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RESEARCH PAPER

Investigation of Heavy Metal Traces during Drilling Operations in the Bibi Hakimeh Oil Field, Iran

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ABSTRACT
Drilling operations release considerable amounts of drilling fluids that contain pollutants such as heavy metals and hydrocarbons. By means of the ICP-OES device, the concentration of metals in the drilling mud deposited in the accumulation pools (around the drill rigs), the drilling mud used in the well (initial, input, and output mud), the soil around the drilling rig, and the control sample can be determined. Comparison of metal accumulation volume with current standards (upper crust and the region's soil) indicates that the initial drilling mud contains As, Cd, Mo, Cu, and Ag metals. Also, the outflow and associated cuttings are contaminated with Sb, showing a geological formation origin for this metal. In case of Pb, Ba, Cr, and Mn, the level of contamination is in a medium range wherein the source of the contamination is recycled mud and drilling rocks in the previous phases. Contamination of other metals is evaluated in the low range. Geo-accumulation index (Igeo), Enrichment Coefficient (EF), and Contamination Factor (CF) are used to determine the level of pollution. Results show that As, Pb, Ba, Mn, Mo, Cd, Co, Cu, and Zn have caused a lot of pollution in the area and their amounts should be controlled.
Keywords: geo-accumulation index, mud waste, contamination Factor, enrichment coefficient

INTRODUCTION

The oil industry has been in existence for over a hundred years in Iran. The drilling industry is one of the main sectors and most specialized industrial activities in the world. Bibi Khakimeh oil field with 70 km long and 5 km wide, is located in the southwestern part of Iran, 210 km southwest of Ahwaz and south of Gachsaran oil field (Fig.1). This field consist of Asmari, Bangestan and Khami that Khami reservoir is included Fahlian, Gadvan and Darian Formation reservoirs (Fig 2).

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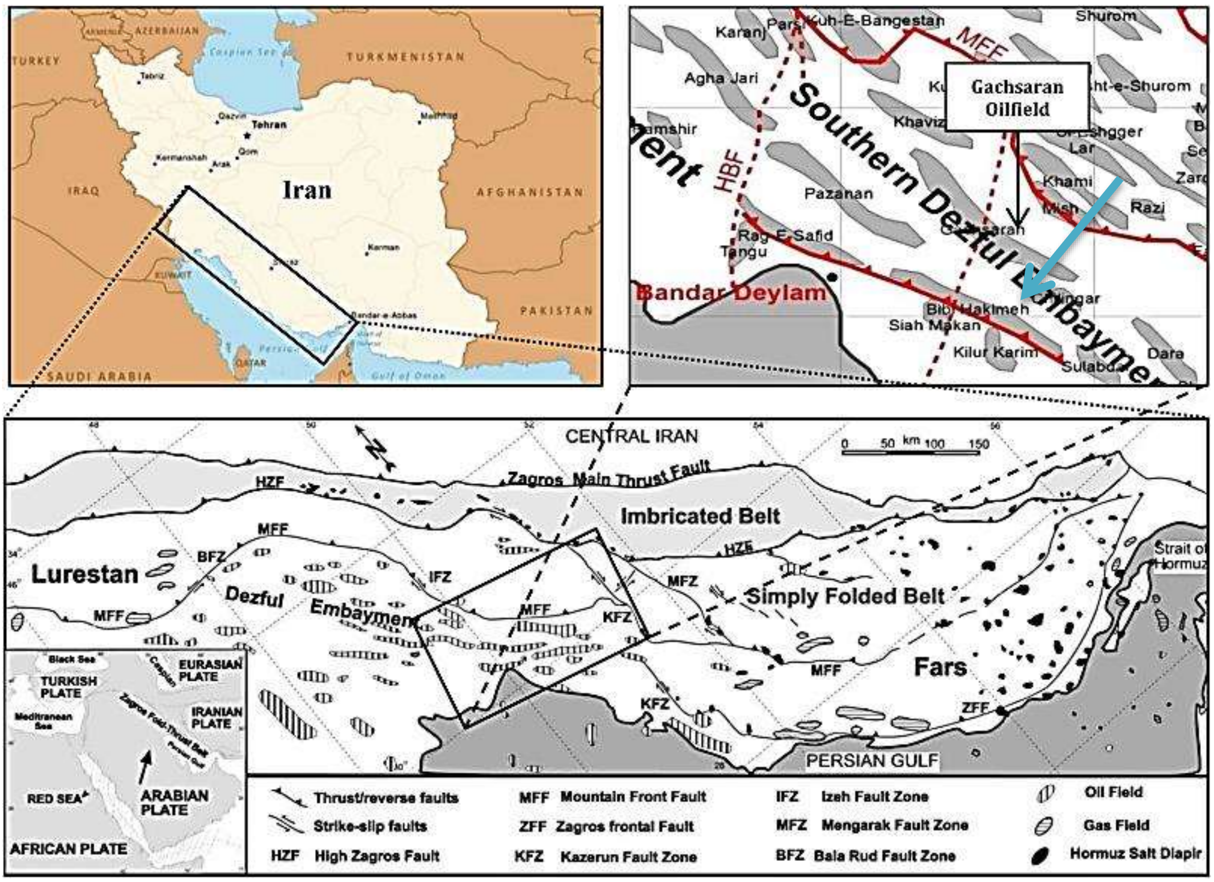

