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Spatio-temporal analysis of pollutants in Karachi coastal water

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Abstract
Karachi coast is heavily polluted due to the regular discharge of domestic and industrial effluents. The present study discloses the occurrence and distribution of anthropogenic pollutants in samples collected from ten different places along the coastline of Karachi. The extent of pollution load for three consecutive years was estimated through the GIS technique. The results of the analysis revealed that physical parameters (pH, DO, salinity) were within the permissible limits whereas the level of chemical pollutants (except cyanide, As, Cr, and Cd) were exceptionally higher as compared to National Environmental Quality Standards (NEQS). On average, the study site Creek Avenue showed a higher pollution load of BOD, COD cyanide, TKN, and oil and grease. The average concentration of all heavy metals for three consecutive years (2015-2017) was found to be in an order of As<Cr<Cd<Pb<Ni. The study concluded that Karachi coastal area is overwhelmed with heavy loads of chemical and metallic pollution that requires strict regulations to secure the aquatic ecosystem.

Keywords
pollutant, heavy metals, toxicity, Karachi, seawater, GIS

Introduction

Rapid urbanization and unsustainable industrialization across the world is responsible for alarming environmental pollutions level that poses continuing threat to environmental health in general and public health in particular (You et al. 2015; Ashaiekh et al. 2019). The marine pollution aggravates from anthropogenic activities is putting tremendous pressure on marine ecosystems. It is therefore imperative to perform regular monitoring of physicochemical pollutants of seawater to protect the marine ecosystems (Bienfang et al. 2009; Zhang et al. 2016). Untreated wastewaters that contain organic waste dumped into the coastal areas around the world impacting the marine ecosystem and disturbing the aquatic biodiversity (Akankali and Elenwo 2015). Moreover, the additional impacts in highly populated coastal areas particularly in developing countries, where the limited facilities available are likely to cause lethal effects to the marine eco-systems (Huang et al. 2013). The wastewater release from various industries contain two major categories of pollutants viz. Organic and Inorganic that are causing serious health and environmental hazards (Arora et al. 2018; Bharagava et al. 2017). The first category includes chlorinated phenols, azo dyes, endocrine-disrupting chemicals, polychlorinated biphenyls, poly-aromatic hydrocarbons, pesticides, etc. whereas, the second category involve toxic metals (cadmium, chromium, mercury, lead, arsenic, etc.) that are highly persistent in the environment and non-biodegradable. The poor biodegradability and high concentration of organic pollutants in wastewaters also

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pose serious health and environmental issues (Saxena and Bharagava 2015; Saxena et al. 2016).
Both the physical and chemical factors within certain limits in the aquatic environments are essential in regulating the aquatic biota that includes temperature, pH, salinity, dissolved oxygen, dissolved solids, mineral and metals etc (Bhatt and Srinivasan 2019). Heavy metals also play an important role, when present as a micronutrient for the growth of living organisms but higher concentrations are extremely toxic for all kinds of terrestrial and aquatic lives (Mamboya 2007).
Coastal environment plays a crucial role in providing multiple resources, habitat and great biodiversity, continuous contamination of effluent in seawater affects aquatic life ultimately affecting the public health through a process of biomagnification (Meiaraj and Jeyapriya 2019). Studies show that contaminated seafood specifically fishes with higher quantities of heavy metals directly influences human health (Erdogrul, 2006; Lim et al. 2008; Fialkowski et al. 2009). The reproductive potential of aquatic organisms is likely to disturb due to higher levels of pollutants leading to the extinction of species (Sridhara et al. 2008). This requires the estimation of the complete pollution profile of the coastal region to protect aquatic life as well as human lives from toxic substances.
Like many other coastal regions, Pakistan is also facing pollution problems owing to an exponential increase in the human population and unsustainable industrialization with limited capacity to control pollution levels in all forms (Khan et al. 2012). Various studies have been conducted in Pakistan to assess pollution levels along coastal regions of Pakistan (Jilani, 2015; Tariq et al. 2016; Alamgir et al. 2017; Jilani, 2018; Alamgir et al. 2019; Ali et al. 2019).
Karachi is the most industrialized and financial capital of Pakistan having a coastal belt of approximately 135km. The region has significant floral and faunal diversity with a wide range of plant species, mammals, invertebrates, amphibians, fishes and migratory birds. This environmental resource-abundant area is heavily polluted due to industrial, commercial, and domestic contamination (Shahzad et al. 2009).
This research aims to evaluate the occurrence of pollutants in the coastal waters of Karachi city and showed the extent of the level of pollution through GIS techniques all along the Karachi coastline. The study covers three consecutive years (2015, 2016 and 2017) to identify the temporal variations of seawater pollutants. The study will be beneficial for decision making and regulating environmental laws by strict monitoring of industrial and domestic discharge into the Karachi coastal environment.

Methodology

Study area and sample location

Karachi city is located at the southern region of Pakistan with a population of around 20 million. This city is an industrial hub of Pakistan comprised of about 10 thousand industrial units in four main industrial zones i.e. the KIA (Korangi Industrial Area), SITE (Sindh Industrial Trading Estate), WWI (West Wharf Industries) and LITE (Landhi Industrial Trading Estate). The majority of these formal industrial units lack wastewater treatment plants (Khan et al. 2012). A sizeable number of industrial units are also located in the informal sectors. Pre-identified sampling sites along the Karachi coast were selected for the present study to analyze the extent of pollution. The sites of sample collection are presented in Table 1 and Fig. 1.

Sample	Latitude °N	Longitude °E	Site adjacent to
S-1	24.843684	66.793658	Arbian Road
S-2	24.859959	66.836882	Jamali Goth
S-3	24.843777	66.900465	Kakapir
S-4	24.797249	66.967858	Manora
S-5	24.815324	66.974437	Kemari Basin
S-6	24.849118	66.975794	Karachi Fish Harbour
S-7	24.844181	66.991664	Under Native Jetty Bridge
S-8	24.841153	66.998628	Channa Creek (near Lalazar)
S-9	24.820482	67.017941	Boat Basin
S-10	24.802205	67.079315	Creek Avenue (near Ghizri Creek)

Table 1. *Sampling sites of the study area at Karachi coast.*

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Samples were collected in clean Amber glass bottles. Seawater samples were collected through the standard Nelson bottle. At 10cm depth, seawater samples with three replictaes were collected in a pre-labeled bottle from each sampling site. For storage of the sample, the icebox was used and immediately transferred to the Institute of Environmental Studies, University of Karachi.

