Baseline distribution of petroleum hydrocarbon contamination in the marine environment around the coastline of Qatar

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Abstract

Levels of organic contaminants (TPHs, PAHs) were simultaneously determined in both abiotic (sediments, seawater) and biotic (*Pinctada radiata* oysters) samples at four sites along the coastline of Qatar (Arabian Gulf) in 2017-2018. TPHs and PAHs were more frequently detected in oyster tissues than sediment and seawater samples collected from the same areas. While levels of TPHs and PAHs in seawater and sediments were lower than previous local studies and worldwide studies, PAHs levels observed in pearl oyster tissue (25.9- 2240 μg/kg) were relatively higher than in previous studies in Qatar. In general, eight PAHs compounds were detected in oyster tissue, with benzo(a)pyrene displaying the highest concentration. The coast of Qatar could be affected by seasonal patterns of pollutants, where TPHs and PAHs levels increased in winter compared to summer. These results provide key information on the use of the pearl oyster as a bioindicator species and Qatar's marine environment.

Keywords:

Total Petroleum Hydrocarbons, Polycyclic Aromatic Hydrocarbons, Bioindicator Species, *Pinctada radiata*, Bioaccumulation, Arabian Gulf

In recent decades, industry and population centers in the coastal areas of Qatar have expanded rapidly (Shandas et al., 2017), impacting Qatar's marine environment. Impacts include wastewater released to coastal waters and sediment resuspension caused by coastline development (Ministry of Development Planning and Statistics, 2017). Some primary point sources of pollution have been identified including the discharge of industrial and sewage effluent within a few kilometers of the Qatari shoreline and wastes released from oil and desalination industries (Bejarano and Michel, 2010). These various activities have resulted in a significant influx of various chemical pollutants in the marine environment of the Arabian/Persian Gulf, including some known to have a variety of adverse (e.g., reproductive and mutagenic) impacts on marine organisms (Rushdi et al., 2017). Chemical contaminants have also been detected in seafood products from the Gulf, at levels sometimes greater than international food safety limits (Freije, 2015). Qatar relies almost entirely on seawater for the production of potable water (Ashfaq et al., 2019), and seafood is a key source of protein in diet, hence, it is important to determine if these marine pollutants pose a threat to national water and food security. Moreover, marine pollution is known to be a threat to ecosystems and species diversity (Kurylenko & Izosimova, 2016). As such, protecting Qatar's marine ecosystems, drinking water and food from chemical contamination has been identified as a core component of the Environmental Development within the National Vision 2030 (MDPS, 2017).

Environmental risk assessments are now recognized to be best undertaken using an integrated approaches including both chemical and appropriate biological parameters in guardian species (EPA, 2019). As such, bivalve molluscs, including mussels and oysters, have been used extensively in marine coastal monitoring programs, from the early U.S. "Mussel Watch" program in the 1970s to more recent European research programs and initiatives in the Gulf in the context of the "*Regional Organization for the Protection of The Marine Environment*" (Tavares et al., 1988). Bivalves are ideal sentinel organisms, with high ecological importance and a wide geographical distribution (Dumbauld et al., 2009). Furthermore, they can bio-accumulate pollutants and exhibit a range of biological responses when stressed or exposed to environmental contaminants (Melwani et al., 2013). In Qatar, the pearl oyster, *Pinctada radiata* is considered an essential part of the nation's cultural heritage and one of the key economic foundations upon which the country developed. Historically, the pearl oyster formed large beds on the western side of the Arabian Gulf extending from Kuwait in the north to Oman in the South (Figure 1A). Due to its biological characteristics, ecological status and sensitivity to anthropogenic stressors this symbolic species of Qatar's maritime history provides an ideal sentinel species to monitor and assess the marine environmental quality as has been studied by Liu et al. (2019); Fowler et al. (1993); Tolosa et al. (2005); and De Mora et al. (2010). Research work has been undertaken in several Gulf countries to assess the contamination levels and bioavailability of organic contaminants in water, sediment and biota tissues. For example, Freije (2015), Lyons et al. (2015), Beg et al. (2003), Bach et al. (2003) and Saeed et al. (1996) reported the levels of Polycyclic Aromatic Hydrocarbons (PAHs) in Kuwait's marine sediment. Tolosa et al. (2005) examined the total contamination level of Total Petroleum Hydrocarbons (TPHs) and PAHs in biota and coastal sediments from Bahrain, UAE, and Oman. TPH levels in marine sediment from Bahrain and oyster tissues from Oman have been reported by De Mora et al. (2010), and there are studies on the bioavailability of TPHs in sediments from Saudi Arabia and Kuwait (Fowler et al., 1993; Lyons et al., 2015). However, although the Qatari marine environment has been impacted by marked industrial expansion and anthropogenic pressure over the last several decades, except for specific studies on trace metals, few studies have investigated other pollutants such as TPH and PAH in Qatar's marine environment. The aim of this study was to quantify the levels of organic contaminants (TPHs and PAHs,) in abiotic matrices (seawater and sediments) and biotic tissues of *P. radiata* from different locations in Qatar in different seasons; and to determine the relationships (correlations) between the organic contaminants among different abiotic and biotic matrices. The applicability of *P. radiata* as an early warning pollution indicator for organic contaminants was also explored.

