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Assessment of heavy metal contamination in intertidal gastropod and bivalve shells from central Arabian Gulf coastline, Saudi Arabia

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ABSTRACT

In order to assess pollutants and impact of environmental changes along the Saudi Arabian Gulf coast, forty specimens of gastropod and bivalve shells belonging to *Diodora funiculata*, *Lunella coronata*, *Cerithium caeruleum*, *Barbatia parva*, *Pinctada margaritifera*, *Amiantis umbonella*, *Acrosterigma assimile* and *Asaphis violascens* from five localities are selected for Fe, Cu, Pb, Mn, Cd, Se, As, Co, B, Cr, Hg, Mo analysis. The analysis indicated that heavy metal values (except Fe) were less than those recorded in molluscan shells from Gulf of Oman, Red Sea and Indian Ocean. *D. funiculate*, *L. coronata*, *B. parva* and *P. margaritifera* are good accumulators of Cu, As, Cr. The other species gave a nearly constant concentration in all the studied areas. Al Jubail coast recorded the highest heavy metal concentrations (except Mn at Ras Al-Ghar and Se at Al Jubail industrial city). Heavy metal contamination is mostly attributed to anthropogenic sources, especially effluents from petrochemical industries, sewage and desalination plants.

1. Introduction

The molluscan capacity to magnify and integrate aquatic pollutants is considered to play a key role in monitoring environmental quality (Bourgoin, 1990). The tolerance and adaptability of the bivalve molluscs have made them a preferred organism as indicator of the quality of ecosystems (Conti and Cecchetti, 2003). Heavy metal contamination level in a given organism results from the net balance between the processes of metal uptake and metal loss (Goodfriend et al., 1989).

Heavy metals can enter into the coastal environments from different sources including natural weathering processes and anthropogenic activities (Sadiq and Alam, 1989; Sadiq et al., 1992; Ziko et al., 2001; Abd El-Wahab and El-Sorogy, 2003; Krishna and Govil, 2007; Badr et al., 2009; Venkatramanan et al., 2012; Garali et al., 2010; Madkour, 2013; El-Sorogy et al., 2012, 2013a, b). Many studies have been dealt with fauna, environmental assessment, and sedimentology of the Arabian and Oman gulfs (Dance and Eames, 1966; Dance et al., 1992; Bosch et al., 1995; Al-Homaidan, 2006; Loughland et al., 2012; El-Sorogy et al., 2015; Almasoud et al., 2015).

The Arabian Gulf is a shallow and semi-enclosed basin, therefore, the impact of pollutants on marine environment as a result of intensive anthropogenic activities may be significant (Pourang et al., 2005; Naser, 2013). Industrial and sewage effluents, and wastewater discharges from desalination plants are considered as anthropogenic sources that may significantly contribute to damage the environment of the Arabian Gulf (Sheppard et al., 2010; Naser, 2013). It has been reported that oil can also be considered as a significant and chronic pollution source in the Gulf environment (Naser, 2013). Additionally, chimney emissions of many industrial complexes may contribute to the metal pollution in the Gulf, leading to disturbance to the coastal environment (Sadiq and Alam, 1989).

There is a rapid industrial development at the coast of the Arabian Gulf in Saudi Arabia, which may be the source of pollution to marine organisms. Therefore, the main objectives of the present study are to evaluate the levels of pollution along the central Arabian Gulf from Ras Abu Aly to Ras Tanura in order to provide a successful management of the marine ecosystems.

2. Material and methods

Gastropod and bivalve shells are collected from the intertidal

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coastal areas of Ras Abu Aly, Al Jubail, Al Jubail Industrial city, Ras Al-Ghar and Ras Tanura along the central Arabian Gulf, Saudi Arabia (Fig. 1). Due to their occurrence throughout the five studied areas, three gastropods species, *Diodora rueppellii* (Sowerby, 1834), *Lunella coronata* (Gmelin, 1791) and *Cerithium caeruleum* (Sowerby, 1855) and five bivalve ones, *Barbatia parva* (Sowerby, 1833), *Pinctada margaritifera* (Linnaeus, 1758), *Amiantis umbonella* (Lamarck, 1818), *Acrosterigma assimile* (Reeve, 1845) and *Asaphis violascens* (Forsskål, 1775) have been chosen for chemical analysis (Fig. 2, Table 1). 40 specimens were analyzed for Fe, Cu, Pb, Mn, Cd, Se, As, Co, B, Cr, Hg, Mo with Inductively Coupled Plasma Mass Spectrometer (ICP-MS): NexION 300 D (Perkin Elmer, USA). Triplicate samples were collected and analyzed. The shell samples were ground, sieved through 2 mm sieve and transferred to plastics bags. 200 mg of samples were placed in a dry and clean Teflon digestion beaker, and 6 mL of HNO_3, 2 mL HCl and 2 mL HF were added to the Teflon beaker. Samples were digested on the hot plate at 120–150 °C for approximately 40 min. The resulting digest was filtered through Whatman filtered paper No. 42. The filtrate was transferred to volumetric flask and the volume was adjusted to 50 mL with deionized water. A blank digest was carried out in the same way. 500 mg of rock-powdered samples were placed in a dry and clean Teflon digestion beaker and 2 mL of HNO_3 and 6 mL HCl were added. Samples were digested, filtered and diluted with deionized water.

3. Study areas

3.1. Ras Abu Aly

This locality lies at about 18 km to the north of Al Jubail city, at N 27° 10′ 00″ and E 49° 33′ 03″ (Fig. 1). The beach is sandy, with high diverse gastropod and bivalve shells belonging to genera of *Diodora*, *Lunella*, *Clanculus*, *Cerithium*, *Nassarius*, *Ancilla*, *Mitra*, *Conus*, *Potamides*, *Calliostoma*, *Barbatia*, *Glycymeris*, *Ctena*, *Divalinga*, *Pinctada*, *Acrosterigma*, *Mactra*, *Asaphis*, *Amiantis*, *Circe*, *Circenita*, *Gafrarium*, *Calista*, *Dosinia*, *Maria*. Field survey indicated that Ras Abu Aly beach is situated under human impacts in the form of crowded diving boats and their supplementary activities (Fig. 3A).

3.2. Al Jubail city

This locality lies at about 18 km to the south of Ras Abu Aly, at N 27° 01′ 46″ and E 49° 38′ 23″ (Fig. 1). The beach is made up artificial rocky blocks, which act as wave breaker to protect neighboring residential areas. This beach is characterized by low diversity of gastropods and bivalves, except some which hide among rocks and in crevices under rocks such as *Diodora*, *Lunella*, *Nerita*, *Cerithium*, *Cypraea*, *Barbatia*, *Brachidontes*, *Madiolus*, *Pinctada*, *Amiantis*, *Plicatula*, *Chlamys*, *Spondylus*, *Chama*, *Asaphis*, *Acrosterigma*. There are accumulations of black, very fine sediments, which may be polluted from nearby petrochemical industries (Fig. 3B).

Fig. 1. Location map of the study areas along the Arabian Gulf coast.

Table 1
Trace metals in seashells of the study areas.

