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Seawater Pollution by Heavy Metals in Coastal Area of Al Hudaydah Governorate, Yemen

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Abstract The surface sea water samples of the eight different stations for the Yemeni western coasts on Red Sea at Al Hudaydah Governorate were collected according to standard methods during winter 2019 and summer 2020. The eight sites chosen represented varying degrees of metal contamination along the coastal area of study area. Heavy metal concentrations of six elements (Zn, Pb, Ni, Mn, Fe, and Cr) of potential environmental concern were determined in seawater by using inductively coupled plasma - optical emission spectrometry technique. The metal of Zn was recorded the highest value (357 ug/L) in the winter season and summer season (432 ug/L), while the Cr metal was recorded the lowest value (0.5 and 0.7ug/L) respectively. Based on the overall mean concentration Zn occupied the first order of abundance, followed by Pb, Fe, Ni, Mn and Cr in summer season and winter season. The contents of heavy metals in the summer were higher than winter as a result of the change in marine phenomena, such as high temperature, increase the flow of sewage, in addition to natural flow of water from valleys during seasonal rainfall in summer. CA was used to interpret relationships between variables. Perhaps the distribution of metal concentrations is by the nature and frequency of discharge. This study was found high positive significant relation between metals and temperature degree.

Keywords Heavy metals pollution, Al Hudaydah, Yemen

1. Introduction

The marine environment in Yemen coastal area is subjected to contamination by metals in largely unknown amounts from untreated domestic, industrial, and agricultural wastewater, in addition to run-off during rainy periods, ship and boat traffic, and atmospheric fallout. Many areas along the coast are used as recreation areas by the public. The coast is successfully used for commercial fish cultivation with a significant market in Yemen. The metals are permanent and toxic to living organisms in excess concentration [1].

Heavy metals are amongst the most serious pollutants within the natural environment due to their toxicity, persistence, and bioaccumulation problems and they pose a risk to humans and ecosystems. The pollution by heavy metals can have adverse effects on marine organisms only after the organisms' uptake these metals and accumulation in their biomass [2, 3]. Urban and industrial activities in coastal areas introduce significant amounts of heavy metals into the marine environment. These amounts of metals are causing permanent disturbances in marine ecosystems, leading to environmental and ecological degradation [4, 5].

The amounts of mostly heavy metals deposited from anthropogenic activities are many times greater than depositions from natural background sources. Metals are one of the serious contaminants in the environment due to their toxicity, persistence and bioaccumulation problems [1].

The toxic effects of heavy metals are long lasting, reason being the non-degradation properties of heavy metals. The heavy metals cannot be degraded whereas organic contaminants decompose into other chemicals with time. Heavy metals have toxic effects even at low concentration, which may prove lethal to any living being.

Their concentration in biota can be increased through bioaccumulations [6,7]. The present study is aiming for investigation of the current status of selected of heavy metals concentration in surface water of Al Hudaydah coastal area. The results of this study will provide a valuable reference data set for environmental managers.

2. Materials and Methods

2.1Sample Collection and Analyzing

Eight sampling(Radio building, Navy College, Four Season, Al – Salif, Ebnabase, Al – Khawbah, Al – Luhayyah and Al – Jabanah)were selected to represent different locations in the study area. Water samples were collected in December 2019 and July 2020. Sampling location was detected using GPS localization (Table 1 and Figure 1).

Table 1: Sampling locations and the results of hydrographic parameter in seawater of winter

Station	Description	Coordinates		Hydrographic Parameter (Winter 2020)						
		Lat	Lon	T °C	S %	pH	DO mg/l	EC Us/cm	TDS g/l	Turbidity NTU
ST-1	Radio building	14° 46' 36"	42° 56' 58"	27.30	38.4	7.90	**4.2**	**61900**	**41.47**	16
ST-2	Navy College	14° 47' 00	42° 56' 59"	26.82	37.8	7.95	5.1	59800	40.07	7.5
ST-3	Four Season	14° 48' 28"	42° 55' 52"	26.91	38.0	8.06	4.7	60800	40.74	9.4
ST-4	Al- Salif	15° 20' 01"	42° 41' 31"	27.00	38.3	8.09	4.3	61700	41.34	14
ST-5	Ebnabase	15° 23' 23"	42° 48' 13"	26.33	37.5	8.11	5.4	59600	39.93	4.3
ST-6	Al- Khawbah	15° 32' 12"	42° 46' 46"	26.93	38.2	8.01	4.9	61200	41.05	12
ST-7	Al- Luhayyah	15° 44' 12"	42° 42' 37"	26.30	37.4	8.21	5.8	59400	39.78	3.1
ST-8	Al- Jabanah	14° 55' 44	42° 56' 06"	26.0	37.3	8.33	**7.0**	**59300**	**39.73**	**1.5**

