



Determination of Poly Aromatic Hydrocarbons (PAHs) Amounts in Anzali Lagoon (Iran) and Assessment of Their Origin

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Accepted on 21st April 2017, Published online on 27th May 2017

ABSTRACT

Biological monitoring of Bandar Anzali lagoon received much attention regarding to its international emphasis and ecosystem condition of aquatic growing and upbringing in south of the Caspian Sea. This research focused on the concentration specification and source of oil pollutants, poly aromatic hydrocarbons (PAHs), in the surface sediments of the aforementioned lagoon. Sampling carried out in two different time periods of June and September in two main stations of Selke and Mahruzeh. Accordingly based on the GC-Mass analyses, PAHs had the mean value of 34.839 ng dg⁻¹ and 61.439 ng dg⁻¹ for Mahruzeh station in June and September, respectively. While represented a mean value of 80.432 ng dg⁻¹ and 45.439 ng dg⁻¹ in Selke station for warm and cold seasons, respectively. In contrast with the other countries in the Caspian Sea border, foregoing stations had the least value of contaminations at their maximum value. In addition it was found that exact source of this kind of oil pollution is not clear regarding to the observation of both pyrogenic and petrogenic origin. What's more based on the statistical analyses, $P\text{-value} > 0.05$, there isn't any significant difference between these two objective stations.

Keywords: Bandar Anzali lagoon, Mass Chromatography Gas, PAHs, Sediment.

INTRODUCTION

Exploration and exploitation of several rich oil reservoirs in the ooze and border of the Caspian Sea generated oil pollution terribly in that zone in the recent decades. Considering the Caspian Sea Environmental Protection agency (CEP) reports, Iran faces sever aquatic and environmental dangers as a result of natural characteristics of the Caspian Sea. While it has the sharpest depth through Iran borders which result in ramping the pollutants. Correspondence of Bandar Anzali to the Caspian Sea, tiding and water fluctuations are responsible for exposure of the aforementioned lagoon to oil contaminants as PAHs. Besides industrial wastewaters of nearly 50 factories and million tons of urban wastewaters which introduced to the lagoon play an important role for contamination via adjacent regions. Furthermore unclean rivers unload huge volume of industrial and urban wastewaters to the lagoon. In the last decades carcinogenic and mutagenic properties of the PAHs increased the worldwide concerns. Most categories of acting as transportation of petroleum fuels, crude oil operational activities, unprincipled discharge of

industrial effluents and some others play the important role as the petrogenic sources which are responsible for entrance of these contaminants to the aquatic environment [1],[31-32]. But definitely pyrogenic sources as combustion of fossil and bio fossil fuels in different industries and human activities are the main parent of this contamination. On the other hand, injection of diesel engines and particularly activities of some unclean industries were accompanied with emission of hazardous particles and finally air pollution occurred. Consequently falling out of different kinds of pollutants to the ground can contaminate aquatic environments. But generally deep PAH contamination occurred in the port towns and near the beach border [2-5]. These compounds can arrive to the food chain easily and so to the human bodies regarding to their lipophilic essence. Thereafter bioaccumulates in the lipids and dominates serious irrecoverable cancerous signs. In addition they simply can penetrate to the sediments because of their heavy mass [6]. These contaminants estimation needs a sharp accuracy while they have sever dangerous effects by permeating to the human bodies via aquatic usage. For the sake of these problems, recent studies all around the world represented a sort of anxiety considering PAH concentrations which is reported in table 1. Evaluation of PAHs concentration in the Bandar Anzali lagoon can open a new window to the future works while provided a deep sight of situation qualification. It should be mentioned that recent researches on the contamination of PAHs in the Bandar Anzali lagoon involved the Siah-Keshim zone and the following understudy stations of the foregoing lagoon had been investigated as the first attempt.