Species	Study area	Fe	Cu	Zn	Pb	Mn	Cd	Se	As	Co	B	Cr	Hg	Mo
Diodora rueppellii	1	1248	0.14	0.15	0.15	0.13	0.003	0.006	0.2	0.1	0.4	0.03	0.02	0.006
	2	2734	0.45	0.41	0.6	0.18	0.006	0.04	17.4	0.3	1.1	0.44	0.07	0.03
	3	1495	0.18	0.17	0.16	0.2	0.0035	0.0075	0.55	0.15	0.9	0.04	0.035	0.007
	4	1712	0.22	0.2	0.18	0.08	0.004	0.009	0.9	0.2	0.7	0.05	0.05	0.008
	5	1503	0.17	0.165	0.145	0.08	0.003	0.0075	0.4	0.15	0.6	0.035	0.035	0.0065
Lunella coronata	1	1802	0.3	0.22	0.18	0.18	0.004	0.01	0.8	0.2	1	0.03	0.05	0.008
	2	2734	0.45	0.32	0.55	0.21	0.005	0.033	16.5	0.3	1.2	0.42	0.07	0.02
	3	1830	0.2	0.21	0.18	0.195	0.004	0.009	0.5	0.2	0.7	0.05	0.05	0.008
	4	1270	0.15	0.11	0.14	4.1	0.003	0.007	0.3	0.1	0.9	0.03	0.03	0.006
	5	2145	0.18	0.01	0.05	3.9	0.001	0.02	15.5	0.2	0.5	0.23	0.05	0.008
Cerithium caeruleum	1	1690	0.2	0.2	0.18	0.18	0.004	0.009	0.5	0.2	0.7	0.05	0.05	0.008
	2	1715	0.21	0.21	0.18	0.73	0.004	0.011	0.5	0.2	0.7	0.05	0.05	0.008
	3	1738	0.2	0.22	0.18	0.1	0.004	0.009	0.8	0.2	1.3	0.05	0.05	0.008
	4	1750	0.2	0.21	0.18	0.22	0.004	0.009	0.5	0.2	0.7	0.05	0.05	0.007
	5	1596	0.18	0.17	0.155	0.21	0.0035	0.008	0.4	0.15	0.8	0.035	0.04	0.0075
Barbatia parva	1	2145	0.17	0.01	0.05	0.18	0.001	0.02	17.6	0.2	0.5	0.23	0.05	0.008
	2	2230	0.9	1.6	0.75	0.185	0.021	0.06	12	0.2	11.4	0.25	0.21	0.06
	3	1695	0.2	0.2	0.18	0.22	0.004	0.009	0.9	0.2	1.2	0.05	0.05	0.008
	4	2132	0.8	1.1	0.71	0.2	0.011	0.05	10.1	0.2	10.2	0.31	0.15	0.02
	5	1750	0.2	0.21	0.16	0.18	0.004	0.009	0.6	0.2	0.7	0.05	0.05	0.008
Pinctada margaritifera	1	2155	0.19	0.01	0.05	1.6	0.001	0.02	17.7	0.2	0.5	0.22	0.05	0.009
	2	2161	0.18	0.09	0.08	0.13	0.003	0.02	18.2	0.2	0.5	0.24	0.05	0.009
	3	2367	0.42	0.09	0.24	0.18	0.008	0.03	17.7	0.2	3.4	0.25	0.06	0.01
	4	2367	0.42	0.09	0.24	0.17	0.002	0.03	12.7	0.2	3.6	0.25	0.06	0.01
	5	1910	0.2	0.21	0.17	0.2	0.004	0.009	0.5	0.2	1.2	0.04	0.05	0.009
Amiantis umbonella	1	1866	0.2	0.22	0.18	0.14	0.004	0.009	0.5	0.2	0.7	0.05	0.05	0.008
	2	1813	0.22	0.22	0.17	0.23	0.004	0.009	0.6	0.2	0.7	0.06	0.05	0.007
	3	1275	0.15	0.13	0.12	0.13	0.004	0.005	0.3	0.2	0.5	0.03	0.03	0.005
	4	1266	0.16	0.12	0.12	1.6	0.002	0.005	0.2	0.1	0.5	0.03	0.02	0.004
	5	1235	0.15	0.14	0.13	0.15	0.002	0.007	0.3	0.1	0.6	0.02	0.03	0.004
Acrosterigma assimile	1	1661	0.22	0.21	0.16	0.21	0.004	0.01	0.8	0.2	0.8	0.05	0.05	0.007
	2	1775	0.21	0.21	0.18	0.185	0.004	0.009	0.5	0.2	0.9	0.05	0.05	0.008
	3	1762	0.2	0.21	0.19	0.16	0.004	0.009	0.5	0.2	0.7	0.05	0.05	0.007
	4	1882	0.22	0.22	0.19	0.08	0.004	0.011	0.7	0.2	1	0.06	0.05	0.009
	5	1256	0.14	0.12	0.13	0.175	0.002	0.006	0.2	0.1	0.5	0.02	0.02	0.005
Asaphis violascens	1	1763.5	0.21	0.215	0.17	0.2	0.004	0.0095	0.65	0.2	0.75	0.05	0.05	0.0075
	2	1760	0.2	0.21	0.18	0.22	0.004	0.009	0.5	0.2	0.7	0.05	0.05	0.008
	3	1252	0.16	0.12	0.14	0.14	0.003	0.006	0.3	0.2	0.5	0.03	0.03	0.006
	4	1510	0.175	0.16	0.16	0.12	0.0035	0.008	0.4	0.15	0.8	0.04	0.04	0.0065
	5	1282	0.16	0.13	0.14	0.13	0.003	0.007	0.3	0.1	0.4	0.03	0.03	0.006
Min.		**1235**	**0.14**	**0.01**	**0.05**	**0.08**	**0.001**	**0.005**	**0.2**	**0.1**	**0.4**	**0.02**	**0.02**	**0.004**
Max.		**2734**	**0.9**	**1.6**	**0.75**	**4.1**	**0.021**	**0.06**	**18.2**	**0.3**	**11.4**	**0.44**	**0.21**	**0.06**
Average		**1787.808**	**0.247**	**0.231**	**0.205**	**0.440**	**0.004**	**0.014**	**4.263**	**0.185**	**1.386**	**0.103**	**0.052**	**0.010**

1, Ras Abu Aly; 2, Al Jubail city; 3, Al Jubail Industrial city; 4, Ras Al-Ghar; 5, Ras Tanura.

3.3. Al Jubail industrial city

This locality lies at about 9 km to the south of Al Jubail city, at N 26° 58′ 37″ and E 49° 40′ 58″ (Fig. 1). The beach is sandy, with intertidal flats. It has numerous gastropods and bivalves such as *Diodora*, *Cerithium*, *Lunella*, *Clanculus*, *Nassarius*, *Ancilla*, *Mitra*, *Conus*, *Potamides*, *Calliostoma*, *Glycymeris*, *Barbatia*, *Ctena*, *Divalinga*, *Cardites*, *Acrosterigma*, *Mactra*, *Asaphis*, *Hiatula*, *Circe*, *Circenita*, *Pinctada*, *Gafrarium*, *Calista*, *Dosinia*, *Maria*, *Amiantis*. Field survey illustrated that Ras Abu Aly beach is situated under human impacts, in the form of crowded factories and desalination units (Figs. 3C and 4A).

3.4. Ras Al-Ghar

This locality lies at about 16 km to the south of Al Jubail city, at N 26° 56′ 30″ and E 49° 43′ 45″ (Fig. 1). The beach is sandy, with high diverse gastropods and bivalves such as *Diodora*, *Lunella*, *Clanculus*, *Nassarius*, *Ancilla*, *Mitra*, *Cerithium*, *Conus*, *Potamides*, *Glycymeris*, *Ctena*, *Barbatia*, *Cardites*, *Acrosterigma*, *Mactra*, *Asaphis*, *Pinctada*, *Circe*, *Circenita*, *Amiantis*, *Gafrarium*, *Calista*, *Dosinia*, *Maria*. Ras Al-Ghar beach is clear from human impacts as it is a military port and closed for public (Fig. 4B).