Collection of water samples was carried out from the surface layers and collected directly in a 1-literpolyethylene previously cleaned with acid (6M HNO_3) and rinsed with deionized water. Various quality control and quality assurance measures were adopted during the data collected and processing to ensure reliability of the results. All materials and equipment were cleaned, and the instruments were calibrated daily before starting work, to ensure the validity and reliability of the results, and the work was repeated three times for each sample. Periodic assessment of instrument drift was undertaken by analyzing standards intermittently between samples. The FAAS was calibrated daily prior to each set of analysis, using standard solutions of the respective metals. All samples were analyzed in triplicate. Furthermore, the limit of detection (LOD) for each metal determined and spiked recovery analysis was carried out. Equipment and methodology Analytical grade HCl and HNO_3 acids (ACL Labson) of ACS and IUPAC standards were used without further purification. DDW was used throughout the study to prepare all solutions. The respective metal stock solution sand their working standards were prepared according to the linear range outlined for FAAS (Perkin Elmer Analyst 400) guidelines.

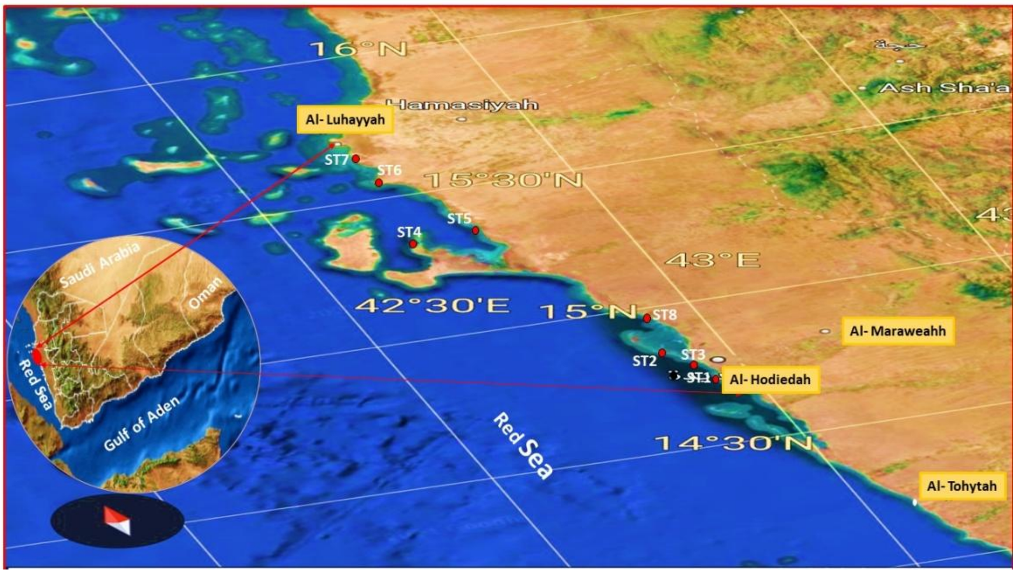

Figure1: Sampling locations in the study area

3. Results & Discussion

3.1. Physicochemical characteristics

The results of physicochemical parameters of the samples of seawater are displaying in (Figure 2). Seawater temperature plays a vital role in the metabolism of aquatic ecosystems. When the water temperature rises, the toxicity of heavy metals becomes more dangerous for marine organisms. The temperature values ranged between 26.00 °C and 27.30 °C during December of winter 2019 with an average value of 26.69 °C, and 32.10 and 33.70 °C in June of summer in 2019 with an average value of 32.10 °C.

