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Assessment of arsenic in coastal sediments, seawaters and molluscs in the Tarut Island, Arabian Gulf, Saudi Arabia

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ABSTRACT

In order to assess arsenic on the Tarut coast, Saudi Arabian Gulf, 38 sediment samples, 26 seawater samples and 40 gastropod and bivalve specimens were collected for analyses by Inductively Coupled Plasma-Mass Spectrometer. The Enrichment Factor (EF), the Geoaccumulation Index (Igeo) and the Contamination Factor (CF) indicated that coastal sediments of Tarut Island are severely enriched, strongly polluted and very highly contaminated with arsenic as a result of anthropogenic inputs. Comparison with arsenic in coastal sediments, seawaters and molluscs in the Red Sea, the Arabian Gulf and abroad coasts suggested that the studied samples have higher concentrations of As. The suggested natural sources of arsenic in the study area are the weathering and decomposition of neighboring deserts. The anthropogenic sources include the land reclamation, petrochemical industries, boat exhaust emissions, oil leakage, desalination plants and sewage effluents. These anthropogenic sources are the dominant sources of As in the study area and mostly came from 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). Mining and smelting, industrial sources (coal, oil, chemicals, fertilizers, pesticides, etc.), urban wastes, wastewater discharges and shipping activities are the major anthropogenic sources which contribute significantly to the natural background of toxic metals in soils and sediments of coastal regions.

Arsenic is ubiquitous in the environment, usually being present in small amounts in all rocks, soils, waters, air and biological tissues (Nriagu and Pacyna, 1988). It is a widely distributed element in aquatic ecosystems derived from both anthropogenic activities and natural sources (Neff, 1997, 2002). The largest sources of pollution from arsenic are agricultural chemicals such as herbicides, fungicides, rodenticides and insecticides. The most important local input of arsenic into marine environment results from smelting of non-ferrous ores and from river drainage of areas with substantial arsenic ferrous ore deposits (Nair et al., 2003).

Previous studies on the Arabian and Oman gulfs have focused mainly on fauna, environment 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; Youssef et al., 2015); however, detailed environmental assessment is still needed. Therefore, the main objectives of the present study are to: 1) evaluate the levels of Arsenic along Tarut coast using coastal sediments, seawaters and bivalve shells, 2) assess the impact of environmental changes along the coast, 3) document the difference between gastropods and bivalves in arsenic uptake, 4) compare between the rate of pollution in the Tarut coast and other coasts in neighboring and abroad coasts.

2. Material and methods

2.1. Study area

Tarut Island is an island in the Arabian Gulf belonging to the Eastern Province of Saudi Arabia, connected to Qatif by two

