinogenic PAHs, it is essential to compute the lifetime average daily dose (LADD) and average daily dose (ADD) for each constituent in the samples. This involves utilizing the following formula.

$$\text{LADD (mg kg}^{-1}\text{ day}^{-1}) = \frac{(Cs \times IR \times CF \times EF \times ED)}{(BW \times ALT)} \quad (3)$$

where Cs means the cumulative of the converted intensities of PAHs in atmospheric particles (ng/m^3) according to TEQ value, BW represents body weight (kg), the air-breathing rate (m^3/day) is represented by IR, CSF stands for breathing cancer slope variable (3.85 mg kg^{-1} day^{-1}), CF for unit conversion factor (1×10^{-6} mg/kg), EF denotes the exposure frequency (day/year), ALT stands for mean period for carcinogens (days) and the lifetime exposure period is denoted by ED (Amit et al. 2020). Table 2 presents the reference values for all constants used in computing LADD and ADD.

The ILCR is determined using particular formulas that include exposure doses, exposure time and exposure frequency. The incremental risk of acquiring cancer during a lifetime as a result of exposure to the pollutant is represented by the unitless ILCR value that these computations provide (Amit et al. 2020):

$$\text{ILCR} = \text{LADD} \times \text{CSF} \quad (4)$$

where cancer slope factor with unit (mg kg^{-1} day^{-1})$^{-1}$ denotes CSF. The CSF value is proposed to be 3.14 (mg kg^{-1} day^{-1})$^{-1}$ (Nor Azura et al. 2019). LADD signifies the potential chemical intake per kilogram of body weight daily, with prolonged exposure posing health risks (Anas Jamhari et al. 2021).

As per U.S. Environmental Protection Agency (USEPA) guidelines, HQ evaluates PAH exposure relative to a concentration considered safe. HQ is crucial for assessing non-carcinogenic PAH exposure, derived from the computed average daily dose (ADD) using Equation (5):

$$HQ = \frac{ADD}{R_fD} \quad (5)$$

Every PAHs compound has a different reference dose (R_fD) value, detailed in Table 3.

RESULTS AND DISCUSSIONS

PAHS SIZE DISTRIBUTIONS IN THE ATMOSPHERE

The summary concentration of 16 PAHs in the gaseous phase (n=48) and particulate phase (n=35) observed in specific areas around Sungai Udang, Melaka from April 2021 to March 2022 is shown in Table 4.

The highest average concentration of gaseous PAHs was measured at Sri Vanathandavar Temple (15.9 ± 27.29) ng/m^3, as this location was the nearest to the petrochemical site at Sungai Udang, Melaka with a radial distance of 2 km away. Sri Vanathandavar has the most chaotic traffic and intensive industrial activities compared to other five locations. The use of petroleum as fuel and the oil and gas industry are the two primary sources of PAH exposure in this area. Following Sri Vanathandavar, Sri Maha Mariamman Temple and Masjid Wadhi recorded greater concentrations of (11.9 ± 28.35) ng/m^3 and (10.41 ± 16.74) ng/m^3, respectively. Both locations are residential and urban areas that are exposed to higher traffic emissions.

The lowest concentration was measured at Tadika Cahaya with a value of (7.47 ± 18.18) ng/m^3. This is likely due to the sampling site's rural location with a smaller population. Table 5 lists a significant comparison of the gaseous level of this study from previous works at some cities around. The gaseous concentration of six passive sampling sites in this study observed to be lower than studies conducted at Harbin, China (68.3 ± 22.3 ng/m^3) (Liu et al. 2021), Osaka, Japan (48.39 ± 16.45ng/m^3) (Kishida et al. 2011) whereas higher than at Gosan, Korea (1.4 ng/m^3) (Kim et al. 2012). The atmospheric parameters significantly affect values of PAHs in all areas, though emission sources at specific study areas may influence their levels in the environment.

The average concentration of particulate PAHs at SK Sungai Udang was measured at 0.24 ± 0.23 ng/m^3, which is significantly lower than concentrations reported from various regions (Table 5). For instance, previous studies have documented higher particulate PAH levels in several countries: Bengbu, China, reported an average of 10.06 ± 8.04 ng/m^3; Nepal showed a concentration of 45.1 ± 32.8 ng/m^3; Kuala Lumpur, Malaysia, recorded 1.74 ± 2.68 ng/m^3; and Ulsan, Korea, measured 2.55 ng/m^3 (Table 5). In a related context, industrial air pollution and vehicular emissions near two elementary schools in northern Portugal resulted in elevated PAH levels, with measurements reaching 20 ng/m^3 and 48 ng/m^3 at a petrochemical site (Oliveira et al. 2017). These findings suggest that PAH pollution in suspended particulate matter at SK Sungai Udang, Melaka is relatively low. Thus, this study's concentrations of gaseous and particulate PAHs comply with the Occupational Safety and Health Administration (OSHA)'s standard of 0.2 mg/m^3 PAH exposure (Lee et al. 2015).

