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ON THE NATURE OF POLYCYCLIC AROMATIC HYDROCARBONS ASSOCIATED WITH SPORTING WALKWAYS DUST: CONCENTRATIONS, SOURCES AND RELATIVE HEALTH RISK

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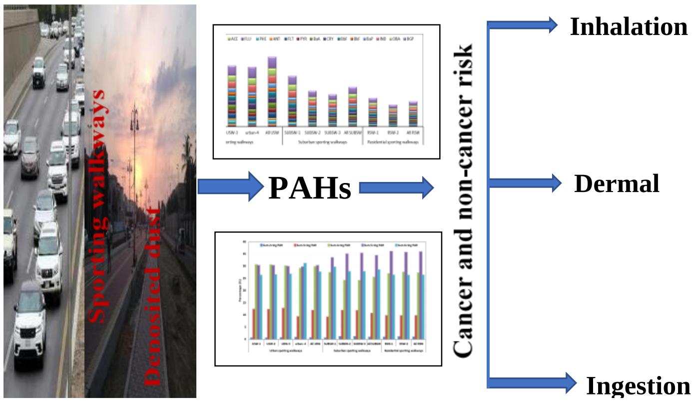
GRAPHICAL ABSTRACT



Caption: Sporting walkways are often close to highways

ABSTRACT

Sporting walkways (SW) are a new innovation which may prove popular in many cities. As there is currently no information on possible health risks associated with their use, concentrations of polycyclic aromatic hydrocarbons (PAHs) associated with deposited dust sampled on SW in Jeddah, Saudi Arabia, have been measured and interpreted in relation to sources and cancer risk. The average ∑PAHs (16 compounds) ranged between 1357 ng/g in residential areas and 3764 ng/g in central urban areas, with suburban areas between. The congener profile and diagnostic ratios of PAHs indicate a predominant source associated with petroleum combustion (pyrogenic source), most probably vehicular emissions. Carcinogenic potential is estimated from the sum of carcinogenic compound concentrations weighted by their individual potency relative to benzo(a)pyrene, and is found to be similar to household dust sampled in the same city, and lower than many other indoor and outdoor (road) dusts sampled across the world.

Keywords: PAHs; Deposited dust; Sporting walkways; Source identification; Carcinogenic compounds; Cancer risk.

1. INTRODUCTION

Atmospheric PM is emitted from both anthropogenic and natural sources, and contains a mixture of organic and inorganic pollutants in solid and liquid particles (Godish, 2005). Airborne particles deposit to surfaces as a result of atmospheric removal processes (dry and/or wet deposition). This deposited dust represents an accumulation of contributions from a variety of urban pollution source types such as traffic emissions, resuspension of road dust and dispersion of construction materials (Gautam et al., 2005; Goddu et al., 2004; Prajapati et al., 2006). Therefore, deposited dust can be effectively used as a monitor of different pollutants, and as an indicator of air pollution sources and air quality. At the same time, deposited dust on surfaces alongside highways acts as an important source for organic pollutants like polycyclic aromatic hydrocarbons (PAHs) which can cause significant risk to human health. Resuspension of deposited dust on the road surfaces by wind and traffic induced turbulence as well as during removal processes can lead to the injection of deposited dust-bound PAHs into the atmosphere.

PAHs are emitted as by-products from incomplete combustion processes and pyrolysis of fuel from a wide range of burning sources as well as from petroleum evaporation (Fang et al., 2003; Froehner et al., 2010). Beside the natural sources, the anthropogenic sources like traffic, domestic combustion, power plants, industrial combustion, accidental combustion sources and the petrochemical and metallurgical industries are considered the major emission sources of PAHs (Ravindra et al., 2008; Mastral and Callen, 2000; Alam et al., 2014; 2015). They are of special interest in the context of public health as some congeners are highly carcinogenic, and the PAH mixture has been classified by International Agency for Research on Cancer (IARC) as a confirmed (Class 1) human carcinogen (IARC, 2010). The USEPA has listed 16 PAH congeners as priority pollutants and classified seven as carcinogenic chemicals (Delgado-Saborit et al., 2011). Previous studies have revealed that human exposure to PAHs has carcinogenic, and mutagenic properties (ATSDR, 2020; IARC, 2010; WHO, 2010; 2013) and can cause toxic effects on the cardiorespiratory, reproductive, immune, and developmental processes (ATSDR, 2020), and restrict intrauterine growth (WHO, 2010).

To promote healthy lifestyles, the Jeddah Municipality (Amanah) has constructed many walkways in the different parts of the city during the last few years (SPA, 2020). They are attracting a great number of city residents of different ages to walk, jog or just relax in the open air especially in the evening. Additionally, it has provided special lanes for bicycling, exercise equipment, seats for the elderly , and special needs accessibility. Unfortunately, most of these walkways are located on busy streets and road intersections and are surrounded by many car parking spaces. Previous studies have shown that running along or near traffic roads during the rush hours can cause a reduction in lung function (Rundell and Caviston, 2008; Strak et al., 2010).

Previous studies in Jeddah city were focused on the characterization, sources and risk assessment of PAHs in the ambient air, street dust and school dust (Harrison et al., 2016a,b; Alghamdi et al., 2015; Shabbaj, et al., 2018; Alghamdi et al., 2020). However, there is no information about the levels, sources and human risk assessment of PAHs in sporting walkways. Therefore, the present study is considered to be the first in-depth investigation of the nature, origin and risk assessment of PAHs in dust deposited from ambient air on different impervious surfaces on both sides of the sporting walkways. The objectives of this study comprised the following: 1) to determine the levels and congener profiles of the dust-bound PAHs in the deposited surface dust of the sporting walkways of Jeddah city; 2) to identify the possible sources of PAHs; and 3) to assess the relative carcinogenic risk from exposure to dust-bound PAHs. Finally, the findings may provide valuable information on the state of environmental pollution of sporting walkways located along the traffic roads for decision makers of Jeddah city, as well as being a useful indicator of pollution sources and air quality.

2. MATERIALS AND METHODS

2.1 Study Area

Jeddah is one of the major cities of the Kingdom of Saudi Arabia and is located in the middle of the east coast of the Red Sea (latitude 29.2^o North and longitude 39.7^o East) (Figure 1). It has an arid climate, warm and humid or moderate in winter, and characterized by high temperature, humidity, and solar radiation in summer, with scarce rainfall. Like other cities in the Middle East and Arabian Peninsula; desert dust strikes the city from time to time (Alharbi and Moied, 2005; Badarinath et al., 2010). Rapid growth of the city (3.8% p.a.) during the last decades, which is higher than the national average, has led to a dramatic increase in population to more than 4 million according to 2016 estimates, and is expected to reach 4.9 million by 2025 (Jeddah CPI Profile, 2018).

