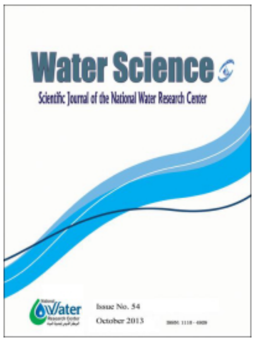

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ARTICLE

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Assessment of polycyclic aromatic hydrocarbon in runoff from different land use at Amman Zarqa Basin-Jordan

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ABSTRACT

A study of 16 polycyclic aromatic hydrocarbons (PAHs) in stormwater runoff was conducted at seven locations in semi-arid Amman Zarqa Basin (AZB) for two winter seasons during 25 October 2012 to 31 May 2014. The selected sites are representing residential, commercial, industrial and agricultural areas. The concentrations of total PAH at all sites were in the range of 0.18–11.03 ppb. The highest average total concentration of PAHs of the runoff samples was found at industrial area (2.42 ppb), while the lowest concentrations was found at residential area (1.66 ppb). Average Daily Traffic was not found to be correlated to total PAHs carried by stormwater runoff from studied land uses (All sites). Isomer pair ratios and individual compound ratios are suggesting that the main source of PAHs in AZB catchment is from a pyrolytic origin especially at commercial, residential, and agriculture area, while industrial area is mainly polluted by PAHs with petrogenic origin. This study is providing a reliable data on PAHs loads carried by storm water runoff from different land uses.

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PAHs; stormwater runoff; land uses; residential; commercial; industrial; agricultural area; amman-Zarqa Basin

Introduction

Pollutant loads carried by stormwater runoff event is a major pollution source that has a great impact of stormwater discharge on water bodies (i.e., dams, rivers) and the ecosystem (Walsh et al., 2016). Therefore, monitoring of storm water quality is always essential to assess and control the potential risks threaten surface water as well as to improve watershed management (Ki et al., 2011; Lee, Swamikannu, Radulescu, Kim, & Stenstrom, 2007).

Various organic and inorganic substances are usually carried by stormwater runoff (Al-Reasi, Wood, & Smith, 2013; Göbel, Dierkes, & Coldewey, 2007; Parajulee et al., 2017). These organic substances such as Polycyclic aromatic hydrocarbons (PAHs) are posing a measure threat to the quality of water bodies particularly if it is used as drinking water source. Polycyclic aromatic hydrocarbons (PAHs) has currently drawn a great attention for stormwater quality monitoring programs as it can negatively impact the composition and quality of surface waters (Chong et al., 2013; McElmurry, Long, & Voice, 2013: McIntyre, Davis, & Hinman et al., 2015). PAHs has a long half- life and potentially carcinogenic nature, therefore, they are widely investigated in the literature (Kawai, Amamoto, & Harada, 1967; NRC (National Research Council), 1983). The PAHs are usually generated during carbonization and incomplete combustion of organic materials (Lin, Chang, Hsien, Chao, & Chao, 2009).

In Jordan, the urbanization areas have continuously increased over the past five decades. The population of Jordan has risen from about 0.9 to 9.71 million between the census dates of 1961 and 2016. The huge increase in population, despite the lower growth rate of 2.2%, is mainly due to many influx waves of refugees from other neighboring countries. Amman Zarqa Basin (AZB) is one of Jordan's important basins as more than the half Jordan population live within the urban areas of AZB. The continuous deterioration of the ecosystem components of the AZB since almost three decades is one of the biggest environmental challenges in Jordan. Many studies indicate that there is a considerable degradation in the quality of the water resources in AZB as a result of general deterioration conditions (Alawi & Haddadin, 2004; Al Kuisi, Al-Qinna, Margane, & Aljazzar, 2012). These conditions are primarily attributed to; general water level declines, human and industrial wastewater along wadi Zarqa, treated wastewater effluents from the existing treatment plants within the basin and the other impacts resulted from the ongoing agricultural activities in the basin. Although a considerable number of studies have focused on the impacts of wastewater pollutants such as heavy metals and pesticides, however, there is a little knowledge about the levels of PAHs carried by stormwater runoff generated from different land uses in AZB.

Therefore, this study aims to investigate the levels of PAHs concentrations generated from different land uses (commercial, residential, industrial, and agricultural) in semi-arid Amman-Zarqa basin at Jordan. PAHs were analyzed for the collected samples from seven sites during two winter seasons (2012–2014).

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Moreover, the correlation between PAHs pollutant concentrations and the average daily traffic (ADT) measured at sampling sites was evaluated.

Methodology

Monitoring sites description

The seven sites are located at different types of land use areas representing four different types of catchments according to anthropogenic activities (residential, commercial, agricultural, and industrial) (figure 1).