Figure 1: Geographic location of Bibi Hakimeh oil field (blue arrow)

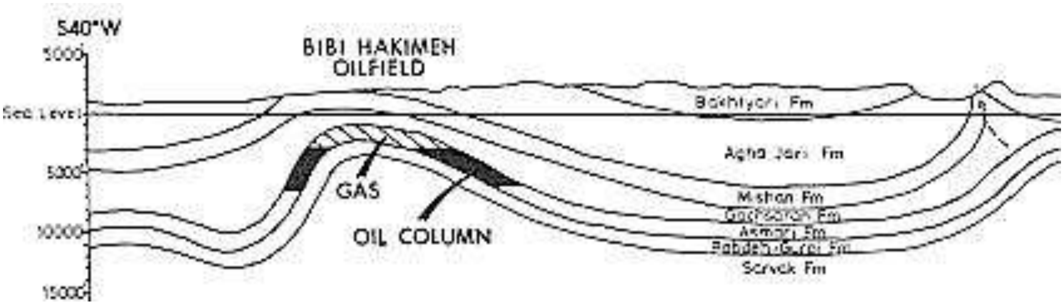

Figure 2: Stratigraphy and Fault Pattern of Bibi Hakimeh oil field

Over the long history of offshore development of oil and gas resources, a large number of different chemical formulations of drilling fluids have been used. The different types of drilling fluids can be separated into two basic types: water based fluids (WBFs) and non-aqueous based fluids (NABFs) (Afkhami et al., 2013). When a liquid is used as a fluid, even if it comes with oil or water, it is generally called a "mud." About 85% of the world's wells are drilled using water-based fluids (Gao & Chen, 2008). In petroleum-based muds, the main composition includes: 46% of petroleum-based materials such as natural oil, refined petroleum products and their products, 36% of barite, 18% of water, which is often saturated, 2% of mixing material and are 1% of detergents. Detergents are used in oil-based flocs to stabilize water-oil emulsions (Pozebon et al., 2005). Barite, which is used as a bulking agent in drilling operations, is insoluble. The presence of large amounts of barium in drilling wastes is due to the use of barite in drilling (Scholten et al., 2000).

In the periodic table, the metals in groups 3 to 16 are called heavy metals in rotation 4 and thereafter (Moore & Ramamoorthy, 2012). In the dictionary of environmental and conservation of natural resources, heavy metal is defined as: A high-atomic metal element that is usually toxic and tends to accumulate in the food chain, including arsenic, cadmium, chromium, lead, and mercury (Park et al..., 2001). The presence of toxic metals in soil can severely inhibit the biodegradation of organic contaminants (Eghbal et al., 2019) Fixed and mobile sources emit large amounts of heavy metals into the air, plants and soil (Ahmed & Ishiga, 2006; Christoforidis & Stamatis, 2009).