PAHs can be categorized into five groups according to the count of aromatic rings they consist of as follows: two-ring PAHs (Nap), three-ring PAHs (Acy, Ace, Flr, Phe and Ant), four-ring PAHs (Flt, Pyr, BaA and Chr), five-ring PAHs (BbF, BkF, BaP and DhA), and six-ring PAHs (IcP and BgP). In this study, the gaseous phase is predominated by three-ring PAHs, while five-ring PAHs dominate the particulate phase. It can be observed that LMW PAHs are highly likely to be found in gaseous forms due to their higher volatility, whereas the opposite would apply to heavier PAHs (Cincinelli et al. 2007; Kamal Hassan & Khoder 2012). Phe was observed to be the most dominant gaseous PAH in this study (Figure 2(a)), contributing about 50% of the total concentration. BaP level was the highest in particulate PAHs followed by BgP and BbF at SK Sungai Udang (Figure 2(b)). Previous research in similar urban settings with high traffic volume also indicates that BaP, BbF, and BgP are the most prevalent PAHs in the particulate phase (Elzein et al. 2020; Mohammad Sadegh et al. 2015).

TABLE 2. Standard values to estimate LADD

Exposure parameter	Unit	Children	Adolescent	Adult
IR	m^3 day^{-1}	12	15.7	15.7
ED	Years	6	6	12
EF	Days year^{-1}	350	350	350
Body weight	Kg	31.2	38	66
Average life span (AT)	Years	70,6	70,6	70,24

*AT for carcinogens (LADD) (fixed at 70 years of exposure)
**AT for non-carcinogens (ADD) (Average years of exposure)

TABLE 3. Reference dose (R_fd) value for each PAH compound (mg/kg/day)

Compound	R_fD (ng/kg day)	R_fD (mg/kg day)
Ace	60000	0.06
Flr	40000	0.04
Ant	300000	0.3
Flt	40000	0.04
Pyr	30000	0.03
BaP	300	0.0003
BgP	30000	0.03

TABLE 4. Mean concentration of gaseous and particulate PAHs in Sungai Udang sampling sites

PAHs	Gaseous (ng/m^3) [n=48]						Particulate (ng/m^3) [n=35]
	Sri Vana Temple	Masjid Wadhi	Tadika Cahaya	Tadika Pasti	Monforth	Sri Maha Mariamman	SK Sungai Udang
Nap	9.09 ± 8.21	3.21 ± 2.84	3.29 ± 3.43	4.34 ± 5.05	3.44 ± 3.43	6.34 ± 6.49	0.02 ± 0.02
Acy	8.58 ± 4.66	4.41 ± 3.36	3.62 ± 3.47	2.57 ± 1.33	3.45 ± 4.18	5.7 ± 5.56	0.29 ± 0.21
Ace	1.34 ± 0.73	1.59 ± 1.90	1.11 ± 0.95	2.48 ± 2.51	1.76 ± 2.12	2.49 ± 2.02	0.02 ± 0.03
Flr	29.51 ± 26.51	4.95 ± 3.45	4.8 ±4.23	5.19 ± 4.04	5.52 ± 4.24	7.52 ± 7.23	0.16 ± 0.15
Phe	114.51± 67.42	69.08 ± 45.23	74.66 ± 48.92	64.55 ± 51.85	80.32 ± 61.43	116.53 ± 129.56	0.07 ± 0.05
Ant	19.13 ± 19.93	11.47 ± 7.72	8.83 ± 3.76	9.86 ± 6.69	12.28 ± 7.40	14.01 ± 17.04	0.33 ± 0.29
Flt	12.43 ± 4.49	12.81 ± 3.51	11.1 ± 4.90	19.64 ± 27.30	15.78 ± 9.63	10.26 ± 6.30	0.09 ± 0.09
Pyr	5.66 ± 6.89	1.45 ± 1.36	0.63 ± 0.82	1.92 ± 1.51	0.96 ± 0.74	1.1 ± 0.92	0.1 ± 0.08
BaA	6.72 ± 8.92	23.03 ± 61.94	0.17 ± 0.48	1.68 ± 1.40	0.93 ± 1.01	0.44 ± 0.48	0.15 ± 0.14
Chr	4.82 ± 3.71	1.67 ± 1.03	2.06 ± 1.19	2.63 ± 1.27	2.57 ± 1.57	2.27 ± 2.20	0.08 ± 0.05
BbF	0.56 ± 0.80	11.74 ± 23.54	0.99 ± 1.61	4.01 ± 5.46	2.86 ± 4.83	1.43 ± 1.69	0.56 ± 1.22
BkF	7.53 ± 17.13	8.93 ± 22.33	1.28 ± 1.02	3.47 ± 4.91	1.92 ± 0.66	1.3 ± 0.97	0.11 ± 0.10
BaP	16.52 ± 14.48	6.79 ± 7.90	3.28 ± 3.10	3.08 ± 1.55	6.69 ± 8.26	5.08 ± 4.98	0.73 ± 1.81
IcP	6.3 ± 15.07	0.49 ± 0.93	0.21 ± 0.23	0.63 ± 0.73	6.54 ±16.37	0.6 ± 1.04	0.07 ± 0.04
DhA	2.87 ± 2.36	2.75 ± 1.90	1.5 ± 1.33	2.73 ± 1.74	2.24 ± 1.44	1.37 ± 1.25	0.42 ± 0.36
BgP	8.85 ± 12.39	2.14 ± 1.98	1.98 ± 1.98	2.19 ± 1.53	3.02 ± 1.60	2.53 ± 3.17	0.66 ± 1.50
Total PAHs	254.43	166.5	119.5	130.97	150.29	178.98	3.86
Mean	15.9	10.41	7.47	8.19	9.39	11.19	0.24
Stdev	27.29	16.74	18.18	15.7	19.35	28.35	0.23