Four coastal sampling sites were selected (Figure 1B) to represent different anthropologically impacted areas. Al-Khor was selected as a relatively pristine inshore site with water depths of 0.5–1.0 m (Leitão et al., 2017). The area is relatively free from industries and urbanization, apart from occasional recreational activity. Al-Wakra, situated in eastern Qatar, 15 km south of the capital Doha, is an inshore location with a water depth of 0.5–1 m with sandy bottom sediment which is very busy with recreational boat traffic and fishing fleet movement. There is a boat yard where minor repairs are carried out on seagoing vessels, fueling activities are also found within the area where fishing and recreational vessels are refueled. Umm Bab is located in western Qatar, only 25 km from the industrial city of Dukhan, and hosts a seawater desalination plant near the coast and a cement company. Simaisma is located on the east coast of Qatar with a water depth of 1–5 m. The area situated within a small bay approximately midway between Lusail and Al Khor. Samples were collected from the four different sites in March 2017, December 2017, May 2018 and November 2018.

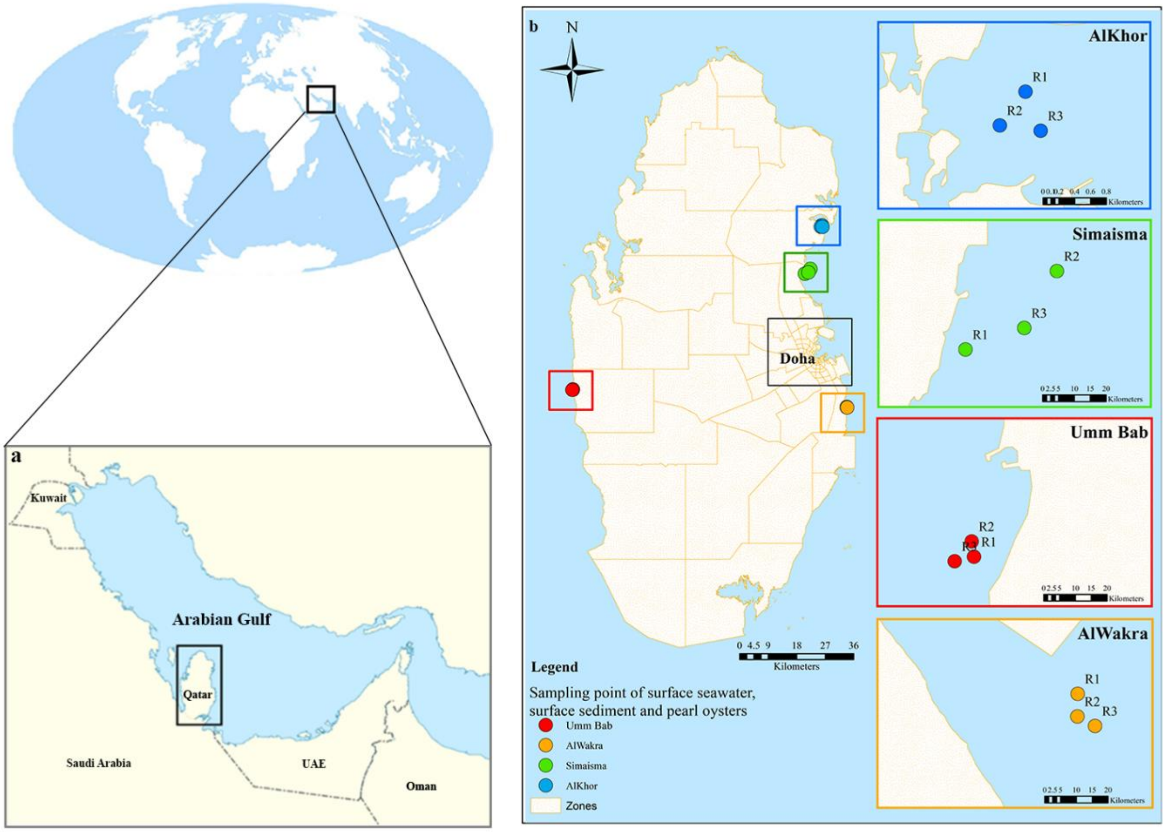

Figure 1: Location of Qatar in the Arabian Gulf (a) and the sampling sites in the Qatar coastal zone (b).

Seawater, sediment and oyster batches were collected in triplicates from each site (Al Khor, Simaisma, Umm Bab, Al Wakra – See Figure 1) over four rounds (March 2017, December 2017, May 2018 and November 2018), resulting in a total number of 12 samples for each matrix from each site. Seawater samples were collected from the sea surface using pumps and seawater temperature, salinity and pH were measured in the field using an EXO3 Multiparameter Sonde (YSI). Composite sediment samples were collected using a Van-Veen grab (0.05 m2). TOC content in seawater, sediment mean organic content and particle size were measured in the laboratory (See Supplemental Methods). *P. radiata* oysters were collected (n=3 composites of >12 oysters per site for each sampling round) by scuba divers at the targeted sites, where water depth did not allow the direct collection of samples. The size (long axis) was recorded for each organism and ranged from 2 to 7 cm. Specimens were transported to the laboratory in seawater and were transferred to a fridge (4˚C) prior to processing. Oysters were subsequently opened, rinsed with ultrapure water and dissected over ice. In total, 576 oysters were analyzed (48 batches). The levels of TPHs (C_8 to C_{32}) and 16 PAHs compounds, listed as priority pollutants