Residual Concentration of PAHs in Seafood from Hormozgan Province, Iran: Human Health Risk Assessment for Urban Population

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Abstract—This study is to determine the concentrations of sixteen poly aromatic hydrocarbons in eighteen pooled samples of fish, shrimp, crab and bivalve from markets of Hormozgan province, Iran. The poly aromatic hydrocarbon levels varied from 16 ± 8.4 to 28.18 ± 3.74 ng/g wet weight. The investigated samples were classified as minimally contaminated. Distribution patterns showed that PAHs with 4, 5 and 6 rings dominated, confirming the pyrogenic source of detected PAHs. Fish contributed more than other biota groups in transforming of PAHs to Hormozgan Province people. The average of B (a) P_{eq} values for the studied biota was 2.71 ± 2.28 ng/g that was greater than calculated local screen value. This finding was implemented in poor quality of studied biota and necessity for risk management.

Index Terms—Hormozgan Province, PAH, risk assessment, seafood.

I. INTRODUCTION

Poly aromatic Hydrocarbons (PAHs) are a large group of lipophilic chemicals [1] with wide environmental distribution [2]. PAHs have been originated from diverse sources, including of the pyrolytic process such as incomplete combustion of organic chemicals [3], [4], land-based sources [5], drilling operation [6] and high contribution of activities related to petroleum industry [7]. US Environmental Protection Agency established a list of sixteen PAHs named as “16 EPA PAHs” [8]. Many of the adverse biological effects are reported for EPA PAHs [9].

Hormozgan Province in south of Iran, is bordered by the Persian Gulf and Gulf of Oman. This strategic province covers the Hormoz Strait with the vital importance in oil exporting. In addition to this, many of the industries have been established along the coastal lines of Hormozgan Province which resulted in release of untreated or imperfect treated urban and industrial wastewater in marine environment. There are no published studies related to human risk assessment from organic pollutants such as PAHs through consuming fish and seafood in southern parts of Iran. The objective of this study was to determine the levels of PAHs in commercial fish and seafood in order to assess the

exposure of Hormozgan Province consumers.

II. MATERIALS AND METHODS

A. Sampling

The common seafood samples which includes fish, shrimp, crab and bivalve, were purchased from fish markets of Hormozgan province between October 2010 and May 2011. The edible parts of collected samples were dissected. Muscle tissues without skin and bone for big size fish, for small size fish the whole edible part and for shrimp, crab and bivalve, the soft bodies were chosen. Three pooled samples for each species were prepared by mixing the transacted parts from at a minimum three numbered individuals or 1,000 g for bivalve. Samples were kept at -20°C until PAHs analysis.

B. PAHs Extraction/ Instrumental Analysis

Analyzed PAHs included naphthalene (NA), acenaphthylene (ACY), anthracene (AN), fluoranthene (FLA), pyrene (PY), benz[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenz[a,h]anthracene (DahA), benzo[ghi]perylene (BghiP), indeno[1,2,3-cd]pyrene (IcdP).

The determination of sixteen EPA PAHs was performed based on microwave extraction [10]. Mixture of about 2-5 g of freeze-dried and smashed sample with methanol was exposed to microwave radiation for 20 minutes at 115°C . Saponification of lipids was performed with addition of KOH and radiation of microwave waves for 10 minimums at 90°C . Clean up and fractionation of extract was performed using silica/alumina column. PAHs were analyzed on a capillary column of DB-5ms in gas chromatography system equipped with a mass selective detector. Injector temperature was kept at 290°C . Column temperature program started from 60°C (1 min hold) and increased to 100°C (with $10^{\circ}\text{C}/\text{min}$ ramp) and finally to 300°C (with $4^{\circ}\text{C}/\text{min}$ ramp) and was maintained for 15 minutes. Helium was used as carrier gas at 1.5 ml / min. Single ion monitoring mode was used at 128, 152, 154, 166, 178, 202, 228, 252, 276, and 278 m/z.

C. Quality Control

With each set of samples, method blanks were run to investigate the possibility of contamination peaks at monitoring ions. To assess the precision of the analytical methods, three duplications of all experiments were performed. The accuracy of analytical methods was inspected through matrix spike technique with surrogate standards. Totally, by using the certified reference material IAEA-406 for fish homogenate, the quality assurance of the procedure was confirmed. The calculated method detection limit and recoveries were 0.5- 0.7 (ng/g) and 85 – 115 respectively.

