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Original Research

Assessing Heavy Metal Contamination in Oil and Gas Well Drilling Waste and Soil in Pakistan

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Abstract

TThe contamination of soil and water with heavy metals is one of the major environmental problems in the world. Oil and gas exploration and production activities contaminate the environment with heavy metals. In this study, heavy metal concentration was assessed in drilling waste discharges of different oil and gas wells at Khyber Pakhtunkhwa (KP), Pakistan. Moreover, the concentration of heavy metals in the soil surrounding the drilling waste discharges was determined. The representative samples were collected from seven oil and gas drilling waste discharges and the surrounding soil. The collected samples were analyzed for selected heavy metals (Ba, Pb Cr, Cd, Zn, Mn, and Ni) by atomic absorption spectrometric standard methods. Analysis showed that oil and gas well drilling operation waste is enriched with determined heavy metals. The high concentration of heavy metals, particularly Ba and Pb, was also found in the surrounding soil samples. In particular, Ba concentration varied from 1050 to 4168 mg/kg soil. Statistical correlational analysis depicted a common origin of the heavy metals in the soil of the study area, potentially the drilling waste discharges from oil and gas wells. The concentration of heavy metals in oil and gas well drilling waste discharge high, and also affects surrounding soils.

Keywords: oil and gas, drilling, waste discharges, soil, heavy metals, contamination

Introduction

Oil and gas well drilling waste discharges contain toxic substances that are potentially harmful to the ecosystem [1]. Drilling waste is one of the largest volumes of waste generated during oil and gas exploration and production activities [2]. The presence of heavy metals in drilling waste discharges poses a risk of contaminating the environment [3-4]. The heavy metal accumulation in soil adversely affects its properties, which could ultimately lead to infertility and lowered crop yield [5]. Moreover, the soil accumulated with heavy metals could restrict biodegradation of organic contaminants, and heavy metals could enter the food web through biomagnification and induce toxicity

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in animals and humans [6-7]. The continuous leaching of heavy metals can also contaminate subsurface water resources. Because heavy metals are highly toxic and carcinogenic [8], heavy metals contamination of soil and subsurface water poses a major risk of affecting human health (through different routes of entry) [9] and other environmental components [10].

In the vicinity of industry, soil contamination because of heavy metal-enriched industrial discharge is a serious environmental issue across the globe [11-12]. In oil and gas well drilling operations, the heavy metals are mainly allied with the composition of drilling fluids and their additives. Many chemicals in the form of drilling fluids injected during drilling operations are consequently disposed of in the waste pit. Heavy metals constituting drilling fluids and additives are of concern because of their tendency to leach from the waste pit into the surroundings, causing environmental contamination [13]. Wastewater from oil and gas well drilling operations is enriched with heavy metals, including barium and lead. Studies have been conducted to assess the impact of drilling waste discharge in the surroundings of the oil and gas well drilling operations [14-16].

Oil and gas exploration and production is one of the major and important industrial activities in KP Province, Pakistan. Waste discharge from oil and gas exploration and production activities have the potential to contaminate the soil, air, surface, and subsurface water that could lead toward serious direct and indirect health problems. The enhanced environmental degradation due to oil and gas well drilling operations adversely affects the surrounding environmental conditions [17].

A large number of industries, including oil and gas installations located in Pakistan, are not equipped with or have no centralized waste treatment and specifically liquid waste treatment provisions, and therefore waste is being discharged into drains, pits, and inland areas [18-19]. Due to heavy metal contamination, outsized land surrounding the industrial sites has become unsuitable for agronomy [20]. Surface and subsurface water contamination due to heavy metals is also a major threat to human health in the vicinity of industrial activities [21-22].

It is important to require assessment of the level of heavy metals in the oil and gas well drilling operation's effluent and in the surrounding contaminated soil. The soil bearing higher concentrations of heavy metals poses serious risks for ecosystem [23]. It is of great importance that soil must be regularly monitored for heavy metal contamination, especially in the vicinity of industrial installations [24]. In an earlier study, Tariq et al. [25] depicted heavy metal pollution levels in subsurface water and soil samples in Peshawar (KP), Pakistan. They showed that industrial waste discharge is one of the main contributing causes of subsurface water and soil contamination in areas surrounding the industrial installations.

