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ORIGINAL PAPER

Assessment of trace inorganic contaminates in water and sediment to address its impact on common fish varieties along Kuwait Bay

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Abstract The impact of the trace elements on selected marine fishes/crustacean in Kuwait (Sheam, Lobster, Speatty, and Nagroor) were investigated (As, Cd, Ni, Pb, and V) using the element concentrations in marine water and sediments. The toxic elements concentrations were measured in water samples (As, Cd, Cr, Cu, Hg, Ni, Pb, V, and Zn) for estimation of toxic levels, heavy metal evaluation index (84–360), and the degree of contamination (77–353). Similarly, sediment samples were analyzed for As, Cd, Cr, Cu, Ni, Pb, V and estimated for contamination factor, I_{geo} index, and ecological risk factor with respect to each element analyzed in the sample. The modified degree of contamination (0.25–3.67), risk index (6.5–282.27), metal pollution index (5.95–18.21), and pollution load index (0.27–1.2) were calculated for the samples. This study demonstrated that the water was medium to high contaminated with Cd, Hg, Pb, and V. The sediment analyses showed that most of the metals were within the toxic limits except for Cd, Cu, and Pb in few samples. Most samples were in between the effect range low–effect range medium and threshold effect level–probable effect level range of most metals, except for Cr, Cu, and Ni. Average trace elements concentration in fishes varieties investigated in this study indicated high As in all varieties irrespective of the season and high Ni in all fish during summer. The bioaccumulation factor showed that the trace elements in sediments contributed more to the fish than water. Concentrations of trace elements were greater in fish sampled in winter than that sampled in summer due to variations in the planktonic population in the sea. The estimated daily intake and the chronic daily intake for the Kuwaiti male and female were calculated. The hazards studied revealed that the consumption of Lobster and Speatty may lead to cancer and non-cancer hazards, in both male and female, Speatty having higher probability. The major sources of toxic elements contamination of Kuwait Bay water and sediment appear to be oil-based contamination, urban sewage, brine from desalination, and the trace elements released due to the natural oxidation–reduction processes.

Keywords Water · Sediment quality · Fish · Contamination · Trace element sources · Health · Trace elements seasonality

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Introduction

Trace element pollution endangers several aquatic species of fish as it plays a key role in the food chain. It was inferred from a 20-year data set that there is a decline in fish catch due to the permanent loss of Nursery grounds in the subtidal and intertidal region (Saenger 1993; Bishop et al. 2002). Fish landings in Kuwait have declined to half during 2007 compared to that of 1995 (CSO 1979–2007). The increased salinity of coastal seawater in the bay due to urbanization and industrialization affected the marine environment, also leading to variation in NO_3 and chlorophyll content in the bay water (Sheppard et al. 2010). Shuaiba Industrial Area (SIA) of Kuwait has a cluster of industries, including fertilizers, paper production, desalination facilities, oil refineries, and other small industries (Beg and Al-Ghadban 2003). This industrial zone produces 51,000 m^3 day^{-1} of effluent, most of which is directly discharged to the open sea (Al-Muzaini 2002; Gevao et al. 2005, 2009). Subsequently, studies have also reported sediment contamination due to the effluent discharges along the coast (Bhandary et al. 2018).

Further, desalinated water serves > 90% of Kuwait's drinking water requirements (Darwish and Abdulrahim 2008). The waste generated by desalination, i.e., brine, contains biocides and other pollutants affecting the marine ecosystem (Sheppard et al. 2010). Al-Muzaini et al. (1991) reported that coastal Kuwaiti suburbs discharge 0.282×106 m^{-3} d^{-1} sewage with high organic content. Kuwait Bay and the Mina Abdulla coast water contain high concentrations of total hydrocarbon content (THC). Sulaibikhat Bay and Ras Al Zour water contained high levels of degraded oil contamination (de mora et al. 2010). A review of the heavy metal pollution due to petroleum hydrocarbon in the Arabian Gulf region (Freije 2015) shows that pollution is least concentrated and stress has been more observed in the coastal land use pattern (Sheppard et al. 2010). The Kuwait Bay is extremely stressful with high evaporation rates, high temperature, poor flushing, elevated salinity, and UV exposure. The residence times of the contaminants are greater in the bay due to low dilution (de Mora et al. 2004).

Fishes are important biomarkers for monitoring metal pollution in their habitat (Rashed 2001; Authman 2008). In the food chain, fishes accumulate metals and transfer them to humans, the next level of consumers (Authman 2015). The earlier studies on Hamoor in Saudi markets revealed minimal values of a few trace elements Al-Bader (2008). The studies on the metal toxicities have proved that the concentration is higher in Pelagic sediments. Hence, the metal concentrations were greater in bottom-dwelling organisms, particularly in the digestive glands of fish and benthic macroinvertebrates (Engel et al. 2001). Increased Pb concentrations were reported in Lobsters of Mazatlan Bay of California associated with arsenic levels (Morales-Hernndez et al. 2004) like Lobster. Studies on Lobster from the Persian Gulf showed (Raissy et al. 2011) excessive levels of Pb, whereas concentrations of As and Cd were also elevated but within the acceptable limits for human consumption. Likewise, excessive Cr concentrations were found in Lobsters from the Red Sea Coast (Wang et al. 2017). Sheam varieties were extensively studied for Hg toxicity (Mortazavi and Sharifian 2011; Babadi et al. 2014). Sheam (A. *latus*) was designated as a bioindicator of metal pollution by studies conducted in Kuwait territorial waters (Bu-Olayan and Thomas 2013). These are considered as target species to monitor coastal pollutants (Hedayati 2016). Kumar et al. (2004) has highlighted the paucity of As levels in fishes of Asia–Pacific species, but this study tries to bring out the As levels of the fish varieties considered.

Similar studies (Table 1) were carried out in Turkey (Türkmen et al. 2005), the Persian Gulf (Mortazavi and Sharifian 2011), Alexandria (Abdel Ghani et al. 2013), India (Velusamy et al. 2014), Malaysia (Bashir et al. 2012), the Mediterranean (Lamiceli et al. 2015; Canli and Atli 2003), Iran (Babadi et al. 2014), China (Liu et al. 2015), Canada (Pyle et al. 2005), and Indonesia (Bentley and Soebandrio 2017).

Sources of metal pollution of Kuwait marine environment and biota have been reviewed by Al-Sarawi et al. (2015), who concluded the relatively low level of metal as compared to that in other heavily industrialized regions of the world. Risk of methyl mercury contamination was linked to the consumption of Hamoor fish; therefore, health advisory is in effect to limit the consumption of this fish (Laird et al. 2017). Chemical and ecotoxicological screening studies (Smith et al. 2015) have concluded that the region of outfalls and certain open water sites were hot spots of marine contamination in Kuwait (Bu-Olayan and

Table 1 Range of trace element concentrations in sea water, marine sediments and fish varities, representing different regions of the world

Country	Region	As	Cd	Cr	Cu	Hg	Ni	Pb	V	Zn	References
Range of trace elements reported in sea water (all values in μg L^{-1})											
Kuwait	Kuwait Bay	0–43	16–73	0	4–7	1–24	0	4–34	0–9	228–365	Present study
Kuwait	Kuwait sea water		0.2–0.7	0.5–1.2	0.6–2.2		1.2–4.7	0.4–3.6	1.6–8.2	3.6–9.4	Bu-Olayan and Al-Yakoob (1998)
Kuwait	Kuwait sea water			0.3–7	0.7–20		0–5	0.1–9	5–50	10–98	Al-Sarawi et al. (2002)
Saudi Arabia	Red Sea Coast, Jizan	0.28	0.17	1.36	7.85		6.37	0.56	–	3.58	Mortuza and Al-Misned (2017)
Nigeria	Qua Iboe Estuary	–	10–300	1.7–31.0			10–500	200–400		10–400	Benson et al. (2017)
Malaysia	Kapar	36	10					10		21	Bashir et al. (2012)
Malaysia	Mersing	30	19					10		15	Bashir et al. (2012)
Range of trace elements reported in sediments (all values in μg g^{-1})											
Kuwait	Kuwait Bay	0–9.7	0–2.43	47.9–1119	9.63–122.24		1.04–49.06	0–32.58	27.3–81.6		Present Study
Kuwait	Sulaibikhat Bay	1–7	2–4	65–190	10–100		25–130	2–32	–	10–290	Al-Sarawi et al. (2015)
Egypt	Abu-Qir Bay	1.60–8.67	0.31–4.89	–	10.24–22.85		–	1.9–16.79	5.57–70.64	25.23–104.08	Abdel Ghani et al. (2013)
Egypt	Eastern Harbor	4.01–16.21	0.3–1.83	–	3.8–129.2		–	1.3–112.09	2.85–23.10	2.9–206.89	Abdel Ghani (2015)
Egypt	Western Harbor	4.7–15	0.61–2.44	–	39–207		–	38–1070	7.0–59.1	58.5–382	Mostafa et al. (2004)
Spain	Algeciras Bay	8–23	0.1–0.7	–	5–25		–	12–39	36–94	33–117	Alba et al. (2011)
Egypt	Mediterranean Coast		0.22	82.74	8.46		25.93	13.17	–	22.19	Soliman et al. (2015)
Saudi Arabia	Red Sea Coast, Jizan	0.34	0.48	5.64	16.39		14.32	3.86	–	24.74	Mortuza and Al-Misned (2017)
Yemen	Gulf of Aden	–	–	17–233.93	8.09–111		16.17–48.07	14.8–138.06	–	21.85–263.49	Saleh (2006)
India	Gulf of Mannar	–	–	148–195			22.63–24.5	15.97–16	–	71–74.06	Jonathan and Mohan (2003)
Mexico	Gulf of Mexico	–	–	3–100	3.82–18.7		0.66–76.9	0.22–20.2	–	0.04–79.6	Macías-Zamora et al. (1999)