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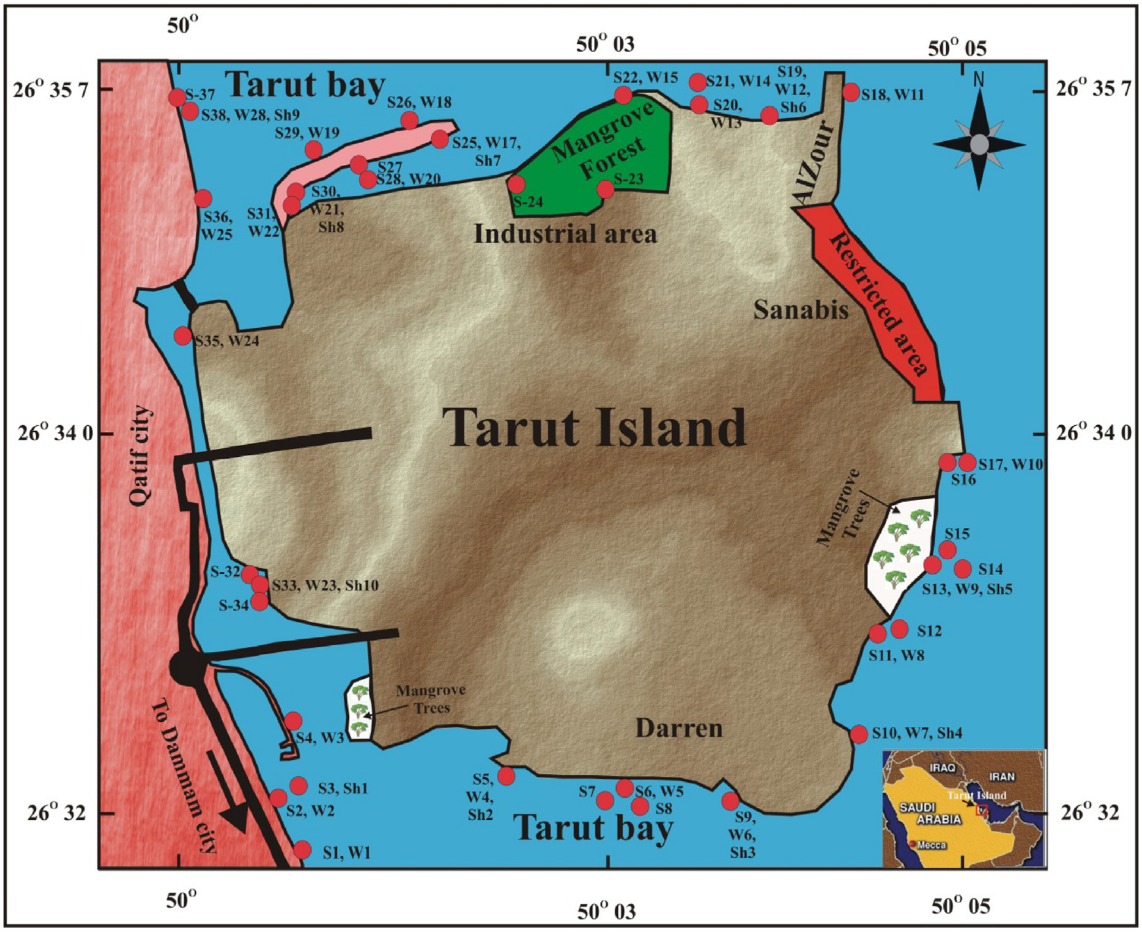

Fig. 1. Location map of the study area showing the position of the collected samples.

causeways (Fig. 1). It is 6 km from the coast, and is the longest island in the Arabian Gulf after Qeshm Island, extending from Ras Tanura in the north to Qatif in the west. The island has an area of 70 square kilometers. It contains a number of populated sectors, including Deyrah, and Darin (Al Ruwaie, 2007).

According to the type of shore materials, the shoreline of Tarut Island could be classified into sandy beaches, mangrove swamps, rocky beaches and tidal mudflats. Sandy beaches (Fig. 2A), formed of friable sandy and locally gravelly sediments as well as shell fragments of molluscs, algae, corals, and foraminifera drifted by storms and high tide above these beaches. Mangrove swamps (Fig. 2B), found in many stations on the Arabian Gulf coast in general and particularly in Tarut coast. The sediments between mangrove stands are very fine due to trapping away from strong waves. Rocky beaches (Fig. 2C), locally man-made landfill beaches formed of moderate to large rocky blocks, used as wave breaks to make artificial lagoons and pools as platforms to fishers harbors. Tidal mudflats (Fig. 2D), are muddy and sandy by turns generally restricted to low energy environment associated with low water movement. These flats provide some of the best conditions for molluscs to flourish. In these flats, cowries, cones, mitres, turrids, bubble shells, pen shells, cockles and scallops all living in close proximity and even the most delicate shells may survive intact.

2.2. Sampling and analytical methods

38 coastal sediments, 26 unfiltered seawaters and 40 gastropod and bivalve specimens were collected in December 2014 from the subtidal zone of the Tarut Island coast (Fig. 1). The sediment samples were stored in a clean polyethylene bags until metals analysis was performed. The grain-size distribution of desalted sediments was determined by wet sieving of sand and gravel and by the pipette technique for silt and clay fractions (Folk, 1974). 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 (Trabzuni et al., 2014). 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. The analytical determination of As and Fe (for Enrichment factor analysis) was carried out by ICP-MS (Inductively Coupled Plasma-Mass Spectrometer): NexION 300D (Perkin Elmer USA). The Statistical Program for Social Sciences (SPSS program) was used for Pearson's correlation coefficient matrix.

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). Molluscs were chosen because they 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.

