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Distribution of mercury in molluscs, seawaters and coastal sediments of Tarut Island, Arabian Gulf, Saudi Arabia

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A B S T R A C T

In order to assess the distribution of mercury along the Tarut coast, Arabian Gulf, Thirty eight (38) sediment samples, twenty six (26) seawater samples, and forty (40) Mollusca specimens were collected from the Tarut coast. The concentrations of Mercury in the sediments of the studied area (average = 0.55 μg/g) are generally high comparing to the reported values from the Gulf of Oman, Red Sea, and the Gulf of Finland. The concentrations of Hg exceeded the wet threshold safety values (median effect concentration (MEC), and probable effect concentration (PEC) indicating possible Hg contamination. According to the Swedish Environmental Protection Agency (SEPA), thirty four (34) samples occur in class 4 and four (4) samples occur in class 5, which means that the sediments of the Tarut Island are largely contaminated with Hg. Enrichment factor (EF) results (average = 1.76) suggested that, the coastal sediments of the Tarut Island are considered to entirely originate from the crustal materials or natural processes. The studied sediments show lower values (Igeo<0) indicating that the sediments are unpolluted. These sediments according to contamination factor (Cf) are considered contaminated with Hg ($1 < CF < 3$). The Hg concentration in water samples (average = 30 μg/g) considered high. Comparison with Hg contents in coastal sediments, seawaters and molluscs in the Red Sea, the Arabian Gulf suggested that the studied samples have higher concentrations of Hg. The suggested natural sources of Hg in the study area are the weathering and decomposition of neighboring deserts. The anthropogenic sources are the land reclamation, petrochemical industries, boat exhaust emissions, oil leakage, desalination plants and sewage effluents exceeded in the study area and in Al Jubail industrial city to the north.

1. Introduction

Coastal environments are often extensively contaminated by receiving various pollutants such as toxic metals, nutrients, and pesticides (Readman et al., 1992; Bhakta and Munekage, 2010; El-Sorogy et al., 2012, 2013a, b). The major anthropogenic sources which contribute significantly to the natural background of toxic metals in soils and sediments of coastal regions are: mining and smelting, industrial sources, urban waste, waste water discharges and shipping activities.

Mercury is a volatile element emitted from both natural and anthropogenic sources (Nishimura et al., 1983). Although the atmospheric Hg input to the ocean is 2-fold higher than that from the rivers (Lantzy and Mackenzie, 1979), all the mercury moving over the surface of the globe is anthropogenic (Moore and Morre, 1976). The main species of Hg in the atmosphere is gaseous elemental Hg or Hg (0), which has a long atmospheric residence time (6–12 months) and can thus undergo long-range transport (Selin, 2009). In the atmosphere, Hg(0) can react with strong oxidants, such as halogen radicals, to form chemical species termed reactive gaseous Hg (RGM) and particulate Hg (PHg), both of which have relatively short atmospheric residence times and therefore are rapidly deposited to underlying surfaces, such as landscapes and water bodies (Steffen et al., 2008).

Many studies in the Arabian and Oman gulfs have dealt with fauna, environmental assessment, and sedimentology (Bosch et al., 1995; Sadiq and Alam, 1989; Pourang et al., 2005; Loughland et al., 2012; Naser, 2013; Almasoud et al., 2015; El-Sorogy and Youssef, 2015; El-Sorogy et al., 2016a,b; Youssef et al., 2015).