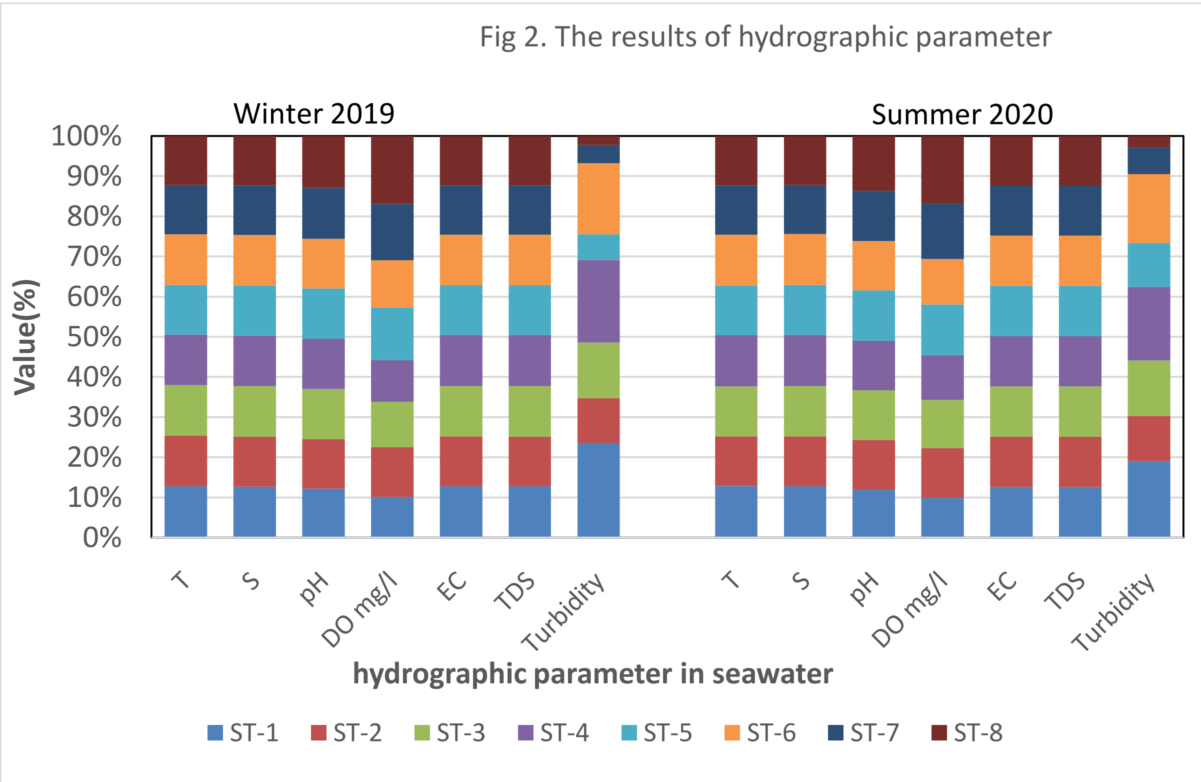

Figure 2: The results of hydrographic parameter in seawater in the study area

The variation that recorded during the seasons of study period in normal rate but above the Limits of WHO 2011(26.00-30.00 °C) [8]. The Salinity values ranged between 37.3 % and 38.4 % during December of winter

2019 with an average value of 37.86 %, and 39.10 and 41.10 % in June of summer in 2019 with an average value of 39.01 %.). The highest values of salinity were recorded at ST4 which affected by waste waters. The average salinity values showed limited changes with an average (1.24) PSU in winter and summer seasons. The pH value of seawater is an important parameter for the biological activities in the marine environment. It is reflecting of the state of pollution and productivity. The pH values ranged between 7.9 and 8.33 during December 2019 with an average value of 8.08, and 7.52 and 8.60 in June of 2020 with an average value of 7.97).The significant differences in surface water pH value where decreased in ST1 (7.9)in winter season related to degradation processes of the dead organic matters by aerobic and anaerobic bacteria, releasing more CO_2 into the water column where increased pH value in ST8 (8.60) this is probably related to the photosynthesis of many algae and sea grasses in this area leading to uptake of CO_2 from seawater and consequently pH increases. This was confirmed from the positive relationship exists in the pH values of water with dissolved oxygen ($r = 0.85$: $p < 0.01$), where the two parameters were used as good indicator for the production level. DO is one of the most important variables in the marine environment. It uses to determine the different water masses and the role of distribution of domestic production and consumption to a large extent. In Al Hudaydah coastal area, the absolute DO values ranged between 4.2 mg /l at ST1 and 7.0 mg/l at ST8 (with an average value of 7.97) during December of 2019 and 4.00 mg /l at ST1 and 6.8 at ST8 (with an average value of 7.97) during June of 2020.The high values of DO in surface water can be attributed to the exchange of O_2 from the air to the surface water as well as to algal photosynthesis [9]. The results of TDS varied from 39.73 g /l at ST8 and 41.47 g/l at ST1 (with an average value of 40.51 g/l) during December of 2019 and from 41.00 g /l at ST7 to 41.54 g/l at ST1 (with an average value of 40.86 g/l) during June of 2020.The turbidity values in the study area ranged between 1.5 NTU and 16.0 NTU during December of winter 2019 with an average value of 8.48 NTU, and from 3.0 to 20.0 NTU in June of summer in 2020 with an average value of (10.78 NTU). ST1 recorded the highest value in both winter and summer seasons and ST8 recorded the less content in the two seasons.