Residential area has only private houses and no offices and factories. For sampling five urban residential homes situated in those areas which have little traffic and lots of greenery in the vicinity, five roadside homes with heavy traffic density. Two sites were selected at Um-Alsummaq and Abu-Nusair areas representing residential land use. Commercial area is located at city center of Amman. Large parts of these areas are residential, but due to its geographical location in the center of the city, it contains several important governmental buildings and businesses. The area is known for containing several bus stations serving routes to many cities in Jordan. Moreover, it contains several companies, banks, hospitals, schools, and governmental buildings. Two sites were selected at Al-Abdali and Downtown areas representing commercial land use. Industrial area is located at King Abdullah II Sahab city. It is the oldest and largest industrial estate in Jordan and is located in Sahab, 12 km south-east of Amman on 253 hectares of land (2.53 million square meters) at the edge of Amman-Zarqa basin. The industrial city was fully occupied in 2010, with 364 enterprises employing 13,694 workers. Factories are diversified across various sectors, including food, metal fabrication and electronics, plastics and rubber, pharmaceuticals, chemicals, textiles, wood and metallic furniture, printing and packaging, leather construction. Two sites were selected at Abdullah II Ibn Hussein Industrial Estate (also refers to the Industrial City of Sahab or Amman Industrial Estate) representing industrial land-use area. Sahab1 is located in the south of the King Abdullah II Sahab city near the wastewater treatment plant. Sahab2 is located in the east of the city. In all the sites, the samples were taken directly from roads and stormwater drains and inlets. Agricultural area is located at Al-Qunyyah catchment. This catchment has intense agricultural activities include farms and greenhouses (vegetables, orchards, and fruit trees) located along the main wadies upstream of Ain Al-Qunyyah, olive trees planted at the hilly areas. Also, there are some areas used for grazing of animals and many chicken farms existed in the catchment area (Subah & Hobler, 2004). There are few springs used as water source for irrigation in addition to the rainfall at the catchment. The main drinking water and irrigation sources at the catchment are natural springs such as Ain Al-Qunyyah spring. Therefore, the runoff will represent impact of agricultural activities without any interference from other sources such as treated wastewater. Al-Qunyyah is located north-west of Zarqa city. One site was selected at Al-Qunyyah catchment area representing agriculture land use at the basin.

The location of sampling point, elevation, and average rainfall for two seasons are shown in table 1.

The elevations of the monitoring sites ranged from 931 m above the sea level at Um-Alsummaq located at the north/west of basin to 522 m south/east at Al-Qunyyah, and all these sites naturally drains toward the King Talal Dam (KTD) at the west of the basin (120 m above the sea level). Daily rainfall data were collected using manual gauge stations installed near the monitoring sites. The highest average rainfall was recorded at Um-Alsummaq site (506.9 mm), while the lowest average rainfall was recorded at Al-Qunyyah site (169.4 mm) (Table 1).

Storm water sampling

Storm water samples were collected from seven sites for two winter seasons (2012/2013 and 2013/2014) Amman-Zarqa basin. Each site was sampled twice during winter season, the first rain event at the

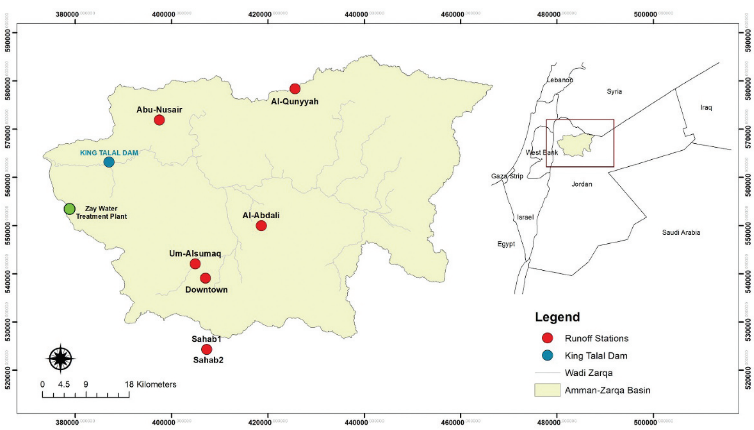

Figure 1. Monitoring runoff sites located in Amman Zarqa basin.

beginning of the winter season and the second event at the end of the winter season.