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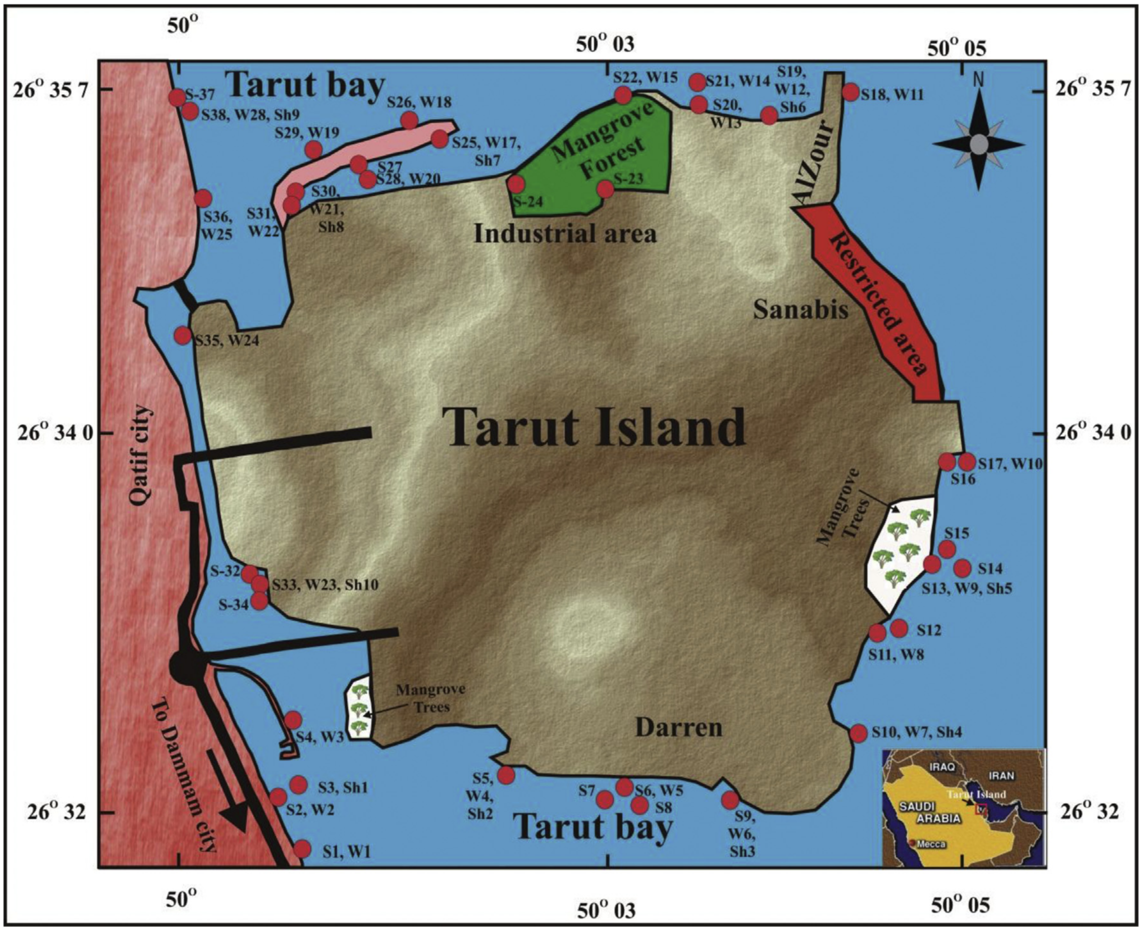

Fig. 1. Location map of the study area and the sites of the collected samples. S = sediments sample, w = water sample, sh = Mollusca sample.

The main objectives of the present study are: 1) evaluate the levels of Hg along the Tarut coast using coastal sediments, seawaters and mollusk shells, 2) assess the impact of environmental changes along the coast, 3) document the difference between gastropods and bivalves in Hg uptake, 4) comparison between the rate of pollution in the Tarut coast and neighboring coasts.

2. Material and methods

2.1. Study area

38 coastal sediments, 26 unfiltered seawaters and 40 mollusk specimens were collected in December 2014 from the subtidal zone of the Tarut Island coast (Fig. 1). Tarut is an island in the Arabian

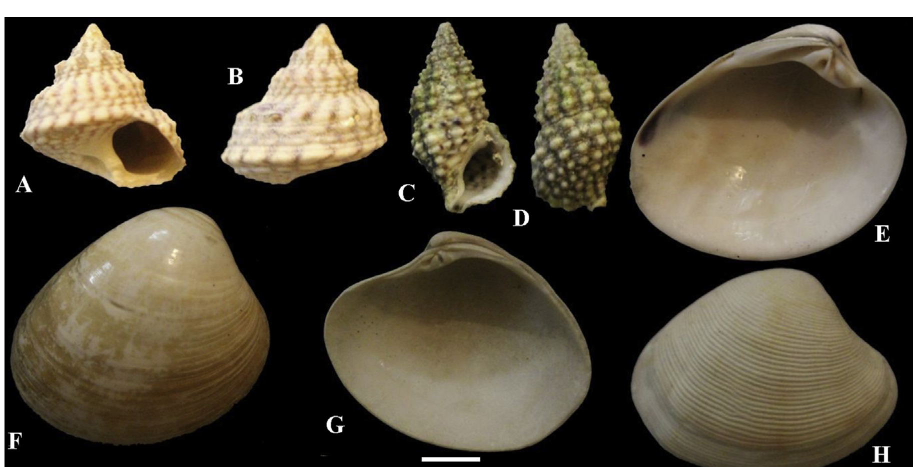

Fig. 2. A, B, *Trochus (Infundibulops) erithreus*; C, D, *Clypeomorus bifasciatus persicus*; E, F, *Amiantis umbonella*; G, H, *Protapes sinuosa*.