3.5. Ras Tanura

This locality lies at about 43 km to the south of Ras Al-Ghar, at N 26° 40′ 13′′ & E 50° 02′ 17″ (Fig. 1). The beach is sandy, with low diverse gastropods and bivalves such as *Diodora*, *Lunella*, *Clanculus*, *Nassarius*, *Ancilla*, *Mitra*, *Conus*, *Potamides and Calliostoma*, *Glycymeris*, *Ctena*, *Divalinga*, *Cardites*, *Acrosterigma*, *Mactra*, *Asaphis*, *Hiatula*, *Circe*, *Circenita*, *Gafrarium*, *Calista*, *Dosinia*, *Maria*. Field survey indicated that Ras Tanura beach is clear due to the absence of human impact with the exception of some touristic activities (Fig. 4C).

4. Results and discussion

4.1. Heavy metals concentrations

Table 1 illustrates the concentrations of B, Cr, Fe, Mn, Cu, Zn, Mo, As, Pb, Cr, Hg, Sr, Co, Ni in three gastropod and five bivalve species. The average metal levels are in the following order: Fe > As > B > Mn > Cu > Zn > Pb > Co > Cr > Hg > Se > Mo > Cd. Table 2 shows the correlation matrix among trace metals. The following is a brief description of the concentration of these metals in comparison with other gastropods and bivalves in other

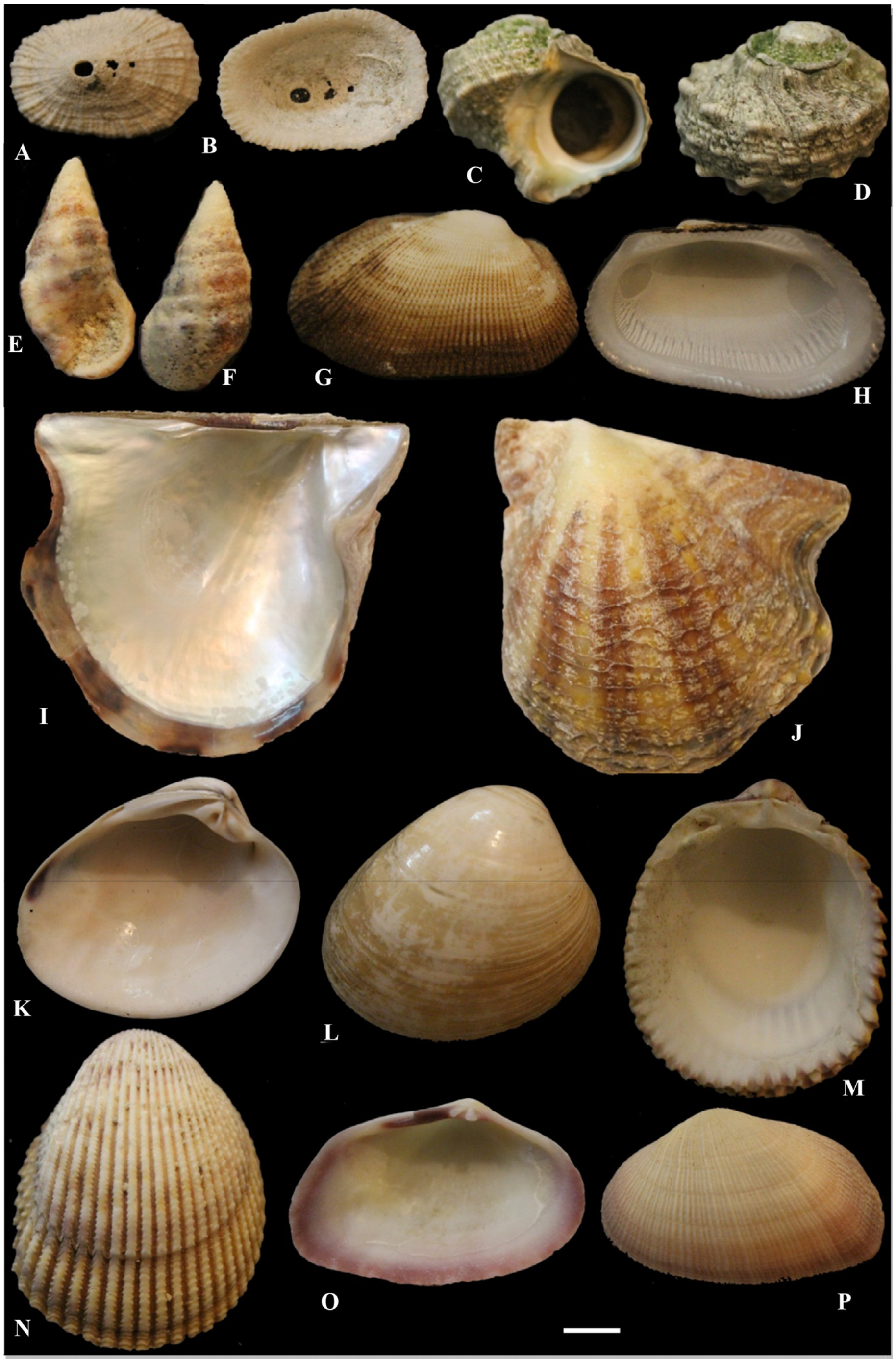

Fig. 2. The examined gastropods and bivalves. A, B, *Diodora rueppellii* (Sowerby, 1834); C, D, *Lunella coronata* (Gmelin, 1791); E, F *Cerithium caeruleum* (Sowerby, 1855); G, H, *Barbatia parva* (Sowerby, 1833); I, J, *Pinctada margaritifera* (Linnaeus, 1758); K, L, *Amiantis umbonella* (Lamarck, 1818); M, N, *Acrosterigma assimile* (Reeve, 1845); O, P, *Asaphis violascens* (Forsskål, 1775).

localities, such as Indian Ocean, Red Sea, Arabian Gulf and Gulf of Oman (Table 3).

4.1.1. Iron

Iron is an essential element in the marine ecosystem, where it plays the most vital role in the biogenic activities (El-Sorogy et al., 2013). Fe values ranged from 1235 to 2734 μg/g, with an average of 1787.8 μg/g (Table 1). The lowest value of Fe is recorded in *A. umbonella* of Ras Tanura area and the highest ones are recorded in *D. rueppellii* and *L. coronata* of Al-Jubail city (Fig. 5). *C. caeruleum* and *A. assimile* have similar Fe uptake along the study areas.

The present Fe values are higher than those recorded from the following molluscans (Table 3): *Nerita lineata* (9.27–53.58 μg/g) from Indonesia and Malaysia (Amin et al., 2006); *Cerithidea cingulate*, *Crassostrea madrasensis* and *Meretrix meretrix* (2.16–18.35 μg/g) from India (Kesavan et al., 2013); *Patella aspera* (35.8 ppm) as estimated by Cravo et al. (2002); *Corbicula fluminalis* (90–106 ppm) from Shatt Al-Arab River (Al-Jaberi, 2014); *Pinctada radiate*, *Saccostrea cucullata*, *Circentia callipyga*, *Pinna muricata and Spondylus*

Fig. 3. Environmental settings at the study areas. A, crowded fishing boats in the intertidal zone of Ras Abu Aly area; B, highly polluted beach at Al Jubail city; C, the beach of Al Jubail industrial city.

sp. (89.9–517 μg/g) in the Gulf of Oman (Mora et al., 2004) and *Tridacna maxima* (216.4–1286.4 ppm) from Red Sea coast (Madkour, 2005). The correlation matrix indicates that Fe is positively correlated with all studied elements except Mn (Table 2).