Table 1 continued

Country	Region	As	Cd	Cr	Cu	Hg	Ni	Pb	V	Zn	References
Nigeria	Qua Iboe Estuary	–	0.55–1.142	9.57–13.78			9.15–13.96	2.00–8.90		0.91.5–161.4	Benson et al. (2017)
Bangladesh		27	1.5	118	82		103	63			Islam et al. (2016)
California	Mazatlan Bay		0.9	18.2	34.5		13.6	57.8		160	Morales-Hernndez et al. (2004)
Egypt	El-Mex Bay		4.41	9.79	41.53	0.2		40		101.8	Masoud et al. (2007)
Saudi Arabia	Ras Tanajib		31.59			0.15		46.3			Sadiq (1992)
UAE	Al Ruweis	2.2	0.11					5.88			Morales-Hernndez et al. (2004)
Bahrain	BAPCO	4.96	0.18			0.22		99			Morales-Hernndez et al. (2004)
Qatar	Doha	6.3	0.07					3.88			Morales-Hernndez et al. (2004)
Oman	Raysut Port	1.09	0.2					0.729			Morales-Hernndez et al. (2004)
Range of trace elements reported in fish (all values in mg g^{-1} except Iran in mg kg^{-1})											
Kuwait Bay		0.04–22.86	0–4.3				0.04–1.11	0.034–0.86	0–10.71		Present study
Kuwait Sea	Kuwait			4.11–5.62	7.36–9.51	1.1–1.36	4.81–6.98	2.1–3.6		19.05–23.97	Bu-Olayan and Thomas (2013)
Saudi Arabia	Red Sea Coast, Jizan		0.63	4.35	3.82		0.19	1.06	–	14.05	Mortuza and Al-Misned (2017)
Nigeria	Qua Iboe Estuary	–	0.162–0.931	3.81–8.62			11.20–17.15	1.90–7.35		125.5–269.75	Benson et al. (2017)
Turkey	Iskenderun Bay		0.95	1.69	1.57		2.9	2.32		4.36	Türkmen et al. (2005)

Table 1 continued

Country	Region	As	Cd	Cr	Cu	Hg	Ni	Pb	V	Zn	References
Korea	Masan Bay		0.01	0.02–0.05	0.18–0.25		0.02	0.04–0.15		6.33–12.9	Kwon and Lee (2001)
	Mediterranean Sea		0.37–0.79	1.24–2.42	2.19–4.44			2.98–6.12		16.5–37.4	Canli and Atli (2003)
Iran	Persian Gulf	118–275	101–401			32–73		379–1120			Raissy et al. (2011)
California	Mazatlan Bay		0.3	0.41	24.4		1.2	1.6		67	Morales-Hernndez et al. (2004)
Egypt	El-Mex Bay		0.38	0.86	2.34	0.1		1.69		9.93	Masoud et al. (2007)
Italy	Mediterranean species	56.7	0.16	0.96	4		0.163	0.093	0.115		Lamiceli et al. (2015)
Malaysia	Kapar	12.58	0.88					0.12		50.99	Bashir et al. (2012)
Malaysia	Mersing	14.2	0.02					0.2		25.39	Bashir et al. (2012)
Saudi Arabia	Maniefa	42.7	0.16					20.01			Al-Saleh and Shinwari (2002)
UAE	Dubai	1.54	0.17			0.05		0.004			Kosanovic et al. (2007)
Bahrain	Badaiya	1.38	0.001			0.82		0.005			Morales-Hernndez et al. (2004)
Qatar	Al Dakhira	10	0.005			0.343		0.108			Morales-Hernndez et al. (2004)
Oman	Raysut Port	2.07	0.11			0.522		0.014			Morales-Hernndez et al. (2004)

Thomas 2012). The nature of trace metal pollution in phytoplankton was studied by Bu-Olayan et al. (2001) in Kuwait coastal waters. Beg et al. (2015) determined that the metal levels and their accumulation in species were identified with respect to oxidative stress due to metallothionein in fishes of Kuwait marine waters.

Sediment quality guidelines (SQG) to safeguard the biota living in or near the sediments were developed with respect to the tolerable levels of pollutants, the quality of sediments, and the bound metals. These guidelines are based on the levels of different metals (Violintzis et al. 2009). The release of metals from the sediments to the overlying aquatic system results in adverse effects on the biota (Wang et al. 2010). Hence, to assess an aquatic environment, water, sediment, and biota should be considered (El Nemr et al. 2012). Goher et al. (2014) studied an index similar to SQG to understand the freshwater sediment pollution.

Metals are absorbed by the sediments after being introduced into the aquatic system and subsequently flow into the food chain (Loska and Wiechula 2003). Thus, metals accumulate in various parts of the fish as they feed on the water and food in this ecosystem. Hence, to examine the risk of metal contamination on fish, concentrations of metals in the medium (water and sediment) provide a comprehensive idea about the sources. So, the water is compared with standards like metal pollution indices, and the sediments with SQG, I_{geo} index, the degree of contamination, Pollution load index (PLI), risk factor, m-ERMQ (effect range median quotient), and m-PELQ (probable effect level quotient) (Goher et al. 2014). Further, the concentrations of As and V and their temporal variations have not been widely investigated in the common fish varieties of Kuwaiti marine waters. The impact on the consumer of the fish is governed by the THQ (target hazard quotient), CDI (chronic daily intake), EDI (estimated daily intake), and other factors like CR (cancer risk) and NCH (non-cancer hazard), margin of exposure (MOE) which is also influenced by gender and age of the consumer. This study tries to bridge the gaps identified, to understand the seasonal variations of trace element concentrations in fish varieties, sediment, water and their mobility with respect to the environment. Furthermore, the risk of trace element contamination in the human food chain was also investigated based on the range of sediment quality and their sources.

Materials and methods

A sampling of fishes, crustacean [Sheam (Acanthopagrus *latus*), Lobster (Thenus *Orientalis*), Speatty (Sparidentex *hasta*), and Nagroor (Pomadasys *argenteus*)], seawater, and sediments were carried out in Kuwait Bay region. The sampling of fish was done in winter (December) and summer (May). Eleven samples of Sheam were collected during winter and 7 in summer; 8 samples of Lobster during winter and 6 in summer; 9 samples of Speatty during winter and 6 in summer; 6 and 5 samples of Nagroor in winter and summer, respectively. Triplicates of each fish sample were washed to remove contaminants, sliced into small pieces, oven-dried (microwave digestive system) at 80 °C for 48 h, and ground into powder by a mortar. Two grams of the dry sample was digested in a mixture of HNO_3 and $HClO_4$, according to AOAC (1995), filtered through Whatman filter, diluted to 50 mL deionized water for analysis. The standard reference material (dogfish liver certified reference material DOLT-3) was used (Supplementary Table 1) and for Vanadium 1566b oyster tissue was considered as reference (Supplementary Table 2). Concentrations of As, Ni, and Pb were analyzed by GF-AAS (PerkinElmer); that of Cd by ICP OES (Agilent) and V by ICP–MS (PerkinElmer) (Table 4). The detection limit for As is 0.02 mg kg^{-1}, and that for Ni, Cd, Pb, and V is 0.01, 0.01, 0.02, and 0.002 mg kg^{-1}, respectively. As and Pb in fish samples were analyzed by PerkinElmer SIMAA-6000 simultaneous multi-element atomic absorption spectrometer. This instrument is also facilitated with transversely heated graphite atomizer and longitudinal Zeeman effect for background correction. Electrodeless discharge lamps were used for As (193.7 nm and slit 0.7 nm) and Pb (247.6 nm and slit 0.7 nm), respectively. Atomic signals were measured in peak area mode. The experiments were conducted in a stabilized temperature platform furnace. Palladium/magnesium modifier solution was used for analysis. Suitable dilution of 10 g L^{-1} of palladium was carried out in HNO_3 and 1 g L^{-1} of Mg in 1% HNO_3 to prepare the solution. 1.25 mg L^{-1} was used as a chemical modifier, and both the elements were analyzed separately. The pyrolysis temperature for Pb and As was 700 °C and 1100 °C, respectively. To avoid clogging at the nebulizer orifice, the samples were centrifuged for 12 min at 5000 rpm. This effect may result in erratic

loss of signal intensity mainly due to the effect of cadmium and lead.

A set of multi-element working standards from 1000 mg L^{-1} single-element stock solutions (Merck, UK) were prepared at 5, 20, 50, and 100 µg L^{-1}. Standards were treated in the same way as samples, with the addition of a pre-reduction solution. Working standards were prepared in 5% HNO_3 and 2.5% HCI. A mixture of 2% L-cysteine and 4% tartaric was used as a pre-reduction solution. To prepare the pre-reduction solution, 20 mL of a 10% L-cysteine solution (in 2% HCl) was added to 4 g tartaric acid and made up to 100 mL with deionized water. Analysis of Ni and Cd was performed using an ICP OES Agilent 5110 Synchronous Vertical Dual View (SVDV). The SVDV design is capable of running in both axes, radial, vertical, and synchronous vertical dual-view modes for maximum versatility in evaluating from higher to lower concentrations. Specific Dichroic Spectral Combiner (DSC) technology provides faster analysis of a sample with lesser consumption of gas. A single-pass glass cyclonic spray container, a SeaSpray nebulizer, a white–white peristaltic pump tube, and a regular 1.8-mm injector torch formed key components of the sample injection system. Blank samples were run after eight samples (Supplementary Table 3). The concentration was derived based on the wet weight basis for As and Hg and on a dry weight basis for other elements.

Seven sediment grab samples were collected, homogenized, and sealed in polyethylene bags for analysis in the laboratory. The samples were stored in icebox and preserved until further analysis. 20 g of each sample was dried in an oven for 24 h at 100 C. The dried sediment sample was ground and digested in HNO_3/$HClO_4$/HF (3:2:1) (Oregioni and Aston 1984). The digested samples for sediment were filtered by Whatman filter, and the filtrate was diluted to 50 mL. The details of the sampling locations are depicted in Fig. 1. Seawater samples were collected, 0.5 m below the surface and 1 L per sample in Teflon bailer, from 15 locations within the region where the fishes were sampled. The water sample was vacuum-filtered using 0.45-µm filter paper and transferred to a polyethylene bottle; the pH was adjusted to below 2 using HNO_3. The Agilent 5110 ICP OES was used for the analysis of trace elements in water and sediment. The hydride forming elements (As and Hg) were analyzed using a Multimode Sample Introduction System (MSIS). In both setups, an SPS 4 autosampler was used. The VistaChip II detector is used, which has a high-speed (1 MHz) CCD detector that enables quick warm-up, high throughput, high sensitivity, with a wide dynamic range of up to 8 orders of magnitude. This setup facilitates full wavelength coverage ranging from 167 to 785 nm through a single slit.