Sample extraction

All runoff water samples were collected in 1000-mL amber glass bottles, which were rigorously cleaned, rinsed with methanol, and dried prior to use. Samples were pre-concentrated using solid phase extraction (SPE) directly or within 24 h after collection to avoid adsorption of PAHs in the glass of bottles. The collected samples were firstly decanted to remove suspended particles and then filtered through Whatman No. 40 filter paper using a vacuum filtration unit. ISOLUTE® ENV+ C18 cartridge from Biotage (Uppsala, Sweden) was connected to a SPE manifold and vacuum pump and preconditioned by passing 6 mL methanol and 6 mL distilled deionized water (DDI H2O) through the cartridge. The filtered sample was then pumped via tube to the conditioned cartridge using a vacuum manifold system. The sample flow through the SPE cartridge was kept at less than 10 mL/min. After the whole sample was extracted, the cartridge was rinsed with 6 mL of DDI H2O. Room air was allowed to flow through the cartridge by continued suction for a minimum of five minutes to help dry the cartridge.

Sample analysis

After extraction, sample cartridges were directly eluted and analyzed using High Performance Liquid Chromatography (HPLC). Cartridges were eluted with 10 mL of high purity dichloromethane into a disposable glass culture tube followed by addition of internal standards and surrogates. The eluent volume was then evaporated in the Turpo-Vap (Biotage TurboVap LV, UK) under a stream of nitrogen gas until dryness. Samples were reconstituted with I ml of acetonitrile, vortexed and then transferred to autosampler vials ready for HPLC analysis. The concentrations of PAHs were determined using HPLC apparatus (Shimadzu, Kyoto, Japan) equipped with ZORBAX Eclipse PAH column, 3.5 µm (4.6 mm x - 150 mm). Lc Solution software was used for data collection and processing (Shimadzu, Kyoto, Japan). The mobile phase consisted of water solution (A) and acetonitrile solution (B). The gradient elution progress was as follows: 50–50% A/B at 0–2 min, 50–50% A/B at 2–22 min, 100–100 % A/B at 22–28 min. The flow rate was 1.5 mL/min. The auto-sampler temperature was maintained at ambient temperature, and the sample injection volume was 50 µL. Evaluation of surface runoff contamination was based on measurements of 16 PAHs during two winter season according from the priority list of US EPA Method No.610. The analyzed PAHs were Naphthalene (NP), Acenaphthylene (ACY), Acenaphthene (ACE), Flourene (FLU), Phenanthrene (PHE), Anthracene (ANT), Flouranthene (FLA), Pyrene (PYR), Benzo(a) anthracene (BaA), Chrysene (CHR), Benzo(b)flourenthene (BbF), Benzo(k)flourenthene (BkF), Benzo(k)pyrene (BkP), Dibenzo (a,h) anthracene (DahA), Benzo (g, h, i) perylene (BghiP), Indeol (1,2,3-c,d) pyrene (IcdP). PAHs are classified into two classes, low molecular weight (LMW) which are made of 2–3 rings and high molecular weight (HMW) consisting of 4–7 rings. All the PAH concentrations are reported on µg/L. Each site was sampled twice at the beginning and the end of winter season.

To assess the linearity of the HPLC results, calibrations were conducted at the following five concentration levels of PAHs in acetonitrile: 10, 20, 50, 100, and 200 µg/L. These calibration standards were tested before and after the collected samples analysis. Additionally, all stock solutions were stored in amber glass bottles and placed in a freezer. To assess any cross contamination that occurred through sample preparation and extraction, a blank sample containing only acetonitrile was processed with the samples. A control sample containing PAHs and blank sample was injected to measure the recoveries of PAHs. The median recoveries were calculated approximately 70–110%, which are comparable to those values reported for PAHs in the literature. Finally, the estimated detection limit of PAHs was 0.01 µg/L.

Average daily traffic data

Traffic volume information were measured at monitoring sites to investigate the correlation of land use (traffic volume) in the catchment and stormwater runoff quality generated from the catchment. Collection of the traffic volume information were conducted by Central Traffic Department in Greater Amman Municipality in five monitored sites for 2013 and 2014 (table 2).

Table 1. Characteristics of the location of monitoring sites.

Site Name	Land use	City	Elevation (m)	Average Rainfall (mm) 2012–2013	Average Rainfall (mm) 2013–2014
Abu-Nusair	Residential	Amman	922	351.8	278.6
Um-Alsummaq	Residential	Amman	931	506.9	372.2
Al-Abdali	Commercial	Amman	787	393.1	276.1
Downtown	Commercial	Amman	750	273.2	192.5
Al-Qunyyah	Agriculture	Jarash	522	291.8	169.4
Sahab1	Industrial	Sahab	600	204.2	196.5
Sahab2	Industrial	Sahab			

Average daily traffic data is collected using an automated traffic counter (pneumatic road tubes). These portable sensors are attached to the road closed to the storm water runoff sampling points and record traffic data typically for 7 days for two years 2013 (26 June to 1 July) and 2014 (12–18 April). Traffic volume was measured at each street next to the sampling point at all monitoring site except Al-Qunyyah site. Daily Traffic were measured two times for continuous seven days (1 week) during April 2013 and May 2014. The measured readings for the 7 days for the two periods (2013& 2014) were averaged as Average Daily Traffic (ADT) and shown in Table 2. The results showed that the highest value of ADT was recorded at Al-Abdali site. This is due to many commercial activities exist at the catchment.