Fig. 2. A, sandy beach; B, mangrove and saline marsh shores; C, rocky shores along the coast. Dark color illustrates the maximum high tide; D, tidal mudflats. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2.3. Estimation of pollution indicators

For calculation of pollutant indicators of As pollution in the coastal sediments of the Tarut Island, the following factors were taken into consideration.

2.3.1. Enrichment factor (EF)

It is a method proposed by Simex and Helz (1981) to assess trace element concentrations. The EF is a good tool to differentiate between the anthropogenic and natural sources of metals in environmental samples (Morillo et al., 2004; Selvaraj et al., 2004; Adamo et al., 2005; Hung et al., 2009; Fang and Chen, 2010). It is mathematically expressed as:

$$\mathrm{EF} = (\mathrm{M}/\mathrm{Fe})\mathrm{sample}/\mathrm{background}$$

where (M/Fe) sample is the ratio of metal and Fe concentrations in the sample, and (M/Fe) crust is the ratio of metal and Fe concentrations in the Earth's crust.

Table 1
Minimum, maximum and averages of As, enrichment factor, geoaccumulation index and contamination factor of the studied samples.

	As			Fe	EF	Geo	CF
	Sediments	Seawaters	Molluscs				
Minimum	53	8.55	16.3	599	53.63	1.69	4.08
Maximum	342	14.88	38.1	12,924	809.44	3.56	26.31
Average	147.95	11.13	26.245	3447.1	227.13	2.61	11.38

2.3.2. Geoaccumulation Index (Igeo)

The Geoaccumulation Index is a common criterion used for evaluating the trace metal pollution in sediments (Leopold et al., 2008). It was originally defined by Müller (1979), where trace metal contamination in sediments was determined by comparing their current concentration levels with those from preindustrial times. The Igeo can be defined as:

$$\text{Igeo} = \ \text{Log}_2(\text{Cn}/(1.5 \times \text{Bn}))$$

where Cn is the measured concentration of metal (n) in the sediments, Bn is the geochemical background concentration of the metal (n) in shale (Turekian and Wedepohl, 1961), and 1.5 is a background matrix correction factor for lithogenic effects.

2.3.3. Contamination factor (CF)

The contamination factor (Cf) was also used to assess the level of contamination and the possible anthropogenic impact of contaminants in sediments (Singh et al., 2002; Gonzales-Macias et al., 2006; Farkas et al., 2007; Çevik et al., 2009). Hakanson (1980) proposed the contamination factor (Cf), which is expressed as;

$$C_f = C_o/C_b$$

where Co is the sediment metal content in the sample and Cb is the normal background value of the metal.

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

Location	Reference	As
Tarut Island, Arabian Gulf	Present study	53–342
Arabian Gulf, Saudi Arabia	Youssef et al., 2015	148
Qatar	Mora de et al. (2004a)	1.0–6.3
UAE		0.7–9.6
Bahrain		3.16–6.88
Oman		0.74–5.01
Gulf of Aqaba	Al-Taani et al. (2014)	12.2–15.1
Azerbaijan	Mora de et al. (2004b)	8.87–22.6
Iran		6.97–20.1
Kazakhstan		2.13–20.2
Russia		0.42–6.71
Gulf of Finland	Vallius et al. (2007)	7.25–19.1
Salaam coast, Tanzania	Rumisha et al. (2012)	0.2–1.3
Background shale	Turekian and Wedepohl (1961)	13
Background continental crust	Taylor (1964)	1.8
Daliao River System, China	Lin et al. (2012)	4.61–19.13

Table 3
Comparison between As in the present seawater samples and other worldwide localities.

Location	Reference	As
Tarut Island, Arabian Gulf	Present study	8.55–14.88
Jordanian Gulf of Aqaba	Al-Taani et al., 2014	0.46–1.55
North Atlantic	Donat and Bruland, 1995	20
North Pacific		20
Kuwait coast, Arabian Gulf	Bu-Olayan and Thomas (2001)	0.01–0.06
Norwegian coast	Lenvik et al., 1978	0.25–2.50
Malaysia coast	Yusof et al. (1994)	0.7–1.8
Spain coast	Navarro et al. (1993)	0.5–3.7
Australia coast	Maher (1985)	1.1–1.6