Table 1
Minimum, Maximum and averages of Hg, enrichment factor, geoaccumulation index and contamination factor of the studied sediments samples.

	Hg			Fe	EF	Geo	CF
	Sediments	Seawaters	Molluscs				
Minimum	0.3	0.19	0.4	599	0.49	−1.22	0.75
Maximum	1.7	0.44	1.2	12924	5.71	1.54	4.25
Average	0.55	0.30	0.763	3447.1	1.76	−0.34	1.38

Gulf belonging to the Eastern Province of Saudi Arabia, now connected by three causeways to Qaṭif. It is six kilometers from the coast; the island has an area of 70 square kilometers, and is the longest island in the Gulf after Qeshm Island, extending from Ra's Tannurah in the north to Qaṭif in the west.

2.2. Methodology

The sediment samples were stored in a clean polyethylene bags until metals analysis was performed. The sediment samples were prepared by accurately weighing around 100 mg of samples into a dry and clean Teflon microwave digestion vessels, 2 ml of HNO_3, 6 ml HCl and 2 ml HF were added to the vessels. Samples were digested using scientific microwave (Model Milestone Ethos 1600). The resulting digest was transferred to a 15 ml plastic volumetric tube and made up to mark using deionized water. A blank digest was carried out in the same way. For Hg and Fe analysis (Enrichment factor analysis) the sediments was analyzed by ICP-MS (Inductively Coupled Plasma-Mass Spectrometer): NexION 300D (Perkin Elmer USA).

Total dissolved solids (TDS), redox potential (Eh), electrical conductivity and dissolved oxygen (DO) were directly measured in seawater samples in-situ. Specimens of the gastropods, *Trochus (Infundibulops) erithreus Brocchi*, 1823; *Clypeomorus bifasciatus persicus* Houbrick, 1985 and the bivalves, *Amiantis umbonella* (Lamarck, 1818); *Protapes sinuosa* (Lamarck, 1818) were collected from 10 representative stations along the studied coast (Fig. 1). Mollusks were chosen because these are: 1) present all over the 10 studied stations and easy to sampling, 2) benthic sessile and abundant, 3) filter feeders and have the potential to bioconcentrate contaminants present in the water or within sediments. The Mollusk shells are illustrated in Fig. 2.

3. Concentration of mercury

3.1. Mercury in sediments

The concentrations of Hg in sediments of the study area (Table 1, Fig. 3, a) are quite high (0.3–1.7 μg/g) comparing to other areas. Within the adjacent areas (Table 2) Hg levels were vary between 0.19 and 2.34 μg/g in coastal sediments from Qatar (Al-Madfa et al.,