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D. Calculation of Screening Value

The screening value for carcinogenic effect of PAHs was calculated based on the following equation [11]:

$$SV = \left[(RL/CSF) \times BW \right] / CR$$

where

BW body weight(73 kg)
 CR consumption rate (55.1 g/d)
 CSF oral cancer slope factor (7.3)
 RL maximum acceptable risk level (0.00001)

The information regarding average body weight and daily consumption of seafood were provided by Hormozgan University of Medical Science and Hormozgan Fishery Office respectively.

E. Calculation of B[a] P Equivalent

B[a] P equivalents of PAHs were determined based on toxic equivalent factors (TEFs) provided by Nisbet and LaGoy (Table I) [12].

F. Determination of Dietary Intakes

Dietary intakes of PAHs were determined based on following equation:

$$\text{Dietary intakes} = \text{PAH concentration} \times \text{consumption rate}$$

TABLE I: TOXIC EQUIVALENCY FACTORS FOR INDIVIDUAL PAHS

Name	TEF	Name	TEF
Naphthalene	0.001	Benzo[a]anthracene	0.1
Acenaphthylene	0.001	Chrysene	0.01
Acenaphthene	0.001	Benzo[b]fluoranthene	0.1
Fluorene	0.001	Benzo[k]fluoranthene	0.1
Phenanthrene	0.001	Benzo[a]pyrene	1
Anthracene	0.01	Indeno[1,2,3-cd]pyrene	0.1
Fluoranthene	0.001	Dibenzo[ah]anthracene	1
Pyrene	0.001	Benzoghi]perylene	0.01

III. RESULTS

A. PAHs Concentration and Distribution Pattern

Average levels of Σ PAHs (ng/g wet weight) in seafood samples are represented in Table II. Concentrations were varied from 16 ± 8.4 to 28.18 ± 3.74 ng/g wet weight. The lowest and highest levels were related to Grunt and Banana Shrimp, respectively. Totally, the detected average of Σ PAHs for the tested biota was 20.9 ± 7.42 ng/g wet weight.

The abundance of PAH components in terms of ring number are shown in Fig.1. The percentages of PAHs abundance for 2+3, 4 and 5+6 rings were 39.3 ± 12.8 , 24.29 ± 15.24 and 36.41 ± 19.82 , respectively. The mean values of AN/ (PHA+AN) and FIA/ (FIA+PY) ratios as indicator isomeric values for identification of PAH origins were 0.48 ± 0.16 and 0.59 ± 0.22 respectively.

TABLE II: BIOLOGICAL DETAILS AND Σ PAHS CONCENTRATION OF ANALYZED BIOTA

Scientific name	English name	Ranges of weight (gr)	Ranges of length (cm)	Σ PAHs
<i>Drepane punctata</i>	Sickle fish	211.6 - 481	18.5 - 25.5	21.6 ± 13.3
<i>Liza subviridis</i>	Mullet	28.1 - 37.6	13.5 - 16	17.9 ± 1
<i>Pumpus argenteus</i>	Silver pomfret	129 - 140	17.5 - 21.6	22.3 ± 4.3
<i>Thunnus tongol</i>	Grouper	289 - 448	27 - 32.5	19.9 ± 13.7
<i>Thunnus tongol</i>	Longtail tuna	2400 - 3800	61 - 75	19.8 ± 5.4
<i>Psettodes erumei</i>	Flounder	177.7 - 374.3	25 - 31.5	24.8 ± 4.7
<i>Sardinella albella</i>	Sardine	20 - 33.7	13.5 - 16	19.6 ± 7.12
<i>Encrasicholina punctifer</i>	Anchovy	2 - 3.44	6.1 - 8	25.7 ± 2.4
<i>Pomadasys kaakan</i>	Grunt	270.3 - 371.4	20.7 - 30.5	16 ± 8.4
<i>Lethrinus nebulosus</i>	Sheiri	244 - 469	24.6 - 31.9	16.4 ± 3.27
<i>Scomberomorus guttatus</i>	King macrel	247 - 459	34.5 - 46.5	20.23 ± 10.22
<i>Otolithes ruber</i>	Tiger tooth croaker	967 - 1278	43.4 - 48.3	16.9 ± 3.9
<i>Sillago sihama</i>	Sillago	21.3 - 90.4	14 - 21.5	18.7 ± 9.1
<i>Lutjanus johnii</i>	Snapper	369 - 509	29 - 33.5	16.9 ± 6.4
<i>Rastrelliger kanagurta</i>	Indian mackerel	136.7 - 170.6	23 - 29.5	21.8 ± 11.9
<i>Penaeus mergiensis</i>	Banana shrimp	8.7 - 42.3	11.0 - 16.7	28.18 ± 3.74
<i>Portunus segnis</i>	Blue crab	112 - 327	11.0 - 15.5	27.6 ± 2.42
<i>Crassostrea gigas</i>	Bivalve			23.6 ± 12.1