4.1.2. Copper

The dissolution of Cu from sediments to water depends on the pH and salinity, while adsorption of Cu from sediments is very low compared with other metals (Forstner and Wittman, 1983). In the studied seashells, Cu values ranged from 0.14 to 0.90 μg/g with an average of 0.25 μg/g (Table 1). The lowest values are recorded in *D. rueppellii* of Ras Abu Aly and *A. assimile* of Ras Tanura and the highest value is recorded in *B. parva* at Al Jubail city (Fig. 6). *C. caeruleum*, *A. assimile* and *A. violascens* have similar Cu uptake along the study areas.

The present Cu values are situated within the values recorded in *Mytilus edulis* (0.10–1.30 μg/g) at Belledune Harbor (Bourgoin, 1990) and within those recorded in *Perna viridis* and *Modiolus metcalfei* (0.13–0.36 μg/g) from southeast coast of India (Ponnusamy et al., 2014). They are correlated to those recorded in *C. cingulate*, *C. madrasensis* and *M. meretrix* (0.10–0.84 μg/g) from India (Kesavan et al., 2013) and correlated to the values recorded *Rapana rapiformis*, *Chicoreus virgineus*, and *Hemifusus pugilinus* (0.21–0.98 μg/g) in Pondicherry, India (Kaviarasan et al., 2012).

Present Cu values are less than those reported from the following molluscs: *Helix aspera* (1.1–3.7 ppm) as indicated by Laskowski and Hopkin (1996); *Mytilus galloprovincialis* and *Ostrea edulis* (0.010–0.040 μg/g) from North Adriatic Sea (Martincié et al., 1984); *Nerita lineata* (2.80–4.40 μg/g) from the intertidal zone between Indonesia and Malaysia (Amin et al., 2006); Spanish bivalves (20 μg/g) as quoted by McLennan and Taylor (1999); *C. fluminalis* (242–326 ppm) from Shatt Al-Arab River (Al-Jaberi, 2014); *P. radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (3.13–276 μg/g) in the Gulf and Gulf of Oman (Mora et al., 2004) and *T. maxima* (12.4–81.4 ppm) from Red Sea coast (Madkour, 2005).

4.1.3. Zinc

Zinc is necessary for normal cell division and growth in both plants and animals but can be unsafe if exist in excessive amounts. Zn values varied from 0.01 to 1.6 μg/g with an average of 0.23 μg/g (Table 1). The lowest concentrations are recorded in *L. coronata*, *B. parva* and *P. margaritifera* at Ras Abu Aly. The highest value is

Fig. 4. Environmental settings at the study areas. A, different factories and desalination units, Al Jubail industrial city coast; B, clean beach at Ras Al-Ghar coast; C, Clean beach at Ras Tanura coast.

recorded in *B. parva* at Al Jubail city (Fig. 7). As in Cu, *C. caeruleum*, *A. assimile* and *A. violascens* nearly have similar Zn uptake along the study areas.

Zn values recorded in three gastropod species *R. rapiformis*, *C. virgineus*, and *H. pugilinus* (0.57–0.79 μg/g) from India (Kaviarasan et al., 2012) are located within the range of the present Zn values. They are less than those recorded from the following molluscs: *P. aspera* (5.5 ppm) by Cravo et al. (2002); *P. viridis* and *M. metcalfei*

Table 2
Correlation matrix of trace metals along the study coasts.

	Fe	Cu	Zn	Pb	Mn	Cd	Se	As	Co	B	Cr	Hg	Mo
Fe	1												
Cu	0.595[a]	1											
Zn	0.290	0.886[a]	1										
Pb	0.542[a]	0.922[a]	0.863[a]	1									
Mn	−0.054	−0.143	−0.178	−0.206	1								
Cd	0.333[b]	0.870[a]	0.942[a]	0.818[a]	−0.208	1							
Se	0.755[a]	0.933[a]	0.745[a]	0.845[a]	−0.024	0.738[a]	1						
As	0.812[a]	0.476[a]	0.155	0.349[b]	0.166	0.199	0.743[a]	1					
Co	0.802[a]	0.394[b]	0.205	0.446[a]	−0.223	0.248	0.471[a]	0.477[a]	1				
B	0.364[b]	0.935[a]	0.911[a]	0.797[a]	−0.095	0.880[a]	0.829[a]	0.325[b]	0.142	1			
Cr	0.880[a]	0.656[a]	0.347[b]	0.635[a]	0.066	0.344[b]	0.857[a]	0.919[a]	0.615[a]	0.445[a]	1		
Hg	0.542[a]	0.937[a]	0.938[a]	0.835[a]	−0.117	0.913[a]	0.872[a]	0.407[a]	0.390[b]	0.929[a]	0.538[a]	1	
Mo	0.536[a]	0.856[a]	0.874[a]	0.845[a]	−0.094	0.884[a]	0.849[a]	0.444[a]	0.387[b]	0.777[a]	0.587[a]	0.893[a]	1

[a] Correlation is significant at the 0.01 level (2-tailed).
[b] Correlation is significant at the 0.05 level (2-tailed).

Table 3
Comparison between heavy metals in the present fauna and other worldwide localities.

References	Locality	Fauna	Fe	Cu	Zn	Pb	Mn	Cd	Se	As	Co	Cr
This study	Central Arabian Gulf	See Materials and methods	1235–2734	0.14–0.90	0.01–1.6	0.05–0.75	0.08–4.1	0.001–0.021	0.005–0.05	0.02–18.2	0.1–0.3	0.02–0.44
Amin et al. (2006)	Indonesia and Malaysia	*Nerita lineata*	9.27–53.58	2.80–4.40	3.38–3.94	38.55–53.35	–	3.74–43.49	–	–	–	–
Kesavan et al. (2013)	India	*Cerithidea cingulate, Crassostrea madrasensis, Meretrix meretrix*	2.16–18.35	0.10–0.84	0.29–3.64	0.03–0.08	0.34–6.19	0.002–0.09	–	–	0.003–0.07	–
Cravo et al. (2002)	South coast of Portugal	*Patella aspera*	35.8	–	5.5	–	29.9	–	–	–	–	–
Al-Jaberi (2014)	Shatt Al-Arab River	*Corbicula fluminalis*	90–106	242–326	110–122	15–22	22–33	–	–	–	4–15	7–14
Mora et al. (2004)	Gulf and Gulf of Oman	*Pinctada radiate, Saccostrea cucullata, Circentia callipyga, Pinna muricata, Spondylus sp.*	89.9–517	3.13–276	69.1–1830	0.089–3.92	2.77–1110	1.17–19.9	2.07–12.8	11.1–156	0.12–12.9	0.01–3.76
Madkour (2005)	Red Sea coast	*Tridacna maxima*	216.4–1286.	12.4–81.4	11.4–32.6	12.3–44.7	29.9–65.2	1.32–1.82	–	–	–	–
Bourgoin (1990)	Belledune Harbor	*Mytilus edulis*	–	0.10–1.30	–	0.19–1.00	–	0.3–49.1	–	–	–	–
Ponnusamy et al. (2014)	South east coast of India	*Perna viridis, Modiolus metcalfei*	–	0.13–0.36	0.95–3.63	0.18–0.36	–	0.014–0.038	–	–	–	0.09–0.15
Kaviarasan et al. (2012)	Pondicherry, India	*Rapana rapiformis, Chicoreus virgineus, Hemifusus pugilinus*	–	0.21–0.98	0.57–0.79	–	–	0.13–0.24	–	–	–	–
Laskowski and Hopkin., (1996)	Experiment in lab.	*Helix aspera*	–	1.1–3.7	2.8–28.0	5.7–27.7	–	0.002–0.127	–	–	–	–
Martincié et al. (1984)	North Adriatic Sea	*Mytilus galloprovincialis, Ostrea edulis*	–	0.010–0.04	1.70–10.0	0.48–1.17	–	0.010–0.040	–	–	–	–
Pérez-López et al. (2003)	Northwest Spain	*Patella vulgate*	–	–	–	3.0–7.6	–	0.04–0.22	–	–	–	–
Davies et al. (2006)	Niger Delta	*Tympanotonus fuscatus var radula*	–	–	–	0.004–0.016	–	0.001–0.006	–	–	–	0.001–0.006
Lotfy (2006)	Egypt (Nile branch)	*Anodonta implicate, Bellamiya unicolor*	0.78–6	0.12–7.71	0.94–4.94	0.12–3.05	0.98–2.28	0.01–0.078	0.08–4.67	0.16–1.33	0.012–0,10	0.01–0.56