TABLE 5. Comparison of gaseous and particulate PAH concentration (ng/m^3) observed in this present study with those from other part of countries

Location	Site type	Type of PAHs	Mean values (ng/m^3)	Total PAHs	References
Sungai Udang, Melaka	Six sites	Gaseous	7.47 ± 18.18 to 15.90 ± 27.29	Σ16 PAHs	Current study
Delhi, India	Urban	Gaseous	37.63 ± 13.62	Σ 17 PAHs	Singh et al. (2023)
Singapore	Urban	Gaseous	5.3 to 277.2	Σ 16 PAHs	He & Balasubramanian (2010)
Gosan, Korea	Urban	Gaseous	1.4	Σ14 PAHs	Kim et al. (2012)
Harbin, China	Urban	Gaseous	68.3 ± 22.3	Σ15 PAHs	Liu et al. (2021)
Osaka, Japan	Urban	Gaseous	48.39 ± 16.45	Σ28PAHs	Kishida et al. (2011)
SK Sungai Udang, Melaka	Urban	Particulate	0.24 ± 0.23	Σ16 PAHs	Current study
Ulsan, Korea	Semi-rural	Particulate	2.5	Σ13 PAHs	Nguyen et al. (2018)
Bengbu, China	Urban	Particulate	10.06 ± 8.04	Σ16 PAHs	Wu et al. (2024)
Nepal	Urban	Particulate	45.1 ± 32.8	Σ16 PAHs	Yadav et al. (2018)
Kuala Lumpur, Malaysia	Urban	Particulate	1.74 ± 2.68	Σ16 PAHs	Hamidah et al. (2021)

SEASONAL VARIATION OF GASEOUS AND PARTICULATE PAHs

The gaseous phase samples were gathered from March 2021 to April 2022. The sampling period was divided into the southwest (SW) monsoon from June to September, inter-monsoon 1 (INTI) from October to November, northeast monsoon (NE), from December to March, and inter-monsoon 2 (INT2) from April to May. SW monsoon observed the highest concentration (13.89 ± 4.69 ng/m^3) in the gaseous phase, followed by IM 2 (10.19 ± 6.828 ng/m^3), NE monsoon (9.00 ± 4.47 ng/m^3) and IM1 (8.22 ± 5.26 ng/m^3) as shown in Figure 3(a).

According to Figure 3(b), the concentration of gaseous PAHs in SW monsoon (33%) is higher than in NE monsoon (22%) which is in line with the research done by a few other studies with several emission sources (Anas Jamhari et al. 2021; Md Firoz et al. 2015; Nor Azura et al. 2019). The PAHs level in the SW monsoon showed that transboundary pollution transport, particularly biomass burning from Southeast Asia always occurs during the dry season (Fujii et al. 2015; Md Firoz et al. 2015). Biomass burning happens in dry months due to agricultural activities in peat soil regions in Southeast Asia (SEA), mainly in Sumatra and Kalimantan, Indonesia (Field, Van Der Werf & Shen 2009; Pongpiachan & Paowa 2015). Pollutants released from biomass-burning sites can spread to neighboring nations, more noticeably to Malaysia, Singapore, and Thailand owing to the airflow of the southwest monsoon (Aouizerats et al. 2015; Balasubramanian et al. 2003). During these events, atmospheric concentration in the ambient air can collect regional pollutants like those from automobile and manufacturing processes (Adelin et al. 2010).

In the NE monsoon, it was found that the gaseous PAH concentration was higher than the particle concentration. The concentration of PAH in the particulate phase was significantly lower, with an average value of (0.24 ± 0.23) ng/m^3 and ranged from 0.02 to 0.69 ng/m^3. The frequent rainfalls during the NE monsoon are linked to the cool conditions at the time, making it ideal for PAHs to condense in the atmosphere (Garban et al. 2002). However, a previous study stated that this condensation mostly affects PAHs with a certain concentration in the gaseous phase rather than those with five aromatic rings (Ravindra, Wauters & Van Grieken 2008). Environmental factors including heat and solar transmission have less effect on PAHs in the gaseous phase than the particle phase (Jia et al. 2021a).