The current study aimed at determining heavy metals (Ba, Pb, Cr, Cd, Zn, Mn, and Ni) in oil and gas well drilling waste discharges and in the waste discharges affecting land by adopting atomic absorption spectrophotometric standard methods. The probable sources of potential heavy metal contamination in soil were identified through statistical correlation between analyzed metals. It is believed that this study will contribute to future monitoring of environmental aspects with respect to environmental pollution, strategies to remedy the contaminated environment and identify cost-effective indigenous environmental management options for oil and gas sector's waste discharges posing the potential to affect soil and subsurface water in KP, Pakistan.

Materials and Methods

Study Area Description

An approximate 143,619.69 km^2 area in Pakistan and 18,890.66 km^2 area (approximately 13.15% of the total area) in KP Province is under oil and gas exploration activities. The total recoverable crude oil reserves in KP are around 148 million barrels, while the recoverable natural gas reserves are around 2.321 trillion cubic feet (TCF), and probable crude oil reserves are more than 500 million barrels and probable natural gas reserves are over 9 TCF. The province is providing low-risk opportunity for oil and gas exploration due to the higher success ratio of oil and gas wells being drilled. Major reservoirs of oil and gas that have been explored in Karak and Kohat Districts depict encouraging oil and gas exploration targets in the region [26]. More than 10 oil and gas fields have been discovered in the districts of Kohat and Karak subbasin areas, with approximate production of 16279021 billion barrels of crude oil and 131207 million cubic feet of natural gas [26-27]. The higher success ratio and the encouraging oil and gas production are positive benchmarks for future oil and gas exploration prospects of KP. This rising trend of oil and gas exploration would also enhance environmental impacts due to the increased oil and gas drilling activities and subsequent waste discharges.

Collection of Drilling Waste Discharges and Affected Soil Samples

Seven different oil and gas well drilling sites located in KP were identified as the sampling area and representative waste discharge effluent samples were collected from each drilling site following the standardized waste discharge sampling guidelines [28]. The waste discharge effluent samples were collected in 1.5 liters PET bottles/containers. The bottles/containers were clearly marked and labeled for the identification of the sample. The liquid samples after collection were stored for preservation in an icebox and shifted to a laboratory on immediate basis for further processing and analysis. In order to attain the accuracy and representativeness, composite waste discharge samples were collected from each sampling point.

A column-sized 35 × 35 × 35 cm was bored using a hand auger to collect soil samples from the discharge-affected site [29]. At each sampling point, soil was homogenized and approximately 1.5 kg of soil sample was withdrawn and placed in a sealable plastic bag. On random basis the sampling sites were selected for soil sampling through visual examination of each site, affected due to oil and gas well drilling operation's waste discharges. Seven soil samples were withdrawn from points suitably distant from each selected oil and gas drilling site to establish heavy the metals' background levels. The sampling points for the collection of oil and gas discharges, affected soil, and background soil samples are depicted in Fig. 1.

Digesting Samples for Heavy Metals Determination

In order to determine realistic heavy metal contents in samples – specifically in soil samples – prior to analysis acid digestion was strongly recommended for soil samples because heavy metals, mostly in soil, are bonded strongly with mineral matrix [30-31]. An earlier study identified that in soil, exchangeable and water-soluble contents hardly surpass 3% of the total concentration of metals [32]. The digestion approach adapted for this study assumed that soil was the most contaminated component of the environment around oil and gas drilling waste discharge sites in KP, Pakistan.

Keeping in view the significant impacts of heavy metal accumulation in soil as degradation in fertility of soil and lowered crop yield, the research like the current study, aimed at the analytical assessment of heavy metal concentration in waste discharges and affected soil cannot be executed simply without digestion. For the précised determination of realistic amount of heavy metals in soil, the acidified digestion filtrate / extract of samples were analyzed [30].