(0.95–3.63 μg/g) from India (Ponnusamy et al., 2014); *M. galloprovincialis* and *Ostrea edulis* (1.70–10.00 μg/g) from North Adriatic Sea (Martincié et al., 1984); *Nerita lineata* (3.38–3.94 μg/g) from Indonesia and Malaysia (Amin et al., 2006); *C. cingulate*, *C. madrasensis* and *M. meretrix* (0.29–3.64 μg/g) from India (Kesavan et al., 2013); *C. fluminalis* (110–122 ppm) from Shatt Al-Arab River (Al-Jaberi, 2014); *T. maxima* (11.4–32.6 ppm) from Red Sea coast (Madkour, 2005); *Pinctada radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (69.1–1830 μg/g) from the Gulf of Oman (Mora et al., 2004) and *Patella vulgate* (3.0–7.6 ppm) from Northwest Spain (Pérez-López et al., 2003).

4.1.4. Lead

The main sources of Pb in the marine environment are from storm water runoff from hinterland and sewage input (Peters et al., 1997; El-Sorogy et al., 2012). Sorption of lead by sediment is correlated to the organic content, the grain size and the anthropogenic pollution (Huang and Lin, 2003; Muniz et al., 2004). Lead varies in the examined shells between 0.05 and 0.75 μg/g, with an average of 0.20 μg/g (Table 1). The lowest limits of Pb content are recorded in *L. coronata* (Ras Tanura) and *P. margaritifera* (Ras Abu Aly). The upper limit is recorded in *B. parva* at Al-Jubail city (Fig. 8). *C. caeruleum*, *A. assimile*, *A. violascens* and somewhat *A. umbonella* nearly have similar Pb uptake along the study areas.

These Pb values are somewhat correlated to those recorded in *C. cingulate*, *C. madrasensis* and *M. meretrix* (0.03–0.08 μg/g) from southeast coast of India (Kesavan et al., 2013) and situated within the values recorded in the following molluscs: *M. edulis* (0.19–1.00 μg/g) at Belledune Harbor (Bourgoin, 1990); *Tympanotonus fuscatus var radula* (0.004–0.016 μg/g) from Niger Delta (Davies et al., 2006); *M. galloprovincialis* and *Ostrea edulis* (0.48–1.17 μg/g) from North Adriatic Sea (Martincié et al., 1984) and *P. viridis* and *M. metcalfei* (0.18–0.36 μg/g) from India (Ponnusamy et al., 2014).

The present Pb values are less than those recorded from the following molluscs: *N. lineata* (38.55–53.35 μg/g) from Indonesia and Malaysia (Amin et al., 2006); *C. fluminalis* (15–22 ppm) from Shatt Al-Arab River (Al-Jaberi, 2014); *Pinctada radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (0.089–3.92 μg/g) from the Gulf of Oman (Mora et al., 2004) and *T. maxima* (12.3–44.7 ppm) from Red Sea coast (Madkour, 2005).

4.1.5. Manganese

Along the coastal areas, Mn is controlled mainly by dissolved and particulate Mn derived from the shelf sediments (Fallon et al., 2002). Mn has been hypothesized to substitute for Ca in the $CaCO_3$ lattice, but may also be adsorbed or occluded within aragonite as an oxide or in some aragonite phase.

Mn values ranged from 0.08 to 4.1 μg/g with average of 0.44 μg/g (Table 1). The minimum concentrations of Mn are recorded in *D. rueppellii* (Ras Al-Ghar and Ras Tanura) and *Acrosterigma assimile* (Ras Al-Ghar). The maximum concentration is recorded in *L. coronata* at Ras Al-Ghar (Fig. 9). *D. rueppellii*, *B. parva*, *A. assimile* and *A. violascens* nearly have similar Mn uptake along the study areas.

These Mn values are somewhat correlated to those recorded in *Cerithidea cingulate*, *C. madrasensis* and *M. meretrix* (0.34–6.19 μg/g) from India (Kesavan et al., 2013) and are much less than those recorded in *P. aspera* (29.9 ppm) by Cravo et al., 2002; *C. fluminalis* (22–33 ppm) from Shatt Al-Arab River (Al-Jaberi, 2014); *Pinctada radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (2.77–1110 μg/g) from the Gulf of Oman (Mora et al., 2004) and *T. maxima* (29.9–65.2 ppm) from the Red Sea coast (Madkour, 2005).

The correlation matrix indicates that Mn is negatively correlated with all studied elements except As and Cr (Table 2). Thermal desalination plants can discharge a variety of heavy metals,

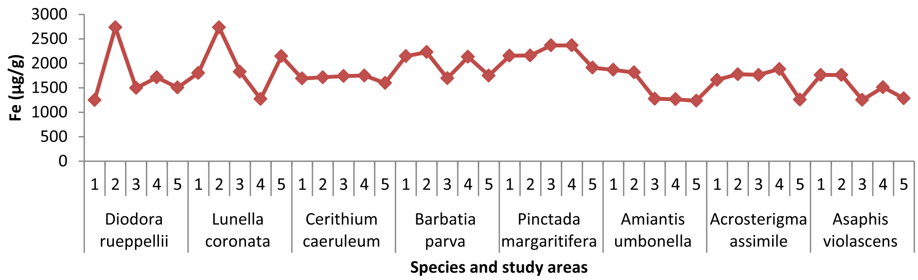

Fig. 5. Fe concentrations in molluscan species of the study areas (1, Ras Abu Aly; 2, Al Jubail city; 3, Al Jubail Industrial city; 4, Ras Al-Ghar; 5, Ras Tanura).

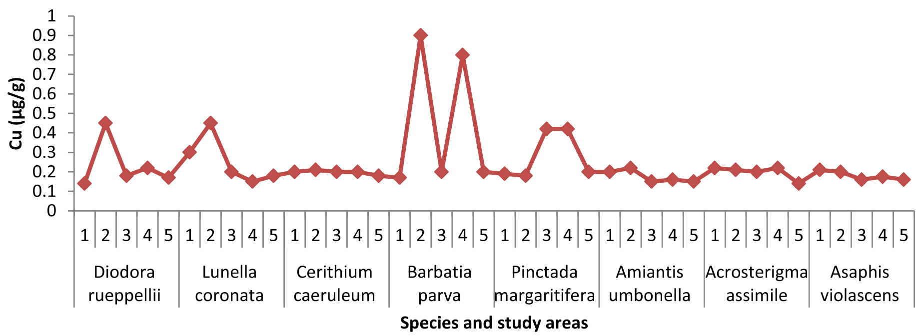

Fig. 6. Cu concentrations in molluscan species of the study areas.