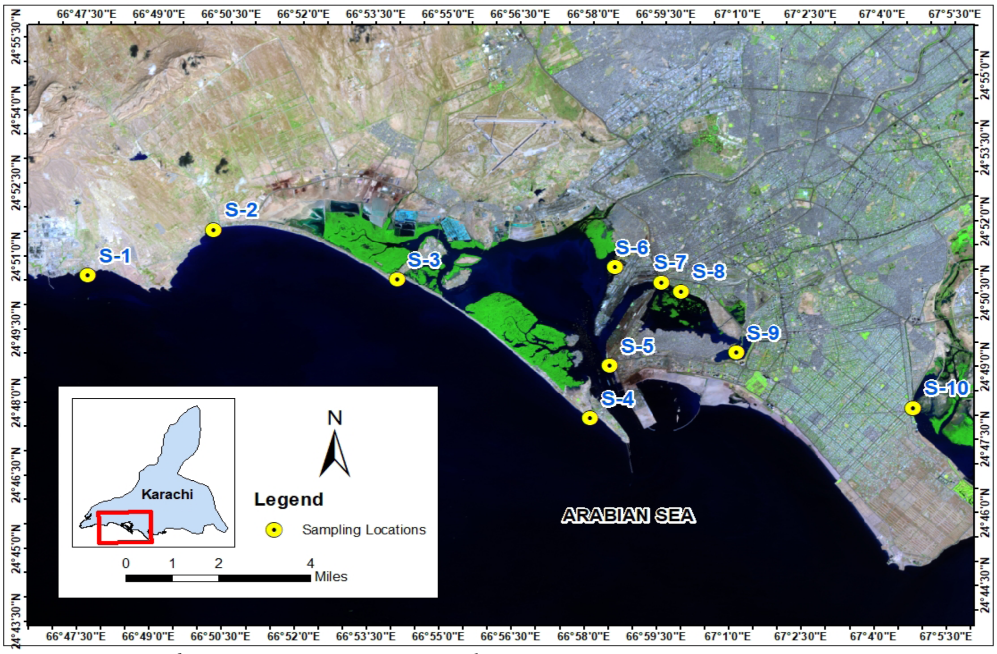

Figure 1. *Study area map represents sampling sites.*

Laboratory analysis of seawater samples

Physicochemical analysis. The in situ measurements of pH, dissolved oxygen (DO) and salinity was carried out by mean of HACH Sension 156 Multiparameter. For chemical parameters, i.e. for Biochemical oxygen demand (BOD_5), the "Alkali-azide method" was used from statndard method of American Public Health Association Methods for estimation of water and waste water, (APHA, 2005) whereas for Chemical Oxygen Demand (COD), dichromate reflux method was adopted from (APHA, 2005). The estimation of cyanide was done the Drechsel bottle method. Gravimetric and photometric method were used to detect the traces of oil, grease and phenol, respectively (APHA, 2005). For nutrient analysis, again APHA (2005) was implemented with titrimetric analysis. The Total Kjehldahl Nitrogen (TKN) estimation was done using distillation method and for Total phosphate, the ascorbic acid method using spectrophotometric analysis by was selected.

Heavy metals Analysis. The sample was taken in a sufficient quantity, oven-dried in China dish (up to a constant weight at 105 ˚C), added HNO_3 (conc.) and performed sample digestion until the sample color became pale yellow or colorless. The addition of 5ml HNO_3 and deionized water was required after digestion. Whatman Filter paper 1 was used after this step to filter out the content and the volumetric flask was used for making up to 25ml by means of deionized water. NOVA 60 (Merck) was used for the final analysis to detect five heavy metals i.e. As, Cr, Cd, Ni and Pb**.**

Statistical analysis

Principle Component Analysis (PCA) was performed using STATISTICA (99 edition) Descriptive statistics was computed for each variable for each year studied. The eigenvalues and eigenvectors were extracted from the original variables through PCA (principal component analysis).

GIS methodology

The spatial distribution of the water quality variables was geo-statistically analyzed using Inverse Distance Weighted (IDW) method by the interpolation tool in ArcGIS 10.1. The IDW has estimated the distribution of each variable based on nearby locations within the area of impact using the following equation (Bhunia et al. 2018).

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$$Z(x_0) = \frac{\sum_i^n \frac{x_i}{h_{ij}^{\beta}}}{\sum_i^n \frac{1}{h_{ij}^{\beta}}} \quad [1]$$

where, $Z(x_0)$ = value for interpolation, n = sample size, x_i = i^{th} data value, h_{ij} = distance between interpolated value and the sample data value, and β = weight.
The weights which were assigned to each interpolating point are the inverse of the distance between the selected interpolating points. This approach has allocated the higher weights with more impacts to those points which are comparatively close to each other than the distant point.

Results and Discussion
The descriptive statistics of physicochemical analysis of the seawater samples at Karachi coast is represented in Table 2 for the year 2015, 2016 and 2017 as three replicates were used for each sample. The pH of coastal water shows neutral to alkaline in all the years i.e. 7.25 to 7.36 (2015), 7.23 to 7.33 (2016) and 7.20 to 7.26 (2017) as presented in Table 2-4. The results are comparable with the recent findings of Meiaraj and Jeyapriya (2019) and El-Hazek and Al-Shiekh, (2019). The results of DO analysis is shown in Table 2-4. The minimum DO recorded in all three years was 1.45 mg/l (2017) while the maximum DO concentration was 5.82 mg/l (2016). The overall trend in the concentration of DO was lower, which depicts high contamination level due to industrial and domestic effluent specifically for the sampling sites S-6 to S-10. Various pollution studies conducted in Asia supported similar results including Titah et al. (2019), Gayathri et al. (2019) and Jilani, (2018) that show lower levels of DO in the Indonesian coast, Indian seawater and Karachi coast respectively. Song et al. (2019) believe that low DO levels could also be attributed to global warming that declines oxygen levels in the seas and oceans but still we can review aforementioned pollution studies where dumping of industrial and municipal waste is responsible for low DO levels.
The salinity levels are 18.67 to 26.08 ‰ for the year 2015, 21.33 to 24.92‰ in 2016, and 22.08 to 23.75‰ in 2017 respectively (Table 2). The salinity factor is extremely important for the survival of aquatic life and it is gradually increasing in three years. High salinity has also been recorded in the eastern Bay of Bengal from Carbines Cove beach, India (Murugan et al. 2020). However, the lowest salinity was found at Chinna creek and Malir River that enters the Arabian sea (Tariq et al. 2016) and western backwater, Karachi (Alamgir et al. 2019). Salinity in a water body is affected by multiple factors including rocks, precipitation and evaporation (Kaymaz and Ozdemir 2019).