Acid digestion of samples was carried out as described earlier [30]. Briefly, air-dried soil (0.5 g) was taken from a thoroughly homogenized soil sample, ground, placed in a conical flask, and 2 ml nitric acid (70%), 3 ml hydrofluoric acid (40%), and 6 ml hydrochloric acid (35%) were added. A transparent solution was attained by refluxing the mixture on a heating plate. The attained solution was cooled and to remove excessive hydroflouric acid, and 15 ml of saturated boric acid was added and after mixing volume was made up to 100 ml with the help of interference free double de-ionized water [30-31]. The obtained solution was filtered on Whatman grade 42 paper (Whatman International, UK) and filtrate was utilized for analytical determination of heavy metals concentration in soil. The drilling waste discharge liquid samples were also digested with nitric acid and hydrochloric acid as mentioned above [30].

Analyzing Oil and Gas Drilling Waste Discharge Effluent and Soil Samples

Atomic absorption spectrophotometry (AAS) was utilized for determining selected heavy metals (Ba, Pb,

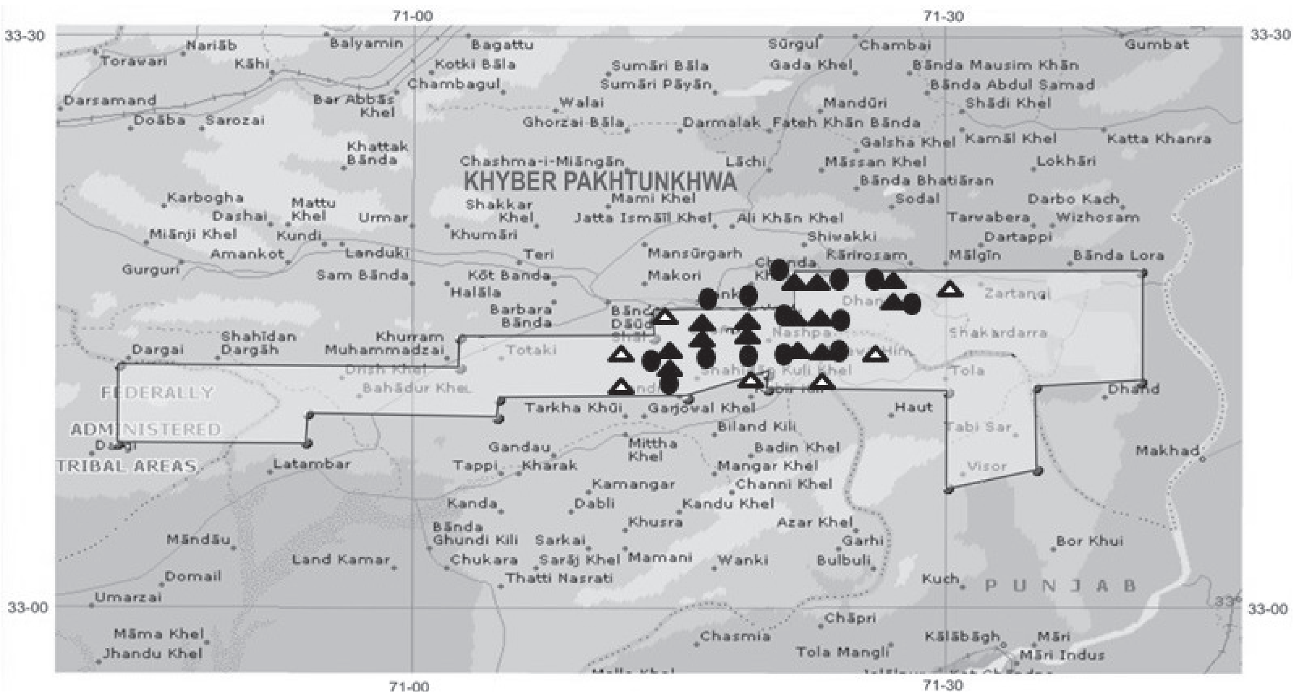

Fig. 1. Identification of sampling points of oil and gas well-drilling waste discharges (●), contaminated soil (▲), and background soil (Δ) in highlighted study area of Khyber Pakhtunkhwa, Pakistan.