Table 1: Different reports of PAH concentrations and origins in different parts of the world

Zone	Σ PAHs (ng/g) ¹	Origin	Ref.
North America			
Entire US coast	13.4-40453		EPA EMAP Program ²
Entire US coast	4.87-30674		NOAA Status and Trends program ³
Pales Verdes Shelf, CA, USA	1252-7037		[7]
Alaska stations	2.17-733		[8]
West Beaufort Sea (Polar Star sediments)	159-1092		
Fraser Estuary (BC, Canada)	180-620	combustion	[9]
Fraser Estuary (BC, Canada)	220-660	petroleum	
Burrard inlet (BC, Canada)	430-91800	combustion	
Burrard inlet (BC, Canada)	70-39500	petroleum	
Strait of Georgia (BC, Canada)	300-8470	combustion	
Strait of Georgia (BC, Canada)	560-4300	petroleum	
San Francisco Bay	40-6300	pyrogenic	[9]
San Francisco Gulf	31-316		[10]
South Carolina estuaries	33-9630		[11]
Narragansett Bay, RI	100-29300		[12]
New Bedford Harbor, MA	14000-170000		[2]
Europe			
Eastern Mediterranean Sea	20-18700		[13]
Baltic Sea	3.16-30100		[14]
Irish estuaries	83-22960		[15]
Gironde Estuary (France)	3.5-853		[16]

¹ ng g⁻¹: nanogram/dried weight

² <http://www.epa.gov/emap/nca/html/data>

³ <http://ccmaserver.nos.noaa.gov/nsandtdata/NSandtdatasets/benthicsurveillance/welcome.html>

Arcachon Bay (France)	293	
Lazaret Bya (central Mediterranean)	86.5-48060	[17]
Near- coastal Spain and France (Mediterranean Sea)	0.32-8400	[18]
White Sea (Russia, Arctic Ocean)	13-208	[19]
Africa		
Cotonou coast (Benin)	80-1411	[16]
Asia		
Kyeonggi Ba (Korea)	9.1-1400	[4]
Surface Sediment (Hong Kong)	7.25-4420	[5]
South china Sea (China)	24.7-275.4	[20]
Bohai Bay (China)	31-2513	[21]
Yellow Sea (China)	20.5734	[22]
Persian Gulf Surrounding Countries		
United Arabic Emirates	4.9-39	[23]
Qatar	55-92	
Bahrain	13-6600	
Oman	6.1-30	
Caspian Sea Bordering Countries		
Azerbaijan	338-2988	[24]
Kazakhstan	6-294	
Russia	6-345	
Caspian Sea (Iran)	84-1789	

Abbreviations: It is necessary to introduce the abbreviations of the whole PAHs compounds which are presented in the result table.

Naphthalene	NAP
Acenaphthylene	ACL or ACPY
Acenaphthene	ACN or ACP
Fluorene	FLU or FL
Phenanthrene	PHE
Anthracene	ANT
Fluoranthene	FLR or FLUR
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	INP
Dibenzo (ah) anthracene	DBA
Benzo(ghi)perylene	BPR

MATERIALS AND METHODS

In this study normal hexane, acetone and active copper powder were obtained from Merck and used without any purification. Based on the strategic policies two stations selected depends on their geographical zone of interception of different seasonal rivers and adjacency of protection zone, Mahruzeh and Selke, respectively (Fig. 1). As the literature [10], [25] only two types of climate condition samplings applied as dry and warm seasons and wet and rainy seasons. As a result based on the continental and

geographical conditions, local sampling facilities and considering the fact that there is n't any remarkable temperature changes during the year in the Bandar Anzali, sampling involved June and September seasons [32]. Sampling done via Van Veen Grab in one kilometer length of the sampling zone which involved three samples for each station. At first samples collected and provided by keeping in the Aluminum vessels at freezer and analyzed based on the MOOPAM, 1999 instruction. The beforehand samples dried completely by the freezer dryer apparatus (VaCo-ZiRBus) and introduced into the micro wave vessels after they were weighted. At this time sediments charged with extractive solvents, normal hexane and acetone, with volume ratio of 1:1 (15 mL respectively) and remained in the macro wave apparatus under 180 °C for residence time of 30 minutes. In the next stage samples had been filtered by suitable filter paper behind the aspirator and provided of 1 mL by volume via addition of N₂ gas. Thereafter 0.1 mL of normal hexane added. Then a mixture of 1 g of active copper powder with 5 mL of normal hexane centrifuged with the present samples. In this way the troublesome sulfur components extracted from the sediment. Above all in the end samples thickened by N₂ gas to 1 mL volume and passed through a 1 cm diameter column of silica gel, alumina and sodium sulfate. This chromatography column filled with three different adsorbents with various polarities which can extract PAHs easily. This effluent solution injected to the GC-Mass analyzer after it was concentrated [26-27]. Finally experiments had been completed by SPSS software employed for data processing. Whereof data proceeded an abnormal manner, non-parametric and Mann-Whitney u test were applied for further analysis.