The As and Hg calibration specifications were designed using traceable standard single-element NIST. The concentrated HCl was added to the samples and 0.5% NaOH along with 0.6% $NaBH_4$, for reduction. The samples prepared were analyzed using the Multimode Sample Introduction System (MSIS) by the Agilent 5110 ICP OES. MSIS is a basic sample introduction system that replaces the ordinary spray chamber and is capable of nebulizing the liquid sample and simultaneously producing volatile hydrides. The detection limit for As and Hg is 0.4 and 0.051 mg L^{-1} (4% HCl and 1% L-cysteine) in multi-element hydride analysis using the MSIS with correct acid matrix selection. NIST multi-element standard was used for the analysis of other elements, and they were diluted with 5 percent HNO_3 50 mL of sample and diluted with 45 mL of deionized water, thus made up to 100 mL. The samples were injected using the single-pass cyclonic spray chamber fitted with the SeaSpray nebulizer. Standard Reference Material 1643d was considered to determine trace elements in water; the accuracy and precision of the process were assessed using the standard (Supplementary Tables 4, 5).

Water

Metal indices

The heavy metal evaluation index (HEI) for water was proposed by Edet et al. (2003) and is calculated as:

$$\mathrm{HEI} = H_{ci}/H_{\mathrm{maci}} \quad (1)$$

H_{ci} is the measured value of *i*th parameter (element), and H_{maci} is the standard allowable limit of the parameter. Ni and Cr were excluded from the calculation as they were below detectable limits in all samples.

The degree of contamination (C_{d}) for metals in water (Backman et al. 1998) was determined by the following equation:

$$C_{\mathrm{d}} = \sum C_{fi} \quad (2)$$

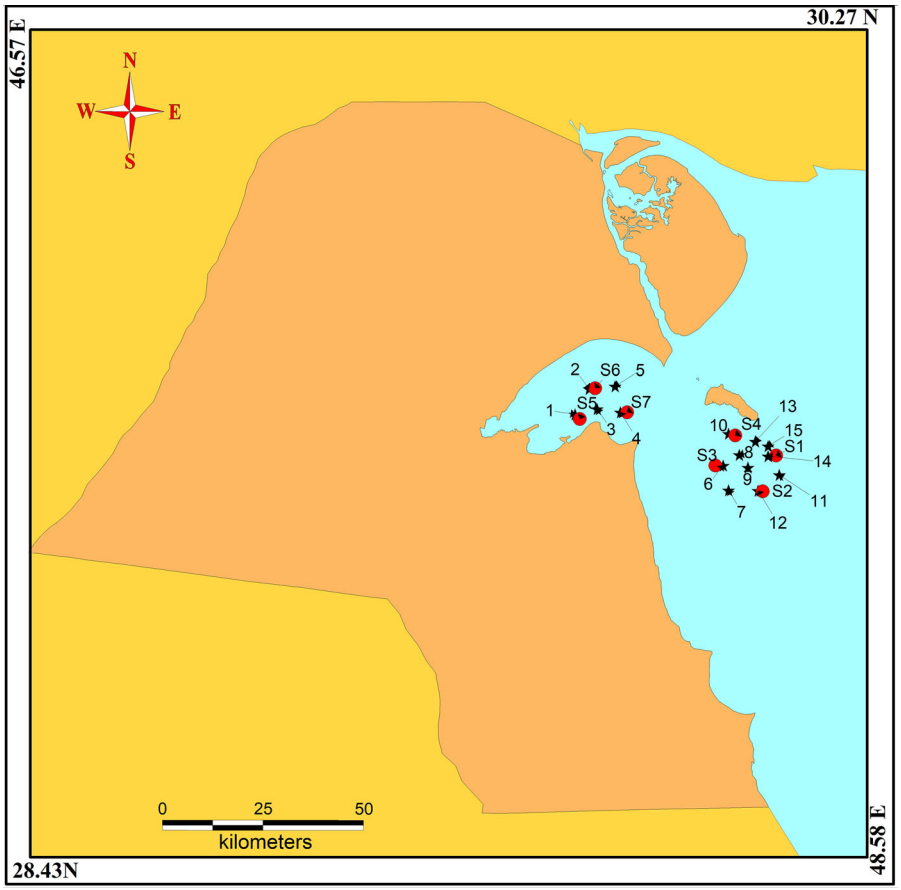

Fig. 1 Locations of the sediments (S) and water samples collected for the study

where $C_{fi} = C_{Ai}/C_{Ni} - 1$.

C_{fi}, C_{Ai}, and C_{Ni} represent contamination factor, analytical value, and upper permissible concentration of the *i*th parameter. *N* denotes the "normative value" in this calculation, and maximum acceptable concentration (MAC) is considered as C_{Ni}. Chidambaram et al. (2012) adopted a similar methodology to calculate and classify the degree of contamination.

Sediments

Metal pollution index

The metal pollution index (MPI) of these four different fish with respect to the season can be obtained by the formula (Usero et al. 1996; Usero 1997):

$$\mathrm{MPI} = (M_1 \times M_2 \times M_3 \times \cdots \times M_n)1/n \quad (3)$$

M_n represents the metal concentration in μg g^{-1} of weight.

Contamination factor

This is used as a parameter to estimate pollution over time period by calculating the values for each metal (Turekian and Wedepohl 1961; Loska et al. 1997). The contamination factor for an individual metal in the sample is measured as $\mathrm{CF} = M_x/M_b$, where M_x stands for the metal concentration and M_b as the background value of the respective metal. Later, in order to represent the total contamination factor of the sample, the CF of all the metals in the sample was summed up. Abrahim and Parker (2007) modified the contamination factor (mC_d) of the sample by considering the average values of each metal CF:

$$mC_{\mathrm{d}} = \frac{\sum_{i=1}^{n} C_{\mathrm{F}}^{i}}{N} \quad (4)$$

Geo-accumulation index

The level of metal concentration above the baseline concentration in the logarithmic component was proposed by Muller (1969) as I_{geo} index:

$$I_{\mathrm{geo}} = \log C_n / 1.5\, B_n \quad (5)$$

C_n and B_n are the concentration of the metal analyzed and its background value, respectively. The B_n is generally considered as pre-industrialization value. Stoffers et al. (1996) showed that a value of 1.5 is introduced to reduce the variation in the background values, resulting due to the difference of lithology (Chakravarty and Patgiri 2009) and anthropogenic influence (Qingjie et al. 2008).

Pollution load index

The representation of pollution load index for a sample was determined as indicated by Tomlinson et al. (1980) by the following equation:

$$\mathrm{PLI} = (\mathrm{CF}_1 \times \mathrm{CF}_2 \times \mathrm{CF}_3 \times \mathrm{CF}_n)^{1/n} \quad (6)$$

where CF and n are contamination factor and number of elements considered, respectively. Evaluation using this parameter was done by several authors (Chakravarty and Patgiri 2009; Seshan et al. 2010; Abel Ghani et al. 2013, 2015; Benson et al. 2017), to determine the pollution status of the sample.

Potential ecological risk factor

The ecological effects of the sediments were studied for the determination of the trace element content in the sediment in relation to the environmental and toxicological impacts. This is studied in conjunction with the contamination factor of a trace element (CF_i) and trace element toxic factor (T_i). Hakanson (1980) derived this potential ecological risk index (E_i) and risk index (RI) by the following relationship:

$$E_i = T_i \times \mathrm{CF}_i \quad (7)$$

$$\mathrm{RI} = \sum E_i \quad (8)$$

Sediment quality guidelines

To evaluate the toxic effect of trace elements on the aquatic organism, the sediment chemistry data, especially metals, have played a significant role. The US National Oceanic and Atmospheric Administration (NOAA) has developed sediment quality guidelines in the past decade based on a large series of biological and chemical data incorporating various methodologies for chemical and biological species. These data sets form the basis for determining the adverse biological effects due to contaminated sediments (Macdonald et al. 2000; Long et al. 1995; Long and Macdonald 1998). Effect range low (ERL) and effect range medium (ERM) are 10th and the 50th percentile of chemical constituents resulting in adverse biological effects. The guideline consists of a threshold effect level (TEL) below which there is no adverse effect in the biota. Probable effect level (PEL) indicates concentration above which adverse effect is expected. The range between TEL and PEL may affect the sensitive organs of an organism, with slight risk (Abdel Ghani et al. 2013; Benson et al. 2017). A large number of samples from the estuaries of the USA have been studied by Long and Macdonald (1998) and Long et al. (2000, 2006) for the probability of toxicity, matching toxicity data with chemical data in relation to m-ERM-Q.

The mean ERM quotient (m-ERM-Q) of the sample is calculated by Long et al. (1995):

$$\text{m-ERMQ} = \frac{\sum_{i=1}^{n} (C_i / ERM_i)}{N} \quad (9)$$

where C_i is the concentration of the trace element and ERM_i is the ERM of the respective metal and N is the number of elements analyzed or included for analysis.

The mean probable effect level m-PELQ is also thus determined by

$$\text{m-PELQ} = \frac{\sum_{i=1}^{n} (C_i / PLI_i)}{N} \quad (10)$$

where PEL_i is the PEL of the respective trace element considered in C_i

Metal bioaccumulation

The bioaccumulation of trace element in the different species of the fishes sampled was determined by comparing with the trace element concentration of

water and sediments collected from the same environment, as it is supposed to be routed into the fish by exposure to this environment (USEPA 2000). This is determined by the formulae adopted by several authors (Abdallah and Abdallah 2007; Islam et al. 2015; Benson et al. 2017). Biological contamination factor (BCF) and biotic sediment accumulation factor (BSAF) are calculated as follows:

$$\text{BCF} = \frac{\text{Trace element concentration in fish}}{\text{Trace element concentration in water}} \quad (11)$$

$$\text{BSAF} = \frac{\text{Trace element concentration in fish}}{\text{Trace element concentration in sediment}}. \quad (12)$$

Target hazard quotient

Target hazard quotient (THQ) (USEP 1989), the non-carcinogenic risk due to the consumption of a specific variety of fish, was calculated, with respect to each trace element analyzed or considered for the study. The ratio of the exposure level of the trace element to the reference dosage determines the potential carcinogenic risk:

$$\text{THQ} = \frac{\text{EF} \times \text{ED} \times \text{FIR} \times \text{Ci} \times 10^{-3}}{R_{fd} \times \text{Bw} \times \text{AT}_n}. \quad (13)$$

Total THQ of the sample is calculated as

$$\text{THQ}_{\text{total}} = \sum_{i=1}^{n} \text{THQ}_i \quad (14)$$

EF is the exposure frequency (365 days year^{-1}), ED is the exposure duration (75.99 years for Kuwaiti women; 73.9 for Kuwaiti men), average life span of people in Kuwait (www.countryeconomy.com/demography/life-expectancy/Kuwait), FIR sea food ingestion rate (30.72 g per capita day^{-1} for Kuwait), (www.FAO.org/faostat/en/#/dataCL), Ci trace element concentration (mg kg^{-1}), Bw average body weight (85.8 for Kuwaiti male and 78.3 for Kuwaiti female), (www.worlddata.info/average-bodyheight.php), AT_n average exposure time for non-carcinogen (365 days years^{-1} × ED), and Rf_D is the oral reference dose, mg kg^{-1} day^{-1}. The oral reference dosage for different elements considered is as follows: Cd 0.003; Ni 0.02; Pb 0.004; As 0.0003 and V 0.007 (Table D1-Appendix D NSQS-USNOAA).