Cr, Cd, Zn, Mn, and Ni) in oil and gas well-drilling waste effluent, background soil, and the affected soil samples [31]. The digested wastewater and soil extract were analyzed after dilution with double-distilled water, where required.

Quality Assurance and Quality Control

Following the sampling guidelines and standard analytical methods [30-31], the concentration of each metal was determined using the standardized analytical conditions and AAS system. The blank was prepared and carried out through the steps of analytical process in the same way as sample solutions/extracts were prepared for analytical determination of heavy metals. Precision of the analytical methods, equipment, and accuracy of the results were checked through standard reference material [31]. The sets of results matched within ±1.0 to ±1.5%.

Heavy Metals Assessment Method

The concentrations of heavy metals in the drilling waste discharge and the affected soil samples were assessed via single pollution index (SPI) for each heavy metal using Equation 1:

$$P_i = \frac{C_i}{L_i} \tag{1}$$

…where *Pi* is the single pollution index, *Ci* represents the determined concentration of *i* parameter, and L_i is permissible value for *i* parameter [4, 33].

The Nemerow composite pollution index (NPI_c) was also determined to evaluate the pollution level of the most contaminated elements using Equation 2:

$$\mathrm{NPI}_c = \sqrt{\frac{(P_{imax})^2 + (P_{i\,avg})^2}{2}} \tag{2}$$

…where P_{imax} is the maximum value of SPI of all tested heavy metals and P_{iavg} is mean of the SPI of all tested heavy metals [34-36]. Being comprehensive pollution indices, SPI and NPI_c were used to assess the heavy metal pollution levels in drilling waste discharge and the affected soil.

Data Analysis

The acquired data was processed and statistically analyzed for the statistical parameters through utilization of IBM SPSS Statistics version 22 software [37]. Basic statistical parameters (mean, median, standard deviation, skewness, minimum, and maximum) were calculated to analyze the data statistically for the selected heavy metals distribution in drilling waste discharges and the affected soil. Item-to-item correlation between studied heavy metals in drilling waste discharge and soil was also determined statistically, and metal-to-metal correlation matrices were also developed.

Table 1. Heavy metal concentrations in oil and gas well-drilling waste effluent.

Parameter	Unit	Mean value	NEQS
Ba	mg/l	51.00	1.5
Pb	mg/l	47.31	0.5
Cr	mg/l	1.69	1.0
Cd	mg/l	0.19	0.1
Zn	mg/l	2.69	5.0
Ni	mg/l	0.20	1.0
Mn	mg/l	1.02	1.5

National Environmental Quality Standards (NEQS), Pakistan

Results and Discussion

Heavy Metal Assessment in Oil and Gas Well Drilling Waste Discharges

Environmental monitoring could assess that the waste discharge regulatory requirements are sufficiently protective or otherwise [38]. Analytical characterization of selected heavy metals in the drilling waste discharge showed that it is enriched with heavy metals and contained high values of Ba, Pb, Cr, and Cd, and these exceed the permissible limits under the provisions of National Environmental Quality Standards (NEQS), Pakistan [39]. The mean values of heavy metals determined in the drilling waste discharge samples and their comparison with Pak-NEQS is shown in Table 1. Among determined heavy metals, Ba (51 mg/l) and Pb (47.31 mg/l) exhibited higher mean concentrations than other heavy metals. The higher concentration of the heavy metals in the drilling waste discharge is attributed to the constituents of the drilling fluid additives used while drilling the oil and gas well [14]. The statistical parameters of data processed depicting the distribution of selected heavy metals in drilling waste discharges in terms of the concentration is presented in Table 2, and statistical metal-to-metal correlation of oil and gas well drilling discharge is given in Table 3.

