






























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5 Arabia

6

7

#### ABSTRACT

8 The coastline of the Arabian Gulf attracts people throughout the year for tourism and  
9 fishing activities.<sup>[5]</sup> The present work aimed to document the contamination and human  
10 health assessment of heavy metals (HMs) in 34 surface sediment samples collected  
11 along Ras Abu Ali coastline, Saudi Arabia.<sup>[22]</sup> Enrichment factor (EF), contamination  
12 factor (CF), and sediment quality guideline (SQG) were calculated to estimate the  
13 sediment contamination, while the hazard index (HI), cancer risk (CR), and total  
14 lifetime cancer risk (LCR) were determined for human health assessment via ingestion  
15 and dermal contact pathways on both adults and children.<sup>[5]</sup> The averages of the HMs  
16 ( $\mu\text{g/g}$  dry weight) were in the following order: Fe (4808) Ni (13.00) Zn (6.89) Cr  
17 (7.86) V (6.67) Cu (4.14) Pb (3.50) As (2.47) Co (1.43). Results of EF  
18 indicated minor enrichment with Ni, Pb, and As, and no enrichment with the remaining  
19 HMs. Based on CF, the coastal sediments of Ras Abu Ali showed low contamination  
20 with HMs.<sup>[5]</sup> Reported values of As, Cr, Cu, Pb, and Zn were lower than the ISQG-Low  
21 values, however, 4 samples of Ni reported values between the ISQG-Low and ISQG-  
22 High values, indicating some anthropogenic effects with Ni. HI values were higher  
23 among children in comparison to adults, suggesting that children were at higher risk of  
24 non-carcinogenic exposure than adults. LCR values indicated that no significant health  
25 hazards for people inhabited the study area from the carcinogenic Pb, Cr, and As.

1 Keywords: Heavy metals, Coastal sediment, Human risk, Chronic daily intake, Arabia  
2 Gulf.

3

#### 4 Introduction

5 Contamination of coastal sediment is widely recognized as a severe  
6 environmental issue, and it is critical to examine the ecological and health consequences  
7 of HMs in coastal sediment.<sup>[39]</sup> The historical development of industrialized and  
8 residential complexes on coastal zones around the world represents a strong pressure  
9 on the capacity of natural systems and human health to assimilate the high amount of  
10 waste derived from human activities (Bellas et al., 2020; Di Cesare et al., 2020; Tonne  
11 et al., 2021; Saavedra and Quiroga, 2021). HMs discharged into aquatic environments  
12 will be accumulated in marine sediments causing an ecological risk to filter-feeder  
13 organisms, and ultimately affecting humans (El-Sorogy and Youssef, 2015; Singovszka  
14 et al., 2017; Ustaoglu et al., 2019; Rajeshkumar et al., 2018, Wu et al., 2014).

15 HMs enter the human body through inhalation from air, sediment/dust  
16 ingestion, and skin contact (Naveedullah et al., 2014; Nazzal et al., 2021). Excessive  
17 HM intake in the human body can cause neurological, cardiovascular, and chronic  
18 kidney diseases, tumors, and even cancers (Song and Li, 2014; Pan et al., 2018).  
19 Children are particularly sensitive to HMs because they experience additional routes of  
20 exposure from breastfeeding, placental exposure, hand-to-mouth activities in early  
21 years, and lower toxin elimination rates (Ma et al., 2016; Rahman et al., 2021).

22 During the last two decades, the coastal sediments along the eastern and western  
23 sides of the Arabian Gulf have been subjected to intensive environmental studies (e.g.,  
24 El-Sorogy et al., 2017, 2018a; Alharbi et al., 2017; Al-Kahtany et al., 2018). These  
25 studies evaluated the HM contamination using different pollution indices and

1 background references.<sup>[19]</sup> Studies on human health assessment using hazard index, and  
2 total lifetime cancer risk via ingestion and dermal contact on both adults and children  
3 are still scares. Therefore, the objectives of the present work are:<sup>[48]</sup> (i) to determine the  
4 levels and document the distribution of V, Fe, As, Co, Ni, Zn, Cr, Pb, and Cu in marine  
5 sediments along Abu Ali Island, Saudi Arabia, (ii) to assess the degree of HM  
6 contamination, and (iii) to determine the potential health risks of these HMs as  
7 cumulative carcinogenic and non-carcinogenic risks.