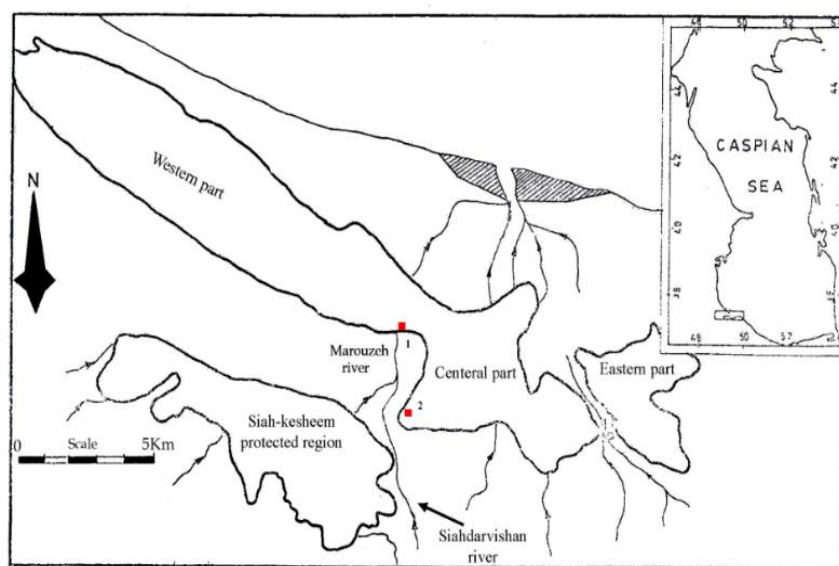


Figure 1: Geographical location of sampling stations (■ 1: Mahruzeh station: 39 S 0359731, UTM 4143193; ■ 2: Selke station: 39 S 0363473, UTM 4139305)

RESULTS AND DISCUSSION

Mahruzeh station in June: Table 2 summarized the analyzed results while Sed. represented sediment with M₁ as the mean value of pollutant concentration in the first sampling point in this station. Similarly M₂ and M₃ shows the mean value of pollutants at second and third sampling points, respectively. Additionally in the following table “J” refers to June. It should be mentioned that repeatability of all data monitored was three times. Also the oil pollution source was determined. That is to say from Table 3 it is clearly obvious that M₁/J and M₃/J have petrogenic and pyrogenic sources, respectively. In contrast, for M₂/J there isn't any specific source according to FLUR/PY and PHEN/AN ratio.

Table 1: PAH concentrations for Mahruzeh station, June 2010

PAH compounds (ng/g)	Sed.M ₁ /J	Sed.M ₂ /J	Sed.M ₃ /J	Mean
NA	0.906	0.785	1.039	0.91
ACPY	2.346	2.006	1.707	2.019
ACP	0.807	0.744	0.703	0.751
FL	0.758	0.666	0.311	0.578
PHEN	6.036	5.259	5.367	5.554
AN	0.455	5.447	5.663	3.855
FLUR	2.163	1.667	1.302	1.710
PY	2.378	1.937	1.214	1.843
BaA	0.991	0.665	0.298	0.651
CHRY	2.925	2.745	1.113	2.261
BbF	7.942	6.325	2.182	5.483
BkF	3.191	2.57	0.538	2.099
BaP	0.528	0.334	0.956	0.606
ICdP	2.993	2.659	2.095	2.582
dBan	1.289	1.087	1.445	1.273
BPe	3.236	2.97	1.786	2.664
Σ PAH	38.944	37.893	27.719	34.839