Table 2. *Descriptive statistics of physicochemical analysis of the seawater samples at Karachi coast.*

Parameters (mg/l)	Min	Max	Mean	Std. error	Variance	Stand. dev	Skewness	Kurtosis	Coeff. var	NEQs
					2015					
pH	7.25	7.36	7.31	0.014	0.002	0.044	0.198	-1.751	0.599	6-9
DO	1.65	5.61	3.82	0.575	3.304	1.818	-0.051	-2.460	47.572	N/A
Salinity ‰	18.67	26.08	22.159	0.806	6.500	2.550	0.373	-1.304	11.506	N/A
BOD	163.08	210.67	184.74	4.256	181.112	13.458	0.098	0.827	7.285	80
COD	376.67	457.42	418.10	10.192	1038.74	32.230	-0.103	-1.953	7.709	400
Cyanide	0.38	0.62	0.48	0.026	0.007	0.082	0.321	-1.107	17.180	1.0
Oil and grease	7.58	20.14	14.42	1.431	20.479	4.525	-0.371	-1.161	31.376	10
Phenol	0.79	1.39	1.11	0.06	0.03	0.18	-0.32	0.17	15.94	0.3
Phosphate	5.96	7.86	6.69	0.202	0.408	0.639	0.464	-0.789	9.556	N/A
TKN	48.71	62.98	55.80	1.622	26.304	5.129	0.045	-1.643	9.192	N/A
As	0.043	0.086	0.054	0.004	0.000	0.012	2.186	5.669	22.589	1.0
Cr	0.03	0.08	0.05	0.005	0.000	0.015	0.254	-0.973	28.872	1.0
Cd	0.06	0.08	0.07	0.002	0.000	0.006	0.576	-0.929	8.405	0.1
Pb	1.74	2.51	2.20	0.085	0.072	0.269	-0.688	-0.743	12.247	0.5
Ni	6.03	8.55	7.12	0.279	0.780	0.883	0.407	-1.189	12.404	1.0

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					2016					
pH	7.23	7.33	7.27	0.010	0.001	0.030	1.141	0.995	0.416	6-9
DO	1.65	5.82	3.84	0.582	3.392	1.842	-0.041	-2.446	47.938	N/A
Salinity ‰	21.33	24.92	22.9	0.376	1.411	1.188	0.168	-1.075	5.187	N/A
BOD	174.42	209.75	190.05	4.120	169.72	13.028	0.326	-1.490	6.855	80
COD	349.75	516.25	437.43	16.041	2573.0	50.725	-0.168	-0.332	11.596	400
Cyanide	0.30	1.11	0.65	0.079	0.062	0.250	0.604	-0.121	38.737	1.0
Oil and grease	15.6	24.19	19.96	1.054	11.104	3.332	-0.133	-1.862	16.692	10
Phenol	1.65	2.21	1.95	0.06	0.03	0.18	-0.20	-0.82	9.26	0.3
Phosphate	6.56	8.05	7.42	0.135	0.183	0.428	-0.670	0.683	5.764	N/A
TKN	49.9	61.29	57.12	1.305	17.039	4.128	-1.030	0.029	7.227	N/A
As	0.046	0.066	0.055	0.002	0.000	0.007	0.205	-0.884	12.604	1.0
Cr	0.05	0.09	0.07	0.004	0.000	0.012	-0.019	-0.967	17.187	1.0
Cd	0.06	0.08	0.07	0.002	0.000	0.007	1.051	-0.068	10.636	0.1
Pb	3.08	4.50	3.67	0.160	0.255	0.505	0.307	-1.566	13.741	0.5
Ni	6.11	7.32	6.92	0.115	0.132	0.363	-1.233	1.812	5.245	1.0
					2017					
pH	7.2	7.26	7.23	0.007	0.000	0.022	-0.166	-1.606	0.306	6-9
DO	1.456	5.127	3.429	0.494	2.436	1.561	-0.068	-2.360	45.517	N/A
Salinity ‰	22.08	23.75	22.966	0.190	0.360	0.600	-0.350	-1.302	2.611	N/A
BOD	169.42	219.25	192.32	6.769	458.132	21.404	0.048	-2.345	11.129	80
COD	389.5	623.75	473.35	27.432	7525.32	86.749	0.541	-1.250	18.326	400
Cyanide	0.77	1.51	1.21	0.083	0.068	0.261	-0.891	-0.647	21.626	1.0
Oil and grease	18.93	27.84	22.48	1.109	12.301	3.507	0.488	-1.647	15.604	10
Phenol	1.76	2.39	2.14	0.06	0.03	0.18	-1.22	1.74	8.31	0.3
Phosphate	7.02	7.83	7.44	0.093	0.087	0.295	-0.291	-1.293	3.964	N/A
TKN	51.15	59.95	55.67	0.921	8.474	2.911	-0.231	-0.947	5.229	N/A
As	0.049	0.072	0.059	0.003	0.000	0.009	0.419	-1.513	15.271	1.0
Cr	0.06	0.08	0.07	0.002	0.000	0.005	-1.136	2.159	6.943	1.0
Cd	0.06	0.13	0.08	0.006	0.000	0.019	2.752	8.239	24.419	0.1
Pb	2.91	7.42	4.37	0.523	2.735	1.654	1.086	-0.232	37.857	0.5
Ni	4.55	7.39	6.60	0.262	0.684	0.827	-1.924	4.242	12.530	1.0

The results of BOD estimation are presented in Table 2. In the year 2015 and 2016, the BOD concentrations were 163.08-210.67 mg/l and 174.42- 209.75 mg/l, respectively. In the year 2017, with a slight increase it became 169.4-219.25 mg/l. Maximum BOD of all years was found at S-7 in 2017 while minimum BOD concentration was recorded from S-2 in 2015. High BOD levels indicating organic pollution through industrial and domestic discharges in the sea affecting the marine life and coastal species. The study area also observed dead fish species and other coal population due to high organic matter. Similar observations were recorded by Kiran and Ramaraju (2019) for Indian coastal regions. The BOD tends to deplete the DO levels that is detrimental to the aquatic lives. The present results corroborate the findings of Khan and Shaukat, (2008) who earlier noticed lower BOD levels at Chinna creek along the Karachi coast while Alamgir et al. (2019) detected higher BOD levels of 347 mg/l from the western backwater of Karachi coastal area. The COD values in 2015 for S-4 showed a minimum concentration of 376.67 mg/l while S-5 show a maximum concentration of 457.42 mg/l (Table 2). On the contrary, in the year 2016 and 2017, higher concentrations of COD are detected that is in the range of 349 to 516 mg/l and 389.5 to 623.7 mg/l, respectively. The samples show high COD levels exceeding the permissible limit i.e. 400 mg/l for seawater. In contrast, Alamgir et al. (2018) noticed higher levels of COD from Korangi creek indicating heavy industrial pollution. However, high levels of COD pointing towards increasing pollution load due to industries entering the Karachi coastal area. The results of the cyanide analysis are shown in Table 2 wherein, the lowest concentration of cyanide was observed at S-1 (0.772 mg/l; 2017) while the highest concentration recorded at S-4 (1.51 mg/l). The results