8

9

## Material and methods

### 10 Study area

11 Ras Abu Ali Island is located at Eastern Province, Saudi Arabia (Fig. 1). The  
12 island has a unique crescent shape with the outer section facing north. The coastline is  
13 mostly sandy-dominated shore, with rocky and mangrove shores in parts. The sandy  
14 shores are mostly bounded by seagrass, dominantly of *Halophila uninervis*, *H.*  
15 *stipulacea* and *H. ovalis*. The mangrove is represented by mono-specific stands of  
16 *Avicennia marina* of less than 2m height (Saderne et al., 2020). Seagrass and mangroves  
17 are under threat due to local dredging activities, land reclamation, and marine pollution  
18 (Almahasheer, 2018). The rocky shores and their inhabited molluscs were bioeroded  
19 by clionid sponges, duraphagous drillers, endolithic bivalves, polychaete annelids,  
20 acorn barnacles, and vermetid gastropods like those previously identified from Al-  
21 Khobar, Al-Khafji, Jazan, and Duba areas along the Arabian Gulf and Red Sea coasts  
22 (El-Sorogy, 2015, El-Sorogy et al., 2018b, 2020, 2021; Demircan et al., 2021).<sup>[9]</sup>

23

### 24 Sampling, analytical methods and data analysis

1           Thirty-four modern surface sediment samples were collected in January, 2021,  
2 from the coastal zone of Ras Abu Ali Island (Fig. 1). Samples were stored in plastic  
3 bags and placed in an icebox. In the laboratory, samples were dried in air temperature  
4 (18-26 °C) for a week after removing sea grass and gravels, then samples subjected to  
5 size fractionation using a nest of sieves to obtain the 63 µm fraction for analysis. A  
6 prepared sample (0.50 g) is digested with HNO<sub>3</sub>- HCl aqua regia for 45 minutes in a  
7 graphite heating block. The resulting solution is diluted to 12.5 mL with deionized  
8 water, mixed and analysed. V, Fe, As, Co, Ni, Zn, Cr, Pb, and Cu were analysed using  
9 inductively coupled plasma-atomic emission spectrometry (ICP - AES) in ALS  
10 Geochemistry Lab, Jeddah branch, Saudi Arabia. The ICP-AES method was validated  
11 in terms of linearity, limits of detection (LOD), limits of quantification (LOQ), accuracy  
12 and precision. Calibration curves for each element were constructed by plotting the  
13 peak area of the optimum emission line to the concentration of the standard solutions  
14 or spike solutions for standard addition curves. Calibration curves showed an excellent  
15 linearity for all elements.<sup>[9]</sup>

16           The enrichment factor (EF) and contamination factor (CF) were used to assess  
17 the HM contamination in sediment samples (Kowalska et al., 2018). The National  
18 sediment quality guidelines (SQG) of ANZECC/ARMCANZ was applied to predict the  
19 adverse effects produced by polluted sediments on benthic aquatic communities  
20 (Simpson et al., 2013).<sup>[20]</sup> The estimation of the health risks via ingestion and dermal  
21 contact pathways on both adults and children can be estimated using of the chronic  
22 daily intake (CDI), hazard quotients (HQ), hazard index (HI), cancer risk (CR), and  
23 total lifetime cancer risk (LCR). These indices are calculated according to the following  
24 formulas (Hakanson, 1980; El-Sorogy and Attiah, 2015; Luo et al., 2012; IRIS, 2020;  
25 Mondal et al., 2021):

1 
$$EF = (M/X)_{\text{sample}} / (M/X)_{\text{background}}$$

2 
$$CF = C_o / C_b$$

3 
$$CDI_{\text{ingest.}}^{[36]} = (C_{\text{sediment}} \times \text{IngR} \times EF \times ED/BW \times AT) \times CF$$

4 
$$CDI_{\text{dermal}} = (C_{\text{sediment}} \times SA \times AF_{\text{sediment}} \times ABS \times EF \times ED/BW \times AT) \times CF$$