Results and discussions

PAHs concentrations for residential area

The anthropogenic activities in the residential area contribute to the emission of PAHs to the street runoff of both petrogenic and pyroletic types such as vehicle emission, leakage of diesel refill abrasion of the asphalt and wheels, oil leakage and others.

Table 3 shows that all individual PAHs were detected at both sites except the NP at Um-Alsummaq site. Total PAHs in stormwater samples from all the sampling sites were in the range of 0.19–3.3 ppb. The total PAHs concentration was lower at Um-Alsummaq site (1.12 ppb) than Abu-Nusair site (2.22 ppb). The average concentration of IcdP (0.5 ppb) (HMW compound) was the highest value at Abu-Nusair site among other PAHs compounds. While the average concentration of ANT (0.27 ppb) (LMW compound) was the highest value at Um-Alsummaq site.

The Average Daily Traffic (ADT) at Abu-Nusair (13881) was much higher than Um-Alsummaq (1542) (~ 10 times higher). This can be attributed to higher anthropogenic activities at the later site than former site. For example, there are a difference in the population density and type of housing between Um-Alsummaq and Abu-Nusair. Actually, Um-Alsummaq is a wealthy residential with a two floors buildings or villas while the type of Abu-Nusair building is mainly four-storey apartment buildings.

It is interested to mention that the total PAHs at Um-Alsummaq (1.12 ppb) was higher than those reported by previous study conducted at the same site 10 years ago (Jiries, Hussein, & Lintelmann, 2003). This study has reported that the PAHs concentration ranged from 0.066 to 0.49 ppb with a mean value of 0.196 ppb which is much lower than the current value reported at this study (1.12 ppb).

PAHs concentrations for commercial area

The main anthropogenic activity contributing to the PAHs in the stormwater runoff at this site which is located in the commercial part of Amman city is the traffic activities with traffic density higher than other sites

Table 2. Average daily traffic (ADT) measured at the monitoring sites.

	Average Daily Traffic (ADT)		
Site	2013	2014	Average
Al-Abdali	52470	56819	54644
Abu-Nusair	13816	13945	13881
UmAlsoummaq	1584	1501	1542
Dwontown	31902	33216	32559
Sahab	5262	6505	5883
Al-Qunyyah	.	.	~500[a]

[a]This value of ADT was not measured but it was estimated based on the literature (Al-Masaeid et al.,1998).

Table 3. PAHs concentrations (ppb) of stormwater runoff generated from Um-Alsommaq and Abu-Nusair sites during two winter seasons.

	Abu Nusair Site			Um Alsoummaq Site		
Typical parameters	Average	Min.	Max.	Average	Min.	Max.
Naphthalene (NP)	0.01	<0.01	0.02	<0.01	<0.01	<0.01
Acenaphthylene (ACY)	0.01	<0.01	0.02	0.01	<0.01	0.02
Acenaphthene (ACE)	0.01	<0.01	0.02	0.05	<0.01	0.15
Flourene (FLU)	0.11	<0.01	0.36	0.18	<0.01	0.65
Phenanthrene (PHE)	0.06	<0.01	0.23	0.16	<0.01	0.59
Anthracene (ANT)	0.19	<0.01	0.53	0.27	<0.01	0.94
Flouranthene (FLA)	0.24	0.01	0.53	0.09	<0.01	0.34
Pyrene (PYR)	0.13	<0.01	0.33	0.01	<0.01	0.01
Benzo(a) anthracene (BaA)	0.04	<0.01	0.12	0.07	<0.01	0.21
Chrysene (CHR)	0.04	<0.01	0.09	0.02	<0.01	0.04
Benzo(b)flourenthene (BbF)	0.13	<0.01	0.29	0.02	<0.01	0.05
Benzo(k)flourenthene (BkF)	0.09	<0.01	0.24	0.01	<0.01	0.01
Benzo(k)pyrene (BkP)	0.41	0.05	1.20	0.03	<0.01	0.10
Dibenzo (a,h) anthracene (DahA)	0.03	<0.01	0.06	0.03	<0.01	0.08
Benzo (g, h, i) perylene (BghiP)	0.21	<0.01	0.79	0.07	<0.01	0.23
Indeol (1,2,3-c,d) pyrene (IcdP)	0.50	<0.01	1.48	0.09	<0.01	0.31
Total	2.22	.	.	1.12	.	.

investigated in this study. Table 4 shows that all the tested PAHs compounds were detected in both sampling sites (Al-Abdali and Downtown). Total PAHs in stormwater samples from these sites were in the range of 0.21–-7.01 ppb.