The statistical parameters of single pollution index (SPI) and Nemerow composite pollution index (NPI_c) for the selected heavy metals in oil and gas drilling waste discharges are summarized in Table 4. The SPI is calculated against the Pak-NEQS for each selected heavy metal in order to assess the degree of contamination of selected heavy metals in oil and gas drilling waste discharges as per SPI categorization [33-35]. The mean of SPIs of the selected heavy metals was found in the order as follows: Pb (94.617) > Ba (33.999) > Cd (1.885) > Cr (1.687) > Zn (0.538) > Ni (0.198). The mean SPI of Pb and Ba falls in Class V (i.e., severely polluted), Cd and Cr falls in Class II (i.e., slightly clean), and Zn and Ni falls in Class I (i.e., clean). The Nemerow composite pollution index (NPI_c) is also calculated to comprehend

Table 2. Statistical parameters of selected heavy metals (mg/l) in oil and gas-drilling waste discharge.

	Ba	Pb	Cr	Cd	Zn	Ni	Mn
Mean	50.999	47.307	1.687	0.188	2.692	0.196	1.024
Median	52.690	48.515	1.827	0.184	2.800	0.179	1.000
Standard deviation	8.285	3.397	0.306	0.029	0.366	0.072	0.140
Skewness	-0.421	-1.368	-0.633	0.188	-0.239	1.423	0.515
Minimum	38.140	39.950	1.190	0.150	2.101	0.120	0.870
Maximum	61.590	51.020	2.010	0.240	3.200	0.350	1.250

Table 3. Metal-to-metal correlation matrix in oil and gas well-drilling waste discharge.

	Ba	Pb	Cr	Cd	Zn	Ni	Mn
Ba	1.000						
Pb	0.827	1.000					
Cr	0.823	0.814	1.000				
Cd	0.690	0.669	0.696	1.000			
Zn	0.837	0.765	0.911	0.726	1.000		
Ni	0.793	0.567	0.689	0.696	0.728	1.000	
Mn	0.703	0.426	0.573	0.561	0.736	0.767	1.000

Table 4. The single pollution index (SPI) and Nemerow composite pollution index (NPI_c) for the selected heavy metals in oil and gas-drilling waste discharge.

	SPI						NPI_c
	Ba	Pb	Cr	Cd	Zn	Ni	
Mean	33.999	94.617	1.687	1.885	0.538	0.198	68.253
Median	35.126	97.030	1.825	1.850	0.560	0.180	70.001
Standard deviation	5.522	6.797	0.306	0.305	0.073	0.071	4.950
Skewness	-0.421	-1.367	-0.630	0.176	-0.239	1.454	-1.332
Minimum	25.430	79.900	1.190	1.500	0.420	0.120	57.610
Maximum	41.060	102.040	2.010	2.400	0.640	0.350	73.710

the collective pollution level of selected heavy metals in oil and gas-drilling waste discharges. The mean NPI_c was 68.253, the value falling within in Class V (i.e., severely polluted) [33-35].

Heavy Metals Assessment in Soil

The heavy metal concentrations in the drilling waste affected soil and their allowable levels prescribed by United States Louisiana Statewide Order (US LSO) 29B Standard [40] (literature review revealed that there is not any standard found in Pakistan) were compared (Table 5). The selected heavy metals (Ba, Pb, and Cd) concentrations were higher than the allowable standard values. The Zn, Cr, and Ni values were found to be below the standard values. The statistical parameters of data processed depicting the selected heavy metals distribution in drilling waste discharge affected soil. The mean values of Ba, Pb, and Cd were 2077.43 mg/kg, 1015.76 mg/kg and 12.77 mg/kg, respectively, in the drilling waste-affected soil (Table 6). The high Ba, Pb, and Cd concentrations were a consequence of the heavy metal-rich drilling waste discharges in the area.

The oil and gas well drilling operations generate mainly two types of waste, i.e. drill cuttings and drilling fluids. The drilling fluids are mainly utilized to facilitate the drilling operation; fluids could be oils (synthetic or natural), air, water, foam, or could be a combination of these components. The additives used in

Table 5. Heavy metal concentration in waste discharge-affected soil samples.