Seasonal values showed that the turbidity values were higher during summer compared to winter. This variation may be due to the abundance of phytoplankton, massive contribution of suspended solids from sewage and the influx of rainwater carrying suspended soil particles via wadies and other substances to sea in summer. The increase in stations 1, 4 and 6, may be due to increase sewage discharge and soil particles into sea water. The statistical analysis shows the positive strong relation between turbidity and TDS (r:0.98) and negative relation with the DO (r: -0.91). The electrical conductivity values ranged between (59300 µs/cm to 61900 µS/cm) with a mean of 60462 µS/cm in winter season as shown in (Figure 2). Whereas it ranged from 60800 µS/cm to 62000 µS/cm with a mean of 60987 µS/cm in summer season. The maximum value of electrical conductivity in summer season was at ST1 and ST4 which was (62000 and 61900 µS/cm). This increasing related to increase sewage discharge that contains ions such as chloride, nitrates, and phosphates that contribute to increase electrical conductivity as well as an increase temperature in these two stations. It is also an indicator of pollution which shows the presence of more inorganic ions in the effluent discharges received by the water [10]. The minimum value of electrical conductivity in winter season was finding at ST8 and ST7 which was (59300 and 59400 µS/cm).This related to rainwater runoff which mixing with seawater and decreasing electrical conductivity. Total dissolved solids values ranged from 39.731 g/L to 41.473 g/L with a mean value 40.509 g/L in winter season **(**Figure 2). The maximum value of total dissolved solids e was recorded at ST1(41.473 g/L) and ST4 (41.339 g/L). Whereas it ranged from 40.736 g/L to 41.540 g/L with a mean of 41.213 g/L in summer season. The maximum value of electrical conductivity was at ST1 and ST4 which was (41.473 and 41.373g/L).

The elevated in values of TDS related to electrical conductivity values, as they increase and decrease with increasing and decreasing electrical conductivity. Figure (4) shows the heavy metals as a function of the turbidity. It has important positive relationships and the turbidity mainly controls the distribution of heavy metals, the same applies in this relationship with salinity, EC and TDS.