Table 2: Pollutant source determination of Mahruzeh station, June 2010

Target compound (ng/g)	Sed.M ₁ /J	Sed.M ₂ /J	Sed.M ₃ /J
FLUR	2.163	1.667	1.302
PY	2.378	1.937	1.214
FLUR / PY	0.9	0.86	1.07
PHEN	6.036	5.259	5.367
AN	0.455	5.447	5.663
PHEN / AN	13.26	0.96	0.94
Source	Petrogenic	Diffuse	Pyrogenic

Mahruzeh station in September: Considering the results, maximum mean value of PAHs appeared in the sediments followed a range of 57.891-66.754 ng/g (Table 4). By the target components ratio diffuse source determined for both M₂/S and M₃/S samples. In comparison sample M₁/S displayed pyrogenic origin (Table 5).

Table 3: PAH concentrations for Mahruzeh station, September 2010

PAH compounds (ng/g)	Sed.M ₁ /S	Sed.M ₂ /S	Sed.M ₃ /S	Mean
NA	3.521	3.964	11.897	6.460
ACPY	7.663	8.169	9.915	8.582
ACP	2.991	2.93	3.325	3.082
FL	1.296	1.236	2.659	1.730
PHEN	4.991	5.428	7.57	5.996
AN	5.025	5.608	1.521	4.051
FLUR	2.515	2.511	2.323	2.449
PY	1.292	4.68	2.891	2.954
BaA	3.424	1.319	1.856	2.199
CHRY	2.402	3.403	4.773	3.526
BbF	5.87	5.598	6.557	6.008
BkF	2.29	2.174	2.765	2.409
BaP	4.914	0.52	1.217	2.217
ICdP	2.965	4.308	0	2.424

dBan	3.141	3.671	3.06	3.290
BPe	3.389	4.372	4.425	4.062
∑ PAH	57.689	59.891	66.754	61.439

Table 4: Pollutant source determination of Mahruzeh station, September 2010

Target compound (ng/g)	Sed.M ₁ /S	Sed.M ₂ /S	Sed.M ₃ /S
FLUR	2.515	2.511	2.323
PY	1.292	4.68	2.891
FLUR / PY	1.94	0.53	0.8
PHEN	4.991	5.428	7.57
AN	5.025	5.608	1.521
PHEN / AN	0.99	0.99	4.97
Source	Pyrogenic	Diffuse	Diffuse

Selke station in June: analysis revealed that maximum and minimum amounts of sediments' contamination varied in the range of 22.54-187.399 ng g⁻¹, table 6. The source determination test clarified that S₁/J had a diffuse source while S₂/J and S₃/J followed a pyrogenic source, table 7. In addition S₁, S₂ and S₃ represented the mean value of pollutant concentration in first, second and third sampling stations, respectively.

Table 5: PAH concentrations for Selke station, June 2010

PAH compounds (ng/g)	Sed.S ₁ /J	Sed.S ₂ /J	Sed.S ₃ /J	Mean
NA	0.702	0.793	1.046	0.847
ACPY	2.505	1.834	1.892	2.077
ACP	0.79	0.706	0.714	0.736
FL	5.691	0.295	0.309	2.098
PHEN	87.732	1.706	4.394	31.277
AN	12.805	1.949	0.46	5.071
FLUR	21.079	0.912	2.279	8.09
PY	26.051	0.801	2.26	9.704
BaA	6.01	0.534	1.143	2.562
CHRY	7.014	0.746	2.103	3.287
BbF	7.56	3.64	5.544	5.581
BkF	3.071	1.465	1.656	2.064
BaP	2.101	0.702	1.923	1.575
ICdP	2.236	2.64	0	1.625
dBan	0.883	1.452	3.184	1.839
BPe	1.169	2.365	2.451	1.995
∑ PAH	187.399	22.54	31.358	80.432