Estimated daily intake (EDI)

The quantity of the daily intake determines the impact of trace element on the biological system. It depends on two parameters: the level of trace element in the food tissue consumed and the bodyweight of the consumer (Zhao et al. 2012; Fu et al. 2008). The EDI (mg kg^{-1}) is calculated for the adult male and female by

$$\text{EDI} = C_i x \text{ DNI } x C_{\text{f}}/Bw \quad (15)$$

C_i is the concentration of trace element mg kg^{-1}; DNI is the daily nutritional intake (30.72 g capita day^{-1}); C_f is the conversion factor of fresh tissue to dry weight (0.2250); and Bw is the bodyweight 85.5 (male) and 78.3 (female).

The chronic daily intake (CDI, mg kg^{-1} day^{-1}) determines cancer and non-cancer hazard; the index is determined by (USNOAA, Appendix B)

$$\text{CDI} = C_i \times FIR \times EF \times ED/Bw \times AT_n \quad (16)$$

Cancer risk is calculated by CDI × SF_i, and SF_i is slope factor for chemical (mg kg^{-1} day^{-1}).

The non-cancer hazard is calculated by

$$C_i = Bw \times AT_n \times Rf_D \times 10^{-3}/(\text{FIR} \times \text{EF} \times \text{ED}) \quad (17)$$

The margin of exposure (MOE) is also used to assess the species-specific non-carcinogenic effects from fish consumption due to trace element concentration (Watanabe et al. 2003) and is determined as follows:

$$\text{MOE} = \text{MCC} \times \text{FIR}/(Bw \times Rf_D) \quad (18)$$

where MCC is the species-specific mean chemical concentration of trace elements (mg kg^{-1}).

Statistical analysis/PCA analysis

Multivariate statistical analysis technique has been used for hydrogeochemical studies on various aspects (Prasanna et al. 2012). Factor analysis is a statistical technique to reduce complex data and to evaluate the relationship among the set of variables (Kumar et al. 2008). It also helps to rank the hydrogeochemical processes according to their order of importance and to detect the most dominant processes in the region. The data were processed by SPSS 16.0 software package.

The normality and homogeneity of the variable were tested by Shapiro–Wilk test and Levine test, respectively. Log transformation of data was performed to improve the distribution for those not meeting the parametric assumptions. The comparison of the mean for each variable was done by the analysis of variance and Tukey–Kramer multiple comparison test. The data set was cleaned initially by removing the variable that was rarely detected in the samples, thus minimizing the colinearity among the variables. The Kaiser–Meyer–Olkin (KMO) measure of the sampling adequacy for overall data is 0.589. Factor extraction was completed with a minimum acceptable eigenvalue of 1. Kaiser normalization and Varimax rotation method were used to extract the final factor along with their percentage of variance and cumulative percentage. The principal component analytical method was used to obtain the initial factor solutions data (Olobaniyi and Owoyemi 2010). The factor scores of each sample were also calculated using this software and were stored as a variable in the input file for further analysis.

Results and discussion

Trace elements in water

The concentrations of Ni and Cr were below the detection limit in all the water samples (Table 2). Vanadium concentration in the seawater sample ranges from 1 to 3 μg L^{-1} (Sepe et al. 2003). The concentrations of the rest of the trace elements analyzed in this study ranked in the following order:

Zn > Cd > As > Pb > Hg > Cu > V

The mean concentrations of Cu, Hg, Pb, and Zn were greater than the respective recommended standards (USEPA 2004). The maximum values of all the trace elements detected in the samples were higher than the recommended water quality criteria, considering the levels of priority toxic pollutants in saltwater (USEPA 2004). The vanadium levels in the present study varied from less than limit of detection (< LOD) to 9 μg L^{-1}, which were greater than those reported in the Saudi coast (2.08–2.60 μg L^{-1}) (Sadiq 1992) and the US marine waters (1.6 μg L^{-1}) (Kennish 1998).

Due to the higher concentration of trace element in seawater, the toxic nature was studied by adopting heavy metal evaluation index (HEI) and degree of contamination (C_d) (Edet and offiong 2002) (Table 3). HEI is evaluated in three categories as low (< 90), medium (90–180), and high (> 180). The HEI value ranges from 84.2 to 360.5 and that of C_d from 77.2 to 353. Among the 15 samples analyzed, the low, medium, and high categories were 2, 8, and 5, respectively. The degree of contamination reveals the representation of 2, 7, and 6 samples in low, medium, and high contamination categories. The metal contamination evaluation studies for water samples show that they are low to highly contaminated.

Trace elements in sediments

The sediment samples reveal the natural and anthropogenic contamination of metals (Kucuksezgin et al. 2006) and are a major sink for the trace elements in the aquatic environment (Masoud et al. 2007). The concentrations of trace elements (Table 4) in the sediments evaluated in this study decreased in the following order: Cr > V > Cu > Ni > Pb > As > Cd. The mean concentrations of all trace elements, except As and Pb, in the sediments, were greater than the threshold effect level (Macdonald et al. 1996) for the respective trace elements. However, the high values of all trace elements are greater than the threshold effect level USEPA guidelines (1999) and the Canadian Environmental Quality Guidelines (2002). The comparison of probable effect levels of Macdonald et al. (1996) and that of Environment Canada (2002) shows that the maximum value of the samples is lesser than the standards except for Cu, Cr, and Ni. The humic substances forming an integral component of organic matter have a high affinity to Cu influencing their concentration levels in sediments. 86%, 71%, 14%, 43%, 29%, and 86% of the samples have their respective trace element (As, Cd, Cr, Ni, and Pb) values below TEL (Table 5). The values of the remaining sample show representation between TEL and PEL, except for Cu, Cr, and Ni with 14%, 14%, and 29% of samples higher than the PEL value, respectively (Table 5). The quantity of the organic matter influences the binding of these trace elements in sediments as it controls their adsorption and desorption capacities (Tomlinson et al. 1980). Cd is more prone to the anthropogenic impacts and shows more association with colloidal materials (Liao et al. 2011). Leakage of gasoline and septic tanks can be possible

Table 2 Evaluation of trace metal content in Marine waters (all values in μg L^{-1})

Samples	As	Cd	Cr	Cu	Hg	Ni	Pb	V	Zn	HEI	Cd
1	43	70	< LOD	7	8	< LOD	27	9	230	360.82	353.82
2	12	54	< LOD	5	< LOD	< LOD	31	< LOD	300	154.44	147.44
3	31	43	< LOD	6	7	< LOD	28	1.3	302	264.02	257.02
4	27	37	< LOD	5	5	< LOD	20	1.2	244	222.37	215.37
5	15	35	< LOD	6	11	< LOD	28	1.1	341	197.07	190.07
6	24	32	< LOD	5	11	< LOD	14	1.61	345	235.12	228.12
7	19	27	< LOD	5	8	< LOD	16	1.52	228	179.91	172.91
8	9	27	< LOD	5	11	< LOD	4	1.01	298	144.45	137.45
9	18	24	< LOD	4	24	< LOD	18	1.24	< LOD	160.84	153.84
10	9	23	< LOD	5	4	< LOD	17	1.62	335	142.38	135.38
11	< LOD	20	< LOD	5	< LOD	< LOD	15	< LOD	344	84.24	77.24
12	12	20	< LOD	6	1	< LOD	18	1.77	365	157.15	150.15
13	10	19	< LOD	4	3	< LOD	11	1.2	353	144.53	137.53
14	9	18	< LOD	4	13	< LOD	7	< LOD	276	136.97	129.97
15	< LOD	16	< LOD	5	3	< LOD	8	1.34	324	86.68	79.68
Avg	15.87	31.00	< LOD	5.13	7.27	< LOD	17.47	1.59	285.67	178.07	171.07
Max	43.00	70.00	< LOD	7.00	24.00	< LOD	31.00	9.00	365.00	360.82	353.82
Min	< LOD	16.00	< LOD	4.00	< LOD	< LOD	4.00	< LOD	< LOD	84.24	77.24
SD	10.40	15.05		0.83	5.99		8.22	2.22	46.21	70.73	70.73
USEPA*1999	36	8.8	50	3.1	0.94	8.2	8.1	–	–	NA	NA

NA not applicable, *LOD* limit of detection

*Recommended water quality criteria for priority toxic pollutants for salt water (in μg L^{-1})

Table 3 Heavy metal evaluation index and degree of contamination in water

	HEI	C_d
Maximum	360.82	353.82
Minimum	84.24	77.24
Average	178.07	171.07
HEI		
Low	< 90	2
Medium	90–180	8
High	> 180	5
C_d		
Low	< 80	2
Medium	80–160	7
High	> 160	6

sources of Pb; it is less soluble and extremely toxic to biota (Sayadi et al. 2009). Pb is also reported to be contributed through atmospheric deposition (Praveen-Kumar et al. 2007).