The culture of walking is increasingly widespread in Saudi society, and engages males, females and children. Integrating with other Saudi cities to achieve "2030 vision" programmes in many sectors, Jeddah has contributed to implementing one of these programmes that is related to the residents: the "quality of life programme". It aims to improve individual lifestyles by supporting and creating new options to encourage Jeddah residents to indulge in cultural, environmental, and sports activities (Saudi Vision 2030, 2016) as well as to promote a healthy lifestyle.

2.2 Sample Collection

Deposited dust (27 samples) was collected during mid-January to mid-February 2020 from 9 different sporting walkways in Jeddah city located within the three various functional classes, namely, residential, urban and suburban areas. Sampling site locations appear in Figure 1. At each sampling site, dust deposited from ambient air on different impervious surfaces of items located on both sides of the sporting walkways, such as lampposts, seats, electric cabins and from windows and doors of public building services, were collected by gently sweeping, put in airtight polyethylene bags and then transferred to the laboratory. All collected dust samples were air-dried at room temperature in the dark, and then any visible hair, grit and soil were removed. The samples were homogenized after placing in a desiccator to eliminate moisture and then kept in sealed polyethylene bags for analysis.

2.3 Sample Extraction and Instrumentation

Deposited dust samples were extracted and analyzed according to methods reported by USEPA (1996), Shabbaj et al. (2018) and Alghamdi et al. (2020). The procedures were carry out as follows: (1) samples (2g) of deposited dust were Soxhlet extracted with a mixture of dichloromethane/n-hexane/acetone (1/1/1,v/v/v), for 16h. (2) The collected organic extracts were concentrated to about 2 ml using a rotary evaporator, (3) the organic extracts were purified with columns containing anhydrous sodium sulfate, silica gel, alumina, sand, and glass wool (Lee et al., 2011), and (4) the extracted eluent from the clean-up procedure further concentrated by a rotary evaporator. The residue was taken up in 1mL hexane, and then stored frozen until analysis. PAHs in the cleaned concentrated organic extracts were analyzed using a Hewlett–Packard (HP6890, Agilent, Santa Clara, CA, USA) gas chromatograph (GC), fitted with a flame ionization detector (FID) equipped with a HP-5 capillary column and hydrogen as the carrier gas. . One microliter (μL) of extract was withdrawn from the samples and blanks, and injected. The concentrations of the target compounds were quantified by an external standard solution of 15 PAH compounds (PAH mixture, Supelco, Inc., St. Louis, MO, USA). Quality assurance/quality control (QA/QC) was implemented throughout, including regular measurements of reagent blanks, analytical standards, standard spike recoveries, GC/FID calibration and detection limits. Retention times and responses of PAH compounds in the standard calibration mixture were checked daily. The PAH mixture standard of 15 compounds (Supelco, Inc., St. Louis, MO, USA) was used for preparation of the calibration curve for the target PAH compounds, and for spiking of extracts to determine recoveries. For all measured PAHs, the relative standard deviation of the replicated analyses of the calibration standard ranged from 4% to 6%. The average recovery of each PAH ranged from 70% to 108% for the 15 measured PAH compounds, and corrections have been applied. The detection limit of each PAH compound ranged from 0.048 ng/g to 0.376 ng/g of dust.. Full details of the quality assurance/quality control (QA/QC) process used to check the precision of the obtained results are reported in Alghamdi et al. (2020).

2.4. Source Apportionments (Diagnostic Ratio) of PAHs in Deposited Dust

In order to identify the source emissions of the target PAHs in deposited dust of sporting walkways, many ratios including PAHs-LMW/ PAHs-HMW, ∑CPAHs/∑PAHs, BGP/BaP, IND/ BGP, BaA/CRY, BaA/BaA + CRY, BaP/BaP + CRY, ANT/ANT + PHE, IND/IND + BGP, FLU/ FLU+PYR, FLT/PYR, and PHE/ANT were evaluated (Błaszczyk et al., 2017; Arnold et al., 2018; Iwegbue et al., 2019; Menezes and Cardeal, 2012; Tobiszewski and Namiesnik, 2012; Zhang et al., 2016; Qi et al., 2014; Slezakova et al., 2013; Kamal et al., 2016; Krugly et al., 2014). Moreover, bivariate plots of ANT/(ANT+PHE) vs. IND/(IND+BGP), ANT/(ANT+PHE) vs. BaA/(BaA + CRY), and BaA/(BaA + CRY) vs. IND/(IND + BGP) diagnostic ratios were constructed to differentiate PAH origins (Davoudi et al., 2020) as from pyrogenic and petrogenic sources.

3. RESULTS AND DISCUSSION

3.1. PAH Levels and Profile at Different Sporting Walkways Areas

The mean concentrations of the individual PAHs (sixteen PAH compounds) measured in deposited dust of different sporting walkways (SW) of Jeddah are presented graphically in Figure 2, and tabulated in Table S1. The main objectives of the work were to identify the PAH sources, and evaluate possible risks to health. The highest PAH concentrations were found at urban sporting walkways (USW) areas, whereas the lowest levels were detected in residential sporting walkways (RSW) areas. The mean concentrations of the total PAHs (∑PAHs) were in the following order: USW (3765 ng/g) > SUBSW (2136 ng/g) > RSW (1358 ng/g). This result indicates that the PAH species in deposited dust at SW of different areas are probably derived mainly from anthropogenic sources. The observed concentrations of ∑PAHs in USW could be attributed largely to the vehicle emissions caused by heavy traffic along roads that run adjacent to SW sites. On the other hand, the lowest detected levels of ∑PAHs at RSW are consistent with residential land use, relatively lower traffic density and absence of industrial activities. Therefore, the spatial distribution pattern of PAH concentrations in the deposited dusts at SW of Jeddah was probably related to the local traffic emissions. Anthropogenic activities that contribute to PAH compounds in urban areas include vehicle exhausts, tyre wear, resuspended soils dust, asphalt, and power plants (Duran and Gonzalez, 2009; Li et al., 2017; Zielinska et al., 2004). A study of airborne PAH in Jeddah (Alghamdi et al., 2015) showed a congener profile of the particle phase quite similar to that of the SW dust, and receptor modelling attributed PAH to three main sources: road traffic, the oil refinery and diesel/fuel oil combustion. The city has point sources including the refinery and a large oil-burning desalination plant. The gradient in concentrations from measurements at multiple sites between central urban, suburban and residential areas, however, is not consistent with a single point source, but is more likely to be explained by local road traffic density.