The total PAHs was higher at Al-Abdali site (3.16 ppb) than at the Downtown site (1.55 ppb). Among the tested PAHs compounds, the average concentration of BghiP was the highest value at Al-Abdali site (0.73 ppb) and at Downtown site (0.38 ppb). BghiP compound is HMW containing five aromatic rings. This is indicating that the predominance source of PAHs in commercial area is mainly the high traffic volume at both sites (See ADT values in Table 2).

The results of this study were compared with an earlier study conducted by Jiries et al. 2003 at the same site (Al-Abdali) in 2002. It is found that there is an increase in PAHs levels from an 0.24 ppb in 2002 to 3.16 ppb in this study (2012). This might be attributed to the sharp increase in number of vehicles in Amman city within the last eleven years. According to the Department of Statistics, the number of operating vehicles has increased in Amman city from 376457 cars in 2002 to 963211 cars in 2012.

PAHs concentrations for agricultural area

The site was chosen at Al-Qunyyah village to represent agricultural activities. It is located around 40 Km North West of Amman, the capital city of Jordan. The concentrations of individual PAHs are shown in Table 5 representing PAHs concentrations during the two winter seasons. All the tested PAHs compounds were detected in Al-Qunyyah site. Total PAHs in stormwater samples from the sampling site were in the range of 0.18–6.25 ppb with the average of 1.84 ppb. Among the tested PAHs compounds, the average concentration of PHE was the highest value at Al-Qunyyah site (0.77 ppb). PHE compound is LMW containing three aromatic rings where is used in manufacturing of resins and pesticides. Actually, pesticides are widely used in this agricultural area. Therefore, this might be explained the high concentration of PHE at this site.

Table 4. PAHs concentrations (ppb) of stormwater runoff generated from Al-Abdali and Downtown sites during two winter seasons.

	Al-Abdali Site			Downtown Site		
Typical parameters	Average	Min.	Max.	Average	Min.	Max.
Naphthalene (NP)	0.09	<0.01	0.33	0.06	<0.01	0.20
Acenaphthylene (ACY)	0.01	<0.01	0.01	0.02	<0.01	0.06
Acenaphthene (ACE)	0.53	<0.01	1.99	0.28	0.01	0.84
Flourene (FLU)	0.24	<0.01	0.58	0.20	<0.01	0.77
Phenanthrene (PHE)	0.10	<0.01	0.34	0.08	<0.01	0.29
Anthracene (ANT)	0.08	<0.01	0.22	0.03	<0.01	0.08
Flouranthene (FLA)	0.12	<0.01	0.37	0.04	<0.01	0.08
Pyrene (PYR)	0.08	<0.01	0.28	0.07	<0.01	0.22
Benzo(a) anthracene (BaA)	0.19	<0.01	0.36	0.04	<0.01	0.08
Chrysene (CHR)	0.07	<0.01	0.18	0.02	<0.01	0.05
Benzo(b)flourenthene (BbF)	0.16	<0.01	0.61	0.01	<0.01	0.02
Benzo(k)flourenthene (BkF)	0.08	<0.01	0.30	0.01	<0.01	0.03
Benzo(k)pyrene (BkP)	0.02	<0.01	0.04	0.03	<0.01	0.08
Dibenzo (a,h) anthracene (DahA)	0.12	<0.01	0.45	0.09	<0.01	0.32
Benzo (g, h, i) perylene (BghiP)	0.73	<0.01	2.86	0.38	<0.01	1.38
Indeol (1,2,3-c,d) pyrene (IcdP)	0.54	<0.01	1.87	0.18	<0.01	0.62
Total	3.16	.	.	1.55	.	.

PAHs concentrations for industrial area

For industrial area, the monitoring sites (Sahab1 and Sahab2) showed different PAHs concentrations as total PAHs was 3.27 ppb at Sahab1 and 1.54 ppb at Sahab2 with a general trend of higher concentrations at Sahab2 compared with Sahab1 site (table 6). The total PAHs concentration was two times higher in the Sahab1 than in the Sahab2.