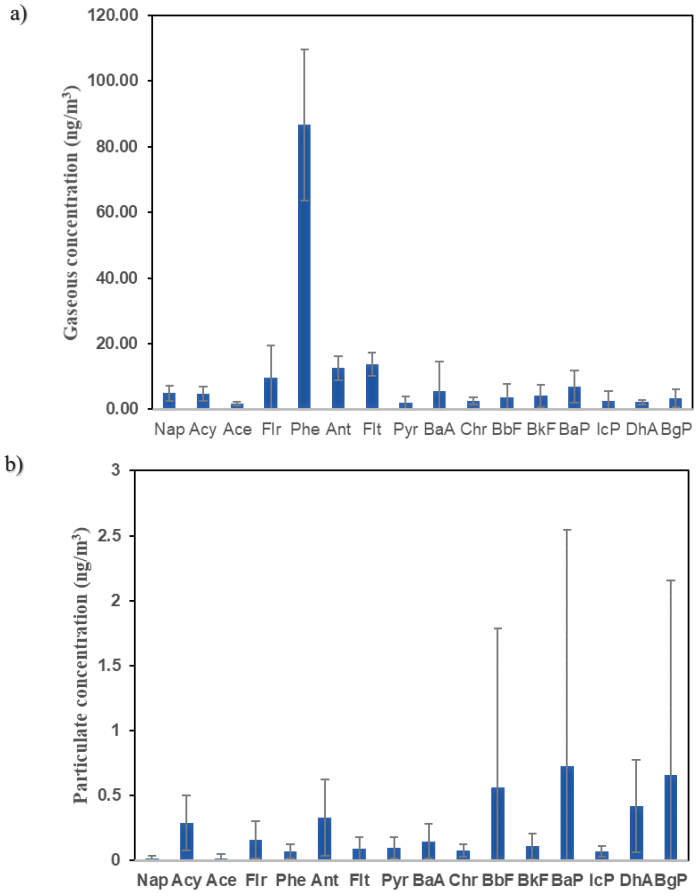

FIGURE 2. Average concentration of (a) gaseous PAHs and (b) particulate PAHs

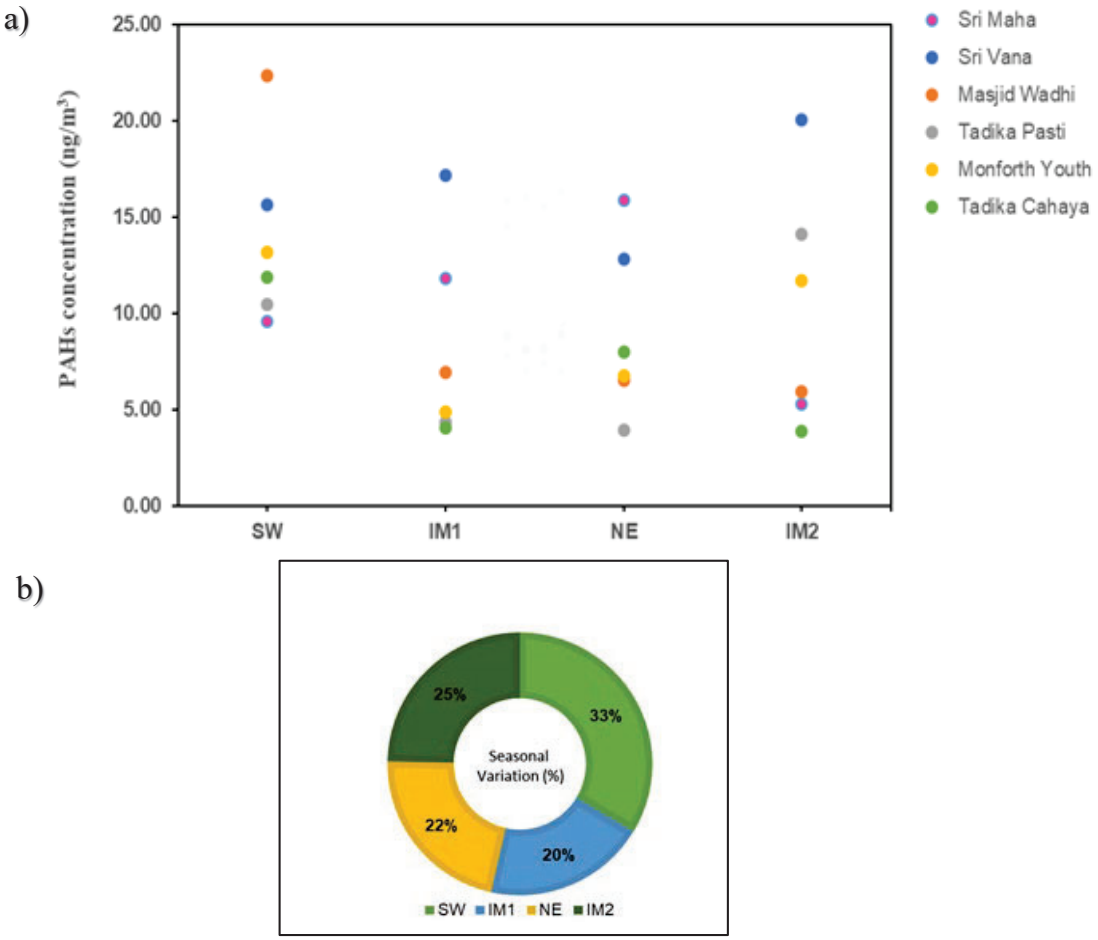

FIGURE 3. (a) The mean concentration (ng/m^3) of seasonal variation for gaseous PAHs and (b) Composition of gaseous PAHs in different seasons

