



FULL LENGTH ARTICLE

Distribution and sources of polycyclic aromatic hydrocarbons in water and sediments of the Soltan Abad River, Iran



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Abstract Distribution of the sixteen polycyclic aromatic hydrocarbons (PAHs) was detected in water and sediments of the Soltan Abad River. Liquid–liquid extraction was used for water samples, while PAHs in sediments were extracted using Soxhlet Extraction and finally analyzed by means of gas chromatography. Results showed that in water samples, the highest concentration was related to acenaphthene (3-ring PAH), whereas fluoranthene (4-ring PAH) was the most important pollutant in sediments. The mean concentrations of PAHs was the highest in autumn and the lowest in summer, in both water and sediment samples. Diagnostic ratios such as phenanthrene/anthracene and fluoranthene/pyrene were achieved to evaluate the emission sources of PAHs. These ratios indicated a pyrolytic source of PAHs for sediments, as well as a pyrolytic or pyrogenic origin for water samples (with a dominant pyrolytic input) in the study area.

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are a ubiquitous group of different organic compounds with fused aromatic rings that are detected in a variety of environmental conditions. Their physical and chemical characteristics are varying with both the number of aromatic rings and the pattern of ring linkage. PAHs have carbon and hydrogen atoms in their structures but some carbons can be substituted by some elements such as nitrogen, sulfur and oxygen, resulting in polycyclic heteroaromatic hydrocarbons, e.g. carbazole and furans

(Bayoumi, 2009). PAHs are lipophilic compounds that their solubility and so their concentration in water is trivial (Nasr et al., 2010; Qiu et al., 2009). PAHs' solubility in water decreases with increase in their molecular weight (Nikolaou et al., 2009). As a consequence of PAHs' hydrophobic properties, they tend to settle out of water and accumulate in the bottom sedimentation. Therefore, concentration of PAHs should be high in the sediments of polluted aquatic environments. Both anthropogenic activities and natural emissions are the PAH sources of coastal sediments. Anthropogenic sources include petrogenic and pyrolytic PAH contaminations. Pyrolytic sources include incomplete combustion of organic matter such as fossil fuels and biomass whereas the petrogenic PAHs are formed by petroleum products (Qiu et al., 2009; Perra et al., 2009). The PAH composition within the sediments reflects the origin(s) from which they were derived (Simpson

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et al., 1998; Yan et al., 2009). Lower molecular weight PAHs most often come from natural resources (petrogenic) of PAHs and higher molecular weight PAHs often originate from combustion (pyrolytic) (Helfrich and Armstrong, 1986)

Since some of PAHs and their metabolites, can form DNA adduct which can induce mutations; therefore their entrance into the bodies of organisms can be dangerous. Their carcinogenic and mutagenic properties cause to classify 16 of them as priority pollutants by the USEPA. Some authors mentioned that PAHs can be synthesized by unicellular algae, bacteria or higher plants but they accumulate PAHs rather than synthesize them (Magi et al., 2002).

The Soltan Abad River is located in the south of Shiraz. It passes through the industrial town of Shiraz, and flows into Maharlu Lake. The industrial town of Shiraz has numerous factories and industries (industrial materials and chemical products, rubber and plastics, metal artifacts, etc.) which all have organic matter like PAHs. Shiraz urban sewage and agricultural lands around Shiraz are other PAH sources in the Soltan Abad River (Kafilzadeh and Moghtaderi, 2015). The purpose of this study is to analyze the distribution of the sixteen PAHs in water and sediments of the Soltan Abad River through four seasons and to determine their source (s) on the basis of their concentrations.

Materials and methods

Water and sediment samples were collected seasonally during the period from April 2013 to March 2014 from four sampling stations. Location of the stations was determined by the global positioning system (GPS) (Table 1). Each sampling was carried out in five replicates. Water samples were taken at the surface and 50 cm below the surface level. Simultaneously sediment samples were taken at a depth of 5 cm of sediment surface from the idem sites. All samples were instantly transferred to the laboratory for further experiments. Prior to analysis samples were allowed to air dry in dark condition for 48 hours. PAHs were extracted using liquid-liquid extraction (LLE) with 100 ml of n-hexane and dichloromethane mixture (1:1 v/v) as described in APHA (APHA, 1998). Before extraction, water sample (800 ml) was filtered with Whatman filter paper (i.d. 70 mm) to remove debris and suspended materials. The extract was concentrated to a final volume of 2 ml under a gentle stream of nitrogen using a rotary evaporator and then analyzed with Gas Chromatograph (Siriwong et al., 2009).