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of the year 2015 and 2016 have also shown similar findings within a range of 0.38 to 0.62 mg/l and 0.30 to 1.11 mg/l, respectively. The NEQs for cyanide is 1.0 mg/l and the present findings show only a few sampling sites have shown the concentration of cyanide higher than the maximum permissible limit. Cyanide is used for fishing in many regions but considered as toxic method whilst many studies have been conducted to identify the potential hazards related to it (Vaz et al. 2017; Calado et al. 2014). In Karachi, there are many anthropogenic sources of cyanide that mainly include shipyard waste and untreated waste of chemical industries. Even the lowest concentrations of cyanide are extremely harmful to living organisms. As compared to the present findings, Titah et al. (2019) recorded less than 0.005 mg/l of cyanide from Kamal Madura port, Indonesia.

The average concentration range of oil and grease were 7.58 – 20.14 mg/l in 2015, 15.60 – 24.19 mg/l in 2016, while maximum concentrations were 18.93 – 27.84 from S-3 and S-9 in the year 2017. Lower values of oil and grease have been recorded from the Tuticorin Harbor area, India (Meiaraj and Jeyapriya 2019). Oil spillage, industrial effluents and domestic discharges are few sources of oil pollution at Karachi coast. Also, there are fishing boats that contribute to oil and grease concentration in the study area. However, the oil degradation rate is challenging to quantify in the oceanic ecosystem owing to its complicated complexity and the components of oil and petroleum products (Zahed et al. 2010).

Seawater can be polluted either from marine-based contamination or land-based that is responsible for higher levels of phenol in the sea. To marine organisms, phenol and related compounds are highly toxic as these are endocrine disruptors (Ramos et al. 2009). The toxicity of phenols arises at a level of 10-24 mg/l for humans and 9-25 mg/l for fishes. The detectable levels of phenols by odor and taste range from 10-100µg/l. Phenol analysis with descriptive statistics is presented in Table 2 to 4. The average phenol concentrations in 2015, 2016 and 2017 were in a range of 0.79-1.39 mg/l, 1.65-2.21 mg/l and 1.76-2.39 mg/l, respectively. These results slightly contradict with Suez Bay seawater study of Phenols and derivatives that shows an average concentration of 0.46 µ/l (Soliman et al. 2017) but the present findings are consistent with the findings of Alamgir et al. (2017) and Sirajuddin et al. (2016). Phenols and related compounds are released at Karachi coast predominantly by industrial wastewaters from petroleum, pharmaceutical, steel manufacturing, chemical industries and paper and pulp mills.

The mean minimum concentration of phosphates in the study period was 5.96 mg/l from the sample site S - 1 in the year 2015, whereas, the maximum value recorded from S-2 i.e. 8.05 mg/l from 2016. A similar study has also been carried out in India that revealed total phosphate concentration in the seawater ranged between 0.086 to 0.779 µmol/l (Murugan et al. 2020). In China, 2.8 µmol/l levels were found in coastal water that is lower than the present findings (Yang et al. 2019). In the study area, the phosphate input is mainly from inorganic fertilizers and industrial activities. One of the typical sources of phosphate is the use of detergents. Nonetheless, phosphates in excess quantities lead to eutrophication in the marine ecosystem that affects the aquatic life with the production of toxins from algal growth (Roy et al. 2013). There are physical, chemical and biological methods for the treatment of phosphates from point sources whereas constructed wetlands, riparian belts and buffer zones are essential for remediation from non-point sources (Sumathi and Vasudevan 2019).

The results of TKN analysis are reported in Table 2. The minimum and maximum mean values for TKN in 2015 were 48.71 (S-1) and 62.98 (S-9), in 2016 were found as 49.9 (S-2) and 61.29 (S-4), while in 2017, these are in a range of 51.15 (S-8) and 59.95 mg/l (S-1). It can be noticed that the TKN concentration has increased in 2017 (S-1, collected from the site adjacent to Arbian road) in three years indicates more pollution at this site. The reason behind high nitrogen content is again the untreated wastewater entering into the sea through Malir and Lyari Rivers. Alamgir et al. (2017) reported higher TKN values for Keti Bunder and Shah Bunder creeks in Sindh that are consistent with the present findings. Kaymaz and Ozdemir, (2019) however, indicated low nitrogen levels from Marmaris Bay, Turkey.

The heavy metals enter into the marine environment through two main sources i.e. natural and man-made and release into the aquatic bodies by means of industrial effluent, atmospheric deposition and community run-off (Förstner and Wittmann 2012). Aquatic biodiversity is disturbed by toxic metals as they can change the properties of water (Yılmaz and Sadikoglu 2011). It can be found in the literature that more soluble forms of heavy metals are present in the aquatic system as compared to terrestrial systems (Kumar et al. 2013). Interestingly, heavy metals in seawater are present in

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dissolved form but higher quantities are complexes with ligands that are either organic or inorganic (Mamboya 2007). The profile of heavy metals of coastal water can be seen in Table 2 covering all five toxic heavy metals such as As, Cr, Cd, Pb and Ni for three years i.e 2015-2017.