PAHs DIAGNOSTIC RATIO (DR)

Table 6 shows the DRs of the chosen PAHs and their associated sources (Akyüz & Çabuk 2010; Brändli et al. 2008; Manoli, Kouras & Samara 2004; Parshetti et al. 2010; Yunker et al. 2002). Cluster analysis of ratios that represent various sources are shown in Figure 4.

The DR value of Ant/(Ant +Phe) at 0.13 indicated that the primary source of gaseous PAHs is pyrogenic. Coal or wood combustion is one of the sources of gaseous PAHs, as indicated by the DR value of Flt/(Flt+ Pyr) at 0.87. PAHs are likely released from both pyrogenic and wood-burning activities according to BaA/(BaA+ Chr) 0.67 DR. The DR values of I[c]P/(I[c]P+ BgP) and B[a]P/(B[g]P) are 0.42 and 2.00, respectively, indicating that the sources of PAHs include traffic emissions (Yunker et al. 2002) and activities such as cracking of petroleum into lighter hydrocarbons and coal combustion. PAH compounds like phenanthrene, fluoranthene, and anthracene were dominant in all gaseous phase samples since sampling regions were located around the petrochemical site which refines and processes crude oil into refined products (Kulkarni et al. 2014). The DR values of Ant/(Ant+Phe), BaA/(BaA+Chr) and Flt/(Flt+ Pyr) in particulate phase PAHs indicated sources from wood or coal combustion, as well as transportation emissions (Jiang et al. 2014; Yang et al. 2013).

BaP EQUIVALENT

The BaP_{eq} values in Table 7 range from (5.273 to 22.016) ng/m^3 for the gaseous phase and 1.176 ng/m^3 for the particulate phase. The order of PAH toxicity of each sampling site ranged as follows Sri Vanathandavar > Masjid Wadhi > Monforth Youth Centre > Tadika Pasti> Maha Mariamman Temple > Tadika Cahaya >Sk Sungai Udang. Among the six places for the gaseous phase, sampling sites Sri Vanathandavar and Masjid Wadhi had higher BaP_{eq} values since these locations were comparatively nearest to the emission sources. The BaP_{eq} value of the particulate phase at SK Sungai Udang was 1.176 ng/m^3 and 5-6 ring PAHs were responsible for almost 95% of the hazard at all sampling sites.

TABLE 6. Potential sources of PAHs based on diagnostic ratios associated with gaseous and particulate phases in areas nearing a petrochemical site

Diagnostic ratio	Indicator sources	Gaseous PAHS	Particulate PAHs
Ant/ (Ant+Phe)	<0.1: Petrogenic	0.13 - Pyrogenic	0.82 - Pyrogenic
	>0.1: Pyrogenic		
Flt/(Flt+Pyr)	<0.4: Petrogenic	0.87 - Pyrogenic, grass, wood, coal combustion	0.47 - Pyrogenic, Fuel oil
	>0.4: Pyrogenic		
	0.4 - 0.5: Fuel oil		
	>0.5: Grass, wood, coal		
	0.6 - 0.7: Diesel		
	0.4: Gasoline		
BaA/(BaA+Chr)	<0.2: Petrogenic	0.67 - Pyrogenic, wood burning	0.66 - Pyrogenic, Wood Burning
	>0.35: Pyrogenic		
	0.2 - 0.35: Coal		
	>0.5: Wood burning		
IcP/ (IcP + BgP)	<0.2: Petrogenic	0.42 - Petroleum, gasoline, diesel	0.10 - Petrogenic
	>0.2: Pyrogenic		
	0.2 - 0.5: Petroleum/ Gasoline		
	>0.5: Grass, wood, coal		
	0.82: Oil combustion		
	0.35 - 0.70: Diesel		
BaP/BgP	<0.6: Nontraffic	2.00 - Traffic	1.10 - Traffic
	>0.6: Traffic		

Sri Vanathandavar and Masjid Wadhi have high BaP_{eq} values, making them harmful to breathe compared to other sampling sites. Although BaP is considered the most prominent PAH in the particulate phase, the BaP_{eq} value was higher in the gas phase than in the particulate phase. Although the TEF value showed that the dominant PAH compounds are low particularly in the gas phase, it should be routinely monitored for evaluating IAQ and the associated health hazards in considering the current WHO IAQ guidelines and its significant abundance (WHO 2010).