Parameter	Unit	Mean value	US LSO 29B standard
Ba	mg/kg	2077.43	2000
Pb	mg/kg	1015.76	500
Cd	mg/kg	12.77	10
Zn	mg/kg	402.44	500
Cr	mg/kg	163.75	500
Mn	mg/kg	14.38	NG
Ni	mg/kg	11.60	72

NG = not given in the list

drilling fluids contain potentially toxic heavy metals and other elements. The major sources of contamination in drilling waste are hydrocarbons, corrosion inhibitors, drilling fluid chemical components, and reservoir fluids, and the remaining or unused drilling chemicals are discarded as drilling waste [41]. The other selected heavy metals (Zn, 402.44 mg/kg; Cr, 163.75 mg/kg; Ni, 11.60 mg/kg; and Mn, 14.38 mg/kg) were found in concentrations lower than the prescribed standards in the study area. However, the levels detected in this study are considerably higher than the levels studied in other effluent-contaminated soils [19-20, 25].

On the basis of statistically processed mean values, the selected heavy metals in waste discharge affected soil found in the following order: Ba > Pb > Zn > Cr > Cd > Mn > Ni. Barium, lead, and other selected heavy metals showed concentrations higher than the normally expected concentration values in soil, raising concern regarding soil fertility in the study area [12]. The statistical processing of data revealed a higher degree of skewness and standard deviation that shows that the selected heavy metals in the discharge affected soil are not following normal distribution. A strong positive metal-to-metal correlation was found through statistical analysis for Cr-Pb, Cd-Pb, Cd-Cr, Zn-Pb, Zn-Cd, Ni-Pb, Ni-Cr, and Ni-Cd pairs (Table 7), showing that the selected heavy metals in discharge-affected soil have the same origin. This proved to be a correlational concentration dependence of metals and their source in the soil of the study area. The statistical correlations matrix highlighted several significant correlations that indicate a common source of heavy metal discharge in the area of study. This might be endorsing the fact that soil sampling was carried out from sites located in close vicinity to oil and gas well drilling waste discharge-affected sites.

The statistical parameters of SPI and NPI_c for the selected heavy metals in the soil contaminated with drilling waste discharge are summarized in Table 8. The SPI is calculated against the U.S. standards for each selected heavy metal in order to assess the degree of contamination of selected heavy metals in drilling waste discharge-affected soil as per SPI categorization.

Table 6. Statistical parameters of the selected heavy metals (mg/kg) distribution in the soil affected by oil and gas-drilling waste discharge.

	Ba	Pb	Cr	Cd	Zn	Ni	Mn
Mean	2077.434	1015.761	163.751	12.773	402.436	11.603	14.376
Median	1870.350	688.395	162.100	11.750	300.500	11.500	14.500
Standard deviation	961.419	710.879	31.233	3.822	232.369	1.059	0.667
Skewness	1.426	0.542	0.123	0.639	0.303	0.747	-0.223
Minimum	1050	116	120	8	160	10.12	13.25
Maximum	4168	2154	215	19	698	14.10	15.25

Table 7. Metal-to-metal correlation matrix in the soil affected by oil and gas well drilling waste.

	Ba	Pb	Cr	Cd	Zn	Ni	Mn
Ba	1.000						
Pb	-0.124	1.000					
Cr	0.074	0.707	1.000				
Cd	0.138	0.860	0.681	1.000			
Zn	0.384	0.821	0.664	0.919	1.000		
Ni	-0.341	0.641	0.790	0.521	0.385	1.000	
Mn	-0.248	0.211	0.199	0.387	0.291	0.100	1.000

Table 8. Single pollution index (SPI) and Nemerow composite pollution index (NPI_c) for the selected heavy metals in the soil affected by oil and gas drilling waste discharge.

	SPI						NPI_c
	Ba	Pb	Cr	Cd	Zn	Ni	
Mean	1.038	3.385	0.327	1.277	0.804	0.161	2.549
Median	0.935	2.294	0.324	1.175	0.601	0.159	1.836
Standard Deviation	0.480	2.369	0.062	0.382	0.464	0.014	1.688
Skewness	1.426	0.542	0.123	0.639	0.303	0.749	0.572
Minimum	0.530	0.390	0.240	0.850	0.320	0.140	0.470
Maximum	2.080	7.180	0.430	1.980	1.400	0.201	5.270

Table 9. Classes of the single pollution index (SPI) and Nemerow composite pollution index (NPIc) of the soil affected by oil and gas-drilling waste.