The mean concentrations of the individual PAH compounds in deposited dust of all Jeddah sporting walkways are shown in Figure 3. The average concentrations of the individual PAH ranged from 18.37 ng/g for NA to 406.91 ng/g for BGP. Moreover, the average total concentration of the PAH species (∑PAHs) was 2420 ng/g for all SW of Jeddah city. This concentration is lower than those reported in dust deposited on leaves of trees in Cairo (Egypt) (18220 ng/g (traffic areas) and 8261 ng/g (residential areas); Hassan et al., 2020), Nanjing (China) (3430 ng/g in urban areas; Zha et al., 2018), in dustfall in

Tianjin (China) (10001 ng/g (heating season) and 6001 ng/g (non-heating season); Wu et al., 2005) and Hubei Province (China) (4001 ng/g; Zhang et al., 2015), and in deposited dust in Mexico city (1390–7960 ng/g; Puy-Alquiza et al., 2016). The concentration of ∑PAHs in the present study was also lower than in street dust in some studies from Jeddah (Saudi Arabia) (3320 ng/g; Shabaj et al., 2018), Xuzhou (China) (6616 ng/g; Wang et al., 2017a), Beijing (China) (3700 ng/g; Li et al., 2017), Guangzhou, (China) (4800 ng/g; Wang et al., 2011), Dalian (China) (7460 ng/g; Wang et al., 2009), Xian (Northwest China) (500-48,000 ng/g; Wang et al., 2016); and Nepal, (747–4910 ng/g; Yadav et al., 2018). On the other hand, the PAH pollution level in deposited dust of SW of Jeddah was higher than those found in street dust in some studies from Bushehr (Iran) (1116 ng/g; Keshavarzi et al., 2017), Mahshahr (Iran) (769.6 ng/g; Mehr et al., 2016), Hanoi (Vietnam) (1900 ng/g; Tuyen et al., 2014), Bangalore (India) (1100 ng/g; Tuyen et al., 2014), New Delhi (India) (1100 ng/g; Tuyen et al., 2014), Kabul (Afghanistan) (427 ng/g; Khpalwak et al., 2019) and in the surface dust of urban areas of Qom (Iran) (478.3 ng/g; Davoudi et al., 2020). The clear differences in the PAH levels between these cities around the world can be associated with urbanization/ industrialization activities, traffic density, and economic development levels as well as the types of fuels consumed in each city (Wang et al., 2016).

3.2. Low and High-Molecular Weight PAHs at Different Sporting Walkways Areas

Based on the molecular weight of PAHs, in the present study, PAHs of low molecular weight (LMW-PAHs) include the sum of compounds with two aromatic rings (NA) and three aromatic rings (ACY, ACE, FLU, PHE, and ANT), whereas PAHs of high molecular weight (HMW-PAHs) include the sum of compounds with four aromatic rings (FlT, PYR, BaA, and CRY), five aromatic rings (BbF, BkF, BaP, and IND) and six aromatic rings (DBA and BGP). The distribution of ∑PAHs, Low-molecular weight (LMW) PAHs, high-molecular weight (HMW) PAHs, $\sum PAH_{carc.,}$ ∑PAH, LMW-PAHs/HMW-PAHs ratio, and $\sum PAH_{carc.}$ /∑PAHs percentage in deposited dust of USW, SUBSW and RSW are summarized in Figures S1 and S2. Based on percent (%) abundance of aromatic rings at various SW locations, the distribution pattern of PAHs in the deposited dust had the following order: 5 > 4 > 6 > 3 > 2 rings at USW, 5 > 6 > 4 > 3 > 2 rings at SUBSW and 5 > 4 > 6 > 3 > 2 rings at RSW (Figure S1). HMW-PAHs (4-6 aromatic ring) were the more abundant PAH compounds in deposited dust of SW, and ranged from 87.1 to 90.0% (with a mean value of 87.65%), 87.2-90.4% (with a mean value of 88.32%), and 89.3 to 89.4 % (with a mean value of 89.34%) of the total PAHs at USW, SUBSW and RSW, respectively (Figure S2 and Figure S3). Whilst the LMW-PAHs (2-3 aromatic rings) represented 10.1 to 13.4% (with a mean value of 12.47%) at USW, 9.7-12.9% (with a mean value of 11.51%) at SUBSW, and 10.5 to 10.7 % (with a mean value of 10.61%) at RSW of the total PAHs. The relatively greater abundance of HMW-PAHs than the LMW-PAHs in deposited dust of SW reflects the lower volatility of HMW-PAHs which tend to be associated with the particulate phase, whereas LMW-PAHs tend to be associated with the gas phase (Hassan and Khoder, 2012; Iwegbue et al., 2019; Nazmara et al., 2020), and therefore do not efficiently partition to dust solids, consistent with Soltani et al. (2015), Škrbić et al. (2019), and Orecchio (2011) who reported that LMW-PAHs were correlated principally with the gas-phase, whereas the HMW-PAHs were correlated mainly with particulate-phase and have a relatively high resistance to degradation. Previous studies (Shabbaj et al., 2018 and Alghamdi et al., 2020) concluded that PAHs with four to six aromatic rings (HMW) were the predominant compounds in street dust and classrooms filter dust of Jeddah schools.