CARCINOGENIC EXPOSURE

INCREMENTAL LIFETIME CANCER RISK (ILCR)

It was observed that the concentration follows the order of adults> adolescents > children. Adults recorded the highest concentration with a total value of 3.91×10^{-5} mg/kg per day for the gaseous phase compared to the particulate phase (1.52×10^{-7} mg/kg per day). Adults may have higher levels of PAHs due to some jobs, smoking, or living in polluted areas. Jobs like construction, manufacturing, and certain agriculture fields may increase exposure to PAHs in adults (Adeyeye et al. 2023).

The ILCR computation proves that adults have a higher risk of developing cancer from PAH inhalation than adolescents and children (Figure 5). It is imperative to recognize that adults are at risk as they may inhale cancer-causing PAHs and be exposed to polluted air outdoors for longer periods compared to other age groups (Chen et al. 2019). Previous studies also reported that the ILCR value in the adult age group is higher than in children and adolescents (Hamidah et al. 2021; Nor Azura et al. 2019).

BaP from particulate phase has the highest ILCR value of 8.91×10^{-8} (Figure 5(b)) in adult age group, indicating that adults who inhale BaP are at an increased risk of cancer at a rate of 8 to 9 in 10^8 adults. In the gaseous phase, Phe showed a high ILCR value with a total value of 5.16×10^{-5}, 5.54×10^{-5}, and 6.38×10^{-5} in children, adolescents, and adults, respectively. Although Phe had

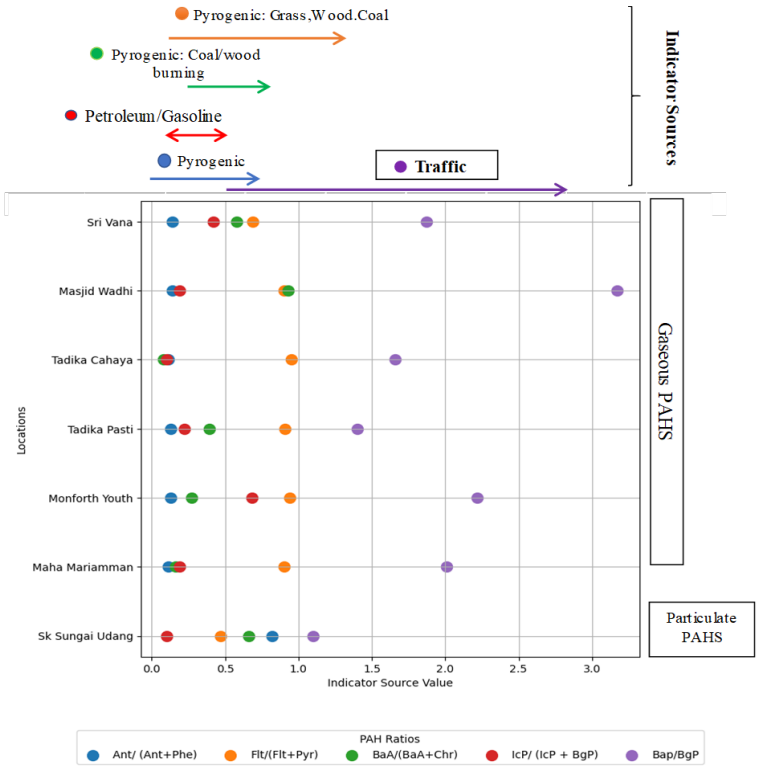

FIGURE 4. Cluster analysis of diagnostic ratios for different emission sources

TABLE 7. The toxicity of individual PAH based on BaP_{eq} concentration (ng/m^3)

PAHs	Concentration (ng/m^3)						
	Gaseous						Particulate
	Sri Vana	Masjid Wadhi	Tadika Cahaya	Tadika Pasti	Monfort Youth	Sri Maha Mariamman	SK Sungai Udang
Nap	0.01	0.00	0.00	0.00	0.00	0.01	0.00
Acy	0.01	0.00	0.00	0.00	0.00	0.01	0.00
Ace	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Flr	0.03	0.00	0.00	0.01	0.01	0.01	0.00
Phe	0.11	0.07	0.07	0.06	0.08	0.12	0.00
Ant	0.19	0.11	0.09	0.10	0.12	0.14	0.00
Flt	0.01	0.01	0.01	0.02	0.02	0.01	0.00
Pyr	0.01	0.00	0.00	0.00	0.00	0.00	0.00
BaA	0.67	2.30	0.02	0.17	0.09	0.04	0.01
Chr	0.05	0.02	0.02	0.03	0.03	0.02	0.00
BbF	0.06	1.17	0.10	0.40	0.29	0.14	0.05
BkF	0.75	0.89	0.13	0.35	0.19	0.13	0.01
BaP	16.52	6.79	3.28	3.08	6.69	5.08	0.71
IcP	0.63	0.05	0.02	0.06	0.65	0.06	0.01
DhA	2.87	2.75	1.50	2.73	2.24	1.37	0.38
BgP	0.09	0.02	0.02	0.02	0.03	0.03	0.01
Total	22.02	14.21	5.27	7.04	10.45	7.17	1.18

the highest concentration of PAHs, its low TEF meant it contributed minimally to the total risk. Flt had the second-highest ILCR value and is mutagenic despite being a mild carcinogen (Ramírez et al. 2011). According to the USEPA, the permissible limits for ILCR values or human exposure to carcinogenic PAHs are 1.00×10^{-6} and 1.00×10^{-4}. Based on the ILCR values derived for all the age groups (children, adults, and adolescents) in both gaseous and particulate, it can be seen that the values are in the range of 1.00×10^{-5} to 1.00×10^{-9}. This indicates that children, adults and adolescents staying near the petrochemical areas have a minimal risk of developing cancer.