by the US EPA, were measured following US EPA Methods 3510C for seawater and Method 8015B for surface marine sediment and oyster tissue. Gas Chromatography / Flame Ionization Detector (GC/FID) and Gas Chromatography/ Mass Spectrometry (GC/MS) were used for the analysis of TPH and individual PAHs (Supplemental Table S1-S2). Quality assurance included duplicate samples and analyses of standard reference materials ("CRM361 *TPH - Sea Sediment*" from Fluka Analytical and "SRM 1941b *Organics in Marine Sediment*" from NIST, for TPHs and PAHs respectively). Recoveries ranged 80-120% (Supplemental Table S3). The statistical analyses were performed using Excel and SPSS software. The Normality test was applied to all data to check the distribution of data. For normal distributions, One-way ANOVA was performed followed by a Tukey HSD (Tukey's Honestly-Significant Difference) post-hoc test to determine which pairs of the factor were significantly different from each other. Whereas for data that was not a normal distribution, the nonparametric test (Kruskal-Wallis test) was performed. Post-hoc pairwise comparisons were only applied where a significant difference was noted. The LOD divided by the square root of 2 was used for samples below the limit of detection.

The physicochemical parameters and contaminant levels in seawater are presented in Table 1 for the four sites and four sampling rounds (PAHs values are not presented in this table because they were negligible). The surface seawater samples from the four different locations showed negligible levels of PAHs, while TPHs ranged from 51.9 to 89.7 µg/L. Low concentration of PAHs observed in seawater samples collected in this study may be due to relatively high hydrophobicity of PAHs (Karthikeyan et al., 2001). In general, TPHs concentrations in seawater in the spring / summer month (March 2017 and May 2018) were higher than the winter months (December 2017 and November 2018) (see supplemental Table S5). The observed TPHs concentrations in seawater (mean 0.160 to 486 µg/L) were lower than those found in a previous study in Qatar (30 to 1700 µg/L) (Leitão et al., 2017). While detectable levels of PAHs (0.06 to 10.76 µg/L) was observed earlier by Leitão et al. (2017), negligible levels of PAHs was noted in this study. A review of the literature from the Gulf region and worldwide environmental studies showed that our result are in line and sometimes lower than most findings (Saeed et al., 2011;El Nemr and Abd-Allah, 2003, Xiang et al., 2018 and Nizzetto et al., 2008; Valavanidis et al. 2008; Ţigănuş et al., 2016 Kim et al. 2013).