Mean concentrations of the trace elements were lower than the respective trace element effect range low (ERL). However, the high values of all the trace elements were greater than the ERL, except for Pb, and were lower than ERM, except for Cr. Table 5 shows that 86%, 71%, 28%, 71%, and 100% of the samples were below the ERL for As, Cd, Cr, Cu, Ni, and Pb, respectively, 14% of the samples were above the ERM for Cr, while rest of the trace elements concentrations were between ERL and ERM.

Contamination factor is a good indicator (Turekian and Wedepohl 1961; Loska et al. 1997) for the initial assessment of sediment contamination, which is grouped into four categories with respect to values < 1, 1–3, 3–6, and > 6 for low, moderate, considerable, and very high contamination. Accordingly, our data showed that all samples were under low contamination for As, Ni, and V; five samples were in moderate contamination for Cd; three and one samples

Table 4 Analytical content of trace elements in sediments (all values in μg g^{-1})

Samples	V	As	Cd	Cr	Cu	Ni	Pb
1	33.01	1.54	0.57	57.70	10.85	17.40	0.84
2	27.30	< LOD	< LOD	47.89	9.63	16.56	10.55
3	80.50	3.79	< LOD	156.30	37.11	42.92	0.56
4	60.49	9.72	< LOD	148.10	14.85	13.97	3.83
5	58.91	< LOD	2.43	1119.00	122.24	1.04	32.58
6	81.16	0.12	1.50	86.10	24.33	49.06	< LOD
7	70.80	< LOD	< LOD	142.20	20.93	41.95	< LOD
Minimum	27.30	< LOD	< LOD	47.89	9.63	1.04	< LOD
Maximum	81.16	9.72	2.43	1119.00	122.24	49.06	32.58
Average	58.88	3.79	1.50	251.04	34.28	26.13	9.67
SD	21.51	4.23	0.93	385.28	39.91	18.27	13.42
TEL	–	7.24	0.676	52.3	18.7	15.9	30.2
PEL	–	41.6	4.21	160	108	42.8	112
ERL	–	8.2	1.2	81	34	20.9	46.7
ERM	–	70	9.6	370	270	51.6	218
ORD	0.007	0.0003	0.003	1.5	0.04	0.02	0.004
CR		1.5					0.0085
NCH	0.007	0.0003	0.0005	0.005	0.0371	0.02	0.004

All values are in μg g^{-1}

TEL threshold effect level, *PEL* probable effect level, *ERL* effect range low, *ERM* effect range medium, *ORD* oral dosage reference, *CR* cancer risk, *NCH* non-cancer hazard

Table 5 Assessment of elements considering the SQG

	< ERL (%)	ERL–ERM	> ERM (%)
As	86	14%	0
Cd	71	28.50%	0
Cr	28.50	57%	14
Cu	71	28.50%	0
Ni	51	43%	0
Pb	100	0.00	0
	< TEL (%)	TEL–PEL	> PEL (%)
As	86	14%	0
Cd	71	28.50%	0
Cr	14.00	71%	14
Cu	43	43.00%	14
Ni	29	43%	29
Pb	86	14.00	0

were in very high contamination for Cr and Pb, respectively; one sample in considerable contamination for Cu and Cd; and one sample is very high contamination for Cd and Cr. In general, the contamination index reveals that predominantly two metals Cr and Cd are responsible for the contamination and sample 5 shows more contamination than other samples.

The pollution load index of greater than one indicates pollution (Chakravarty and Patgiri 2009; Seshan et al. 2010). Accordingly, in our study, only sample 5 was polluted. The metal pollution index (MPI) ranged from 5.95 to 18.21, with the highest value for sample 5 (Table 6).

I_{geo} index is categorized into six groups (Muller 1969): Accordingly, our results showed that (1) unpolluted (< 1) for most trace elements in all the samples, especially As, Ni, Pb, and V; (2) very low polluted (1–2) for Cu and Cd in samples 5 and 6, respectively; (3) low polluted (2–3) for Cd in sample 5; (4) moderately polluted (3–4) for Cr in sample 5.

Table 6 Evaluation of sediment pollution by different parameters

	Contamination factor (CF)						
	As	Cd	Cr	Cu	Ni	Pb	V
1	0.12	1.90	0.64	0.27	0.26	0.04	0.25
2			0.53	0.24	0.24	0.53	0.21
3	0.29		1.74	0.93	0.63	0.03	0.62
4	0.75		1.65	0.37	0.21	0.19	0.47
5		8.10	12.43	3.06	0.02	1.63	0.45
6	0.01	5.00	0.96	0.61	0.72		0.62
7			1.58	0.52	0.62		0.54
	Ecological risk factor (E_i)						
	As	Cd	Cr	Cu	Ni	Pb	V
1	1.18	57.00	1.28	1.36	1.28	0.21	0.51
2			1.06	1.20	1.22	2.64	0.42
3	2.92		3.47	4.64	3.16	0.14	1.24
4	7.48		3.29	1.86	1.03	0.96	0.93
5		243.00	24.87	15.28	0.08	8.15	0.91
6	0.09	150.00	1.91	3.04	3.61		1.25
7			3.16	2.62	3.08		1.09
	Igeo index						
	As	Cd	Cr	Cu	Ni	Pb	V
1	− 3.66	0.34	− 1.23	− 2.47	− 2.55	− 5.16	− 2.56
2			− 1.50	− 2.64	− 2.62	− 1.51	− 2.84
3	− 2.36		0.21	− 0.69	− 1.25	− 5.74	− 1.28
4	− 1.00		0.13	− 2.01	− 2.87	− 2.97	− 1.69
5		2.43	3.05	1.03	− 6.62	0.12	− 1.73
6	− 7.34	1.74	− 0.65	− 1.30	− 1.06		− 1.26
7			0.07	− 1.52	− 1.28		− 1.46

	mC_d	RI	MPI	PLI	m-ERM-Q	m-PEL-Q
1	0.50	62.82	5.95	0.27	0.62	1.05
2	0.25	6.54	8.05	0.45	0.53	0.87
3	0.60	15.56	12.30	0.47	1.45	2.42
4	0.52	15.54	13.18	0.51	0.88	1.66
5	3.67	292.27	18.21	1.20	3.90	9.02
6	1.13	159.90	7.63	0.53	1.43	2.27
7	0.47	9.95	9.83	0.83	1.27	2.06

Furthermore, there is no representation of samples in groups ranging between values 4 and 5 and > 5. Similar to earlier observation Cd, Cr and Cu show low to moderate pollution by I_{geo} index (Table 6). Our results show that contamination was much greater in sample 5 as compared to that of the other samples.

The ecological risk factor was proposed by Hakanson (1980) to determine the heavy trace element contamination in sediments. The threat to ecological health due to the release of toxic elements from the sediment to the water could be assessed by this method based on the contamination factor and toxicological response factor. There are five categories based on the range of these factors: E1 (< 40; low); E2 (40–80; moderate); E3 (80–160; considerable); E4 (160–320; high); and E5 (> 320; very high). Accordingly, all trace elements irrespective of the sample, except Cd, represent the E1 category. For Cd, samples 1, 6, and 5 were in E2, E3, and E4 categories, respectively, suggesting potential ecological risk with respect to Cd (Table 6).

The risk factor (RF) represents the sum of all potential ecological risks of all trace elements in a sample. Using this index, the ecological risk can be grouped under four categories: low, moderate, considerable, and high for RF values less than 95, 95–190, 190–380, and greater than 380, respectively. In our study, RF varied from 6.5 to 292.3. Accordingly, all samples represented a low risk, except samples 5 and 6 which showed considerable and moderate risks (Table 6).

The mean ERMQ of the samples varied from 0.53 to 3.8, while mean PELQ ranged between 0.86 and 9.0. These quotients were high in sample 5. The classification is based on the percent probability of being toxic, i.e., 12, 30, 40, and 74 percent for < 0.1; 0.11–0.5; 0.51–1.5; > 1.5, respectively. All samples have m-ERM-Q in the 0.5–1.5 range and thus indicate a 40 percent chance of being toxic, except sample S5 that was in the category of 74% chance of toxic. Similarly, m-PEL-Q are classed into low, moderate, toxic, and high (Table 6).

Trace elements in fish

The accumulation of trace elements in fish is generally governed by the efficiency of biotic binding sites and concentration in the feed, metabolism, swimming behavior, and the environment. This is also influenced by various other physiological parameters like weight, body mass, osmoregulation and not through ventilation (Zheng et al. 2007). So, it varies in smaller and larger varieties of fishes (Lamiceli et al. 2015). Fishes living in contact with sediments or in neritic zones show the possibility of more contamination due to anthropogenic pressure than the pelagic fishes (Suárez-Serrano et al. 2010). Copper, Mn, Ni, and Zn perform vital functions in all living organisms as they form the essential elements. These elements are required for the secretion of different enzymes and the development of cell components, but in higher concentrations, they are still toxic (Demirezen and Uruç 2006). The average concentration of the trace elements in fishes show (Table 7) the following order during winter: As > Cd > Ni > Pb > V and during summer it is As > Ni > Cd > Pb > V, irrespective of the fish species. It is interesting to note that Ni concentration increases in the fish during summer. As Cd and Pb have no role in the biological process even at low levels, they are harmful and toxic, so they are considered as nonessential elements (Bahnasawy et al. 2009). Cadmium bioaccumulation is generally hazardous as it is a nonessential trace element (Kalman et al. 2010; Liao et al. 2011). Cd and Pb can be concentrated in the food web and tend to be toxic as they can be stored and assimilated.

Arsenic, nickel, and lead are detected in all fishes unlike the other elements like Cd and V. It is observed that high average concentrations of Cd, Ni, and Pb are noted in Speatty and Nagroor during winter (Supplementary Fig. 1). Arsenic concentration is higher in Lobster irrespective of the season in varying magnitude. In Sheam, Nagroor, and Lobster, As concentration is observed to be higher during winter, but in Speatty, it is higher during summer. Earlier studies by Bu-Olayan et al. (2001) in Nagroor and Sheam observed that most of these marine organisms have lower arsenic levels than those from other parts of the world, but the present result shows, on the contrary, the higher arsenic levels in these fishes. Arsenic does not form a part of the natural biogeochemical process; it is considered as a very toxic and hazardous element (Kosanovic et al. 2007). The concentration of As is due to the difference in biological metabolism. The methylation of inorganic arsenic produces the organic arsenic forms, which are excreted much faster during the metabolism (Mandal 2002). The essential element concentrations are regulated in the tissues and other parts of the body by the process of biomechanism in crustaceans. Hence, the concentration of nonessential trace elements like As in the body is regulated in these species (Rainbow 1988, 2002; López et al. 2004), by an enhanced mechanism to accumulate more than the essential elements (Suarez-serrano et al. 2010).