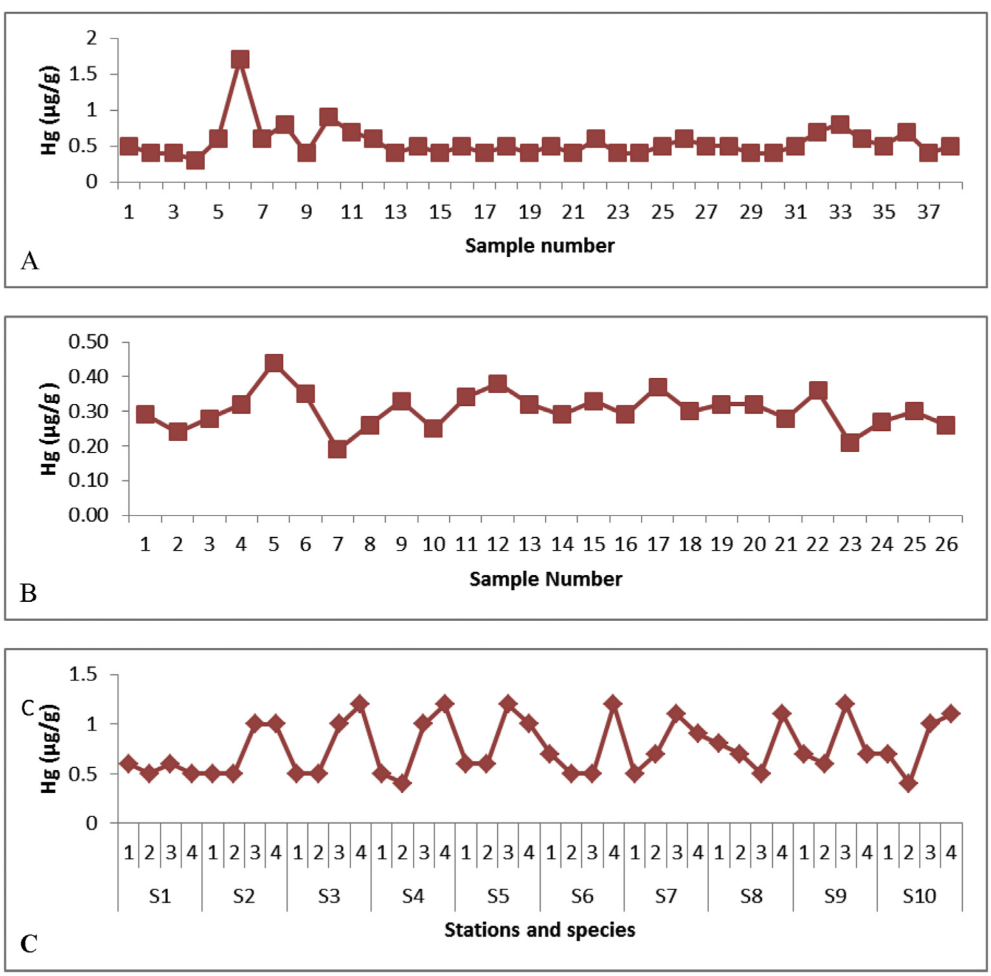

Fig. 3. Concentrations of Hg in coastal sediments (A), seawaters (B) and mollusc specimens (C), (1 *Trochus (Infundibulops) erithreus*; 2 *Clypeomorus bifasciatus persicus*; 3 *Amiantis umbonella*; 4 *Protapes sinuosa*).

Table 2
Comparison between Hg in the present coastal sediments and other worldwide localities.

Location	Reference	Hg ($\mu g\ g^{-1}$)
Tarut Island, Arabian Gulf	Present study	0.3–1.7
Red Sea, Saudi Arabia	Youssef et al., submitted	0.01–0.02
Qatar	de Mora et al. (2004)	0.0015–0.0167
UAE		0.0006–0.0022
Bahrain		0.0025–0.220
Oman		<0.0001–0.011
Gulf of Aqaba	Al-Taani et al. (2014)	2.36–2.37
Gulf of Finland	Vallius et al. (2007)	0.09–0.32
Background shale	Turekian and Wedepohl (1961)	0.4
Background continental crust	Taylor (1964)	0.08
Daliao River System, China	Lin et al. (2012)	0.01–0.1

Table 3
A- Number of samples that had metal concentrations above the sediment effect data of ERL and ERM in the collected samples. Data after Long et al., 1996, ERL = Effect range low, ERM = effect range median. B- Classification of sediment Hg contamination according to the Swedish environmental Protection Agency (μg g-1 dry weight). from Vallius et al., 2007.

Metal ($\mu g\ g^{-1}$)	Guidelines		Study area	No. of samples below the ERL	No. of samples between ERL and ERM	No. of samples above the ERM
	ERL	ERM	Average			
Hg	0.15	0.71	0.55	0	34	4

Metal ($\mu g\ g^{-1}$)	Class 1 little or none	Samples in class 1	Class 2 slight	Samples in class 2	Class 3 significant	Samples in class 3	Class 4 large	Samples in class 4	Class 5 very large	Samples in class 5
Hg	<0.04	0	0.04–0.10	0	0.10–0.27	0	0.27–0.7	34	>0.7	4

Data after Long et al., 1996.
ERL = Effect range low, ERM = effect range median.