Table 1. Physicochemical parameters and contaminant levels in seawater samples (n=12 for each site)

Site		**Temperature (ºC)**	**pH**	**Salinity (psu)**	**TOC (mg/L)**	**TPHs (µg/L)**
Simaisma	Mean ± SD	24.4 ±3.10	8.17 ±0.05	43.0 ±0.7	27.8 ± 0.7	51.9 ± 84.1
	Min.	20.4	8.08	42.1	26.7	2.70
	Max.	29.2	8.24	44.6	28.5	250
Umm Bab	Mean ± SD	24.8 ±3.80	8.01 ±0.16	56.1 ±2.2	29.5 ± 0.7	52.6 ± 68.9
	Min.	19.0	7.81	52.6	28.7	1.90
	Max.	29.2	8.24	58.2	30.9	190
Al Khor	Mean± SD	25.3 ±4.6	8.20 ±0.12	43.3 ±0.5	28.3 ±0.9	89.7 ± 139
	Min.	18.0	7.99	42.3	26.6	1.00
	Max.	30.4	8.33	44.3	29.9	486
Al Wakra	Mean	27.0 ±2.00	8.11±0.08	41.4 ±1.8	28.4 ±1.5	65.1 ± 64.8
	Min.	23.8	7.92	38.5	25.6	0.160
	Max.	29.2	8.18	43.5	30.3	170

Significant differences were observed between the percentage of sand, silt and clay in the surface sediments and sites ($p < 0.05$; Supplemental Table S4). Surface sediments were identified as sand in Umm Bab and Al Wakra, and as silty sand in Simaisma and Al Khor. Sandy sediment generally has less retention with regards to TPH and PAH. The mean OC content in surface marine sediments from the four sites in Qatar ranged from 0.079 to 0.219 % (Supplemental Table S4). Relatively higher sediment OC was observed in Umm Bab and Al Khor in comparison to Simaisma and Al Wakra, though this difference was only significant for Umm Bab (*p*=0.002). The mean concentration of TPHs ranged between 75.0 to 1750 µg/kg in surface sediments (Figure 2, Supplemental Table S5). Significantly higher TPHs concentrations were observed in Al Khor (*p*=0.01) compared to Umm Bab, Simaisma and Al Wakra which was not expected as all area have similarly industries. The average values of the Σ_{16}PAHs concentrations in surface marine sediments from various stations were between 4.25 to 36.7 µg/kg (Supplemental Table S6). The highest concentration was found in Al Khor, followed by Simaisma, then Al Wakra with lowest levels at Umm Bab. From all the 16 tested compounds, only six were detected in the marine sediment of which pyrene was the most prominent within all sites (mean 10.1 µg/kg), benzo(a)pyrene (B[a]P; mean 4.44µg/kg) was detected mostly in winter while indeno(1,2,3-cd)pyrene (mean 2.23µg/kg) was detected within the summer month in all sites. There was a weak positive correlation between the levels of TPHs and levels of PAHs in marine sediment samples collected from Qatar's marine environment (Figure 2-a). While a stronger positive correlation was observed at Umm Bab (Figure 2-c) followed by Simaisma (Figure 2-a), no relationship could be observed at Al Wakra (Figure 2-e) and surprisingly a negative, though weak correlation, was noted for Al Khor (Figure 2-d).

In this study, TPHs measurement ranged from 75.02 to 1751.82 µg/kg whereas the range in previous local studies of marine sediment from Qatar spanned 17.3- 79.3 µg/kg (Rushdi et al., 2017); 160- 1700 µg/kg (Leitao et al., 2017); and 2200- 84000 µg/kg (Tolosa et al., 2005). The high levels reported by Tolosa et al. (2005) where from Qatari sites impacted by the oil terminal and refinery plant (Umm Said, Dukhan, Ras Lafan). Moreover, the concentrations of total PAHs ranged from 4.25 - 36.7 µg/kg and were in the same range (Leitao et al., 2017; Soliman et al. 2019). ΣPAHs reported in Qatar's marine sediments were low compared to data from the wider Gulf

(7200- 80000 μg/kg, Readman et al., 1996; 1333.6 μg/kg, Beg et al., 2003; 12.9-190 μg/kg, Lyons et al., 2015). Moreover, concentrations of ΣPAHs from Qatar's marine sediments were much lower than those reported in other international studies conducted in China, Russia and the UK (13.59-166.5 μg/kg, Zhang et al., 2016; 61.2-368 μg/kg, Readman et al., 4171- 79648 μg/kg, 2002 Brettell, 2013). This may be related to the high proportion of sand in the sediments in the current study.