At Sahab1 site, FLU, ACE, and BkF were the most abundant compounds which accounted for more than 50% of the total PAHs, while ANT, PYR and BghiP were the most abundant compounds at Sahab2. It is well documented in the literature that the FLU, ACE, ANT, and PYR are widely used in manufacturing of pigments, dyes and plastics. Actually, there are some factories in Sahab1 and Sahab 2 areas which are manufacturing pigments and dyes. Total PAHs in stormwater samples from all the sampling sites were in the range of 0.26–11.03 ppb. All PAHs compounds were detected in collected samples except NP and DahA, which were under the limit of detection in both sites. FLU was the dominant compound and contributed to 53% of the total PAHs at Sahab1, while BghiP was the dominant compound and contributed to 30% of the total PAHs at Sahab2. This is consistent with previous studies which showed that fluoranthene typically have higher reported concentration in stormwater runoff than other PAHs (DiBlasi, Li, Davis, & Ghosh, 2009; Hwang & Foster, 2006; Menzie, Hoeppner, Cura, Freshman, & LaFrey, 2002. Stein, Tiefenthaler, & Schiff, 2006).

Relation between the PAHs and traffic volume

Traffic volume is one of the prominent sources of polycyclic aromatic hydrocarbons (PAHs) (Gunawardena et al., 2014). Different monitoring locations showed different concentrations of the total PAHs in stormwater runoff samples (sum of the two- to six-ring PAHs) which reflects the emission of these compounds due to different anthropogenic activities. The highest total PAHs in stormwater runoff was generated from industrial catchment (2.42 ppb) while the lowest generated from agriculture catchment (1.66 ppb). The total PAHs generated

Table 5. PAHs concentrations (μg/L) of stormwater runoff generated from Al-Qunyyah during two winter seasons.

	Al-Qunyyah Site		
Typical parameters	Average	Min.	Max.
Naphthalene (NP)	0.01	<0.01	0.01
Acenaphthylene (ACY)	0.01	<0.01	0.01
Acenaphthene (ACE)	0.02	<0.01	0.03
Flourene (FLU)	0.03	0.02	0.05
Phenanthrene (PHE)	0.77	<0.01	3.07
Anthracene (ANT)	0.01	<0.01	0.01
Flouranthene (FLA)	0.41	<0.01	1.44
Pyrene (PYR)	0.15	<0.01	0.54
Benzo(a) anthracene (BaA)	0.21	<0.01	0.65
Chrysene (CHR)	0.02	<0.01	0.03
Benzo(b)flourenthene (BbF)	0.01	<0.01	0.02
Benzo(k)flourenthene (BkF)	0.04	<0.01	0.12
Benzo(k)pyrene (BkP)	0.02	<0.01	0.04
Dibenzo (a,h) anthracene (DahA)	0.06	<0.01	0.20
Benzo (g, h, i) perylene (BghiP)	0.04	<0.01	0.12
Indeol (1,2,3-c,d) pyrene (IcdP)	0.03	<0.01	0.08
Total	1.84	.	.

from the four catchment (industrial, agricultural, commercial, and residential areas) were compared with ADT measured at each land use as shown in figure 2. The results showed that there was no particular trend seen between total PAHs and ADT at each different land uses. This is suggesting that there are other sources also found to contribute to the total PAHs carried by stormwater runoff. However, the sites in residential and commercial areas showed a positive linear relationship between PAHs levels and traffic volume.

Table 7 shows the concentrations of detected PAHs generated from the four studied land use areas; Residential (Abu-Nusair & Um-Alsummaq), Commercial (Al-Abdali & Downtown), Industrial (Sahab1 & Sahab2) and Agriculture (Al-Qunyyah). For all catchments, the concentrations of detected PAHs were relatively low in concentrations and mostly of low molecular weight which can be attributed to the low solubility of these compounds in water especially with increasing their molecular weight as they often bind to particles and tend to accumulate in the upper layer of soil (Yonge, Hossain, Barber, Chen, & Griffin, 2002).

Petroleum and combustion-related sources of PAH emission were identified on the basis of ∑LMW/∑HMW, FLA/(PYR + FLA), An/178, PHE/ANT and BaA/228 analysis in the stormwater runoff samples collected from four areas.

Isomer pair ratios and individual compound ratios were used to identify the possible emission sources of PAHs in stormwater runoff. The results of various PAHs ratio in the four areas is given in Table 7. For mass 178 (ANT/178), which is the ratio of anthracene to anthracene plus phenanthrene concentrations. Ratio of less than 0.10 usually is taken as an indication of petroleum such as lubricating oil, diesel fuel, gasoline, asphalt and kerosene while a ratio of above 0.10 indicates a dominance of combustion (Budzinski, Jones, Bellocq, Pi´erard, & Garrigues, 1997). ANT/178 ratio in the four areas ranged from 0.27 to 0.54 where it was above 0.1 limit indicating that the main source of PAHs in this monitoring catchment is from combustion origin. Moreover, the highest value of ANT/178 ratio was produced from industrial catchment while the lowest was produced from agriculture catchment. For FL/FL+PYR

Table 6. PAHs concentrations (ppb) of stormwater runoff generated from Sahab1 and Sahab2 sites during two winter seasons.