3.3. Source Apportionment (Isomeric Ratios) of PAHs in Deposited Dust of SW Areas

Identification of PAH sources is fundamental to the control of pollution, and for risk management. The major anthropogenic emission sources of PAHs in the environment are classified as pyrogenic and petrogenic sources (Qi et al., 2014). The values of concentration ratios of the individual PAH compounds (diagnostic ratios) are a basic indicator of PAH emission sources (Oliveira et al., 2017; Tue et al., 2014). These may be difficult to interpret in a complex emissions scenario with multiple sources, but can nonetheless be a useful indicator of PAH sources. Source apportionment by Positive Matrix Factorization or Principal Component Analysis would be preferable, but was precluded by the small number of samples in relation to the number of PAH congeners. In the present study, twelve specific PAH ratios in samples from SW areas were computed and are presented graphically in Figure S4. These ratios were PAHs-LMW/ PAHs-HMW, ∑CPAHs/∑PAHs, BGP/BaP, IND/ BGP, BaA/CRY, BaA/BaA + CRY, BaP/BaP + CRY, ANT/ANT + PHE, IND/IND + BGP, FLU/ FLU + PYR, FLT/PYR, and PHE/ANT. The first diagnostic ratio used to distinguish between pyrogenic sources and petrogenic sources is LMW-PAHs/HMW-PAHs concentration ratios. Ratio values less than one indicate pyrogenic sources are the major contributor, whereas ratios above one indicate that petroleum is the main source (Błaszczyk et al., 2017; Rogula-Kozłowska, 2015). In the present study, LMW-PAH/HMW-PAH ratios were 0.12 (USW), 0.13 (SUBSW) and 0.14 (RSW) (Figure S4), revealing that the PAHs in deposited dust are emitted mainly from pyrogenic sources (high-temperature burning of fuels) including the emissions of traffic. The LMW-PAHs/HMW-PAHs ratio values in this study were in line with the results of classroom filter dust of Jeddah schools (Saudi Arabia) (0.14 - 0.20, Alghamdi et al., 2020), urban street dust of Jeddah (Saudi Arabia) (0.21 - 0.22, Shabaj et al., 2018), and lower than those found in street dust of Steel city (China) (0.32 - 0.88, Zhang et al., 2016), and an urban-industrial area of Dąbrowa Górnicza (Poland) (0.62, Błaszczyk et al., 2017). Previous studies (Arnold et al., 2018; Iwegbueet al., 2019; Menezes and Cardeal, 2012; Tobiszewski and Namiesnik, 2012; Zhang et al., 2016) have reported that the value of FLU/(FLU + PYR) ratio is more than 0.4 for petroleum/ petrogenic sources, less than 0.5 for pyrogenic sources or between 0.4 and 0.5 for traffic emission/incomplete combustion of fuels. This ratio in the present study was 0.16 for USW, 0.31 for SUBSW and 0.17 for RSW, and is suggestive of pyrogenic sources of PAHs. PHE/ANT ratios are lower than 10 for PAHs that mainly arise from pyrogenic sources and vehicular traffic emission, while ratios above 10 characterise PAHs that originate from petrogenic sources (Liu et al., 2007; Qi et al., 2014). In this study, the values of PHE/ ANT ratio are less than ten, ranging from 1.61 at USW to 2.81 at RSW, confirming that pyrogenic sources and vehicular emission are probably the main sources of PAHs in deposited dust of SW in different areas. Our ratios of PHE/ANT are similar to those of Shabbaj et al. (2018) in street dust of Jeddah (1.18-1.33), and lower than those found in household floor dust, car dust and air conditioner filter dust (Jeddah-Saudi Arabia) (4.2-5.63; Ali et al., 2016), rural and urban dust (China) (8.26-9.91; Qi et al. 2014), in road dust PAHs (Shanghai) (China) (0.45-5.5; Liu et al., 2007). Moreover, Błaszczyk et al. (2017), Kamal et al. (2014) and Zhang et al. (2016) reported that the value of the ANT/(ANT + PHE) ratio is more than 0.1 for pyrogenic sources and traffic emissions, whereas values lower than 0.1 are suggestive of petrogenic sources. The values ANT/(ANT + PHE) ratios in the present study were more than 0.1 (ranged from 0.36 - 0.38), which also confirms that pyrogenic sources and traffic emissions are the major sources of the PAH concentrations in the deposited dust of SW in Jeddah. The ANT/(ANT + PHE) ratios in the present study are consistent with previously reported data in street dust of Jeddah (Saudi Arabia) (0.43-0.48; Shabbaj et al., 2018), classroom filter dust of Jeddah schools (Saudi Arabia) (0.43-0.49; Alghamdi et al., 2020) and lower than in air-conditioning filter dust from high-rise apartments (Hefei-China) (0.7; Wang et al., 2017b). In addition, if the ratio of IND/(IND+ BGP) has values > 0.5, it suggests the major source of PAHs was coal or wood combustion, whereas ratios < 0.2 and between 0.2 and 0.5 indicate that the dominant sources of the target compounds were petroleum and traffic emissions, respectively (Qi et al., 2014; Slezakova et al., 2013). The IND/(IND + BGP) ratio in this study was 0.40 in USW, 0.44 in SUBSW and 0.41 in RSW, indicating that traffic emissions are the main source of PAHs. In addition, this ratio value (IND/(IND + BGP) = 0.40 - 0.44) is comparable with previous studies including in indoor dust of Jeddah (Saudi Arabia) and Kuwait (0.04 - 0.42; Ali et al., 2016) and in road dust PAHs in Shanghai (China) (0.5 - 0.8; Liu et al., 2007). In addition, another PAH ratio used for source apportionment is the ratio of BaA/(BaA + CRY), which is reported to be lower than 0.2 for petrogenic sources, more than 0.35 for combustion sources/vehicular emissions, and between 0.2 and 0.35 for both combustion and petrogenic sources (Kamal et al., 2016; Zhang et al., 2016). The values of the BaA/(BaA + CRY) ratios in the present study ranged from 0.44 (SUBSW and RSW) to 0.50 (USW) which are more than 0.35, suggesting that PAHs arise from combustion sources/traffic emissions. This is consistent with previous reports of data for air conditioner filters, household floor and car dust (Jeddah-Saudi Arabia) (0.38 - 0.61; Ali et al., 2016), urban street dust (Jeddah, Saudi Arabia) (0.40-0.46; Shabbaj et al., 2018), an urban-industrial area (DąbrowaGórnicza Poland) (0.49; Błaszczyk et al., 2017), and street dust (Steel industrial city- China) (0.21 - 0.57; Zhang et al., 2016).