Table 6: Pollutant source determination of Selke station, June 2010

Target Compound (ng/g)	Sed.S ₁ /S	Sed.S ₂ /S	Sed.S ₃ /S
FLUR	21.079	0.912	2.279
PY	26.051	0.801	2.26
FLUR / PY	0.8	1.13	1
PHEN	87.732	1.706	4.394
AN	12.805	1.949	0.46
PHEN / AN	6.85	0.87	9.55
Source	Diffuse	Pyrogenic	Pyrogenic

Selke station in September: This zone had the mean maximum and minimum values of 16.276-69.832 ng g⁻¹ which are depicted in Table 8. Also fingerprinting test clarified that S₁ and S₂ had the origin of diffuse in comparison of pyrogenic source of sample S₃ (Table 9). Unfortunately in comparison of values calculated in Selke zone in June and September there is a sharp increasing in PAH concentrations in June, Sed.S₁/J= 187.399 (Table 6), since analytical results didn't represent any significant variations as P=1. As statistical analysis investigated there weren't any meaningful differences between PAH concentrations in Mahruzeh and Selke station both in June and September (P>0.05). Although Figure 2 demonstrated significant increase of PAH concentrations in June respect to September in Selke station considering Sed.S₁/J. This incrementing can arise from increasing in FLUR, PY, PHEN and AN concentrations for Sed.S₁/J (Table 6). It seems that as a result of temporary and point wise pollution this increasing occurred while it didn't display in Sed.S₂/J and Sed.S₃/J. It can be concluded that aforementioned sudden incrementing of PAH concentrations in Sed.S₁/J is abnormal and the rest concentration values followed the same trend in both Mahruzeh and Selke stations in September (Table 4, 8).

Table 7: PAH concentrations for Selke station, September 2010

PAH Compounds (ng/g)	Sed.S ₁ /S	Sed.S ₂ /S	Sed.S ₃ /S	Mean
NA	3.285	3.711	2.463	3.153
ACPY	6.931	11.125	0	6.018
ACP	2.808	3.239	0	2.015
FL	0.882	0.953	0.522	0.785
PHEN	2.139	2.373	0.284	1.598
AN	2.254	1.448	0.411	1.371
FLUR	1.669	2.15	0.709	1.509
PY	2.42	3.858	0.384	2.220
BaA	1.6	1.971	0.957	1.509
CHRY	2.153	2.708	0.752	1.871
BbF	7.442	5.506	2.994	5.314
BkF	2.934	2.84	1.064	2.279
BaP	0.605	11.943	0.545	4.364
ICdP	5.382	6.506	1.013	4.300
dBan	4.079	5.009	2.144	3.744
BPe	3.635	4.483	2.034	3.384
∑ PAH	50.218	69.832	16.276	45.439

Table 8: Pollutant's source determination of Selke station, September 2010

Target Compound (ng/g)	Sed.S ₁ /S	Sed.S ₂ /S	Sed.S ₃ /S
FLUR	1.669	2.15	0.709
PY	2.42	3.858	0.384
FLUR / PY	0.68	0.55	1.84
PHEN	2.139	2.373	0.284
AN	2.254	1.448	0.411
PHEN / AN	0.94	1.63	0.69
Source	Diffuse	Diffuse	Pyrogenic