The organic forms of As is highly toxic and formed as a result of interaction with organic matter and sulfur present in wastewater. It has higher affinities to form complexes with the mentioned compounds and produce thioarsenic species (Sharma and Sohn 2009). ASTDR, (2019) ranked Arsenic as first in the 'Substance Priority List' for toxicological profiles of hazardous substances. In the coastal water of Karachi, the mean concentrations of As are 0.043 to 0.086 mg/l in the year 2015, 0.046 to 0.066 mg/l in 2016 and 0.049 to 0.072 mg/l in 2017, respectively. It can be noticed that the minimum concentration has increased gradually in these years. Similar results can be observed in the studies conducted worldwide in which moderate to high levels of As were found in water bodies of South Korea (Hong et al. 2016), seawater and aquatic species of Greek coast (Kalantzi et al. 2017), India (Vardi and Chenjiv 2020), China (Rao et al. 2018), Iran (Abidi et al. 2018) and Bangladesh (Ayers and Patton 2018). The major anthropogenic sources of As release in the environment include smelting and mining activities, fertilizers, glass manufacturing, fossil fuel combustion and cancer drugs (Mandal and Suzuki 2002). Chromium is frequently used in metallurgical industries for metal production and alloy making and less used in chemical industries, tanneries and foundries (Dhal et al. 2013). These sources are also typical for the contamination of Karachi coastal water. The concentration of Cr^{3+} is in a range of 0.030 (2015) to 0.088 mg/l(2016) with the minimum concentration is found at S-2 and maximum concentration is found at S-9, respectively. In contrast to the present study, lower levels of Cr were detected in Arabian Gulf i.e. 0.70 µg/l (Alharbi and El-Sorogy 2019). In the aquatic ecosystem, Cr is available in the trivalent and hexavalent form in reducing and oxidizing state, respectively. The hexavalent form of Cr is present in various species under different pH levels (Barrera-díaz et al. 2012). Cr is carcinogenic and mutagenic causing tubular necrosis and renal failure (Rahman and Singh 2019). The seawater of Karachi coast shows slight variations in the concentration of Cd during the study period that varies from 0.060-0.077 mg/l (2015), 0.058-0.079 mg/l (2016) and increase remarkably in 0.063-0.133 (2017). The maximum concentrations are found at S-6 in 2017. Jilani (2015), recorded similar values for high Cd in Karachi coastal area. Hong et al. (2011) considered that in highly alkaline pH of water, sediments release the cadmium. Combustion of fossil fuel, Ni-Cd batteries, stabilizing, mining, incineration and fertilizers are the major anthropogenic sources of Cd. Bronchitis, pneumonitis, pulmonary edema, osteoporosis and anemia are the major toxic effects of Cd (WHO, 2010). Lead (Pb) has a high tendency for the formation of complexes in seawater with anions and organic ligands (Kumar et al. 2013). The average Pb concentration is in the range between 1.738 - 2.513mg/l in 2015. However, the conditions are lethal in 2016 where Pb concentration is in between 3.081 to 4.497 mg/l. Surprisingly, in the year 2017, the lowest mean value is found as 2.913 mg/l and the highest is 7.419 mg/l which is alarming. Tariq et al. (2016) recorded 4.66 mg/l of Pb in Chinna creek and 4.707 mg/l in Malir River. The important source through which Pb enters the seawater is mainly the industrial effluent (Oymak et al. 2009). The concentration of nickel is far exceeding the NEQs and recorded in a range of 6.03 to 8.55 mg/l in 2015, 6.11 to 7.32 mg/l in 2016 and 4.54 to 7.38 mg/l in 2017. Another study conducted along the coastline of Karachi revealed the higher levels of Ni threatening the aquatic life (Ali et al. 2019). These results deviate from findings of Corales-Ultra et al. (2019) that show low Ni concentration in the Philippines seawater indicating better compliance of regulations.

The results of the principal component analysis is given in Table 3.

In the year 2015, the first, second and third components explained 51.9624%, 17.7292% and 10.0489% of the total variance inherent in the data matrix. Together the first three PCA components retained 79.7424% of the total variability. The first component is primarily a function of phosphate, Pb, TKN and DO and largely represents the nutrient levels plus lead toxicity. The second component is governed by pH, BOD, As and salinity and to a certain extent the seawater reaction and its entropy. The third component that is chiefly regulated by salinity, As, COD and Cd somewhat repeats the trend exhibited by the second component.

In 2016, the first, second and third components explained 58.9261, 12.8614% and 8.7132% of the total variance in the data set. Together they explained 80.5008% of the total variance. The first component is basically a function of BOD, oil and grease, COD and cyanide representing the degree of organic and

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biological pollution. The second component is largely governed by TKN, phosphate, pH and As represents mostly the nutrient gradient and As toxicity. The third component is largely a function of heavy metals (Ni and Cr) and total nitrogen.

In 2017, the first three principal components explained 46.9183%, 22.9662% and 79.4567% of the total variance inherent in the data matrix. The first component is primarily a function of COD, oil and grease, BOD and Pb. This trend resembles a lot with the first PCA component of 2016 data set. The second component is fundamentally governed by Cr, Cyanide, TKN and pH representing chemical pollution. Whereas, the third component is chiefly a function of phosphate, cyanide, TKN and Pb and practically repeats the trend of the second component.

Fig 2 a,b,c show the three dimensional PCA ordination of 10 stations (sites) for the year 2015, 2016 and 2017 respectively. All three ordinations basically depict the same trends with slight variation.

However, two groups (clusters) can be recognized easily in the 3 dimensional configuration, a group of station 1,2,3,4,5 and the second group of stations 6, 7, 8, 9, 10. Within group distribution for the years differs somewhat through the two groups can readily be distinguished in each case. The first group (Left of the Fig 2 ,a, b, c) is characterized by high DO and salinity , while low COD, cyanide low in oil and grease and lower levels of Cd and Pb whereas, pH and TKN showed irregular pattern. The second group (to the right of Fig 2a, b, c) exhibited low DO and salinity and high levels of cyanide , oil and grease, phenols and high levels of Cd and Pb.