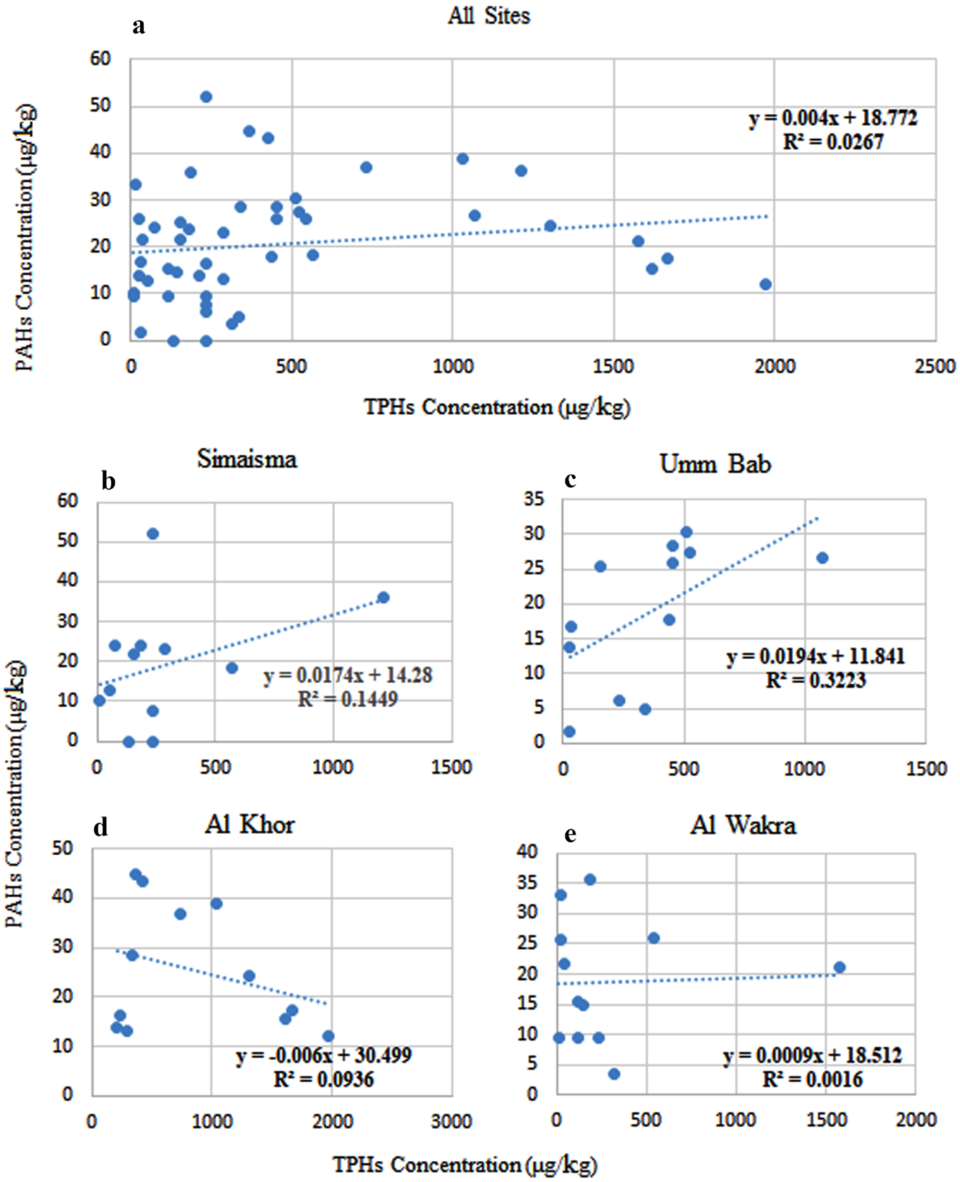

Figure 2: Correlation between TPHs and PAHs "µg/kg dry weight (dw)" in surface sediments for all sites (a) and individual sites (b-e).