HAZARD QUOTIENT

The ADD level in adolescents was higher than in children and adults. The ADD level measured for the adolescent age group in the gaseous phase (6.89×10^{-5}) was higher than the particulate phase (1.60×10^{-6}).

Figure 6 proved that the adolescent age group has the highest HQ value compared to adults and children in the gaseous phase and particulate phase with an average value of 0.01 and 1.01×10^{-3}, respectively. This indicates that adolescents are more likely to develop non-cancer illnesses as a result of inhaling PAHs than adults. HQ for BaP was measured to be the highest among other compounds in every sampling site for three age groups. Thus, BaP has the most significant potential to harm health or to alter body functions. These findings suggested that BaP specifically played a significant role in increasing health hazards associated with high molecular weight PAHs.

The hazard index (HI) can be used to indicate the hazard quotients of several toxins that have comparable harmful effects on health which can be figured by adding up all the HQ values. The USEPA only permits HI levels below 1. The HI values for all age groups in gaseous and particulate calculated were 5.51×10^{-2} and 9.44×10^{-4} for children, 5.92×10^{-2} and 1.01×10^{-3} for adolescents, and 4.1×10^{-2} and 3.57×10^{-4} for adults, respectively. The computed values of HI for all sampling sites were below 1. As a result, inhaling PAHs in this study location has a less negative impact on adults, children and adolescents' health.

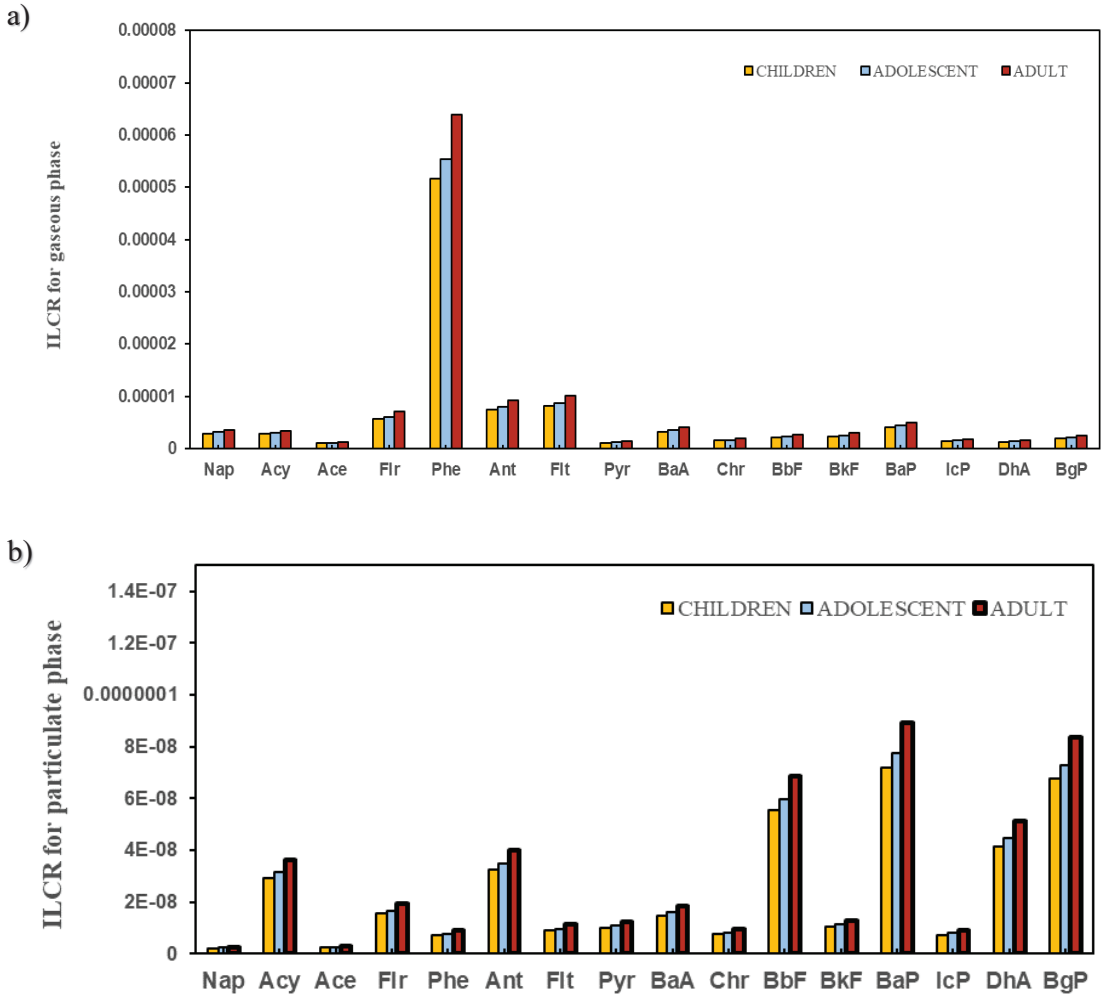

FIGURE 5. Total ILCR values of (a) gaseous phase and (b) particulate phase for children, adolescent, and adult age groups, respectively