Table 7 Range of trace element content in fish varieties considered (all values in μg g^{-1})

Species	Season	Range	As	Cd	Ni	Pb	V
Sheam	Winter (n = 11)	Maximum	10.40	0.37	0.54	0.20	0.06
		Minimum	1.20	< LOD	0.04	0.08	< LOD
		Average	4.67	0.23	0.18	0.12	0.06
		SD	2.53	0.06	0.14	0.04	0.02
	Summer (n = 7)	Maximum	6.90	0.18	0.76	0.90	< LOD
		Minimum	0.04	< LOD	0.16	0.05	< LOD
		Average	4.10	0.15	0.38	0.20	< LOD
		SD	2.35	0.04	0.21	0.31	< LOD
Lobster	Winter (n = 8)	Maximum	43.60	0.76	0.76	0.29	0.04
		Minimum	12.40	0.17	0.06	0.05	0.03
		Average	22.86	0.43	0.29	0.14	0.04
		SD	9.74	0.18	0.26	0.13	0.01
	Summer (n = 6)	Maximum	27.30	0.00	0.75	0.29	0.08
		Minimum	9.05	< LOD	0.08	0.07	0.08
		Average	14.74	0.00	0.26	0.14	0.08
		SD	6.91		0.24	0.09	
Speatty	Winter (n = 9)	Maximum	14.90	4.30	1.10	0.66	10.71
		Minimum	3.60	0.20	0.21	0.03	5.43
		Average	7.29	1.05	0.81	0.16	7.26
		SD	3.28	1.25	0.29	0.20	2.31
	Summer (n = 6)	Maximum	26.00	0.53	0.71	0.26	0.26
		Minimum	17.00	0.14	0.13	0.06	0.10
		Average	21.82	0.35	0.41	0.13	0.20
		SD	4.08	0.14	0.28	0.09	0.08
Nagroor	Winter (n = 6)	Maximum	26.00	1.40	1.11	0.86	< LOD
		Minimum	2.90	0.17	0.23	0.04	< LOD
		Average	7.20	0.78	0.49	0.24	< LOD
		SD	9.22	0.87	0.33	0.31	< LOD
	Summer (n = 5)	Maximum	3.20	0.20	0.51	0.33	< LOD
		Minimum	1.60	0.20	0.15	0.09	< LOD
		Average	2.26	0.20	0.31	0.20	< LOD
		SD	0.68		0.14	0.12	< LOD
MAC WHO	2004		1	0.1	0.8	0.5	0.5
FAO	FAO 1983		0.26	0.05	–	0.5	–
UK	MAFF 2000		–	0.2	–	2	–
EC	EC 2005		–	0.05	–	0.2	–
Saudi	SASO 1997		–	0.5	–	2	–

Species-specific levels were significant for As in Lobster and Speatty. Arsenic present in the fish tissues is the least hazardous form of arsenobetaine (Eisler and Peschel 2010). V studies in fish are scarce (Lamiceli et al. 2015). V is detected in Sheam, Lobster, and at higher levels (7.25 μg g^{-1}) in Speatty (Fig. 2). The role of vanadium in physical anatomy or the biological metabolism is not clear so far, though it is not reflected as an essential trace element (Abdel Ghani 2015). On the contrary, Cd, Ni, and Pb do not show species-specific relationships, but element-specific distribution is observed in this study. Cd is toxic as it is ingested, accumulated, and concentrated through the food chain. Ni is an essential heavy metal

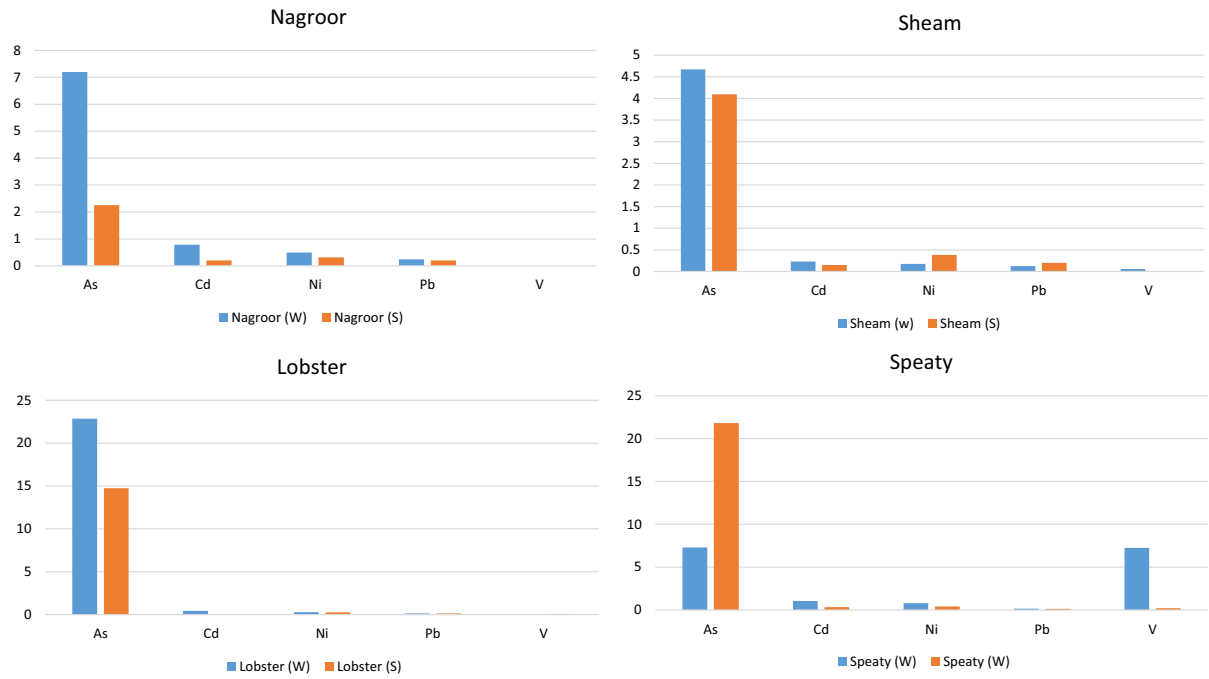

Fig. 2 Temporal variation of trace element concentration with respect to fish varieties (all values in μg g^{-1})

to aquatic life at low levels (Magyarosy et al. 2002) and forms complexes with many ligands, thus becoming more mobile than other toxic elements (Palaniappan and Karthikeyan 2009).

It is also inferred that all the maximum values of trace elements irrespective of species are higher than the standards and mostly during winter than the summer. Certain metals like Ni and Pb in Sheam and As in Speatty show higher concentration during summer (Fig. 2). The toxic level of metals was noted in Sheam in Kuwait Governorates GI, GII, and GIV (Bu-Olayan and Thomas 2013). Further, it was inferred as an authentic bioindicator of coastal pollution. The study on the temperature variations and metal contamination in fish indicated that the levels of metals increase when there is an increase in the temperature of the environment (Grasset et al. 2016). Arsenic in Speatty, and Ni and Pb in Sheam show dominance during winter, when the temperature goes < 11 °C in the Kuwait territorial waters (Bhandary et al. 2018), but due to the sluggish movement of the water mass in the bay, and it is a shallow environment, with several outlets of domestic sewage, desalination brines, treated wastewater, and municipal dump; the temperature of the bay is comparatively higher than the territorial waters (Chidambaram et al. 2019). The movement of water mass, transport of sediments, and shift in prey concentration in the sampling area during various sampling periods influence the metal concentrations in the fish. Metal concentration in fish is mainly due to metal-binding proteins such as metallothioneins (Canli and Atli 2003), and Nussey et al. (2000) inferred that the concentration of metal in the fish is inversely proportional to the size of the fish. Conversely, Douben (1989) reported no relationship between fish size and concentrations of some metals. Metal toxicity in fish is mainly governed by external parameters like pH, salinity, hardness, temperature, amount of metal concentration in water, and exposure time.

The higher levels of trace element concentration in the fishes collected during winter may be due to different reasons. Furthermore, trace element toxicity is also governed by individual metabolism, behavior, growth, and sex (Kalay et al. 1999; Kalay and Canli 2000; Al-Yousuf et al. 2000). Elevated metal concentrations in water affect the size, hatching time, growth and development of fish embryos (Weis and Weis 1989; Heath 1987; Friedmann et al. 1996). Later, Asuquo et al. (2004) inferred that fish species, trophic level, mode of feeding, pollutant type, and the sampling location govern the degree of contamination.

This may be attributed to the dilution of the marine aquatic system by the vertical mixing of the extensive evaporated dense sea surface water with the benthic waters. The rise of the cool waters to the surface results in the circulation of water from the surface to bottom, and TSW gets diluted during summer, but this is less significant during summer. High trace element concentration may also be attributed to the untreated waste discharged into the bay containing higher levels of trace elements during winter than summer (Bu-Olayan and Thomas 2013). Furthermore, it was identified that phytoplanktons bloom during late summer, and subsequently, its consumption results in bioaccumulation. It adds to the fact that due to lesser metabolic activity during winter, the assimilation of trace elements is low. This enhances the trace element levels in the fish tissues during winter. The phenomenal reduction in freshwater flow from the Shatt-Al-Arab River and the spatial distribution of phytoplankton's have also impacted the marine fish environment along with vertical mixing. It may also be due to the migration of fishes from territorial waters into the bay, i.e., from low-temperature regions to warmer regions during winter. The heat is induced into the water mass from various anthropogenic drivers like desalination reject, thermal plant, and effluents from cooling towers.