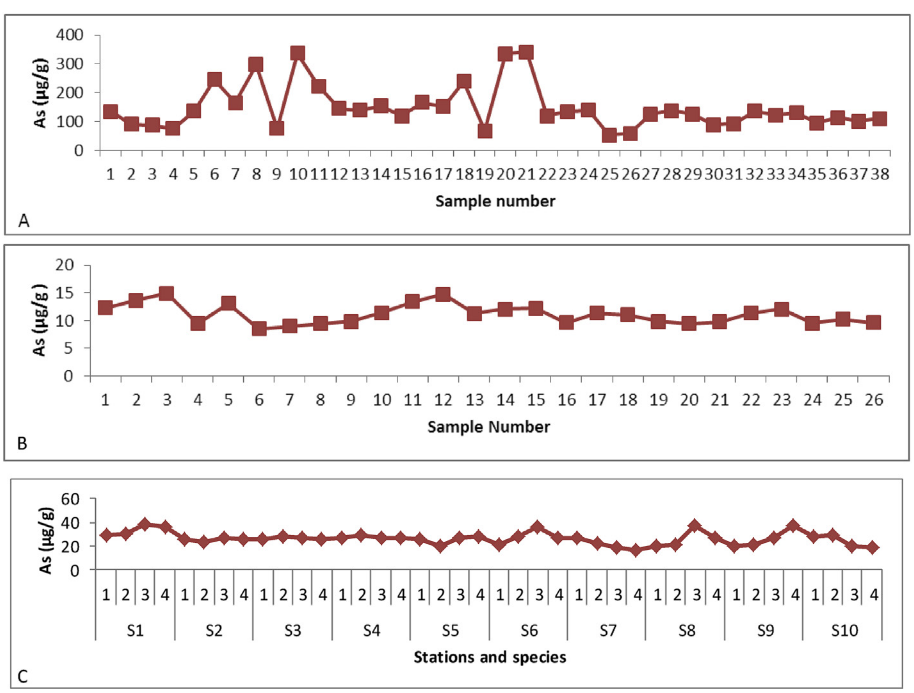

Fig. 3. A, Concentrations of As in coastal sediments; B, seawaters and C, mollusc specimens (1, *Trochus (Infundibulops) erithreus*; 2, *Clypeomorus bifasciatus persicus*; 3, *Amiantis umbonella*; 4, *Protapes sinuosa*).

Table 4
Comparison between As in the present molluscs and other worldwide localities.

Localion	References	As
Tarut Island, Arabian Gulf	Present study	16.3–38.1
Gulf of Oman and Arabian Gulf	Mora de et al. (2004a,b)	11.1–156
Egypt (Nile branch)	Lotfy (2006)	0.62–1.33
Kuwait coast, Arabian Gulf	Bu-Olayan and Thomas (2001)	0.15–0.43
Spain, South, Atlantic coast	Jose et al., 1996	1.10–3.00

Table 5
Correlation coefficients between As and physical properties of seawater in the study area.

	Do	Eh	TDS	PH	EC	As
Do	1.00					
Eh	0.19	1.00				
TDS	−0.21	−0.11	1.00			
PH	0.19	−0.35	0.439*	1.00		
EC	−0.20	−0.13	0.998**	0.493*	1.00	
As	−0.22	−0.18	0.21	0.25	0.23	1.00

**Correlation is significant at the 0.01 level (2-tailed).
*Correlation is significant at the 0.05 level (2-tailed).

3. Results

3.1. Arsenic in sediments

As values ranged from 53 to 342 µg/g, with an average of 147.95 µg/g (Table 1, Fig. 3). The lowest As value is recorded in sample 25 at the northernwest of the Island coast and the highest As concentration is reported in sample 20 at the northerneast. Table 2 illustrates comparisons between As in the present study values with those from coastal sediment of neighboring and abroad gulfs and seas. Present As values are very high in comparison with those recorded from the Gulf of Aqaba (Al-Taani et al., 2014), from coasts of Qatar, UAE, Bahrain, Oman along the Arabian Gulf (Mora de et al., 2004a) and from the Gulf of Finland (Vallius et al., 2007). Our As values also exceed those recorded in shale and continental crust backgrounds of Turekian and Wedepohl (1961) and Taylor (1964).