Mean TPH content in oysters ranged from 633 to 6670 µg/kg with the highest concentrations measured in Simaisma. There was a significant seasonal difference in the TPHs in oyster tissue samples at Simaisma and Al Wakra. The mean concentrations of PAHs in oyster tissue samples showed an extreme variation from 25.9 to 2240 µg/kg. Significantly high levels of PAHs were measured in Al Wakra ($p < 0.05$). In general, ten PAHs compounds were detected in oyster tissues (Supplemental Table S7). Benz(a)pyrene was the highest PAH concentration detected (1510 µg/kg) observed in Al Wakra. BaP is a known carcinogenic compound and has recently been flagged by the International Agency for Research on Cancer (IARC). Another carcinogenic, dibenz[a,h]anthracene, was detected in Al Wakra samples collected in winter seasons (both 2017 and 2018). In general, fluctuations in the patterns of PAHs in oyster tissues suggest that the coast of Qatar could be affected by seasonal patterns of pollutants. Notably, PAHs levels increased in winter compared to summer.

Considering all the sites, TPHs and PAHs concentrations in oyster tissues showed a weak positive association (Figure 3). However, at Simaisma and Al Khor there was a strong, positive, linear correlation between the two organic contaminants (R^2=0.815 and R^2=0.885 respectively). TPHs concentrations reported in *P. radiata* tissues in this study (633- 6670 µg/kg) were in the range of previous study from Qatar (10 to 7410 µg/kg) (Leitão et al., 2017). TPHs concentrations in other Gulf countries were significantly higher than levels found in this study and previous studies in Qatar (De Mora et al. 2010; Tolosa et al. 2005). Levels reported ranged from (22900 to 69700 µg/kg) in Bahrain, (20300 to 33900 µg/kg) in UAE, and (37600 to 632000 µg/kg) in Oman as described by de Mora et al. (2010). Whereas Tolosa et al. (2005) reported high levels of TPHs in oyster tissues in the same Gulf countries, concentration ranges were (38500 to 98000 µg/kg, 25600 to 237000 µg/kg, and 23200-130000 µg/kg) in Bahrain, UAE and Oman, respectively. Furthermore, high TPHs range were observed in different areas around the world. Vaezzadeh et al. (2017) studied TPHs in oyster tissues from Malaysia and reported a level from 56700 to 263000 μg/kg, while Inam et al., 2012 reported levels in the range of 6370- 8440 μg/kg in oyster tissues from Nigeria. Overall, the organic contaminants (TPHs and PAHs) were more readily detected in oyster tissue samples than marine sediment and seawater samples collected from the same areas (Figure 4, Table 1).

Levels of PAHs observed in this study in P. *radiata* oyster tissue samples (25.9- 2240 μg/kg) were higher than the previous observation by Leitão et al. (2017) who reported 0.150- 23.1 μg/kg in *P. radiata* tissues from Qatar's marine environment. The levels of PAHs reported in this study were close to levels reported in other Gulf countries. De Mora et al. (2010) reported PAHs concentration in oyster tissues between 80.0- 335 μg/kg from Bahrain, 19.0- 161 μg/kg from UAE, and 103-13700 μg/kg from Oman. These levels were slightly higher than a study reported five years earlier in same areas, where the PAHs ranged from 58.3-105, 36.6-251, and 17.0-173 μg/kg in Bahrain, UAE and Oman respectively (Tolosa et al., 2005). Concentrations of PAHs noted in P. *radiata* oyster tissue samples collected from Qatar marine environment appear to exceed some other countries at least based on the highest values reported (Hong et al., 2016; Barhoumi el al., 2016; Acosta et al., 2015; Nakata et al., 2014; Ramdine et al. 2012, Oros and Ross, 2005).