Fig. 1. Percentages of PAHs in fish and seafood from Hormozgan Province

B. Health Risk Assessment

Among the PAHs components, benzo (a) pyrene and high molecular weight PAHs have an enormous importance in ecotoxicological studies according to their carcinogenic potential [14]. Table III shows the calculated B (a) P_{eq} values. The highest values related to Indian mackerel, Silver pomfret and Anchovy. The mean B (a) P_{eq} values for studied fish, shrimp, crab and bivalve from Hormozgan Province were 2.87, 1.84, 2.82 and 1.1 ng/g respectively.

TABLE III: B (A) P_{eq} VALUES (NG/G) FOR TESTED SEAFOOD

Name	Mean $\pm$ SD	Name	Mean $\pm$ SD
Sicklefish	3.08 $\pm$ 2.35	Sheiri	0.37 $\pm$ 0.24
Mullet	3.31 $\pm$ 2.6	King macrel	2.02 $\pm$ 2.25
Silver pomfret	5.04 $\pm$ 2.58	Tiger tooth croaker	1.42 $\pm$ 2.01
Grouper	3.27 $\pm$ 2.8	Sillago	2.84 $\pm$ 2.40
longtail tuna	2.35 $\pm$ 3.17	Snapped	0.79 $\pm$ 0.62
Flounder	3.43 $\pm$ 2.11	Indian mackerel	6.59 $\pm$ 2.23
Sardin	2.93 $\pm$ 2.2	Banana shrimp	1.84 $\pm$ 0.4
Anchovy	4.71 $\pm$ 0.37	Blue crab	2.82 $\pm$ 2.02
Grunt	0.93 $\pm$ 0.75	Bivalve	1.1 $\pm$ 1.45

The health risk is assessable through dietary intakes of pollutants. Table IV indicates the means dietary intakes of BaP, seven and eight carcinogenic PAHs.

Hormozgan people receive more of BaP and PAHs with carcinogenic property through fish consuming, due to higher consuming rate.

Moreover, screening value (SV) is known as an useful tool in risk management. In the established screening value by USEPA, the body weight and consumption rate were supposed as 70 kg and 142.4 g/day respectively [11]. These values were different for Hormozgan Province consumers. The calculated screening value for Hormozgan Province population based on local information regarding to body weight and consumption rate was 1.825 ng/g.

The comparison of obtained B (a) P_{eq} values with calculated SVs for consumers of Hormozgan Province showed that except bivalve, the studied seafood contained B[a] P equivalents higher than screening value, offering the intensive need to risk management.

TABLE IV: DIETARY INTAKES OF PAHs FOR HORMOZGAN PROVINCE URBAN POPULATION

PAH	Concentration (ng/g wet wt.)	Consumption rate (g/day/person)	Mean dietary intake (μ /day/person)
<u>Fish</u>			
BaP	1.63		0.089
sum of seven congeners ^a	8.6	55.1	0.47
sum of eight congeners ^b	9.1		0.5
<u>Shrimp</u>			
BaP	0.77		0.017
sum of seven congeners	10.79	21.9	0.24
sum of eight congeners	10.82		0.24
<u>Crab</u>			
BaP	0.28		0.00045
sum of seven congeners	11.3	1.6	0.018
sum of eight congeners	15.4		0.025
<u>Bivalve</u>			
BaP	0.56		0.000112
sum of seven congeners	7.9	0.0002	0.00158
sum of eight congeners	7.94		0.001588

^a Seven congeners refer to seven congeners with carcinogenic property, including of BaA, CHR, BbF, BkF, BaP, DahA and IcdP.