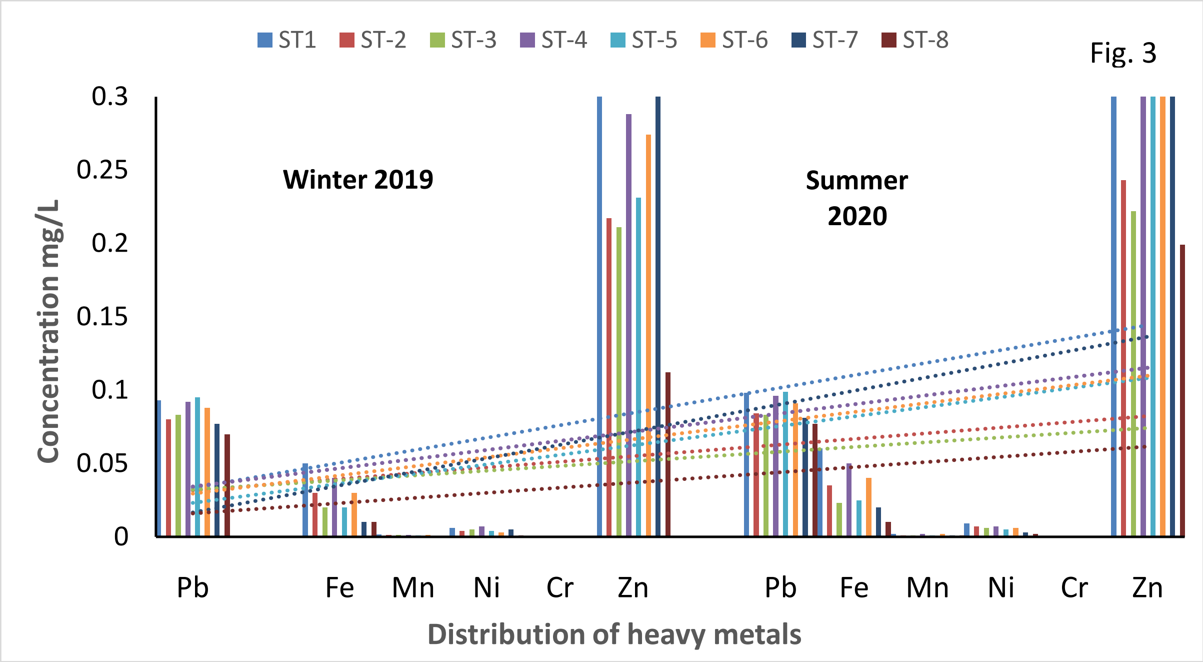

Figure 3: Distribution of heavy metals in seawater in the study area

3.2. Metal Concentrations

The Seasonal variations of heavy metals concentrations in Seawater in the study area summarized in Figure 3. The level of dissolved metals in the seawater showed different spatial variations during winter and summer season along the studied coastline. The order of the heavy metal level in the surface water according to average value in winter and summer respectively was Zn (0.254 and 0.317 mg/L) > Pb (0.085 and 0.089 mg/L) > Fe (0.026 and 0.033mg/L) > Ni (0.004 and 0.006 mg/L) > Mn (0.001 and 0.001) > Cr (0.0003 and 0.0004 mg/L). The highest concentration of all metals was recorded in ST1 (0.43and 0.36 mg/L) in winter and summer respectively. Whereas ST8 was recorded the less concentration for all metals. Cr was recorded the less concentration in all stations in winter and summer season. All metal was under the maximum limit WHO (2011) except Pb was above maximum limit [8].

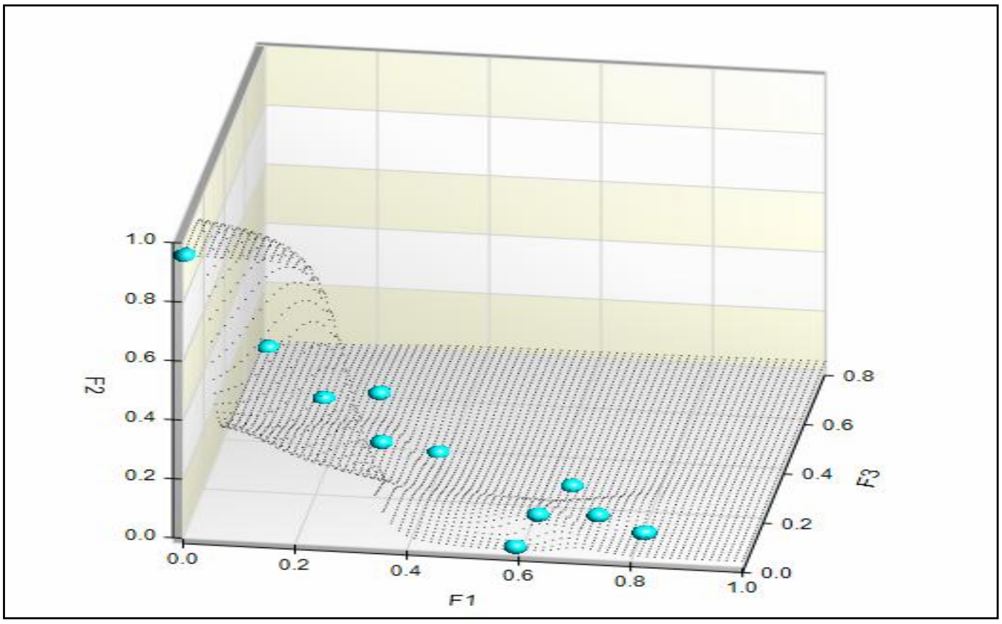

Figure 4: Factors Loading and communalities Varimax Rotation.