5 
$$HI = \Sigma HQE = HQ_{\text{ing}} + HQ_{\text{dermal}}$$

6 
$$HQE = CDI/RfD$$

7 
$$\text{Cancer Risk (CR)} = CDI \times CSF$$

8 
$$LCR = \Sigma \text{Cancer Risk} = CR_{\text{ing}} + CR_{\text{dermal}}$$

9 where M and C<sub>o</sub> are the analyzed metal, X and C<sub>b</sub> are the level of a normalizer element  
10 (Fe), RfD is the reference dose for each HM, and CSF is the carcinogenic slope factor  
11 values (mg/kg.day) for Cr, Pb and As (0.5, 0.0085 and 1.5, respectively). The exposure  
12 factors used in the estimation of CDI are presented in Supplementary Table 1.  
13 Hierarchical cluster analyses (HCA) and Pearson correlation coefficient were  
14 performed to identify the potential sources of HMs.<sup>[25]</sup>

15

## 16 Results and discussion

### 17 Concentration and assessment of heavy metals

18 The coordinates of the selected coastal sediments and the concentrations of HMs  
19 (µg/g, dry weight) were presented in Supplementary Table 2. HMs showed the  
20 following ranges: Fe (1600–11300), Ni (5.0–31), Zn (1.0–35), Cr (3.0–19), V (2.0–19),  
21 Pb (1.0–14), Cu (1.0–11), As (1.0–8), and Co (0.5–4). The highest concentrations of  
22 HMs were recorded in S2 (Cu), S4 (Cr and Fe), S6 (As and Ni), S7 (Co, and V), and  
23 S20 (Pb and Zn) (Fig. 2). The higher accumulation of HMs in these samples may be  
24 attributed to their occurrence in the south western shallow isolated area from the open  
25 sea (except S20) and characterized by fine and very fine sized composition (Vieira et



1 al., 2021). In contrast, the samples of the lower HM levels, such as S10, S14, S16, S18,  
2 S19, and S21, are characterized by medium to coarse size and occurred in the north of  
3 the study area faced to the open sea. The Q-mode HCA subdivided the investigated 34  
4 samples into three clusters (Fig. 3). The first cluster includes S4, which reported the  
5 highest values of Cr and Fe. The second cluster accounts 10 samples (S1-S3, S5-S7,  
6 S9, S20, S33, and S34), which showed the highest levels of Cu, As, Ni, Co, Pb, Zn, and  
7 V.<sup>[5]</sup> The third cluster contains the remaining 23 samples (S1, S8, S10-S19, and S21-S32),  
8 which recorded most of the lowest HM values in the study area.<sup>[18]</sup>

9 The average values of the HMs in the sediment samples are listed in Table 1,  
10 along with their comparison to background references, SQGs, and some worldwide  
11 coastal sediments.<sup>[5]</sup> Average values of Zn, V, Co, Cu (except Gulf of Suez, Egypt), Fe  
12 (except Duba, Red Sea coast, Saudi Arabia) were less than those reported in the  
13 worldwide background references, national sediment quality guidelines (when  
14 available), and worldwide coastal areas (Table 1).<sup>[8]</sup> Differently, our average values of  
15 Cu, Ni, and Pb were greater than those recorded in the Gulf of Suez, Egypt (Nour et al.,  
16 2021). Furthermore, As ( $2.47 \mu\text{g/g}$ ),<sup>[10]</sup> was greater than the average earth's crust of  
17 Yaroshevsky (2006) and Taylor (1964). As, Cr, Cu, Pb, and Zn exhibited values less  
18 than the ISQG-Low values (Simpson et al., 2013), indicating a low risk of these HMs  
19 in Ras Abu Ali coastal sediment. However, 4 samples (S4, S6, S7, and S20) reported  
20 values of Ni between the ISQG-Low and ISQG-High values, indicating some  
21 anthropogenic effects with Ni.<sup>[28]</sup>

22 Enrichment factor is used to distinguish between elements contributed by  
23 human intervention from those of geological origin (Reimann and de Caritat, 2005;  
24 Kahal et al., 2020). Average EF values in the study area indicated minor enrichment  
25 with Ni, Pb, and As, and no enrichment with the remaining HMs (Table 2). S19 showed

1 moderately severe enrichment with Ni (EF = 5.64), while S29 and S19 revealed  
2 moderate enrichment with As and Pb (EF = 4.64 and 4.43, respectively). All HMs  
3 showed low contamination factor (CF < 1)<sup>[14]</sup>. The results of Pearson's correlation  
4 revealed high positive correlations between many elemental pairs (Table 3), such as  
5 between Fe and each of Co, Cu, Pb, Cr, As, Ni, V and Zn, suggesting natural sources  
6 for these HMs due to the presence of Fe, which is a well-defined marker for natural  
7 weathering and erosion of crustal materials (Mil-Homens et al., 2014; Mao et al., 2020).  
8 In the other hand, the weak correlations between Cu-As, Pb-As, and Pb-Cu may be  
9 indicated some contribution from other anthropogenic source for these HMs, such as  
10 municipal and domestic discharges (Tepe et al., 2022)<sup>[32]</sup>.