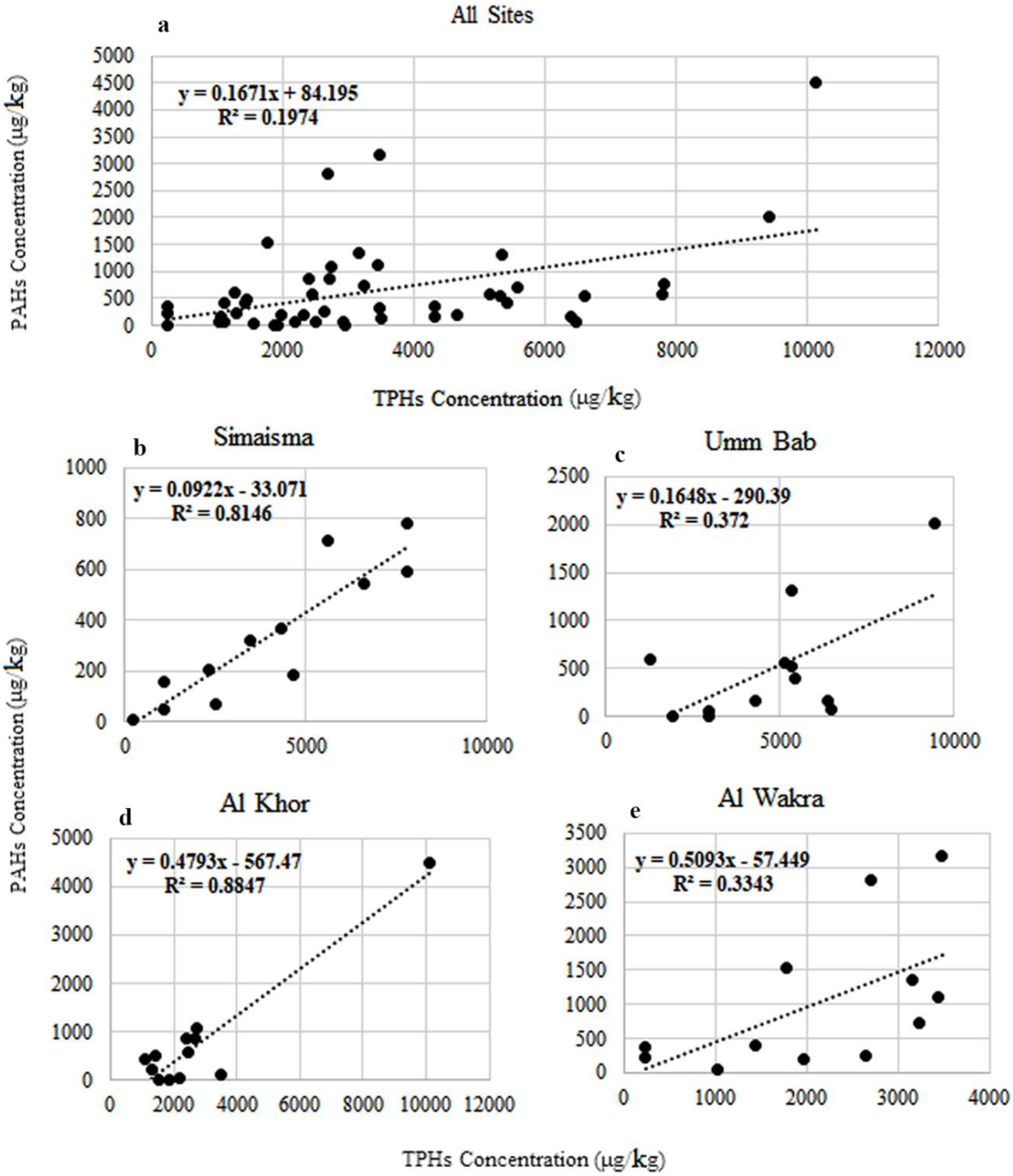

Figure 3: Correlation between TPHs and PAHs "µg/kg dry weight (dw)" in oyster tissues (all seasons) for all sites (a) and individual sites (b-e).