^b Eight congeners refer to eight congeners with carcinogenic property, including of BaA, CHR, BbF, BkF, BaP, DahA, IcdP and BghiP.

IV. DISCUSSION

According to the Σ PAHs variations and representations of categories in Table V [15], the tested samples were classified as minimally in polluted biota.

TABLE V: CRITERIA FOR PAH CONCENTRATION IN FISH

Category	PAH concentration (ng/g)
Not contaminated	<10
Minimally contaminated	10 -99
Moderately contaminated	100-1000
Highly contaminated	>1000

Diagnostic ratios of $AN/(PHA+AN)>0.1$ and $FIA/(FIA+PY)>0.4$ indicate the pyrogenic origin of PAH components [16]. The dominate with high molecular weight PAHs and diagnostic ratios confirmed the pyrogenic source of PAH pollution in studied biota. In past two decades, Hormozgan province had a rapid economic and urban expansion. Urban and industrial wastewaters have been discharged in marine environment. Furthermore, high traffic of marine transportations in studied area results to release of PAH with Pyrogenic source. With regard to studies associated with PAH pollution in edible seafood, there are so limited published data, especially in Southern Iran. The detected average value for Σ PAHs in clams from Bushehr Province was 421.86 ng/g, this value is much higher than our result, attributing to species of studied biota and area, because Bushehr is the mail area in Iranian oil export [17]. Sixteen individual PAH components were studied in 2 types of fish from Musa Inlet in the northwest of the Persian Gulf. Molecular ratios showed that in studied area, PAHs have been originated from both pyrogenic and petrogenic sources, however the pyrogenic sources were predominant [18]. In comparison with the international results, our findings were in good agreement with range of Σ PAHs (73-135 ng/ g dry weight) reported for fish from Kuwait [19]. Dietary intakes of PAHs were determined for Kuwaiti people on 2009. Σ PAHs varied from 30 to 247 ng/g dry weight, whereas the maximum value was detected in fish imported from Iran to Kuwait. Mean dietary intakes of B (a) P_{eq} was reported as 0.00133 μ g/day [20].

Values of Σ PAHs in mussels from Istanbul Strait and Marmara Sea varied from 43 to 601 ng/g wet weight and mainly originated from pyrogenic source [4]. Σ PAHs in marine fish samples from Hong Kong markets ranged from 15.5 to 118 ng/g wet weight, and the petrogenic source was known as the major polluting origin [3]. The concentrations of parent PAHs in bivalves from RSA (ROPME Sea Area) showed the wide range of 2 to 419 ng/g dry weight, and the maximum level was attributed to oyster from Mina Al Fahal, whereas the fish samples contained the much lower amounts from 0.4 to 13 ng/g) [10].

Taiyuan city is one of the most contaminated cities in China, suffers from unjustly planned industrial structure, which causes heavy contamination. Σ PAHs and B (a) P_{eq} for fish from Taiyuan city reported as 160 and 5.71 ng/g wet weight respectively [21] which were much higher than our results.

A comprehensive study was carried in Catalonia, Spain, in order to estimate the PAH intake through various kinds of

foods. Dietary intakes of Benzo[a] pyrene, Σ seven and Σ eight carcinogenic PAHs were 0.003, 0.026, and 0. 029 μ g/day, respectively for adult men [14]. The calculated values for people from Hormozgan Province were much higher than for Spanish. The total intake of B (a)P was estimated at 0.15 μ g/d/person for fish from Red Sea coast of Yeman which was higher than ours [5].

V. CONCLUSION

According to undesired biological effects of PAHs, there is a special concern about their health risk assessment. Unfortunately, there was no data on human risk assessment from PAHs in Iran. The Σ PAHs concentrations in collected biota showed that PAH contamination in fish and seafood from Hormozgan Province were relatively high. The worry would be more serious when PAHs with high molecular weight dominate to light ones. The risk of poly aromatic hydrocarbons intake through fish consuming was noticeable. However, for final risk assessment of PAHs, more investigations on the contribution of other food groups (meat products, oil and fats, fruits and vegetables), cooking methods and rate consuming of various food categories with regards to sex and age are suggested. Risk management has to apply in order to support the health of consumers in Hormozgan Province. Control and environmental management of urban and industrial discharges have not been ignored.

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