11

## 12 Human health risk assessment

13 Table 4 presented the results of the CDI, HQ and HI for non-carcinogenic risk  
14 of HMs from ingestion and dermal contact pathways on adults and children. About  
15 adults, the maximum CDI values of the non-carcinogenic risk values were  $6.454 \times 10^{-3}$   
16 mg/kg.day and  $2.575 \times 10^{-5}$  mg/kg. day through the ingestion and dermal pathways,  
17 respectively. In the other hand, the maximum CDI for children were  $6.024 \times 10^{-2}$  mg/kg.  
18 day and  $1.202 \times 10^{-4}$  mg/kg. day through the ingestion and dermal pathways,  
19 respectively. This difference indicated that children were at higher risk of non-  
20 carcinogenic exposure than adults.

21 The HI values varied from  $2.859 \times 10^{-5}$  to  $1.079 \times 10^{-2}$  for Adults, and from  
22  $2.663 \times 10^{-4}$  to  $1.005 \times 10^{-1}$  for children. This means that the cumulative hazard index  
23 was higher among children compared to adults regarding the non-carcinogenic risk.  
24 However, our HI values for the HMs were less than 1.0, suggesting there is no  
25 significant non-carcinogenic risk to the people inhabiting the coastline of the Abu Ali

1 Island (Tian et al., 2020).<sup>[47]</sup> The HI values of HMs for both adults and children exhibited  
2 the following descending order: As Fe Cr Pb V Ni Cu Co Zn. However,  
3 the value of HI for As was greater than 0.1 for children, indicating the need to protect  
4 their health.

5 The accumulation of toxic HMs in human bodies may cause harmful  
6 complications. The excessive accumulation of Cr, As, and Pb in human bodies may  
7 trigger lung cancer, stomach cancer, dermal lesion, skin cancer, harmful to the  
8 respiratory system and can impact the nervous system and lead to renal failure (IARC,  
9 1994; Mao et al., 2019; Rahman et al., 2020). The carcinogenic risks for Cr, Pb, and As  
10 were estimated in the studied samples. About adults, the maximum carcinogenic risk  
11 values were  $4.835 \times 10^{-6}$  and  $1.929 \times 10^{-8}$  through the ingestion and dermal pathways,  
12 respectively. The maximum carcinogenic risk values for children were  $4.512 \times 10^{-5}$  and  
13  $9.002 \times 10^{-8}$  through the ingestion and dermal pathways, respectively. The LCR values  
14 for adults ranged from  $3.820 \times 10^{-8}$  in Pb to  $4.850 \times 10^{-6}$  in As, and from  $3.560 \times 10^{-7}$   
15 in Pb to  $4.520 \times 10^{-5}$  in As for children. LCR values revealed that no significant health  
16 hazards from the carcinogenic Pb, Cr, and As in the study area (Mondal et al., 2021),  
17 in spite of the risk in children is higher than that in adults due to their finger sucking  
18 behavior (Zhao et al., 2013; Pan et al., 2018).

19

20

## Conclusions

21 This study highlighted HM contamination and human health risks along the Ras  
22 Abu Ali Island, Saudi Arabia.<sup>[5]</sup> The averages of the HMs were in the order: Fe Ni Cr  
23 Zn V Cu Pb As Co. Ni, Pb, and As showed minor enrichment, while Fe,  
24 Cu, Co, Cr, Zn, and V determined no enrichment.<sup>[18]</sup> Results of cumulative hazard index  
25 (HI) for non-carcinogenic risk of HMs and the carcinogenic risks for Cr, Pb, and As

1 from ingestion and dermal contact pathways indicated no significant health hazards and  
2 the studied coastline is safe for vacationers, tourism, and the marine activities. Future  
3 studies will be needed to document the food chain uptake of contaminants and their  
4 human health implications along the Arabian Gulf.

5

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#### 11 Declarations

12 Conflict of interest The author(s) declare that they have no competing interests.

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