Class	I	II	III	IV	V
Category of heavy metal contamination	Clean	Slightly clean	Soil pollution exceeds background level	Moderately polluted	Severely polluted
SPI	≤1.0	>1.0 ≤ 2.0	>2.0 ≤ 3.0	>3.0 ≤ 5.0	> 5.0
NPIc	≤0.7	>0.7 ≤ 1.0	>1.0 ≤ 2.0	>2.0 ≤ 3.0	> 3.0

The mean of SPIs of the selected heavy metals in wastewater-affected soil is in the order: Pb (3.38) > Cd (1.27) > Ba (1.03) > Zn (0.80) > Cr (0.32) > Ni (0.16) (Table 9); the mean SPI of Pb falls in Class IV (moderately polluted), Cd and Ba are in Class II (slightly clean), and Zn, Cr, and Ni fall in Class I (clean).

The NPI_c is also calculated to comprehend the collective pollution level of selected heavy metals in the soil affected by oil and gas drilling waste. The mean NPI_c was 2.54 and the value falls in Class III (pollution exceeds background level), while the maximum NPI_c is 5.27 and the value falls in Class V (severely polluted).

A comparison of concentrations of heavy metals in background soil samples and in the soil affected by oil and gas drilling waste is presented in Fig. 2.

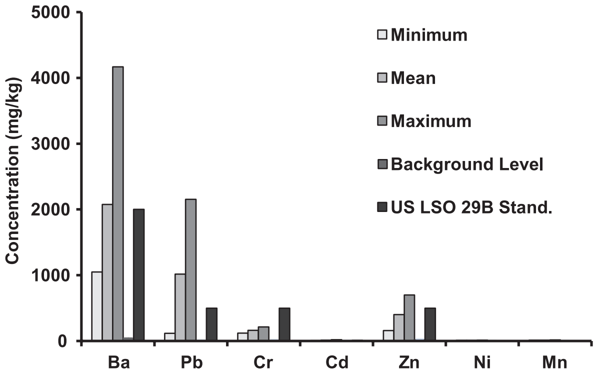

Fig. 2. Heavy metal concentrations in waste discharge-affected soil and background soil samples in comparison with U.S. LSO 29B standard.

The comparison showed that determined heavy metals were in higher concentrations in oil and gas well-drilling waste discharge-affected soil as compared to background soil samples. This data revealed that the contamination of surrounding soil was due to waste discharged from oil and gas well-drilling activity. Environmental pollution in the area under research, specifically due to accumulation of Ba and Pb, needs indigenous cost-effective environmental management and remediation planning for the treatment of soil to improve its fertility [42-43]. In developing regions where limited finances are designated for environmental management, cost-effective and sustainable natural soil remediation and environmental restoration options are required [44]. Bioremediation is an appropriate and cost-effective indigenous remediation technique for the treatment of such environmental contaminants [45-46].

To our knowledge and literature review, this study is the first where a detailed assessment of terrestrial oil and gas drilling waste discharge and affected soil of KP, Pakistan has been conducted. This study identified the current concentration of heavy metals in soil of the area. A high concentration of heavy metals can harm soil fertility and, eventually, human health through bioaccumulation and biomagnification in the food web [47].

Conclusions

In the study area of KP, where oil and gas well-drilling operations release wastewater in the environment without any treatment, the concentrations of heavy metals in wastewater exceed the NEQS. The concentrations of heavy metals (i.e., Ba and Pb) were found to be 34 to 94 times higher in wastewater than the permissible limits and mean SPI values endorsed in the severely polluted category of the wastewater. The mean NPI_c values of selected heavy metals also depicted a severely polluted class of wastewater. The soil in the close vicinity of oil and gas drilling waste discharge sites were found to be potentially affected by higher concentrations of selected heavy metals. The concentrations of Pb, Ba, and Cd were found to be significantly higher as compared to other selected heavy metals. Metal-to-metal statistical correlation analysis established that selected heavy metals in waste discharges and affected soil have a similar source. Additional studies are required to address the further issues of the study area for the assessment of other contamination in the surface and subsurface water resources.

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Conflict of Interest

The authors declare no conflict of interest.

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