1994), from 0.032 to 0.27 μg/g in surface sediments of Arabian Gulf (Kureishy and Ahmed, 1994), and from 0.042 to 0.375 μg/g (Al-Majed and Rajab, 1998). The Hg content in the surface sediments of Oman Gulf also is very low ranges from 0.0001 to 0.22 μg/g (de Mora et al., 2004). Hg concentration is very low also in the Red Sea coastal sediments range 0.0003–0.133 μg/g (Pan et al., 2011). Gulf of Aqaba surface sediments have very high Hg concentration (2.36–2.37 μg/g) and these values were interpreted by Al-Taani et al. (2014) as a geologic source of Hg to seawater. They reported higher levels of Hg with average values of 105 μg/g and 264 μg/g in the rock samples. The average concentration of Hg in Northeastern Gulf of Finland is 0.22 μg/g (Table 2). The concentration of Mercury in some samples in the studied area exceeded the ERM, indicating possible Hg contamination (Table 3) (see Fig. 4).

3.2. Mercury in seawaters

The distribution of Hg is nearly random along the studied coast, in spite of recorded high values in the southwestern and northeastern samples. Hg content in seawater samples vary from 0.19 to 0.44 μg/L with an average of 0.30 μg/L (Fig. 3, b). Comparison made between averages of Hg content in coastal sediments and seawater reveal that sediment samples contain 2-fold higher mercury. Table 4 illustrates comparisons between Hg values in the present study with others from surface seawaters in neighboring and abroad gulfs and seas. These values are much higher than those recorded from seawaters of Gulf of Aqaba (Al-Taani et al., 2014), Red Sea coast (Youssef et al., submitted).

3.3. Mercury in mollusk shells

Hg content in the studied seashells varies from 0.4 to 1.2 μg/g with an average of 0.763 μg/g (Fig. 3, c). The average of Hg content in mollusk specimens are 2-fold higher mercury comparing to the water sample. Low Hg values are recorded in *Clypeomorus bifasciatu* in station 4, 10 and the high Hg concentration is reported in *Protapes sinuosa* in station 3, 4. Bivalves are good accumulators for Hg than gastropods with high concentrations in *A. umbonella* (stations 2, 3, 4, 7, 9, 10) and *P. sinuosa* (stations 2, 3, 4, 5, 6, 8, 10).

Table 5 illustrates comparisons between present Hg values with others mollusks in neighboring gulfs and seas. The present Hg values are higher than the values recorded in *Pinctada radiata*, *Saccostrea cucullata*, *Circentia callipyga*, *Pinna muricata* and *Spondylus* sp. from coasts of Qatar, UAE, Bahrain, Oman along Arabian Gulf (de Mora et al., 2004).

4. Contamination level

Using the Effect range low (ERL) and the effect range median (ERM) (Table 3), thirty four (34) samples lie between the ERL and ERM. According to the Swedish Environmental Protection Agency (SEPA), thirty four (34) samples are largely contaminated, 4 samples are considered very large contaminated (Vallius et al., 2007).

Enrichment factor (EF) analysis (Simex and Helz, 1981; Zhang et al., 2012) suggested that mercury in coastal sediments of the Tarut Island is considered to entirely originate from natural processes. The Minimum, maximum and average EF values for Hg are tabulated in Table 1. Based on Muller scales (Muller, 1981) of geo-accumulation index (Igeo), the studied sediments show lower values (Igeo<0) indicating that the sediments are unpolluted. Also according to contamination factor (Cf) of Hökanson (1980) and Savvides et al. (1995), these sediments are considerable contaminated with Hg (1 < CF < 3).

The sources for Hg in the studied sediments, seawaters and mollusks are natural and for low extent anthropogenic. The weathering and decomposition of rocks from neighboring deserts, in the form of dust storms repeated especially annually in April and May natural are believed to be the sources of Hg. Anthropogenic source of Hg are believed to a wide range of potential effects of the coastal ecosystems, particularly from point and non-point sources