Body accumulation index

The body accumulation index (BAI) is assessed to understand the proportion of trace elements migrated from the sediment or water to the fish; it is generally expressed in percentage. BAI shows a higher percentage of As noted in fish than the sediment and water values. Speatty (S, summer) and Lobster (W, winter) reflect higher concentrations than the trace element content in water (Supplementary Fig. 2). As is higher than the other metals studied in fish and also higher than the average concentration of metals in sediment and water irrespective of the fish variety. But other metals like Pb, Cd, Ni, and V are less than the average composition of sediment and water. Arsenic in Nagroor samples during summer is lesser than those in sediments. Vanadium is found to be higher in Speatty; BAI reveals that the seawater has contributed more fraction of V during summer and from sediments during winter. The biological accumulation of Cd shows more contribution of sediments than from marine waters. Cadmium is also considered as endocrine disruptors and results in variations in morphometry, size, and mortality (Low and Higgs 2014). Among the water samples analyzed in this study, Ni concentrations were below the detection limit but were detected in sediments. A higher fraction of Pb in fish is noted to be derived from the sediments than that of the marine waters. Hence, BAI in fish species shows that the metals in sediments play a significant role than that of metals in marine waters. The sediment metal levels require a long-term process for the reestablishment of the species in the contaminated environment (Pyle and Mather 2003). The higher As and V concentrations in the fish than that of sediment and water may be due to several factors, including environmental and biomass variables (Lamiceli et al. 2015). Trace element uptake by fishes is generally affected by the pH and competing cations like Ca and Mg present in seawater (Pyle et al. 2002). The lack of species-specific patterns for Cd, Ni, and Pb could be related to the difference in background contamination levels. Bioavailability of metals decreases at higher pH due to the formation of inorganic complexes with carbonates and hydroxide, while at lower pH, the metals are predominantly in free form which is readily available (Kushner 1993; Stumm and Morgan 1981). Metal accumulation in aquatic species is primarily due to ingestion of food or intake rather than direct contact with sediments (Beg et al. 2015). Thus, BAI infers that most of the Cd is derived from the sediments and that of Pb and V mainly from marine waters. Lead is adsorbed onto the organic contents in the sediment, but in water, it is influenced by pH, hardness, and alkalinity (Zhang et al.2018). Hence, the study proves that the accumulation of Pb in fish is mainly through contaminated water than food or sediment (Cretì et al. 2009). Both essential and nonessential trace elements are toxic (Ebrahimpour and Mushrifah 2010) due to their biomagnification, bioaccumulation, and long persistence in the food chain. The species in the high part of the food web tend to accumulate more metals (Terra et al. 2007). Lack of biomagnification of trace elements with respect to species length and weight was observed by several authors (Nfon et al. 2009; Lamiceli et al. 2015). The metabolic activity also governs the accumulation, as the activity in younger fish is higher than the older ones; the concentration of trace elements is also observed to be higher in younger

Table 8 Average estimated daily intake (mg kg^{-1} day^{-1})

	As		Cd		Ni		Pb		V	
	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female
Sheam - Winter	0.38	0.41	0.02	0.02	0.01	0.02	0.01	0.01	0.00	0.00
Sheam - Summer	0.33	0.36	0.00	0.00	0.03	0.03	0.02	0.02	0.00	0.00
Lobster - Winter	1.84	2.02	0.03	0.04	0.02	0.03	0.00	0.00	0.00	0.00
Lobster - Summer	1.19	1.30	0.00	0.00	0.02	0.02	0.01	0.01	0.00	0.00
Speatty - Winter	0.57	0.62	0.08	0.09	0.07	0.08	0.01	0.01	0.32	0.36
Speatty - Summer	1.60	1.75	0.02	0.03	0.03	0.03	0.01	0.01	0.01	0.01
Nagroor - Winter	0.58	0.64	0.02	0.02	0.04	0.04	0.02	0.02	0.00	0.00
Nagroor - Summer	0.18	0.20	0.00	0.00	0.03	0.03	0.01	0.01	0.00	0.00

fish (Widianarko et al. 2000), but it was inferred to reach a steady state after a certain age (Douben 1989). Lamiceli et al. (2015) proved that biochemical accumulation could increase the trace element levels in the fish even when there is no source of exposure.

Health hazards on human consumption

The impact of the migration of these trace elements from fish to the human (next level in the food chain) is measured by understanding certain parameters like estimated daily intake, chronic intake, hazard quotient, and margin of exposure. Estimated daily intake is higher with respect to arsenic both in male and in female, specifically in Lobster irrespective of the season and due to the consumption of Speatty during summer (Table 8). The daily intake and other hazard parameters are higher in Kuwaiti female than male, as their life expectancy is more and the average body weight is lesser. This higher estimated daily intake in a female is also reflected in chronic daily intake with respect to arsenic, as its THQ is higher than unity. The THQ in a female is higher than male, especially due to the consumption of Lobster and Speatty in summer. The HQ or the THQ is higher than unity in all the samples due to higher levels of arsenic in all fishes (Supplementary Figs. 3, 4).

The non-cancer hazard due to the metal in all the fish varieties was studied for both male and female. The NCH value ranges from 1.89 to 19.86 in the male for arsenic, and the highest value was observed in Lobster (Table 9). The Cd value ranges from 0.33 to 8.7, and a higher value is noted in Speatty. Ni shows values ranging from 9.81 to 48.5, and higher values were observed in Speatty. Pb ranges from 0.57 to 2.73 with a higher value in Nagroor, and vanadium is higher in Speatty with NCH values ranging from 0 to 78.8 (Supplementary Figs. 3, 4). A similar trend was observed in values for non-cancer hazards for a female with a slightly higher degree than male. Furthermore, it is inferred that the consumption of Speatty has the highest non-cancer hazard quotient in the fish varieties considered for the study. There is a probability of cancer hazard due to arsenic levels in all the fish considered, and it tends to be higher in Lobster and Speatty, with lesser risk in Nagroor and Sheam (Table 10). Though the toxicity calculations generally consider the total analytical value of the element, much of the element (As) is not absorbed by the humans. The margin of exposure (MOE) is greater than 1 for As and < 1 for all the other elements. MOE < 1 indicates a safe daily dose, considering both chronic and non-carcinogenic effects. Similar to other results, Lobster and Speatty have a higher margin of exposure, resulting in health effects due to the long-term consumption of these varieties of fish. Wide variability in concentration with species and season (Table 11) reveals that they are affected along the coastal regions than the pelagic, as arsenic is naturally present in the sediment of reducing environments (Bhandary et al. 2018).

Statistical analysis

Statistical data reduction by factor analysis was carried out for the metals analyzed in all the fish

Table 9 Average non-cancer hazard

	As		Cd		Ni		Pb		V	
	Male	Female	Male	Female	Male	Female	Male	Female	Male	Female
Sheam W	3.91	3.57	1.57	1.43	9.81	8.95	1.36	1.24	0.50	0.45
Sheam S	3.43	3.13	0.36	0.33	21.32	19.46	2.25	2.05	0.00	0.00
Lobster W	19.16	17.48	3.62	3.30	16.02	14.62	0.57	0.52	0.17	0.16
Lobster S	12.35	11.27	0.00	0.00	14.62	13.34	1.31	1.19	0.26	0.24
Speatty W	5.88	5.36	8.77	8.00	48.85	44.58	1.69	1.54	78.80	71.91
Speatty S	16.60	15.15	2.42	2.20	21.23	19.37	1.32	1.20	2.61	2.38
Nagroor W	6.03	5.50	2.19	2.00	27.48	25.08	2.73	2.49	0.00	0.00
Nagroor S	1.89	1.73	0.33	0.30	17.52	15.99	1.82	1.66	0.00	0.00

W winter, *S* summer

Table 10 Average toxicity values in different fish varieties

	THQ	EDI	CDI	NCH	As-Can
Sheam W	5.57	0.38	1.67	3.91	2.51
Sheam S	4.89	0.33	1.47	3.43	2.20
Lobster W	27.29	1.84	8.19	19.16	12.28
Lobster S	17.60	1.19	5.28	12.35	7.92
Speatty W	8.37	0.57	2.51	5.88	3.77
Speatty S	23.64	1.60	7.09	16.60	10.64
Nagroor W	8.59	0.58	2.58	6.03	3.87
Nagroor S	2.69	0.18	0.81	1.89	1.21

W winter, *S* summer

Table 11 Species-specific margin of exposure (MOE)

	As	Cd	Ni	Pb	V
Sheam W	5.84	0.029	0.003	0.011	0.003
Sheam S	5.12	0.019	0.007	0.019	0.000
Lobster W	28.59	0.054	0.005	0.013	0.002
Lobster S	18.44	0.000	0.005	0.013	0.004
Speatty W	9.12	0.131	0.015	0.015	0.389
Speatty S	27.29	0.043	0.008	0.012	0.011
Nagroor W	9.00	0.098	0.009	0.023	0.000
Nagroor S	2.82	0.025	0.006	0.019	0.000

W winter, *S* summer

samples. Three factors with eigenvalues greater than 1 were extracted as significant factors that accounted for 81.5% of TDV. The first factor was represented by Cd, Ni, and V with 38% of TDV, the second factor by Ni and Pb (22.3% TDV), and the third factor by arsenic with 21.2% of TDV (Fig. 3).

The association of Cd, Ni, and V is attributed to the contamination from the effluents of oil-based pollution (Bhandary et al. 2018). Cd is mainly anthropogenic, hazardous, and extremely toxic (Kosanovic et al. 2007) and generally released into the environment through anthropogenic sources (from refineries, batteries, plastic stabilizers, electroplating, and pigment) resulting in the adverse biological effects in the aquatic habitats (Järup 2003). Nickel in fish is due to predominantly anthropogenic and natural sources. It is released due to oil-burning power plants, coal-burning power plants, and trash incinerators. Sediments also receive effluents from the oil industry, and the present study shows that Ni concentrations are higher in sediments. This metal is significant as it gets easily absorbed onto the biological membranes and it also enhances the binding capacity of organic matter in the dissolved form. Vanadium is mostly available in the environment due to the industrial sources, predominantly due to oil refineries and due to the power plants operated by oil or coal (Minelli et al. 2000). The leaching of V from the natural sources like rock weathering and from deposited sediments also contributes as a natural source (Sepe et al. 2003).