3.2. Arsenic in seawaters

The spatial distribution of As in seawaters did not show an obvious general trend, in spite of the recorded high values in the southwestern and northeastern samples. As content in seawater samples varied from 8.55 to 14.88 µg/L with an average of 11.128 µg/L (Table 1, Fig. 3). Comparison between average values of As in coastal sediments and seawater revealed that sediment samples

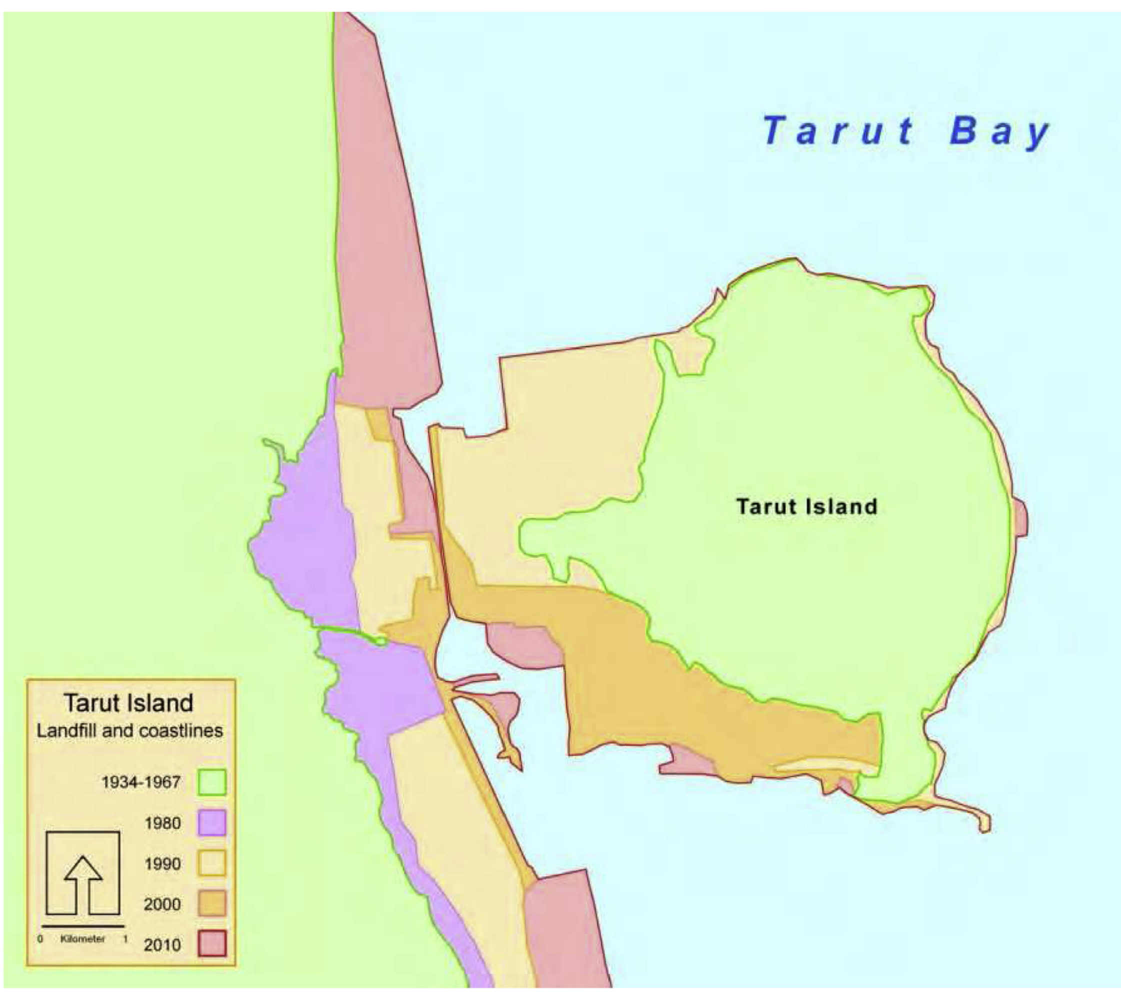

Fig. 4. Changes along Tarut Island and the adjacent coastline since 1934 (After Loughland et al., 2012).

contain 13-fold higher arsenic. Table 3 illustrates comparisons between our As values and others from surface seawaters in neighboring and abroad gulfs and seas. These values are much higher than those recorded from seawaters of the Gulf of Aqaba (Al-Taani et al., 2014), the Arabian Gulf (Bu-Olayan and Thomas, 2001), the Norwegian coast (Lenvik et al., 1978), the Malaysia coast (Yusof et al., 1994) and the Spain coast (Navarro et al., 1993). Our As values are less than those recorded from surface seawaters of north Atlantic and north Pacific (Donat and Bruland, 1995).