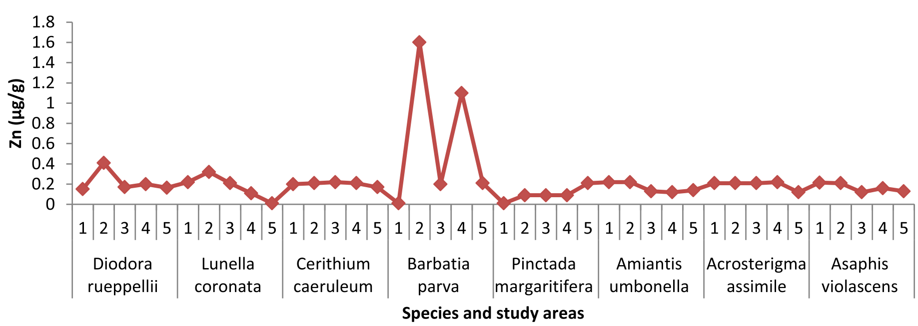

Fig. 7. Zn concentration in molluscan species of the study areas.

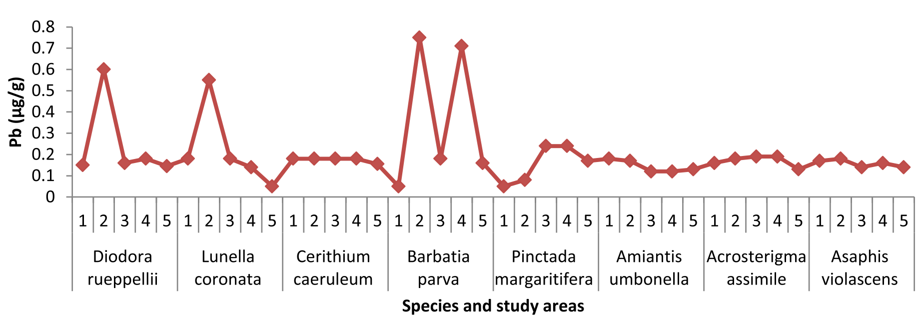

Fig. 8. Spatial distribution of Pb in molluscan species of the study areas.

including Mn, depending on the alloys present in the process line, which may affect water quality (Hoepner, 1999; Lattemann and Hopner, 2008). It can be transported to the marine environment across landfill, marine paintings, construction residuals, and corrosion of steel constructions and pipelines (Fallon et al., 2002; El-Sorogy et al., 2013a).

4.1.6. Cadmium

Cadmium can move between air and water and once reaches the water; it will find its way into sediment (Tessier et al., 1980). Cd concentrations in seashells showed generally low values. Cd content in the studied samples varied from 0.001 to 0.021 µg/g, with an average of 0.004 µg/g (Table 1). The minimum value of Cd is recorded in *B. parva* at Ras Abu Aly and the maximum one is recorded in *B. parva* at Al Jubail city (Fig. 10). *L. coronata*, *C. caeruleum*, *A. umbonella*, *A. assimile* and *A. violascens* nearly have similar Cd uptake along the study areas.

These Cd values are somewhat similar to those recorded in *C.*

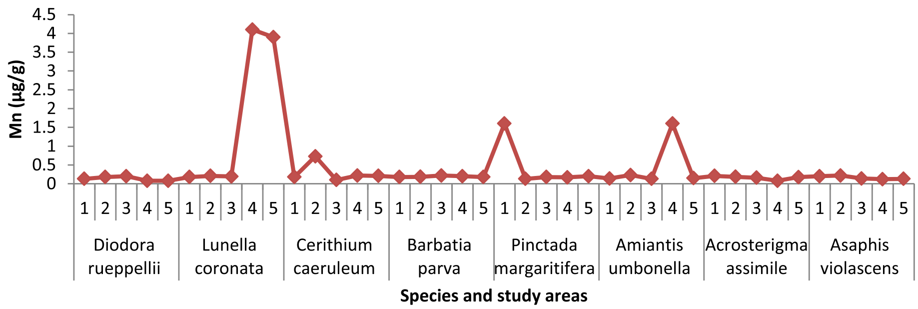

Fig. 9. Concentrations of Mn in molluscan species of the study areas.

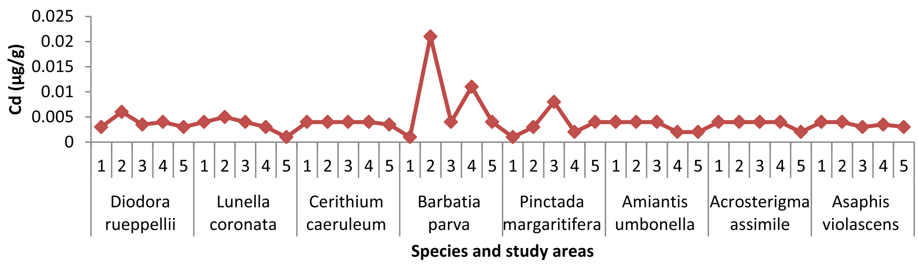

Fig. 10. Concentrations of Cd in molluscan species of the study areas.

cingulate, *C. madrasensis* and *M. meretrix* (0.002–0.09 µg/g) from India (Kesavan et al., 2013). They are situated within Cd values recorded in *T. fuscatus var radula* (0.001–0.006) from Niger Delta (Davies et al., 2006) and are less than those recorded in *P. viridis* and *M. metcalfei* (0.014–0.038 µg/g) from India (Ponnusamy et al., 2014); *M. galloprovincialis* and *Ostrea edulis* (0.010–0.040 µg/g) from North Adriatic Sea (Martincié et al., 1984); *R. rapiformis*, *C. virgineus*, and *H. pugilinus* (0.13–0.24 µg/g) in India (Kaviarasan et al., 2012); *T. maxima* (1.32–1.82 ppm) from the Red Sea coast (Madkour, 2005); *P. vulgate* (0.04–0.22 ppm) from Northwest Spain (Pérez-López et al., 2003) and *M. edulis* (0.3–49.1 µg/g) at Belledune Harbor (Bourgoin, 1990). Higher values of Cd were reported in *N. lineata* (3.74–43.49 µg/g) from Indonesia and Malaysia (Amin et al., 2006) and in *P. radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (1.17–19.9 µg/g) in the Gulf and Gulf of Oman (Mora et al., 2004).

4.1.7. Selenium

Se is commonly found in marine sedimentary deposits (Al-Taani et al., 2012). Se values range from 0.005 to 0.05 µg/g with an average 0.01 µg/g (Table 1). The lowest Se value is recorded in *B. parva* at Ras Al-Ghar and the highest one is recorded in *B. parva* at Al Jubail industrial city (Fig. 11). *C. caeruleum*, *A. umbonella*, *A. assimile* and *A. violascens* have the same spatial distribution of Se all over the study areas. These Se values are less than those recorded in *P. radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (2.07–12.8 µg/g) in the Gulf of Oman (Mora et al., 2004).

4.1.8. Arsenic

The values of As content in the studied seashells varied from 0.02 to 18.2 µg/g with an average of 4.14 µg/g (Table 1). Low As values are recorded in *D. rueppellii* (Ras Abu Aly), *A. umbonella* (Ras Al-Ghar), *A. assimile* (Ras Tanura) and the high As concentration is reported in *P. margaritifera* at Al Jubail city (Fig. 12). *C. caeruleum*, *A. umbonella*, *A. assimile* and *A. violascens* have the same spatial distribution and the minimum uptake of As all over the study areas. These As values are less than those recorded in *P. radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (11.1–156 µg/g) in the Gulf of Oman (Mora et al., 2004).