3.2.1. Pb

The highest levels of Pb were at ST1 (0.093 and 0.098), ST5(0.095 and 0.099) and ST6 (0.088 and 0.091) mg/l in both seasons respectively. The increase in Pb levels related to the load of Pb in wastewater to marine environment. These stations close to untreated sewage outlets. Pb recorded the highest content in summer (Figure 3).This raising of Pb content related to increase the amount of sewage and Lead reaches into the aquatic environment through precipitation, fall-out of lead dust. While the less concentration was at ST8 (The referenced sample) that was farther from pollution sources. The concentration of Pb in present study shows no big differences with other studies such as [9] who found the highest concentrations (Pb) values(0.08 mg/L) in the coastal water of the Red Sea, Yemen, and [10] who found the highest concentrations (Pb) values (0.008 mg/L) in Jeddah Coast, Saadia Arabi Results of the present study were lower than that obtained with by [11] who found the range of Lead was (0.07-0.16 mg/L) in Khawr area in Mukalla Coast in the east of Yemen.

3.2.2. Fe

Fe is one of the first-born metals that was identified by humans. It largest uses in industry and human activities also the Iron reach founding in the crust of the earth. Iron is the fourth most plentiful element in the earth's crust, and it may be present in natural waters in varying quantities depending upon the geology of the area and other chemical components of the surface seawater [12]. From Figure 3, in the surface water of the present study areas, the concentrations of (Fe) ranged between (0.01–0.05 mg/L) in winter season and (0.01- 0.06 mg/L) in summer season. The result of the analysis of (Fe) in the surface seawater showed that the highest levels were at stations close to untreated sewage outlets such as ST1, ST4 and ST6 which recorded (0.06, 0.05, and 0.04 mg/L) respectively (Figure 3).The results of this study were higher than the other studies, such as [13] in Al-Al Hudaydah coast, who found Fe values ranging from (0.0034-0.0085 mg/L). The average of Fe concentration in surface seawater was (30 mg/L) which is lower than the recommended maximum concentration of Fe (0.3 mg/L) set by [8].

3.2.3. Mn

Mn is transported to the marine environment in the same way as iron. However, since (Mn) is related to and associated with iron in the conditions of iron, accumulation and dissolution [14]. The concentrations of Mn ranged between (0.005- 0.0015 mg/L) in winter season and (0.0017- 0/0019 mg/L) in summer season (Figure 3).The result of the analysis of in the surface seawater showed that the highest levels were at ST1, ST4 and ST6 which recorded increasing in Mn levels (0.0019,0.0015, and 0.0017 mg/L) respectively. The result of Manganese in the surface seawater showed that the highest levels in summer season (Figure 3). The results of Mn in this study were lower than the studies observed by [13] in Al-Al Hudaydah Coast, Yemen who found (Mn) values ranging from (0.0004-0.001 mg/L) and [15] who found the range of Mn was (0.00001-0.00002 mg/L) in habitats of the Red Sea coast of Yemen. The average of Mn concentration in surface seawater was (0.0013 mg/L) which is lower than the recommended maximum concentration of Mn (0.5mg/L) set by [8].

3.2.4. Ni

Ni plays an important role in normal metabolism of N_2-fixing microorganism by affecting the Hydrogenase enzyme [16]. The concentrations of Ni ranged between (0.001- 0.007 mg/L) in winter season and (0.002- 0/009 mg/L) in summer season. Figure (3) shows that, Ni recorded the highest levels at ST1, ST2 andST4 which recorded increasing in Ni levels (0.009, 0.007, and 0.006 mg/L) respectively. The results of Ni were higher than the studies conducted by [13] in Al-Al Hudaydah coast (0.002 to 0.006) mg/L, and [15] who found the range of Ni between 0.0001 and 0.002 mg/L in Hurghada Coast, Egypt. The average of Ni concentration in surface seawater of this study was (0.005 mg/L) which is lower than the recommended maximum concentration of Ni (0.07 mg/L) set by [8].

3.2.5. Cr

From the results displayed in (Figure 3), the concentration of Ni ranged between (0.0001- 0.005 mg/L) in winter season and (0.0001- 0/0007 mg/L) in summer season. The highest levels of Cr were at ST1, ST4 and ST6

(0.0007, 0.006, and 0.0006 mg/L). It has the highest levels in summer. Therefore, the data findings from this study suggest that the industrial effluents and power plant wastes might be the principal source of Cr contamination in the study area. The results of this study were lower than those observed by [11] in Jizan coast, Saudi Arabia (0.00013 mg/L) which is lower than the recommended maximum concentration of Cr (0.05 mg/L) set by [8].