The concentration ratio of BaA/CRY can also be applied to identify the traffic sources and degradation of PAHs under the effect of photochemical reactions and the aging of the air mass (Krugly et al., 2014; Lohmann et al., 2000). Values of BaA/ CRY concentration ratios range from 0.28 to 1.20 for gasoline engines and from 0.17 to 0.36 for diesel engines (Fang et al., 2004; Bourotte et al., 2005). Moreover, if the value of BaA/ CRY concentration ratio is higher than 0.40, it indicates that the pollution is freshly released and the photochemical processing of the air mass is relatively low, while a ratio of lower than 0.40 indicates that the air masses are aged or the major sources of PAH are not regional (Lohmann et al., 2000). In this study, the values of BaA/ CRY concentration ratios were 1.01 at SW, 0.77 at SUBSW and 0.85 at RSW, suggesting that gasoline engine emissions are the predominant source of PAHs, and the PAH pollution in deposited dust of different SW areas are freshly released from local sources around the SW which has undergone little photochemical processing of the air mass. These results are consistent with those found in street dust and classroom of filter dusts from Jeddah (Shabbaj et al., 2018; Alghamdi et al., 2020). The diagnostic ratios for identifications of PAHs in deposited dust (27 samples) of Jeddah SW based on ANT/ (ANT + PHE) vs. IND/ (IND + BGP), ANT/ (ANT + PHE) vs.

BaA/ (BaA + CRY), and BaA/ (BaA + CRY) vs. IND/ (IND + BGP) (Figure 4) indicate that PAHs come from petroleum combustion, most probably largely vehicular emissions. This is supported by an analysis reported by Alam et al. (2013) in which ratio data for the roadside traffic increment (roadside concentration minus local background) were plotted onto bivariate plots such as Figure 4. These conformed closely to the diagnostic guidelines referred to for pyrogenic (gasoline combustion) sources.

The profiles of individual PAH showed similarity between the three areas (Figure S3). Correlations between the individual PAHs were calculated and studied as a way of evaluating whether particular compounds were likely to have been produced from the same sources (Yang et al., 2012). Table S2 illustrates the correlation coefficients between the individual PAH compounds in the deposited dust at all SW of Jeddah. From the correlation coefficients matrix, it is clear that significant positive correlation coefficients were found between the individual PAH compounds and also between the individual PAH compounds and LMW-PAHs and HMW- PAHs and ∑PAHs. This is an indication that all PAHs in deposited dust of Jeddah SW probably originated from the same source, or sources whose emissions are at a similar altitude, and closely correlated in time.

Overall, diagnostic ratios and the correlation coefficients between the individual PAHs indicate that PAHs in deposited dust at the SW areas are emitted mainly from the same sources, and a petroleum combustion (pyrogenic) source, probably traffic emissions, around the SW were the dominant sources of PAHs in deposited dust.

3.4 Consequences for Human Health

Many papers report risk assessments, especially in regard of cancer, for PAH exposures (e.g. Gope et al., 2018; Iwegbu et al, 2019; Nazmara et al, 2020). In the case of respiratory exposure, there are wellaccepted cancer slope factors, derived from occupational exposure studies, which relate excess lung cancer risk to exposure concentration, usually using BaP as a marker for the entire mixture of PAH congeners. However, no such cancer slope factors are recognized by international organisations such as the World Health Organization or the European Food Standards Agency for the risk of cancers resulting from ingestion via the gastrointestinal tract. Since the route of intake and target organs are different, it is inappropriate to employ cancer slope factors which derive from studies of inhalation exposure to estimate cancer risks from ingestion of PAH. Nonetheless, the relative potency of individual PAH congeners in the induction of cancer in experimental animals has been established (Nisbet and LaGoy,1992; Delgado-Saborit et al., 2011), and this information may be extrapolated to humans, within wide uncertainty limits. This is useful in judging the relative carcinogenic potential of different PAH mixtures, but does not provide a guide to absolute cancer risk.

The dusts analysed in this work were sampled from the surfaces of the Sporting Walkways, and are analogous to indoor floor dusts and outdoor road dusts in relation to human exposure. Since only small amounts become resuspended into the atmosphere, respiratory exposure is negligible. Contact with skin is also very limited for a surface dust in a space occupied by clothed individuals, and hence skin absorption is very unlikely to be a significant exposure pathway. This leaves only ingestion as potentially significant, and it is this pathway which is normally quantified as a route of intake for toxic substances in surface dusts. While estimates are available for daily dust intakes of different age groups, in the absence of a cancer slope factor, these cannot be interpreted quantitatively as a cancer risk. In their review article on PAH in indoor dusts, Ma and Harrad (2015) implicitly acknowledged this and simply compared relative cancer risks of different dusts via a measure of BaP equivalent concentration, derived from:

$$BaP_{TEQi} = C_i \times TEF_i \quad (1)$$

$$BaP_{TEQ}\,(\text{mixture}) = \sum^{i} BaP_{TEQi} \quad (2)$$

in which, BaP_{TEQ} (mixture) is the BaP equivalent concentration derived from summing the masses of individual compounds (C_i), each weighted by its relative cancer potency, TEF_i, in this case taken from the tabulation by Nisbet and Lagoy (1992). The same method has been used in calculating values of BaP_{TEQ} for the dust mixtures in Table 1, which compares this, as well as PAH concentrations, with those from other published data. Table 1 includes both a summary of the many studies of indoor dusts from around the world reported by Ma and Harrad (2015) together with more recent data from Jeddah and Kuwait, as well as selected representative data from studies of outdoor dusts. Some caution is needed as not all studies measure the same suite of PAH, but most have measured a similar list (usually based upon the USEPA 16 priority compounds), and in most cases the compounds with high relative potencies have all been measured, so the values of BaP_{TEQ} (mixture) are reliable. From the data in Table 1, it may be inferred that the carcinogenic potential, as represented by the mean BaP_{TEQ}, for Sporting Walkway dusts from Jeddah (283-780 ng/g) is comparable to that measured in household dusts in the same city (663 ng/g) by Ali et al (2016), and less than the worldwide average for indoor dusts taken from Ma and Harrad (2015), and road dusts from various parts of the world. Consequently, any exposure to PAH in dusts on the Jeddah Sporting Walkways is only the same as that experienced by remaining in the home and the associated exposed to household dusts.

4. CONCLUSIONS

This is the first study that concentrated on the nature of polycyclic aromatic hydrocarbons, as indicator of air quality and pollution sources, in deposited dust of nine sporting walkways in three different areas, namely: urban, suburban and residential areas in Jeddah, the second largest city of Saudi Arabia.