Table 3. *Results of the Principal component analysis of seawater samples collected from Karachi coast.*

Component	Eigenvalue	% total variance	% cumulative variance	First 4 eigenvector coefficients	Associated variables
			2015		
1	7.7946	51.9624	51.9624	0.9842	Phosphate
				0.9398	Pb
				0.9316	TKN
				-0.9248	DO
2	2.5693	17.7292	69.6935	0.8353	pH
				0.7761	BOD_5
				0.6417	As
				0.5458	Salinity
3	1.5073	10.0489	79.7424	0.5660	Salinity
				0.5595	As
				0.5483	COD
				-0.5171	Cd
			2016		
1	8.8389	58.9261	58.9261	0.9505	BOD_5
				0.9280	Oil and grease
				0.9228	COD
				0.9159	Cyanide
2	1.9292	12.8614	71.7875	0.7211	TKN
				-0.7164	Phosphate
				0.5617	pH
				0.3754	As
3	1.3069	8.7132	80.5008	0.7117	Ni
				-0.6269	TKN
				0.2941	pH
				0.2623	Cr

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			2017		
1	7.0377	46.9183	46.9183	0.9792	COD
				0.9636	Oil and grease
				0.9460	BOD_5
				0.9272	Pb
2	3.4434	22.9662	69.8746	-0.8999	Cr
				-0.7450	Cyanide
				0.7251	TKN
				-0.6885	pH
3	1.4373	9.5821	79.45567	0.8712	Phosphate
				0.5142	Cyanide
				0.4197	TKN
				0.2209	Pb

Pollutants distribution and spatial analysis

The GIS interpolation tool is used to represent the distribution of three years the average concentration of each parameter. Fig 2 to 5 shows the mapping of physical, chemical, nutrients and heavy metals pollution distribution in the study area. For pH, the highest average concentration is found at S-4 and S-6 i.e. Manora and Karachi fish harbor which is also discussed by Alamgir et al. (2019). The three year average salinity values are high i.e. 24.86‰ (Fig.3) but higher salinity values has also been recorded in the northern Arabian Sea during the events of cyclones (Wang et al. 2013). Oil and grease and phenol depict higher concentration i.e. 23.55 mg/l at S-10 and 1.85 mg/l at S-9 (Fig. 4) due to various industrial discharges, petroleum industries, ship activities and oil spills (Soliman et al. 2017). Surprisingly, for the sampling site S-10 i.e. Creek Avenue near Gizri Creek, higher mean concentrations of BOD (204.92 mg/l), COD (528.97 mg/l), cyanide (1.01 mg/l) and TKN (59.77 mg/l) are reported that show heavy loads of pollution in this area due to combined domestic, commercial and industrial effluent (Fig 4 and 5). For heavy metals assessment, the trends show higher mean levels of As from S-7 (0.072 mg/l), Cr from S-9 (0.078 mg/l), Cd from S-6 (0.087 mg/l), Pb from S-9 (4.69 mg/l) and Ni from S-2 (7.42 mg/l) as illustrated in Fig. 6. Each heavy metal appears to be in the higher concentration for different sampling locations due to different sources of effluent discharge in the sea as mentioned in the previous section. The marine ecosystem is affected by elevated levels of heavy metal release from industrial discharges, petroleum products, waste release from desalination plants and the untreated domestic wastewater (Pitchaikani et al. 2016; Viji and Shrinithivihahshini 2017).

This represents that for three years, these sites are considered to be constantly contaminated with such specific pollutants. The maps also show the lowest concentration of each parameter that reflects as comparatively safer sites.

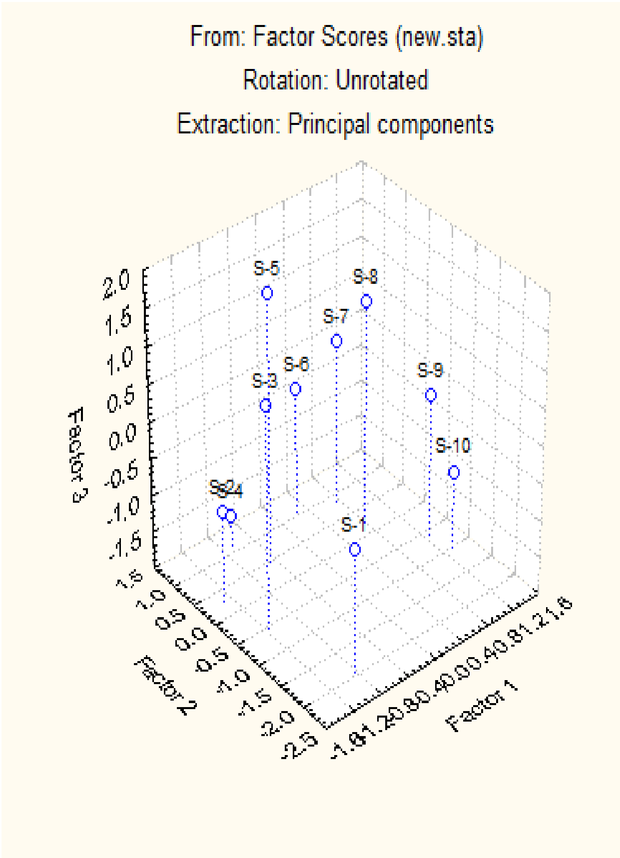

Figure 2a. Principal component analysis ordination (3D) of physico-chemical parameters of seawater at 10 sampling sites at Karachi coast (2015).

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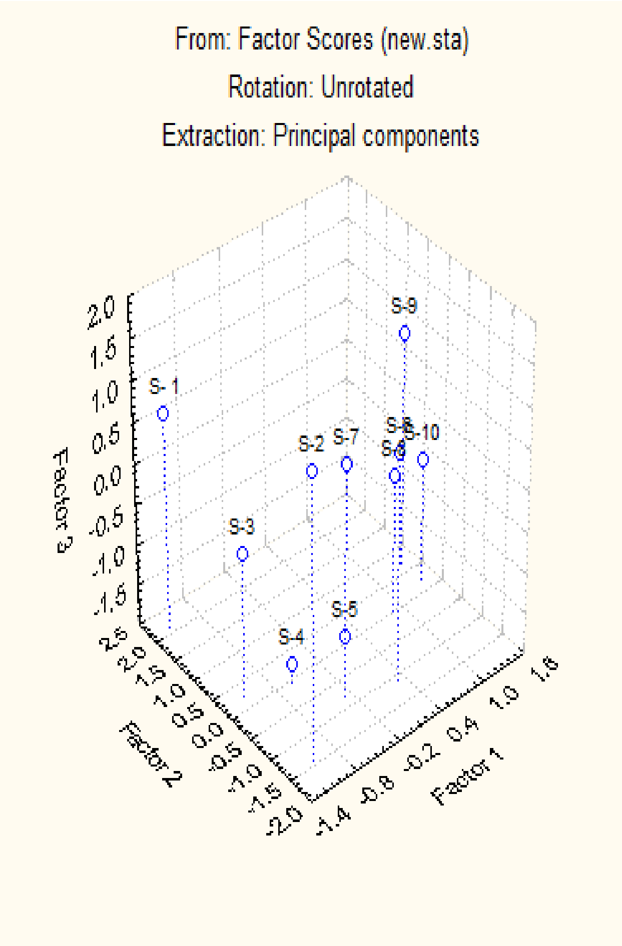

Figure 2b. Principal component analysis ordination (3D) of physico-chemical parameters of seawater at 10 sampling sites at Karachi coast (2016).