	Sahab1 Site			Sahab2 Site		
Typical parameters	Average	Min.	Max.	Average	Min.	Max.
Naphthalene (NP)	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Acenaphthylene (ACY)	0.07	<0.01	0.25	0.04	<0.01	0.11
Acenaphthene (ACE)	0.33	<0.01	1.24	0.03	<0.01	0.08
Flourene (FLU)	1.74	0.05	6.71	0.03	<0.01	0.08
Phenanthrene (PHE)	0.03	<0.01	0.09	0.09	0.02	0.25
Anthracene (ANT)	0.09	<0.01	0.33	0.24	<0.01	0.79
Flouranthene (FLA)	0.08	<0.01	0.20	0.19	0.04	0.53
Pyrene (PYR)	0.06	<0.01	0.18	0.23	0.02	0.84
Benzo(a) anthracene (BaA)	0.04	0.02	0.08	0.11	<0.01	0.21
Chrysene (CHR)	0.03	<0.01	0.10	0.02	<0.01	0.06
Benzo(b)flourenthene (BbF)	0.10	<0.01	0.34	0.02	<0.01	0.03
Benzo(k)flourenthene (BkF)	0.46	<0.01	1.82	<0.01	<0.01	<0.01
Benzo(k)pyrene (BkP)	<0.01	<0.01	<0.01	0.03	<0.01	0.08
Dibenzo (a,h) anthracene (DahA)	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Benzo (g, h, i) perylene (BghiP)	0.16	<0.01	0.63	0.46	<0.01	1.44
Indeol (1,2,3-c,d) pyrene (IcdP)	0.04	<0.01	0.14	0.02	<0.01	0.05
Total PAHs	3.27	.	.	1.54	.	.

ratio was also used to find the source of PAHs. Li et al. 2006 reported that when the ratio (FL/FL+PYR) of less than 0.4 indicates petrogenic origin and especially of 0.4–0.5 are more characteristic for liquid fossil fuel and those of above 0.5 pyroletic origin. For stormwater runoff samples collected from four areas, the ratios ranged from 0.67 to 0.9. These values are slightly above 0.5 which is indicating that the source could be a mixture of both petrogenic and pyroletic. The petrognic is mainly from the abrasion of the asphalts, lubricating oils and tire abrasion and leakage from factories, while the pyroletic origin from burning fossil fuel in factories, automobile emission and diesel emission which is widely used in farming activities. The above source is supported by using BaA/228 ratio as ratios of less than 0.20 imply petroleum, from 0.20 to 0.35 indicate either petroleum or combustion, and more than 0.35 imply combustion (Yunker et al., 2002). For the industrial and agriculture areas, the value of BaA/228 ratio was 0.2 for both areas. This is suggesting that the PAHs source might be petroleum or combustion origin. However, the BaA/228 ratio values for commercial and residential were 0.64 and 0.58, respectively. This is suggesting that the PAHs source might be the combustion origin. HMW of PAHs are usually sourced from pyrolytic origin, whereas PAHs from LMW parent compounds have combustion and petrogenic origins (Mai et al., 2002). Therefore, LMW/HMW ratios (two- and three-ring PAHs/four-, five-, and six-ring PAHs) >1 indicates a petrogenic origin, <1 indicates pyrolytic origin (Magi et al., 2002). HMW is higher than LMW in the commercial, residential and agricultural as LMW/HMW PAHs ratio were 0.58, 0.48, and 0.86 (<1), while it is 1.30 (>1) in industrial area. This is suggesting that the sources of PAHs at industrial area are clearly different from those at the commercial, residential, and agricultural area and mainly polluted by PAHs with petrogenic origin. In conclusion, isomer pair ratios and individual compound ratios are suggesting that the main source of PAHs in AZB catchment is from pyrolytic origin especially at commercial and residential area, while a petrogenic of PAHs sources are generated in industrial area.

Polycyclic Aromatic Hydrocarbons levels were reported in some surface water and runoff in other parts of the world. The PAH concentrations found in this study were moderate compared to PAH concentrations found in run-offs in other studies. For example, PAHs in urban stormwater runoff flowing into the tidal Anacostia River ranged from 1.51 to 12.50 μg/L. (Hwang & Foster, 2006). Concentrations of total 16 PAHs (ΣPAHs) in surface water of the Hungarian upper section of the Danube River samples ranged from 0.025 to 1.208 μg/L, which were predominated by two- and three-ring PAHs (Nagy, Simon, & Szabó et al., 2013). Polycyclic aromatic hydrocarbons (PAHs) were measured in the run-off from three kinds of impervious surface in the Shanghai urban area, China. The mean sum of the 16 PAH concentrations measured in run-offs from ceramic tiles, asphalt roofs, and asphalt roads were 1.404, 1.743, and 4.023 μg/L, respectively (Hou, Bian, & Li, 2013). It can be concluded that there is heterogeneity between the PAHs results reported in the literature. The variability in results supports the need for long-term field monitoring to better understand catchment responses and to improve the calibration of currently used simulation models (Guzha, Rufino, Okoth, Jacobs, & Nóbrega, 2018).