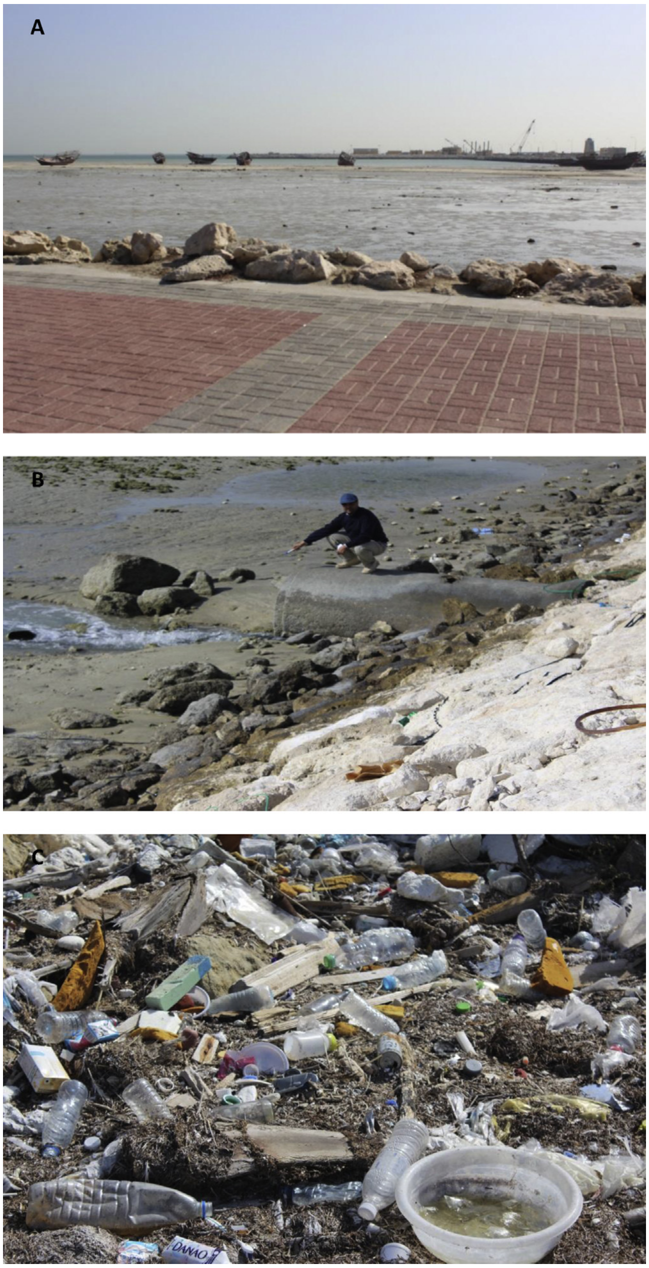

Fig. 4. A, crowded fishing boats in the intertidal zone, station 3; B, Wastewater and desalination units upon the coast, station 8; C, solid wastes in the form of plastic bottles in the intertidal zone, station 6.

of pollution. Discharge of industrial effluents, desalinization plants, sewage, irrigation, and urban runoff are the main sources of Hg from Tarut Island itself or from Al-Jubail industrial city, about 60 Km to the north of the study area, which is highly populated with intense industrial activities.

Table 4
Comparison between Hg in the present seawater samples and other worldwide localities.

Location	Reference	Hg (μg g^{-1})
Tarut Island, Arabian Gulf	Present study	0.19–0.44
Jordanian Gulf of Aqaba	Al-Taani et al., 2014	0.018–0.123
Red Sea	Youssef et al., submitted	0.003–0.01

Table 5
Comparison between Hg in the present molluscs and other worldwide localities.

Location	References	Hg (μg g^{-1})
Tarut Island, Arabian Gulf	Present study	0.763
Qatar	de Mora et al. (2004)	0.315
UAE		0.130
Bahrain		0.074
Oman		0.116

5. Conclusions

Analyses of mercury in coastal sediments, seawaters and mollusks along the Tarut coast indicated that the present Hg values are high in comparison with those recorded from the adjacent areas of Arabian Gulf, Gulf of Aqaba, and Red Sea and exceed those recorded in shale and continental crust backgrounds. The enrichment factor, geoaccumulation index and contamination factor indicated that the coastal sediments are enriched with Hg as a result of natural and anthropogenic inputs. The Tarut coast are considered large to very large contaminated with Hg. The natural source of Hg in the study area is believed to be the dust storms result from weathering and decomposition of rocks from neighbouring deserts. Anthropogenic sources of Hg (Fig. 3) are believed to discharge of industrial effluents, desalinization plants, sewage, irrigation, and urban runoff especially from Al-Jubail industrial city to the north.

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