3.3. Arsenic in mollusc shells

As content in the studied seashells varied from 16.3 to 38.1 μg/g with an average of 26.25 μg/g (Table 1, Fig. 3). Comparison made between averages values of As in mollusc specimens and seawaters revealed that mollusc specimens contain 2-fold higher arsenic. The lowest As value is recorded in *Protapes sinuosa* in station 7 and the highest concentration is reported in *Amiantis umbonella* in station 1. Bivalves are good accumulators for As than gastropods with high concentrations in *A. umbonella* (stations 1, 6, 8) and *P. sinuosa* (station 9).

Table 4 illustrates comparisons between present As values with other molluscs in neighboring and abroad gulfs and seas. The present As values are located within 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 (Mora de et al., 2004a). Our As values in seashells are much higher than those of the Kuwait coast, the Arabian Gulf, the Spain, South Atlantic coast and the Nile branch of Egypt (Bu-Olayan and Thomas, 2001; Jose et al., 1996; Lotfy, 2006).

Fig. 5. Different solid pollutants along Tarut coast. A, crowded fishing boats in the intertidal zone; B, Wastewater and desalination units upon the coast; C, solid wastes in the form of plastic bottles in the intertidal zone.

4. Discussion

According to Enrichment Factor analysis, the coastal sediments of Tarut Island are extremely severe enriched with As (EF > 50) as a result of anthropogenic inputs. Based on Müller scales (Müller, 1981) of the Geoaccumulation Index, the studied sediments are moderately to strongly polluted with As (2 < Igeo < 3). According to Contamination Factor, the studied coastal sediments are very high contaminated with As (Cf $\geq$ 6). The Minimum, maximum and average values of Enrichment Factor, Geoaccumulation Index and Contamination Factor for As are tabulated in Table 1.

Table 5 showed Pearson correlation coefficients between As and physical properties in seawater. However, As does not show correlation with any physical properties. It shows a negative correlation with dissolved oxygen and redox potential.

There are natural and anthropogenic sources for As in the studied sediments, seawaters and molluscs. The natural source of As is believed to be the weathering and decomposition of rocks from neighboring deserts, in the form of dust storms repeated especially annually in April and May. Anthropogenic sources of As which have an enrichment factor ˃2 are believed to a wide range of potential effects of the coastal ecosystems, particularly from point and non-point sources of pollution. Discharge of industrial effluents, desalination plants, sewage, irrigation, and urban runoff are the main sources of As 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.

Fig. 4 shows successive landfill operations around Tarut Island as a result of land reclamation, urbanization, dredging and erosion caused by extensive land use. Also, during the field trip, the research team recorded different human solid pollutants along many stations of construction remnants and waste accumulation of plastic, wood, metals, concrete, tar balls and sheets (Fig. 5).

5. Conclusions

1 Analyses of arsenic in coastal sediments, seawaters and molluscs on the Tarut coast indicated that the present As values are very high in comparison with those recorded from the Gulf of Aqaba, the Gulf of Finland, the Norwegian coast, the Malaysian coast, the Spain coast, the Australian coast and exceed those recorded in shale and continental crust backgrounds.
2 Enrichment Factor, Geoaccumulation Index and Contamination Factor indicated that the coastal sediments are extremely enriched with arsenic as a result of anthropogenic inputs. The Tarut coast is moderately to strongly polluted and very highly contaminated with As.
3 The natural source of As in the study area is believed to be the dust storms resulting from weathering and decomposition of rocks from neighboring deserts. Anthropogenic sources of As are believed to be derived from the industrial effluents, desalinization plants, sewage, irrigation, and urban runoff especially from Al-Jubail industrial city to the North.

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