3.2.6. Zn

Zn happens in the number of minerals Zn blends (ZnS, $ZnCO_3$, Zn_2SiO_2, ZnO). Brass is working in a variety of applications from decorative hardware to plumbing and heat exchange units. Rolled Zn is required for battery production, photo engraving, lithographic printing plates, roofing, zinc oxide is also required for prints and other end-products such as photocopy paper, agricultural products, cosmetic and medical products [18]. From the (Figure 3), the concentrations of Zn ranged between (0.211- 0.357 mg/L) in winter season and (0.199- 0.432 mg/L) in summer season. The highest level of Zn was at ST1, ST4 and ST7 (0.432, 0.344, and 0.422 mg/L) respectively. The highest levels of Zn in summer related to increase public sewage that content high level of Zn. The results of Zn were higher than the studies conducted by [13] in Al-Al Hudaydah coast of Yemen, who found Zn values ranging from (0.001-0.0047 mg/L) with the average concentration (0.00032 mg/L) which is lower than the recommended maximum concentration of Zn (0.3 mg/L) set by WHO [8].

4. Factor Analysis

The results of this study were distributed in different PCA factors based on Varimax Rotation. According to the results of the PCA, the original variables could be reduced to three components with eigenvalues less than 1, which accounted for 11.32 (% Var: 0.48) of the total variances (Table3 and Figure. 4). Factor 1 (F1) accounting for 6.195 of the total variances is characterized by very high positive loadings in Fe (0.881) turbidity (0.851), Cr (0.751), Ni (0.774), Mn (0.731), and Pb (0.731), with statistically good and significant loadings in EC and TDS (0.662), and moderate loading with Zn (0.374) and salinity (0.321). F1 is strongly association controlled by the turbidity (Figure 4), that plays an important role of metals in the samples. Turbidity showed very good positive correlations with Fe and Cr (0.91), Mn (0.87), Ni (0.82) and Pb (0.72). Mn and Fe mostly originated from an identical source (Figure 3). The sources of these metals most probably come from lithology sources. Factor 2 accounts 3.51 (%Var: 0.27) of the total variance. F2 is formed by pH and DO. It shows poor positive loadings in pH (0.324) and DO (0.10). pH and DO Factor 3 accounting for 1.62 (%Var: 0.13) of the total variance is characterized by high positive loading of Zn (0.79) poor positive loading of Cr (0.415), Ni (0.315), temperature (0.29) and Mn (0.28), F3 represents the mixed of F1 and F2 and indicates to occurs more than one of anthropogenic sources show moderate to high negative correlations with all parameters such as turbidity (-0.83 and -0.87) respectively. This Factor mainly related to anthropogenic inputs.

Table 2: Factors Loading and communalities Varimax Rotation

Variables	Factor1	Factor2	Factor3	Communality
T	0.065	-0.941	**0.291**	0.974
S	**0.321**	-0.889	**0.267**	0.965
pH	-0.67	**0.324**	-0.477	0.781
DO	**-0.942**	**0.088**	-0.214	0.942
EC	0.662	-0.686	0.01	0.909
TDS	0.662	-0.685	0.005	0.908
Turbidity	**0.851**	-0.472	0.156	0.971
Pb	**0.731**	-0.164	**0.358**	0.689
Fe	**0.881**	-0.257	**0.243**	0.902
Mn	**0.731**	**-0.379**	0.281	0.756
Ni	**0.774**	-0.259	0.315	0.766
Cr	**0.751**	**-0.436**	**0.415**	0.926
Zn	**0.374**	-0.256	**0.793**	**0.835**
Variance	6.195	3.51	1.62	11.3233
% Var	0.477	0.27	0.125	0.871

Correlation matrix (Table 3 and Figure 5)shows that, there are significance positive relation between all heavy metals with each other, ranging from medium to very high, and these metals are associated with both turbidity, salinity, TDS and temperature with significance positive relations, while the relationship appears to be negative with both DO and pH.