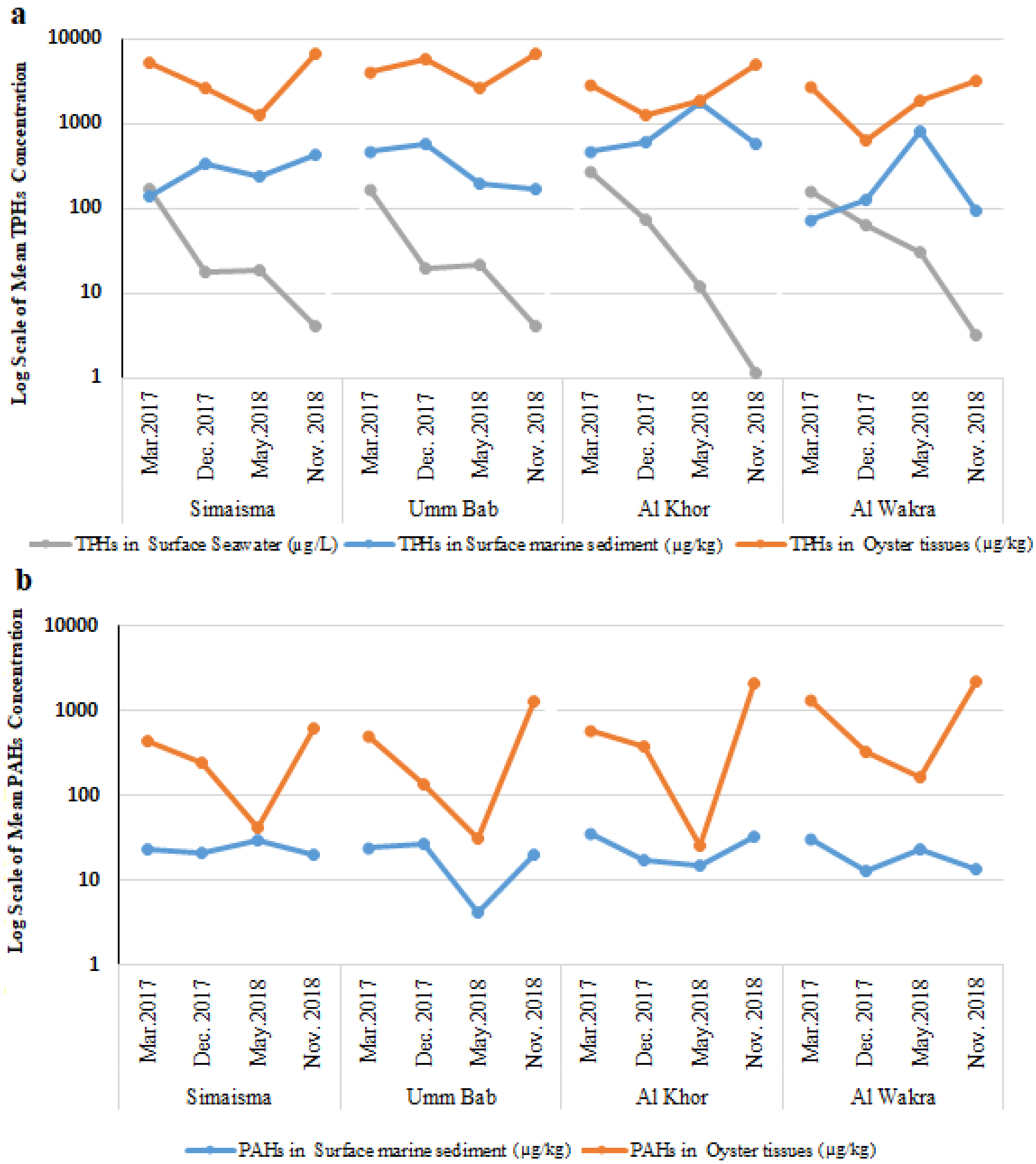

Figure 4: Seasonal variations of TPHs (a) and PAHs (b) in seawater (grey), sediments (blue) and oysters (orange). (Note: PAHs were not detected in seawater)

Based on our findings, while levels of TPHs and PAHs in seawater and sediment samples were lower than previous local studies and examples worldwide, the PAHs levels observed in pearl oyster tissue samples (25.9- 2240μg/kg) were higher than previous studies in Qatar and even higher than levels recorded in different areas around the world based in our selected studies. The Al Khor site showed a higher range of TPHs concentration in seawater samples (1.20-272 μg/L) than in a previous local study (40.0-50.0 μg/L), and higher levels of TPHs and PAHs in surface sediment than other three sites (480- 1750 μg/kg and 15.0- 36.7 μg/kg respectively). On the other hand, the highest PAHs ranges in oyster samples were observed from Al Wakra and Al Khor (167- 2240 μg/kg, and 25.9- 2080 μg/kg respectively) with higher levels reported in winter season than summer. Therefore, although the sediments did not seem to have been affected by organic contaminates, high levels of PAHs recorded in oysters collected from Al Wakra and Al Khor, and in seawater collected from Al Khor might reflect residual hydrocarbons in these east coast sites of Qatar. Al Wakra reported the highest level of PAHs, which may suggest chronic contamination due to the boat traffic, fishing or fueling activities around this location. The present study provides useful background information on the use of the pearl oyster as a bioindicator species and for further investigations to understand the presence of organic contaminants in Qatar's marine environment.

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