The results indicated that the levels of PAHs varied among different sporting walkways areas, with highest total PAH (∑PAHs) levels in deposited dust of sporting walkways of urban areas (USW) and lowest levels in deposited dust of sporting walkways of residential areas (RSW). The average concentration of ∑PAHs (#PAH=16) was 2420 ng/g at all sporting walkways of Jeddah. The individual PAH contribution in the deposited dust increases with the molecular weight. Based on aromatic rings, the distribution pattern of PAHs in the deposited dust had the following order: 5 > 4 > 6 > 3 > 2 rings at USW, 5 > 6 > 4 > 3 > 2 rings at SUBSW and 5 > 4 > 6 > 3 > 2 rings at RSW. The predominant HMW-PAHs (4-6 aromatic ring) in deposited dust of SW accounted 87.65%, 88.32% and 89.34% of the total PAHs at USW, SUBSW and RSW, respectively, indicating that HMW-PAHs tend to be associated with the particulate phase. Various diagnostic ratios of PAHs and cross-plots of ANT/ (ANT+PHE) vs. IND/ (IND+BGP), ANT/ (ANT+PHE) vs. BaA/ (BaA + CRY), and BaA/ (BaA + CRY) vs. IND/ (IND + BGP), and the correlation coefficients between the individual PAHs indicated that PAHs in deposited dust in the SW areas are emitted mainly from the same sources, and petroleum combustion (pyrogenic source) in road vehicles is the dominant source of PAHs in deposited dust. Estimation of the carcinogenic potential of the dusts using the calculated BaP-equivalent concentration indicates a potential very similar to that of household dusts sampled in Jeddah, and smaller than the mean for household dusts and road dusts from around the world. This indicates a probable low level of risk, similar to that experienced for dust ingestion within the home.

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CONFLICTS OF INTEREST

The authors have no actual or potential conflict of interest including any financial, personal or other relationships with other people or organizations within three years of beginning the submitted work that could inappropriately influence, or be perceived to influence, their work.

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TABLE LEGENDS:

Table 1: Comparative measurements of PAH in dusts and BaP-equivalent concentrations.

FIGURE LEGENDS:

Figure 1: Map of Jeddah city showing districts and the sporting walkways sampling sites.

Figure 2: Concentration of the individual PAH compounds in deposited dust of SW in the different areas of Jeddah.

Figure 3: Average concentrations of the individual PAH compounds in deposited dust of Jeddah sporting walkways.

Figure 4: Bivariate plots of diagnostic ratios for identifications of PAHs in deposited dust of Jeddah SW: (a) ANT/(ANT + PHE) vs. IND/(IND + BGP); (b) ANT/(ANT + PHE) vs. BaA/(BaA + CRY); (c) BaA/(BaA + CRY) vs. IND/(IND + BGP).

Table 1: Comparative measurements of PAH in dusts and BaP-equivalent concentrations.

Location	Sample type	N	#PAH	∑PAH (ng g^{-1})	Mean BaP_{TEQ} (ng g^{-1})	Reference
Worldwide	Indoor dust (range)	1 - 616	2 - 19	127 - 115,187	19 - 15,530	Ma and Harrad (2015)
Worldwide	Indoor dust (average)	79 ± 24	15 ± 1	14,150 ± 4,025	1,897 ± 552	Ma and Harrad (2015)
Shanghai (winter)	Road dust	27	16	9,176 - 32,573	2,500	Liu et al. (2007)
Shanghai (summer)	Road dust	27	16	6,875 - 27,766	1,000	Liu et al. (2007)
Ulsan, Korea	Road dust	11	16	19,690 - 154,640	16,550	Lee and Dong (2010)
Shanghai, China	Road dust	68	18	1,020 - 138,990	2,440	Zheng et al. (2016)
Lahore, Pakistan	Wind-blown dust	5	17	170 - 1,150	101	Smith et al. (1995)
Birmingham, UK	Road dust	4	17	2,020 - 99,600	4,033	Smith et al. (1995)
Kuwait	Household dusts	15	16	450 - 9,100	205	Ali et al. (2016)
Jeddah, Saudi Arabia	Household dusts	15	16	950 - 11,950	663	Ali et al. (2016)
Jeddah, Saudi Arabia	Sporting walkway (urban)	12	16	2,732 - 4,985	780	This work
Jeddah, Saudi Arabia	Sporting walkway (suburban)	9	16	1,530 - 2,961	471	This work
Jeddah, Saudi Arabia	Sporting walkway (residential)	6	16	1,125 - 1,567	283	This work

Note: N = number of samples; #PAH = number of PAH analysed; Value of BaP_{TEQ} are reported by Ma and Harrad (2015), but are estimates calculated from data presented in the other studies.

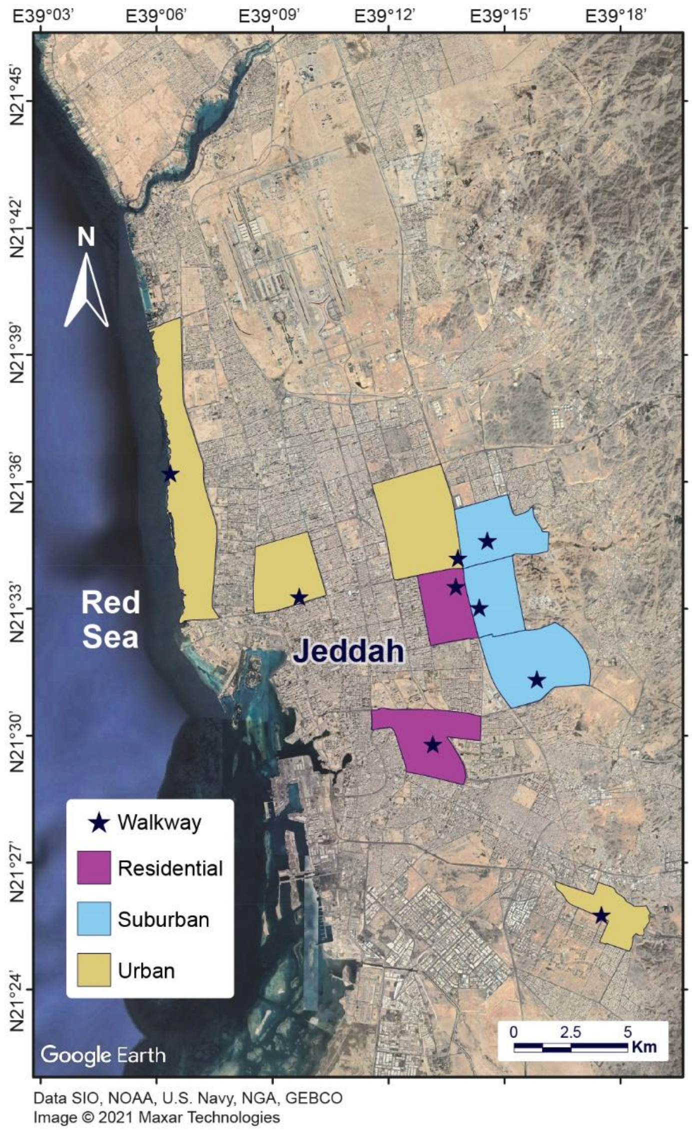

Figure 1. Map of Jeddah city showing districts and the sporting walkways sampling sites.