4.1.9. Cobalt

Co levels varied from 0.1 to 0.3 µg/g with an average of 0.19 µg/g (Table 1). Low Co values are recorded in *D. rueppellii* (Ras Abu Aly), *L. coronata* (Ras Al-Ghar) *A. umbonella* (Ras Tanura), *A. assimile* (Ras Tanura), *A. violascens* (Ras Tanura) and the high Co concentrations are reported in *D. rueppellii*, *L. coronata* at Al Jubail city (Fig. 13). *B. parva* and *P. margaritifera* have the same spatial distribution and uptake of Co all over the study areas.

These Co values are somewhat correlated to those recorded in *C. cingulate*, *C. madrasensis* and *M. meretrix* (0.003–0.07 µg/g) from southeast coast of India (Kesavan et al., 2013). They are less than those reported in the bivalve *C. fluminalis* (4–15 ppm) from Shatt Al-Arab River (Al-Jaberi, 2014) and those recorded in *P. radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (0.12–12.9 µg/g) in the Gulf of Oman (Mora et al., 2004).

4.1.10. Boron

Boron is a common element in marine water with a typical concentration of 4.5 mg/L (Moss, 1981) which may vary depending on the location and seasonal effects. Patterns of B levels along the studied seashells varied from 0.4 to 11.4 µg/g with an average of 1.38 µg/g (Table 1). Low B values are recorded in *D. rueppellii* (Ras Abu Aly), *A. violascens* (Ras Tanura) and the high Mo concentrations are reported in *B. parva* at Al Jubail city (Fig. 14). *D. rueppellii*, *L. coronata*, *C. caeruleum*, *A. umbonella*, *A. assimile* and *A. violascens* have the same spatial distribution and uptake of B all over the study areas.

4.1.11. Chromium

Cr levels varied from 0.02 to 0.44 µg/g with an average of 0.10 µg/g in molluscan samples (Table 1). Low Cr value is recorded in *A. umbonella*, *A. assimile* at Ras Tanura and the high Cr concentration is reported in *D. rueppellii* at Al Jubail city (Fig. 15). *A. umbonella*, *A. assimile* and *A. violascens* have the same spatial distribution and uptake of Cr all over the study areas.

The present values are higher than those in *Tympanotonus*

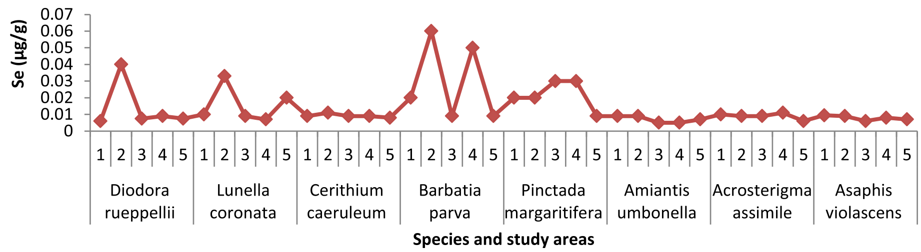

Fig. 11. Se concentrations in molluscan species of the study areas.

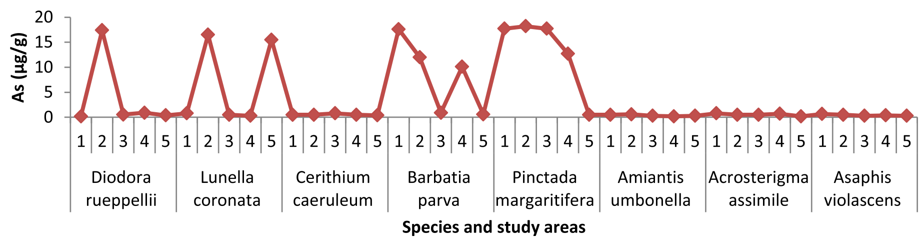

Fig. 12. Arsenic levels in molluscan species of the study areas.

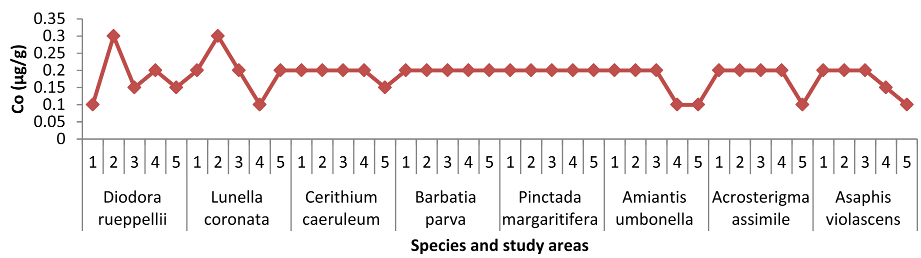

Fig. 13. Co levels in molluscan species of the study areas.

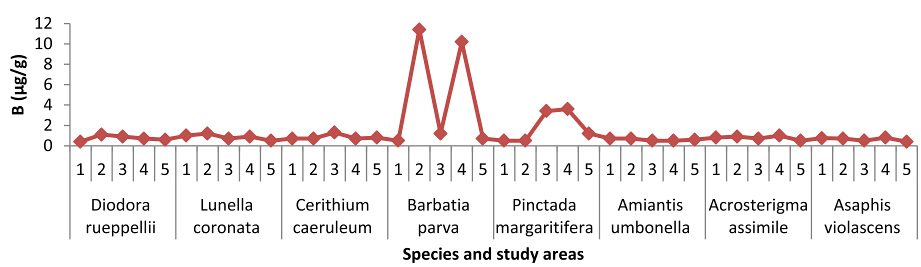

Fig. 14. Boron concentrations in molluscan species of the study areas.

fuscatus var radula (0.001–0.006) from the Elechi Creek, Niger Delta (Davies et al., 2006) and less than those reported in *C. fluminalis* (7–14 ppm) from Shatt Al-Arab River (Al-Jaberi, 2014) and those recorded in *P. radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (0.01–3.76 µg/g) in the Gulf of Oman (Mora et al., 2004). The values recorded in *P. viridis* and *M. metcalfei* (0.09–0.15 µg/g) from Vellar Estuary, south east coast of India (Ponnusamy et al., 2014) are situated within present values.

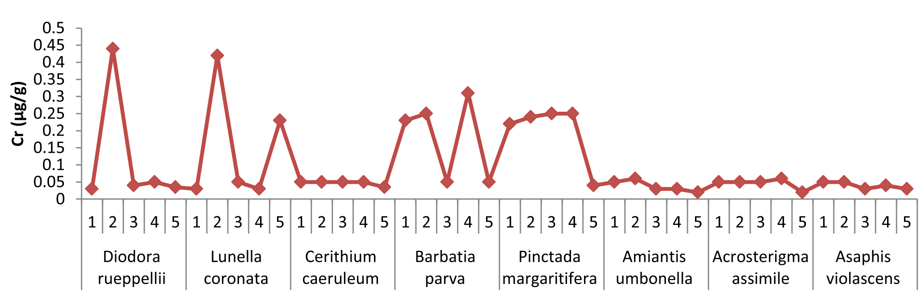

Fig. 15. Cr levels in molluscan species of the study areas.