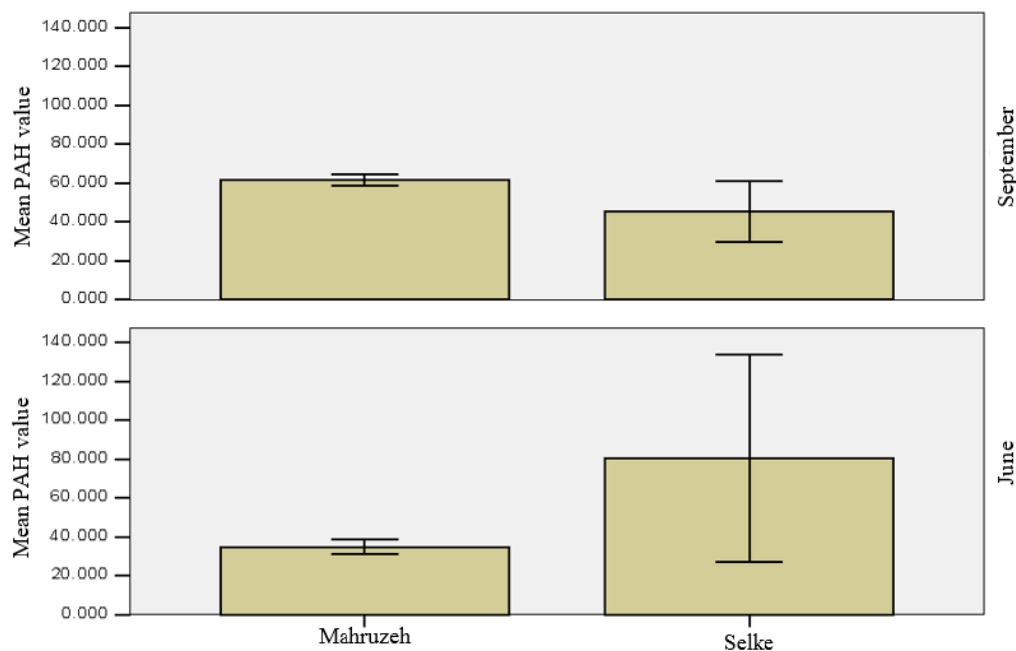


Figure 2: Comparison of PAH concentrations (ng g^{-1}) in Mahruzeh and Selke stations in June and September

APPLICATIONS

The results showed the great efficiency of such a concentration specification of the oil pollutants, poly aromatic hydrocarbons (PAHs), in the surface sediments. Hence, the method is one of the advanced processes for specifying the concentration of poly aromatic hydrocarbons in the same lagoons.

CONCLUSIONS

Regarding to the PAH hazardous identity most of the aquatic environments encounter the dangerous effects of these pollutants. In this research it was found that mean value of PAH concentration in Mahruzeh station was 34.839 ng g^{-1} and 61.439 ng g^{-1} in June and September, respectively. In comparison Selke represented mean values of 80.432 ng g^{-1} in June and 45.439 ng g^{-1} in September in this period of time. Recent reports demonstrated that maximum values of PAH concentration in the two stations were much less than values represented by the other countries except some countries in the Persian Gulf border. That is to say Oman and the United Arab Emirates had less PAH contamination than aforementioned stations in Iran. Also Qatar followed the same pollution pattern as Iran while Bahrain represented more contaminated environment. In addition Bandare Anzali lagoon in maximum amount of pollution, $34.839\text{-}80.432 \text{ ng g}^{-1}$, had lower contamination grade than other countries around the Caspian Sea such as the northern and southern countries, Kazakhstan and Azerbaijan, because of their vast petroleum activities. Fortunately Mahruzeh and Selke station didn't touch the critical values of contamination while recent studies clarified pollution levels. As critical risk values, the amount of ERL was $552 \mu\text{g g}^{-1}$, the ERM was $3160 \mu\text{g g}^{-1}$, MPL ranged from 100 to $1000 \mu\text{g g}^{-1}$ [3] and the TPL was $1000 \mu\text{g g}^{-1}$ [28]. In the case of pollution source that was evaluated in this research it seems that estimation of exact source of contamination in these understudy sediments was not possible. Namely Mahruzeh didn't represent an exact parent based on the results showed in tables 3 and 5 both in June and September. Similarly Selke had the same condition while there wasn't a distinct origin for all three samples collected for each season, Tables 7 and 9. Consistently the same results approved for Tanzania beach sediments [29]. It can be raised from different vast categories of parameters that they are responsible

in sediment formation during years. Moreover, while rainy seasons showed high value of PAH contamination it is estimated that petrogenic and pyrogenic parents of contamination strengthened with other pollution sources as industrial wastewaters combined with flowage [30].

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