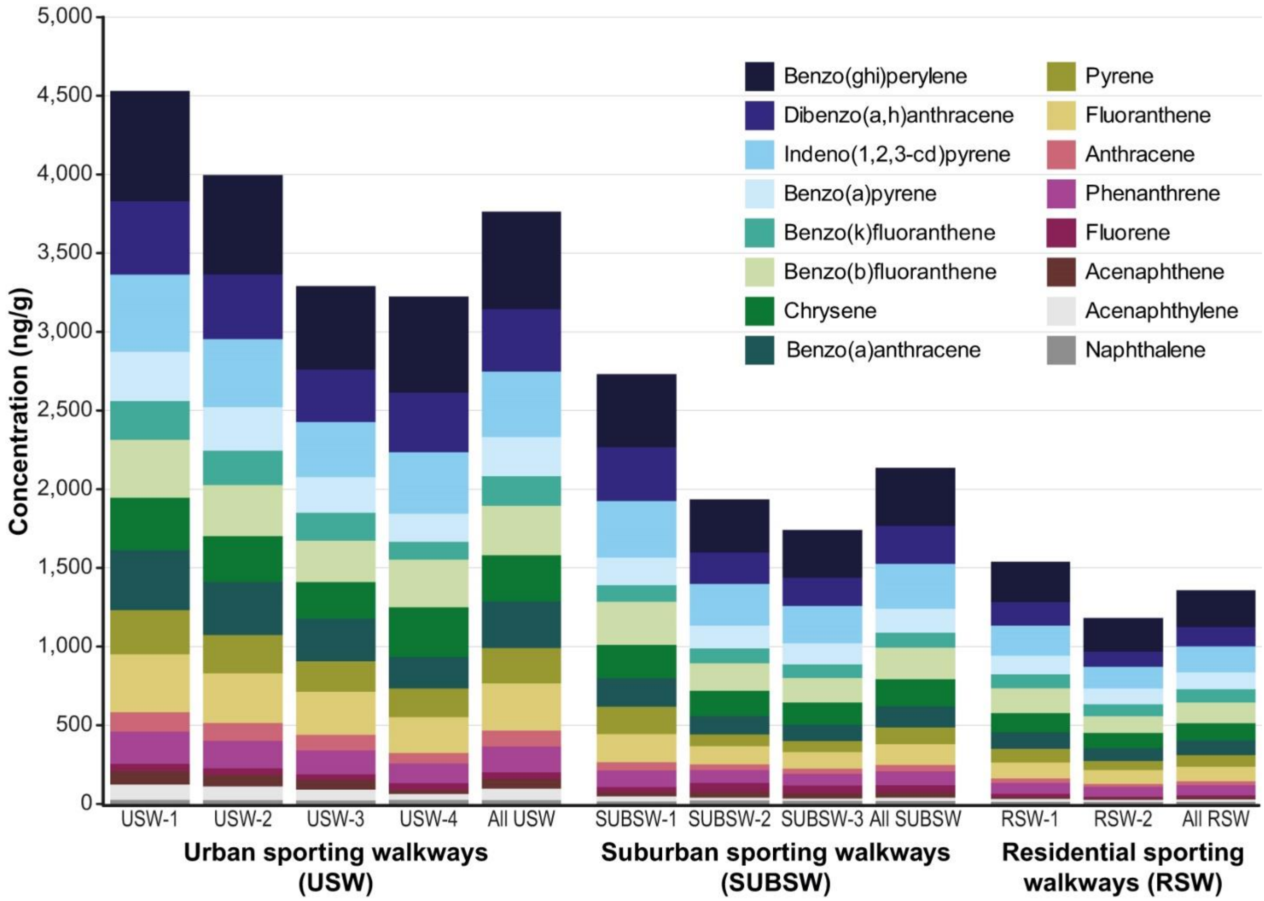

Figure 2: Concentration of the individual PAH compounds in deposited dust of SW in the different areas of Jeddah.