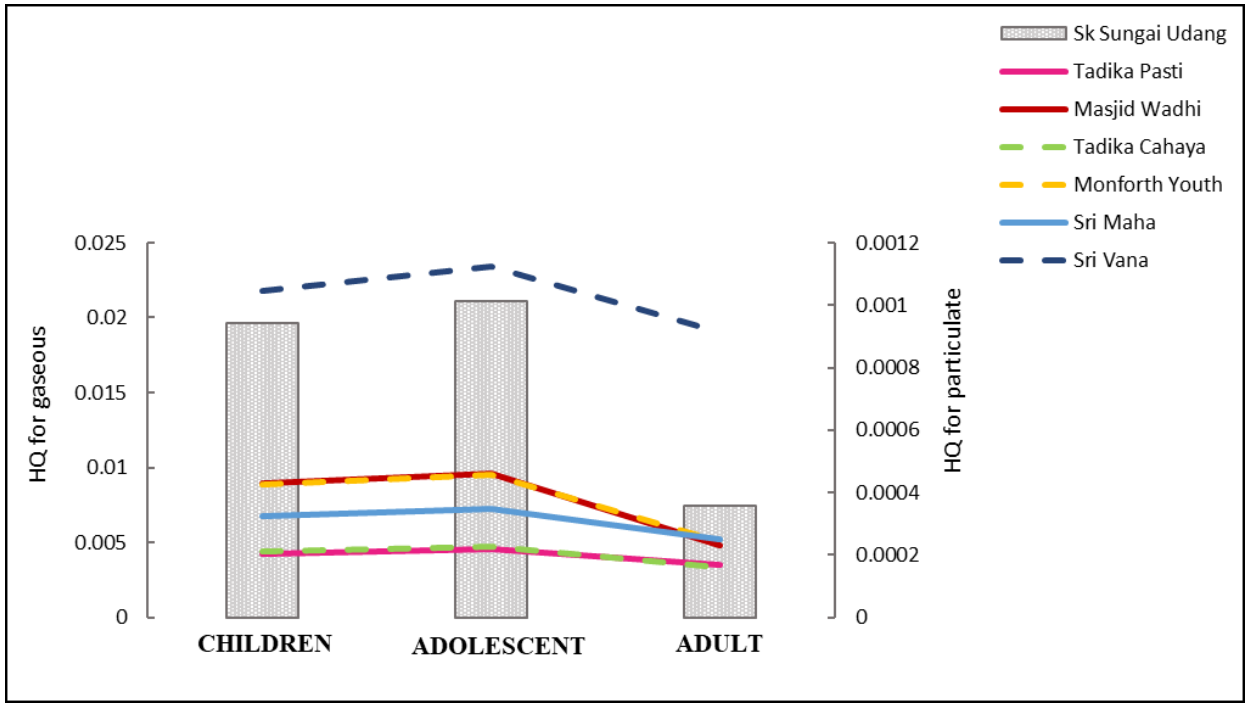

FIGURE 6. Total HQ values for gaseous sampling sites and particulate sampling sites for three age groups

CONCLUSION

This study determined the level of PAHs, their sources, and human exposure to PAHs in both gaseous and particulate phases around a petrochemical site in Melaka. According to the diagnostic ratio (DR), the gaseous PAHs were contributed by vehicle emissions, burning of petroleum, and coal combustion whereas particulate PAHs were contributed by wood burning and transportation emissions. Phe, Flt, and Ant were the most prevalent PAHs in the gaseous phase, whereas BaP, BgP, and BbF dominated the particulate phase. When the health risks of PAHs were evaluated for children, adolescents, and adults, it was discovered that the carcinogenic risks were minimal, with incremental lifetime cancer risk (ILCR) values ranging from 1.00×10^{-6} to 1.00×10^{-4}. The study analyzed that in both the gaseous and particle phases, ILCR for adults was higher than for children and adolescents. Hazard Index (HI) values computed for all sampling locations, including gaseous and particulate sampling sites, were below 1, indicating minimal detrimental effects on health. However, caution is advised to ensure that the residual cancer risk does not go above this level when the total cancer risks of all possibly carcinogenic components are considered. In a nut shell, establishing a baseline on gaseous PAHs in Malaysia is a necessity to evaluate the progress and efficacy of pollution control measures implemented by local authorities over time.

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