Table 3: Correlation matrix of the results in the study area

Variable	T	S	pH	DO	EC	TDS	Tur	Pb	Fe	Mn	Ni	Cr	Zn
T	1												
S	0.957	1											
PH	-0.50	-0.66	**1**										
DO	-0.21	-0.44		**1**									
EC	**0.66**	**0.78**	-0.62	-0.70	**1**								
TDS	**0.65**	**0.77**	-0.62	-0.70	**0.9997**	**1**							
Tur	0.54	**0.75**	-0.83	-0.87	0.87	0.87	1						
Pb	0.33	0.48	-0.63	**-0.76**	**0.62**	**0.62**	**0.72**	**1**					
Fe	0.37	0.58	**-0.77**	**-0.85**	**0.74**	**0.73**	**0.91**	**0.73**	**1**				
Mn	0.48	**0.66**	-0.67	-0.74	**0.69**	**0.69**	**0.87**	**0.66**	**0.87**	**1**			
Ni	0.42	**0.60**	-0.75	**-0.84**	**0.66**	**0.65**	**0.82**	**0.66**	**0.80**	**0.65**	**1**		
Cr	0.59	**0.75**	**-0.86**	**-0.79**	**0.76**	**0.76**	**0.91**	**0.72**	**0.93**	**0.85**	**0.84**	**1**	
Zn	0.44	0.49	-0.61	-0.56	**0.55**	0.54	0.55	0.58	0.57	0.58	0.55	**0.67**	**1**

Note: T: Temperature., S: Salinity., Tur: Turbidity

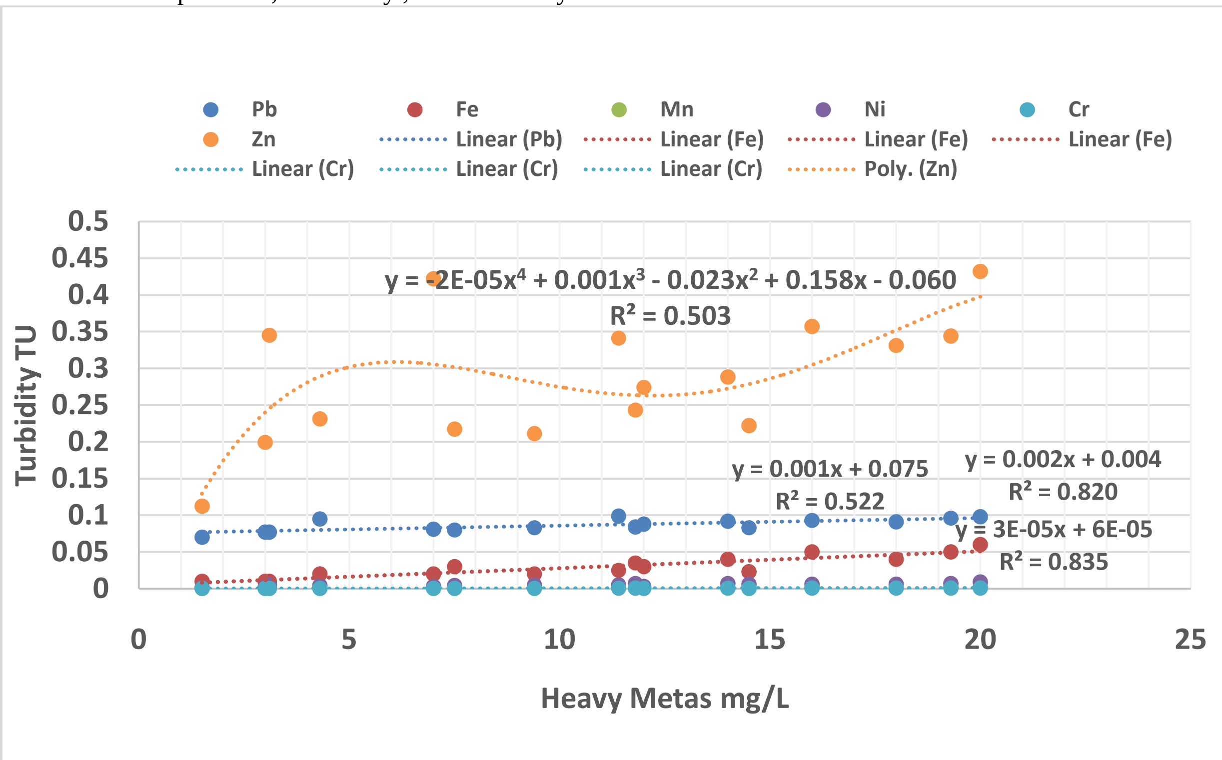

Figure 5: Heavy Metal as a Function of Turbidity

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