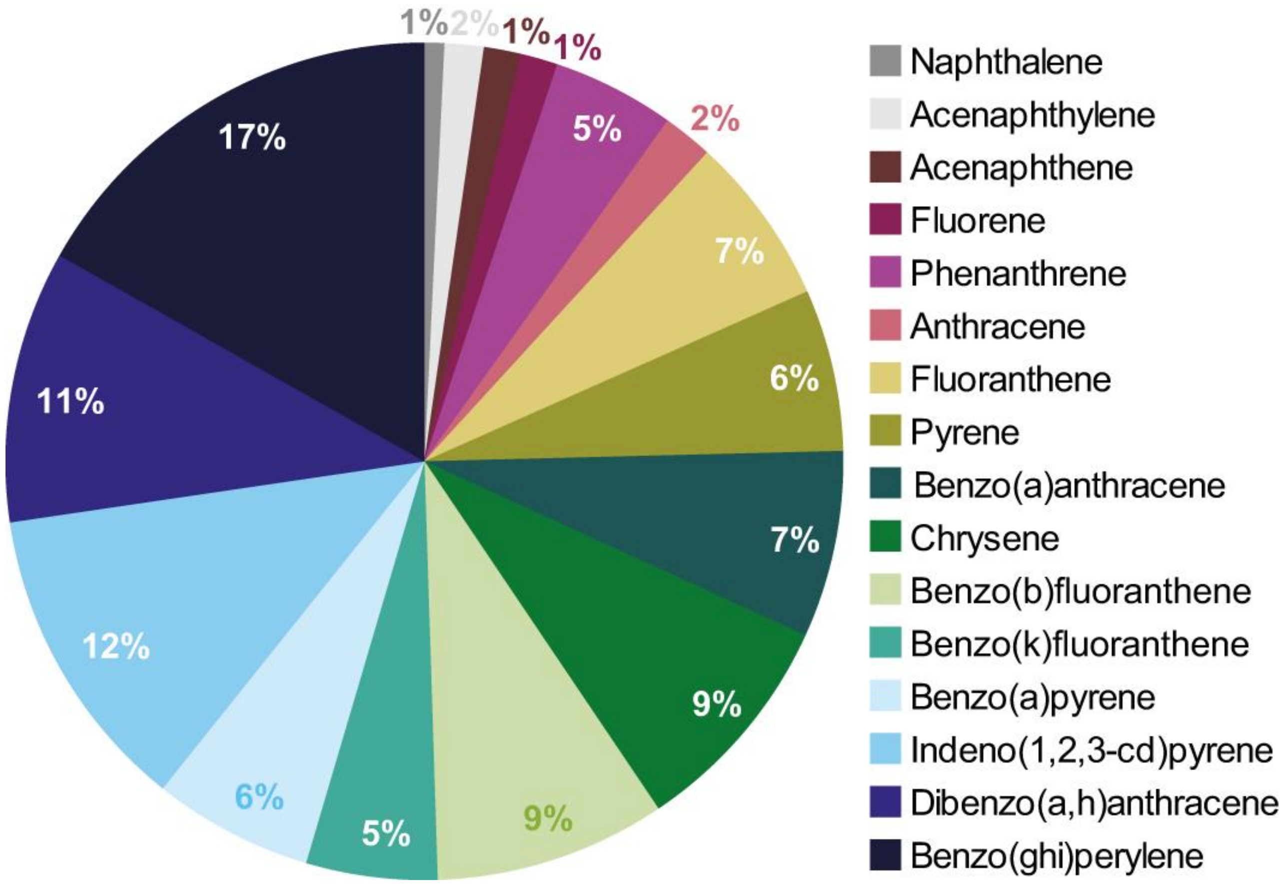

Figure 3: Average concentrations of the individual PAH compounds in deposited dust of Jeddah sporting walkways.

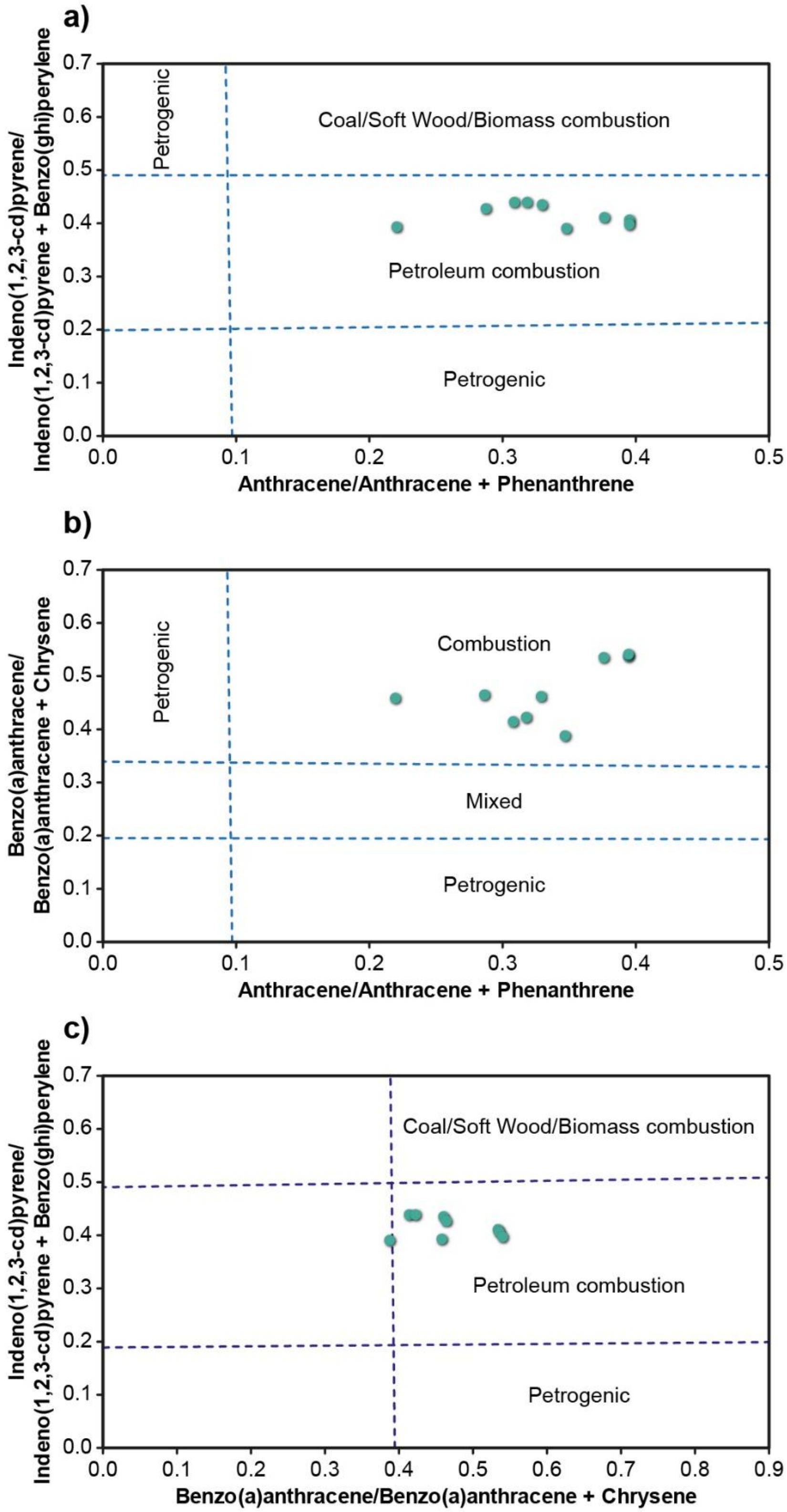

Figure 4: Bivariate plots of diagnostic ratios for identifications of PAHs in deposited dust of Jeddah SW: (a) ANT/(ANT + PHE) vs. IND/(IND + BGP); (b) ANT/(ANT + PHE) vs. BaA/(BaA + CRY); (c) BaA/(BaA + CRY) vs. IND/(IND + BGP).