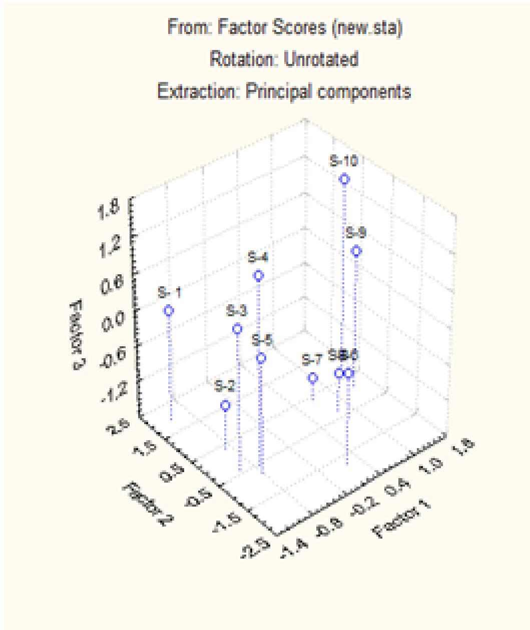

Figure 2c. Principal component analysis ordination (3D) of physico-chemical parameters of seawater at 10 sampling sites at Karachi coast (2017).

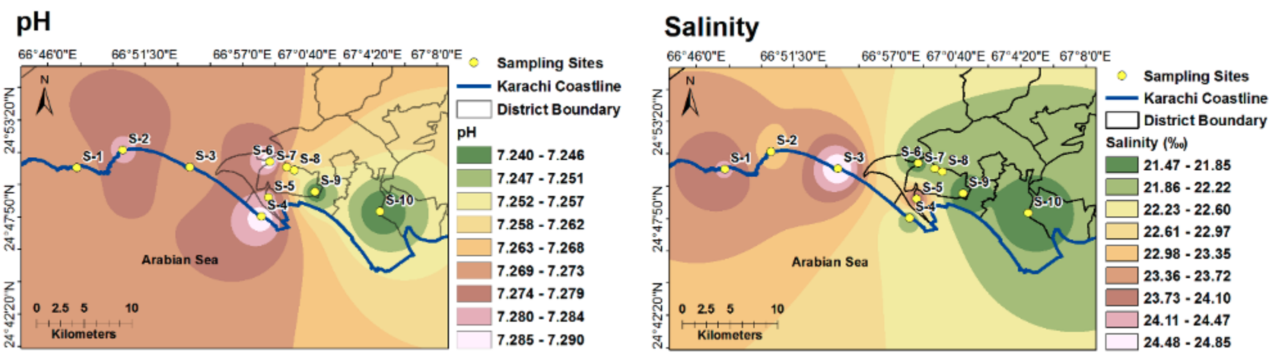

Figure 3. *Three years (2015-2017) average concentration of physical parameters i.e. pH and salinity based on GIS.*

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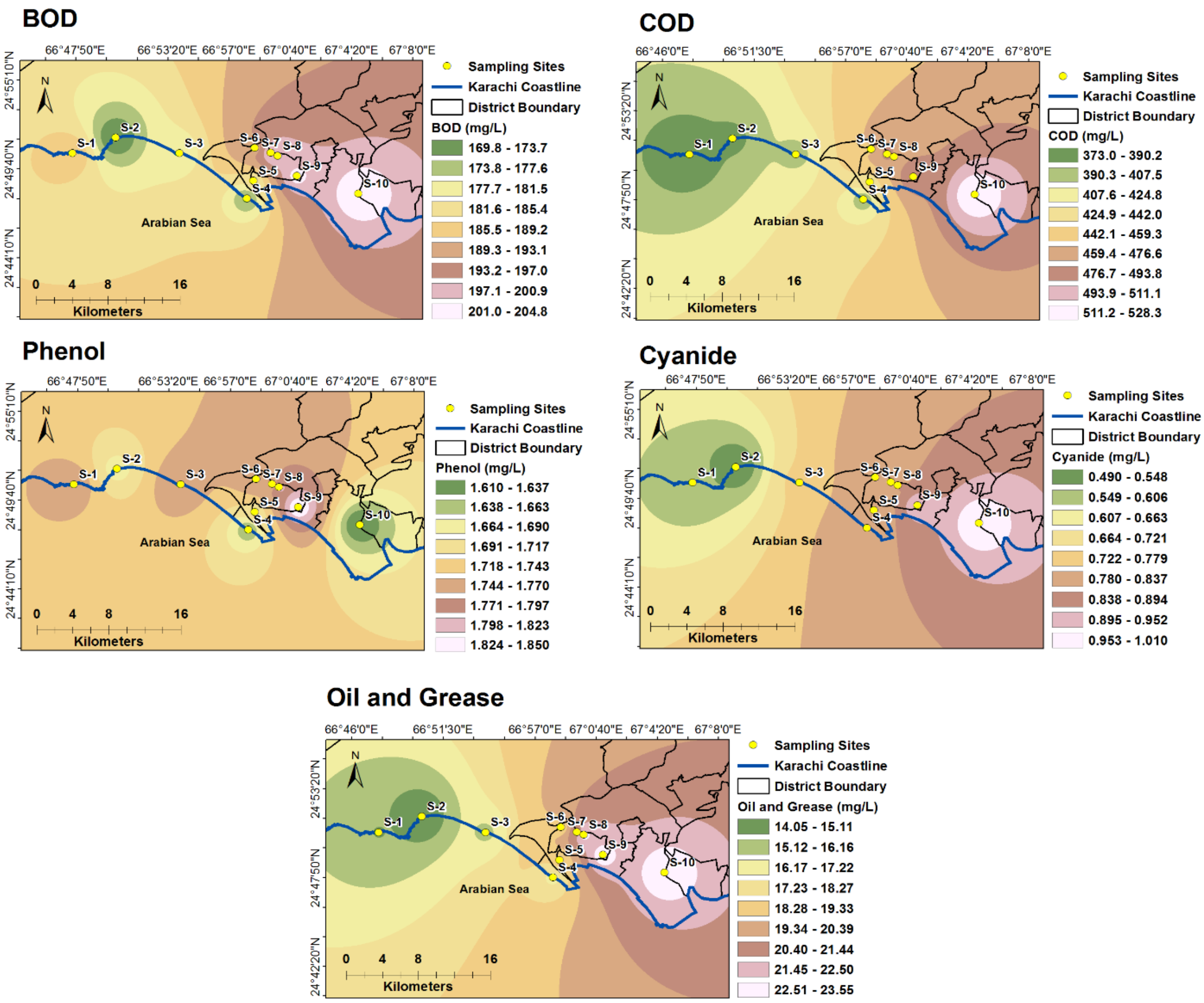