Air-dried sediments were sieved (mesh size 500 μm) and homogenized in mortar. PAHs in sediments were extracted using Soxhlet Extraction. A 10 g homogenized sample was

extracted with 250 ml of dichloromethane for 16 h and concentrated to 2 ml using vacuum rotary evaporator (USEPA, 1996).

Silica/alumina column chromatography was used for clean-up and separation of water and sediments. Saturated aliphatic and aromatic hydrocarbons were washed with 20 ml of n-hexane and 30 ml of a mixture of hexane and dichloromethane (90:10) (v/v) respectively. To detect 16 PAH components, 1 ml of aromatic hydrocarbon fraction was injected into a gas liquid chromatography equipped with a flame ionization detector (GC/FID) (Nasr et al., 2010). GC analysis was implemented on a fused silica capillary column of 60 m length, 0.25 mm ID and 0.5 μm film thicknesses. The following PAHs were used for quantitation: naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene, benzo [a] anthracene, benzo [b] fluoranthene, benzo [k] fluoranthene, benzo [a] pyrene, dibenzo [a,h] anthracene, benzo [g,h,i] perylene, Indeno [1,2,3-cd] pyrene.

For correction of the data, recoveries were fulfilled by the addition of 1, 5 and 10 μg of PAH standard mixtures. The average percentage recovery of total PAHs achieved 96.80% and 91.26% for water and sediment samples, respectively. An analysis of variance (ANOVA) was performed using SPSS software (version 16), and mean comparison was conducted with Duncan test.

Results and discussion

The concentrations of the 16 detected PAHs in surface water and sediments of Soltan Abad River are shown on Tables 2 and 3. Most PAHs were detected at water and sediments of all stations. As can be seen in water samples, the highest concentration was related to acenaphthene (3-ring PAH), whose mean value (ng L^{-1}) in four sites was 16.20, 22.66, 45.58 and 66.30, respectively. ANOVA and Duncan tests showed a significant difference between mean concentration of acenaphthene and other detected PAHs ($P \leq 0.01$). At the same time, the lowest concentration of PAH components in water samples of Soltan Abad River was related to Indeno [1,2,3-cd] pyrene. The associated mean values (ng L^{-1}) were 0.40, 0.63, 0.95 and 1.12. According to our results, the concentration of low molecular weight (2–3 ring) polycyclic aromatic hydrocarbons (LPAHs) in water samples of Soltan Abad River is higher than high molecular weight (4–6 ring) PAHs (HPAHs) (Fig. 1A). The mean values of total concentration (mg L^{-1}) of LPAHs and HPAHs in four sampling sites were 120.23 and 28.58 respectively, which are significantly different ($P \leq 0.01$).

As far as the sediment samples are concerned, fluoranthene (4-ring PAH) was the most important pollutant with mean concentrations (ng g^{-1}) of 27.33, 52.15, 67.59, and 65.24 in four stations, respectively. The difference between fluoranthene and other PAH concentration was highly significant ($P \leq 0.01$). Meanwhile, in terms of the lowest concentration, fluorene (3-ring PAH) ranked the first position, followed by phenanthrene (3-ring PAH). The respective Figures (ng g^{-1}) in four stations are 6.75, 7.55, 8.39, and 9.85 for fluorene and 6.82, 7.53, 7.18, and 9.76 for phenanthrene. Contrary to the observed compositions of PAHs in surface water, HPAHs are more dominant in the sediments of the Soltan Abad River rather than LPAHs (Fig. 1B). The mean total concentration values (ng g^{-1}) were 270.27 for the former and

Table 1 Location of sampling stations.

Station	Region	Coordinates
1	River sediments under the Soltan Abad entrance bridge	N29° 32' 11.1' E52° 33' 12.2'
2	River sediments under the Soltan Abad town center bridge	N29° 31' 12.7' E52° 32' 26.2'
3	River sediments under the bridge of Fasa Road Traffic Police	N29° 29' 07.6' E52° 38' 17.4'
4	Entrance of Maharlu Lake	N29° 32' 15.2' E52° 40' 28.7'

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