4.1.12. Mercury

Low Hg contents were found with a relatively stable pattern across the studied samples with slightly increase in ones. Hg levels varied from 0.02 to 0.21 μg/g with an average of 0.01 μg/g (Table 1). The Lowest Hg values are recorded in *D. rueppellii* (Ras Abu Aly), *A. umbonella* (Ras Al-Ghar), *A. assimile* (Ras Tanura) and the high Hg concentration is reported in *B. parva* at Al Jubail city (Fig. 16). *C. caeruleum*, *P. margaritifera*, *A. umbonella*, *A. assimile* and *A. violascens* have the same spatial distribution and uptake of Hg all over the study areas. These Hg values are less than those recorded in *P. radiate*, *S. cucullata*, *C. callipyga*, *P. muricata and Spondylus* sp. (0.0087–0.315 μg/g) in the Gulf and Gulf of Oman (Mora et al., 2004).

4.1.13. Molybdenum

Molybdenum is an important element that occurs as sulfides (e.g., MoS_2) and is commonly associated with Cu sulfides (El-Sorogy et al., 2013a). It ranged in concentration between 0.004 and 0.06 μg/g with an average of 0.01 μg/g in the studied samples (Table 1). The Lowest Mo values are recorded in *A. umbonella* at Ras Tanura and the high concentration is reported in *B. parva* at Al Jubail city (Fig. 17). Like Hg, *C. caeruleum*, *P. margaritifera*, *A. umbonella*, *A. assimile* and *A. violascens* have the same spatial distribution and uptake of Mo all over the study areas.

4.2. Sources of pollutants

Recently, many localities along the Saudi Arabian coast have been subjected to deterioration as a result of land reclamation, urbanization and dredging. Dredging and reclamation processes are typically associated with elevated levels of heavy metals which are mobilized during dredging and reclamation activities (Hedge et al., 2009; El-Sorogy et al. 2013a; Guerra et al., 2009). One of the expected sources of the metals concentration in the studied areas along Arabian Gulf coast and the eastern Saudi Arabia in general, is the dust storms, which repeat several times a year and contain the erosion products of the surrounding rocks.

El-Sorogy et al. (2015) recorded high Fe values in sediment in comparison with seawater (about 504 times) along Tarut coast, to the south of the study area and suggested that Fe-born dust conveyed to the Arabian Gulf from the surrounding deserts is a potential source of elevated Fe levels. Also, Al-Jubail industrial city is highly populated with intense petrochemical industrial activities and desalinization plants (Figs. 3 and 4). Discharge of industrial effluents, desalinization plants, and mineral dust from fertilizer and cement factories are potential contributors to the land-based sources of pollution affecting coastal waters in the study area.

Hoepner and Lattemann (2002) indicated that Cu is not the only corrosion product discharge from desalination plants but also Ni, Cr, Mo and Fe among others. Possible sources of pollution by Cu, as well as other heavy metals, are renewing the old ships, removing rust, painting the ship bodies and the presence of shipyards (El-Sorogy et al., 2012, 2013a). These sources are available in the study areas, especially at Al-Jubail industrial city.

On his study on assessment and management of heavy metal pollution in the marine environment of the Arabian Gulf, Naser (2013) attributed the contamination of marine environment with Zn, Cu, Cr, and Pb to boat exhaust emissions, antifouling chemicals used in boat paints, sewage effluents, chemical and oil leakage, and discharges of reject water and effluents from a desalination plant. In the study area, dust storms, intense industrial activities, desalinization plants, municipal sewages and chemicals from the northward may be a potential source of Zn in seawater and sediments.

Possible sources of pollution by Pb and Co along the study areas are the emissions from industrial chimneys of petrochemicals, chemical, petrochemical industries and landfilling. Also it has been reported that heavy metals such as Cd, Pb, Cr, and As are the most abundant metals found in soils around chemical and petrochemical sites (Kabir et al., 2012). All these sources are available in the study area (Figs. 3 and 4).

5. Conclusions

1. Analysis of B, Fe, Mn, Cu, Zn, Mo, As, Pb, Cd, Hg, Sr, Co and Se in *D. rueppellii*, *L. coronata*, *C. caeruleum*, *B. parva*, *P. margaritifera*, *A. umbonella*, *A. assimile* and *A. violascens* in Abu Aly, Al Jubail, Al Jubail Industrial city, Ras Al-Ghar and Ras Tanura areas along central Arabian Gulf indicated that all elements (except Fe) yielded concentrations less than those recorded in other molluscan species from the Red Sea, Indian Ocean, Arabia Gulf

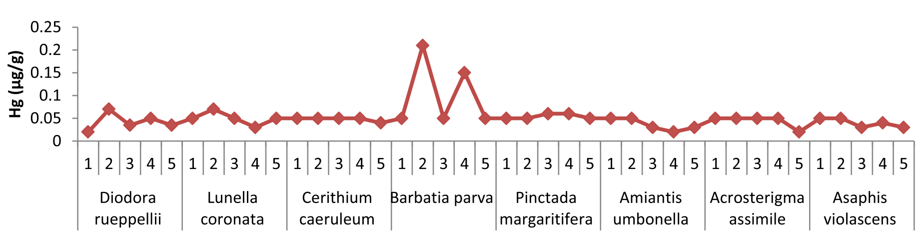

Fig. 16. Spatial distribution of Hg in molluscan species of the study areas.

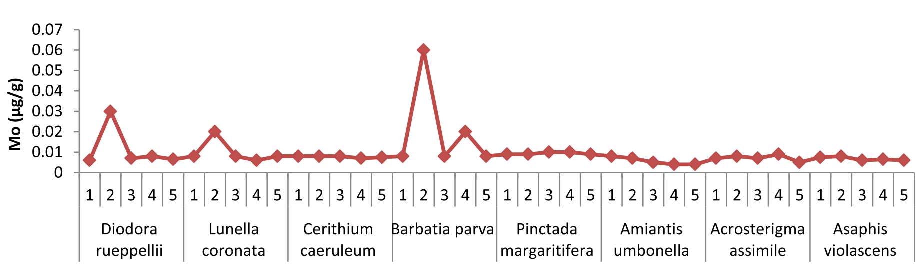

Fig. 17. Mo levels in molluscan species of the study areas.

and Gulf of Oman. The average metal levels were in the following order: Fe > As > B > Mn > Cu > Zn > Pb > Co > Cr > Hg > Se > Mo > Cd.

2. Certain species gave similar spatial distribution and uptake for some elements all over the study areas and therefore consider independent species for such elements. Ex. *Cerithium caeruleum* and *A. assimile* (Fe); *C. caeruleum*, *A. assimile* and *A. violascens* (Cu, Zn); *C. caeruleum*, *A. assimile* and *A. violascens*, *A. umbonella* (Pb, Se, As); *D. rueppellii*, *B. parva*, *A. assimile* and *A. violascens* (Mn); *L. coronata*, *C. caeruleum*, *A. umbonella*, *A. assimile* and *A. violascens* (Cd); *C. caeruleum*, *P. margaritifera*, *A. umbonella*, *A. assimile* and *A. violascens* (Hg, Mo).
3. Certain species consider good accumulators for certain elements. Ex. *Diodora rueppellii* (Fe, Cu, Pb, Mo, Se, As, Co, Cr), *L. coronata* (Fe, Cu, Pb, Mo, Mn, As, Co, Cr), *B. parva* (Cu, Pb, Mo, Cd, Se, AS, B, Cr, Hg), *P. margaritifera* (Cu, Mn, As, Cr).
4. Al Jubail coast recorded the highest concentrations of all the studied elements (except Mn at Ras Al-Ghar and Se at Al Jubail industrial city). Industrial chimneys of petrochemicals industries, boat exhaust emissions, antifouling chemicals used in boat paints, oil leakage, effluents from desalination plants and sewage effluents are the main anthropogenic sources for metal pollutants in the study area in general and at Al-Jubail city in particular.

Acknowledgment

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