Figure 4. *Three years (2015-2017) average concentration of chemical parameters (BOD, COD, Phenol, Oil and grease and Cyanide) based on GIS.*

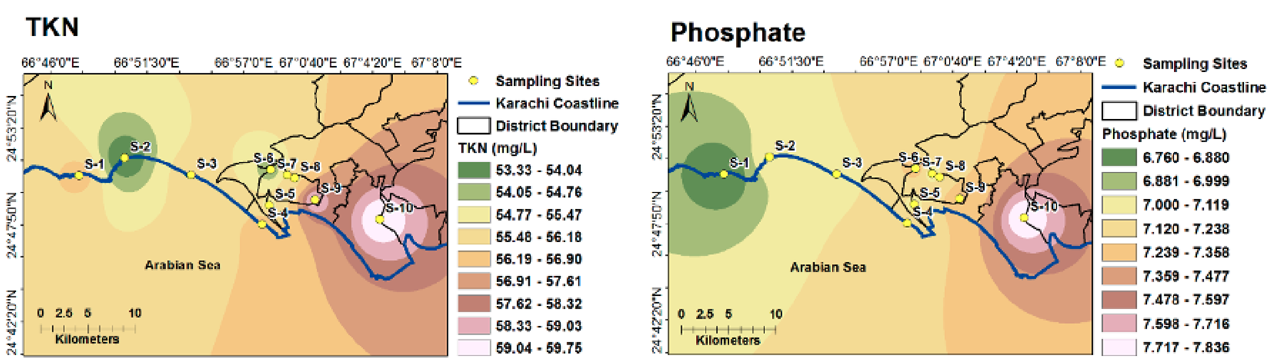

Figure 5. *Three years (2015-2017) average concentration of nutrients (TKN and Phosphate) based on GIS.*

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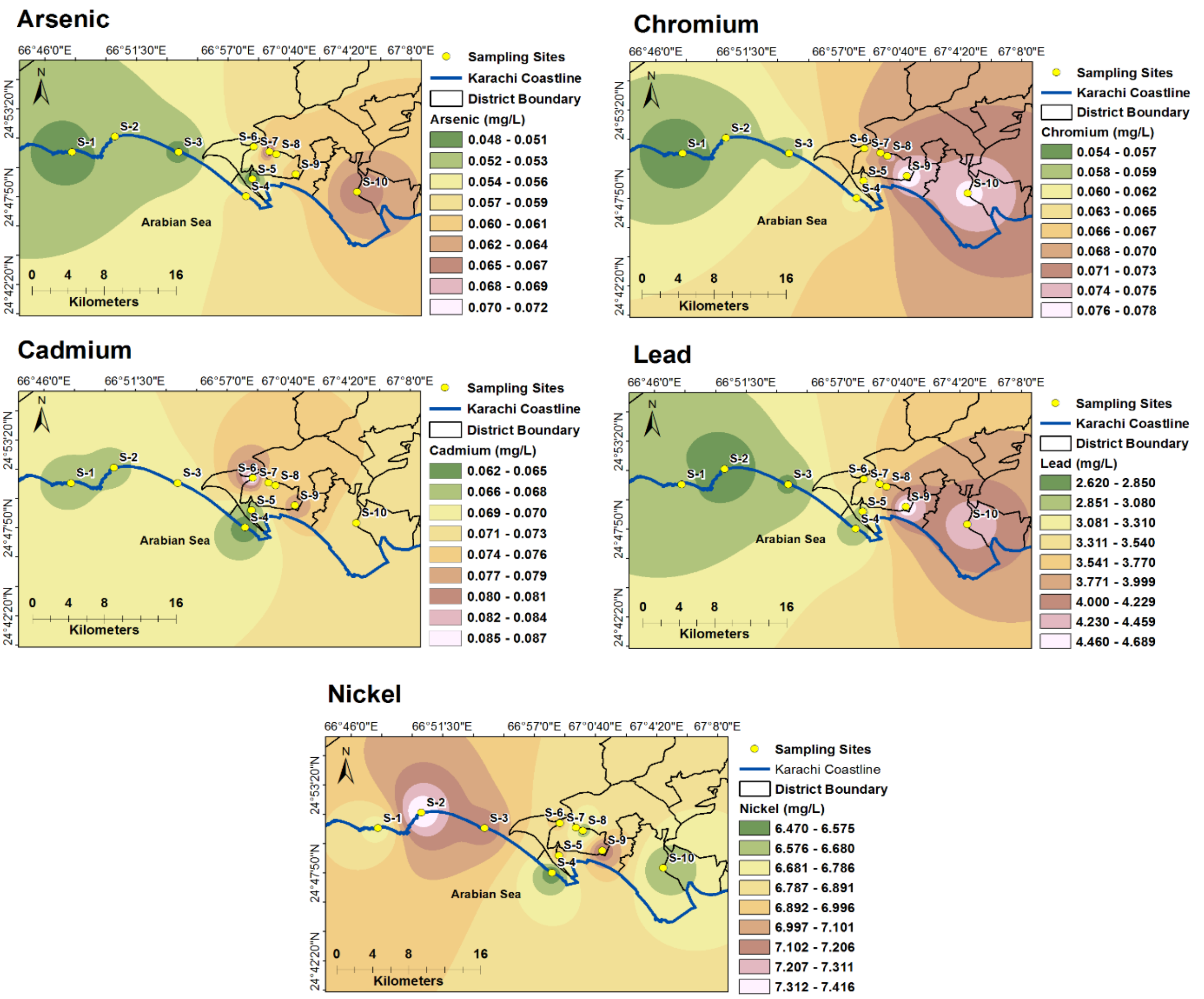

Figure 6. *Three years (2015-2017) average concentration of heavy metals (As, Cr, Cd, Pb, Ni) based on GIS.*

Conclusions

The present research concluded that the Karachi coast is contaminated with industrial and domestic discharges leading to heavy metals toxicity in the food chain. Not only aquatic life is disturbed by chemical and metal contamination but also human being is affected through contaminated seafood. Low dissolved oxygen, high organic pollution and inorganic nutrients in seawater represent a bad picture that adversely affects the ecosystem and needs strict action and remediation strategies.

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