Conclusions

A study of 16 polycyclic aromatic hydrocarbons in stormwater runoff was conducted at seven locations in semi-arid Amman Zarqa Basin for two winter seasons (October 25, 2012 to May 31, 2014). The selected sites are representing residential, commercial, industrial, and agricultural areas. Total PAHs concentrations in stormwater samples from all the sampling sites were in the range of 0.18–11.03 ppb. The results showed that the average total concentration of PAHs of the runoff samples was the highest from industrial area (2.42 ppb), while the residential area showed significantly the lowest concentrations (1.66 ppb). There is no clear correlation between the Average Daily Traffic and the total PAHs

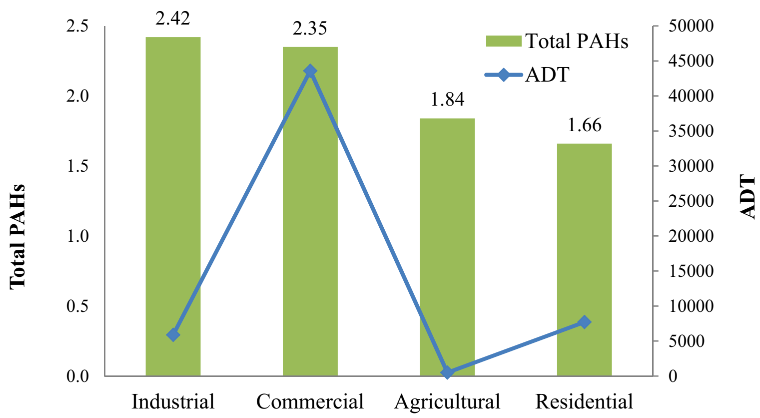

Figure 2. Relation between total PAHs concentration with ADT for different land uses.

Table 7. Individual PAHs concentrations (ppb) and ratios for surface runoff samples collected from areas with different anthropogenic activities during two winter seasons.

Typical parameters	Commercial	Residential	Industrial	Agricultural
Naphthalene	0.07	0.01	0.01	0.01
Acenaphthylene	0.02	0.01	0.06	0.01
Acenaphthene	0.41	0.03	0.18	0.02
Flourene	0.22	0.15	0.89	0.03
Phenanthrene	0.09	0.11	0.06	0.77
Anthracene	0.05	0.23	0.17	0.01
Flouranthene	0.08	0.16	0.13	0.41
Pyrene	0.07	0.07	0.15	0.15
Benzo(a) anthracene	0.12	0.06	0.07	0.21
Chrysene	0.05	0.03	0.03	0.02
Benzo(b)flourenthene	0.09	0.07	0.06	0.01
Benzo(k)flourenthene	0.05	0.05	0.24	0.04
Benzo(k)pyrene	0.02	0.22	0.02	0.02
Dibenzo (a,h) anthracene	0.1	0.03	0.01	0.06
Benzo (g, h, i) perylene	0.55	0.14	0.31	0.04
Indeol (1,2,3-c,d) pyrene	0.36	0.29	0.03	0.03
Total PAHs	2.35	1.66	2.42	1.84
LMW/HMW	0.58	0.48	1.30	0.86
ANT/178	0.36	0.68	0.74	0.01
FLA/FLA+PYR	0.53	0.70	0.46	0.73
BaA/228	0.71	0.67	0.70	0.91

carried by stormwater runoff from studied land uses (All sites). However, the sites in residential and commercial areas showed a positive linear relationship between PAHs levels and traffic volume. Isomer pair ratios and individual compound ratios are suggesting that the main source of PAHs in AZB catchment is from pyrolytic origin especially at commercial and residential area. while a petrogenic of PAHs sources is generated in industrial area. The PAH concentrations found in this study were moderate compared to PAH concentrations found in run-offs in other studies.

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Highlights

- PAH concentrations of stormwater runoff at AZB are comparable with other basins.
- PAH concentrations of industrial areas are higher than other land uses.
- Stormwater runoff contains multiple pyrolytic and pyrogenic PAH sources.
- Average Daily Traffic was not correlated to total PAHs generated from studied land uses.

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