The second factor is represented by Ni and Pb reflecting the influence of urban sewage and wastewater drains (Bu-Olayan and Thomas 2012). Higher Pb concentration in fish is mainly attributed to

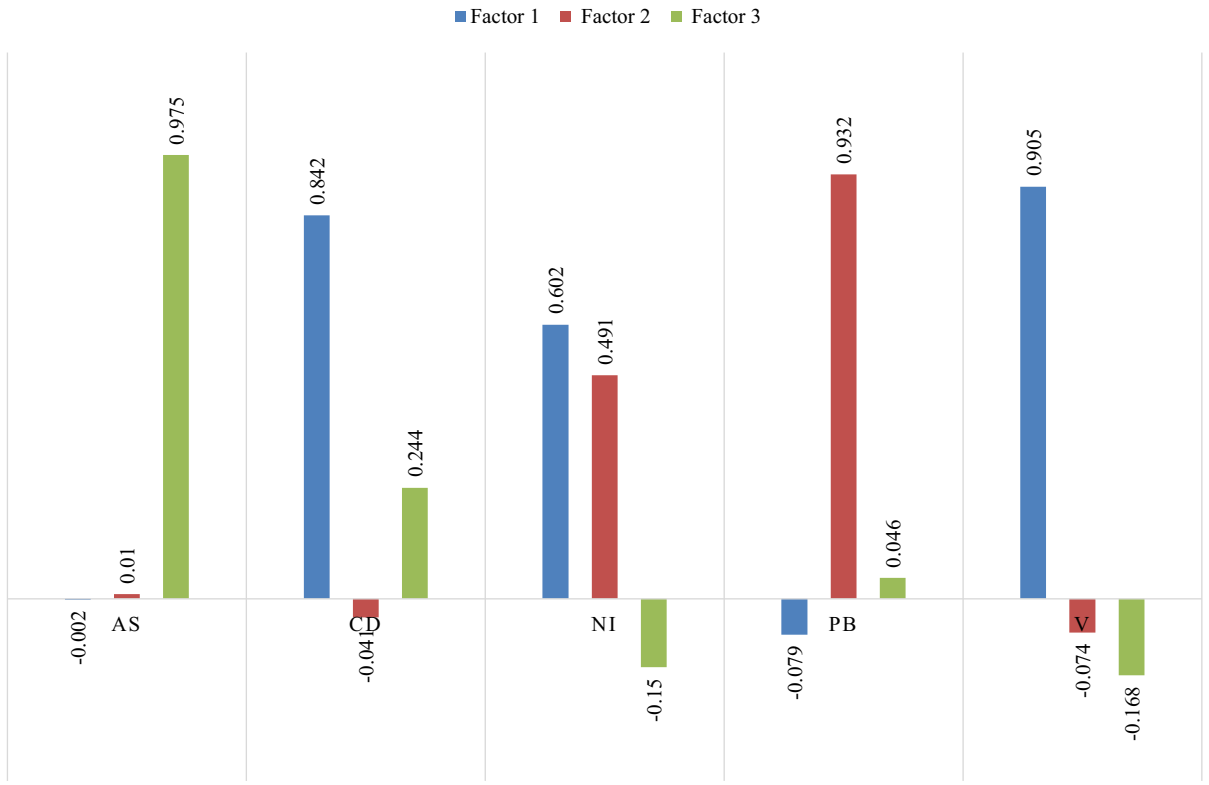

Fig. 3 The principal component analysis of trace element analyzed from studied varieties of fishes

commercial ship traffic and industrial sewage. The concentration of metals in various organisms is dependent on their exposure to assimilate the metals form a mixed pollutant source (Boening 2000). The high lead levels reported in fishes of Bahrain (Madany et al. 1996) were attributed to the urban land-based sources like automobile industry effluents and due to fuel spills from the ship in harbors. Lead is also considered toxic and predominantly released into the environment due to anthropogenic processes like dissolution form lead plumbing, lead dust fallout, and municipal wastewater (Sorensen 1991). The pollution effects of Ni and Pb are mainly due to land-based effluents, desalination brine recreation sailing boat, fishing, and yacht sport. The crude oil spill, industrial effluent, municipal sewage runoff, untreated waste dumping, and urbanization (Bu-Olayan and Thomas 2012) are also associated with this factor, as they are responsible for Ni and Pb content in the marine environment.

The arsenic factor is mainly attributed to the natural source than the anthropogenic source, governed by the redox condition of the environment and adsorption–desorption capacity of the sediments. Fish is contaminated with arsenic due to their long-term exposure through gills and feeding of As-enriched food (Ahmed and Naim 2008; Freije 2015). Arsenic accumulates on the finer sediments which are generally found either in suspension or gets deposited in the seabed (Svobodová 1993). Arsenite [As(III)] form is more adsorbed than the arsenate [As(V)] form. The earlier form is more toxic; hence, the redox states determine the nature of the toxicity of the element. Though oxidation states determine the toxicity of the As, there are classes of compounds found in seafood, like arsenolipids, which are recently considered to be more toxic (Witt et al. 2017).

The factor score representation (Fig. 4) reveals that all the three factors are represented in Speatty. It is also clear that during winter it is mainly influenced by oil-based industrial pollution, but in summer it is influenced by both urban sewage (Pb and Ni) and natural contamination by arsenic. Lobster shows the predominance of the third factor irrespective of seasons.

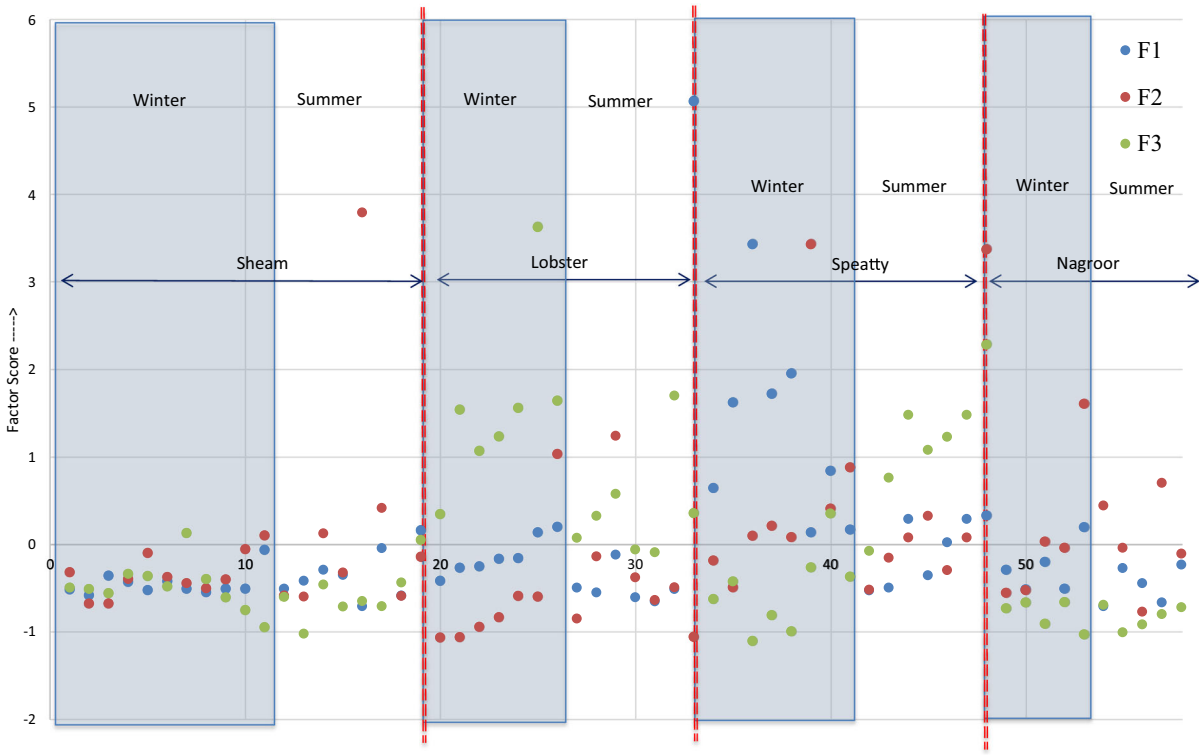

Fig. 4 Factor score representation of PCA loading for studied marine fishes

Raissy et al. (2011) inferred that there had been no statistical variations in the metal concentrations of Lobster irrespective of carapace length and weight. It is a fact that there are numerous pathways for bioaccumulation of metals in Lobster like ingestion of sediment and water, absorption at gills and surface, and consumption of contaminant-accumulated organisms (1993a, b).

There is no significant representation of fuel-based contamination in Sheam and Nagroor, though there are random signatures of this factor in Sheam during winter and in Nagroor during summer. Sheam is targeted to determine environmental degradation due to industrial, coastal, or urban activities and to understand their adaptive mechanism due to chemical exposure (Hedayati and Hosseini 2012). It is inferred that there is practically less influence of oil contamination and the influence of arsenic in Sheam and Nagroor varieties.

Conclusion

This study demonstrated that metals in marine water are moderate to heavily contaminated with some metals like Cd, Cu, Hg, and Pb. The sediments also show Cr greater than ERM and Cr and Cu, along with Ni greater than PEL. Subsequent observations by adopting the contamination factor, I_{geo} index, ecological risk factor, risk index, and PLI, show that the samples range from low- to high-risk category. Cd and Cr levels in the samples were in toxicity range in the sediments. The m-ERM-Q reveals that the samples have 40–74% toxic nature, and by considering m-PELQ it is 21–73%. Concentrations of As and Cd were greater in all fish samples than that of the rest of the trace elements. The toxic values of As were noted in Lobster, Cd, Ni, V in Speatty, and Pb in Sheam. Arsenic in fish is inferred to be due to biomagnification, Cd is chiefly from sediment, and Pb from marine waters. Concentrations of trace elements were greater in winter than those in summer. Consumption of Lobster and Speatty, especially during winter, appears to contribute to increased EDI, CDI, MOE, and non-

cancer and cancer hazards. The hazard impact was greater in Kuwaiti female than that in male. Effluents from oil-based industries, urban sewage, and wastewater drains (land-based effluents) and the redox conditions of the natural environment are identified as chief sources of trace elements in the fish varieties. It has also been identified that Sheam is mainly affected by land-based effluents during winter and Lobster by natural factors irrespective of the season. During winter, the trace element concentration in Speatty is mainly due to oil and land-based effluents, but in summer its land-based effluents and arsenic factor. Among the fish considered in this study, Speatty is exposed to more risk of contamination from all sources. Hence, it is suggested that the consumption of Lobster and Speatty may be regulated to prevent migration to the next level of the food chain.

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