The first step in reducing the effects of these pollutants is to identify the factors that produce them and the amount of each pollutant. It is only after identifying these factors that necessary and appropriate decisions can be made to eliminate or reduce their effects (Doelsch et al., 2006; Jordan et al., 2007; Tijani et al., 2006). Afkhami studied the concentration of heavy metals (Arsenic, Cadmium, Iron, Vanadium and Mercury) in the drill mud waste used in Gachsaran Formation in Ahwaz oilfield (Afkhami et al.,2013).

Due to the activities of the drilling industry, many environmental pollutants is caused to the environment, which can cause pollution in air, water, soil and effects on human health. Heavy metal pollution in industrial environments has become a global phenomenon due to the diversity of human activities (Deepalakshmi et al., 2014). Heavy metals are one of the most important environmental pollutants (Nyarko et al., 2008). These elements have accumulating and carcinogenic properties and can cause various health and environmental problems. The high stability of these metals in water, soil and even the bodies of living things poses major risks to the environment and human health (Solgi et al., 2012). In fact, heavy metals are not excreted after entering the body and accumulate in the tissues, which causes many diseases and complications in the body (Absalon & Ślesak, 2010). For example, in adipose tissue, then nerve tissue and the endocrine system, muscles, bones and joints accumulate, disrupting the body's immune system and cellular metabolism. (Li et al., 2015; Saeedi et al., 2012). As stated, human exposure to heavy metals can occur through various pathways such as swallowing, skin contact and inhalation, among which the swallowing pathway is of particular importance (Meza-Figueroa et al., 2007; Olawoyin et al., 2012; Wu et al., 2015; Zhao et al., 2014). For example, in a number of case studies, the entry of soil into the body through swallowing has been a major source of lead exposure, especially in children, and elevated levels of lead in their blood (Abrahams, 2002).

The waste generated from drilling operations has a large volume and requires environmental studies to understand the waste and how it is formed. The aim of the study is identification and understanding of pollutants that improved functions can be developed to eliminate and reduce any environmental pollution.

MATERIALS AND METHODS

Objectives of sampling and analysis of environmental samples are determined based on the type of project such as control measures, routine monitoring, urgent and urgent surveys and research studies. If sampling is not done well, no matter how accurate the analysis of the samples is, the work done will not be worth it. On the other hand, no matter how well the sampling is done, but if the analysis of the sample is not done well or the correct method is not used, all the activities performed will still be in vain. It is usually necessary for the analyzer to be aware of the nature of the samples and the sampling environment as much as possible in order to obtain results.

After reviews, sampling operations were carried out in winter ٢٠١٧ The well was drilled at a depth of 1635 m using water based mud. In order to identify heavy metals and determine their concentration, as well as determining the origin of these metals, the samples were made from mud tanks, flow in and mud flow out of the well.

To determine the role of the deposited layers in increasing the concentration of metals, samples were taken from shale shaker and waste collected in the pond in different points. In order to do this, from the point of entry into the pond, the middle part of the pond, as well as the side of the pond from 0 to 5 centimeters, samples weighing half a kilogram were removed.

To investigate the effect of drilling operations on the soil, samples were taken from the soil around the drilling rig. Finally, for comparing the concentration of these metals with the amount in the natural area, a sample of the control soil was removed far from the region.

Samples of muds that were jelly and fluffy were placed in glass containers of 250 cc. The samples were then transferred to National Iranian South Oil Company Laboratory. Mud and soil samples were dried at room temperature in the laboratory.

After drying, they were milled and passed through a mesh of 230 mesh and 10 grams of each sample were selected for analysis with the ICP-OES machine for the determination of heavy metals. Samples were prepared for analysis by ICP-OES by dissolution with four acids (HF, HCL, HNO3, HCLO4). After weighing 0.25 g of sample weight, 8 ml of 40% HF and 1 ml of 70% HCLO4 were added and place the prepared solution in a special plastic container in water at a temperature of 200 ° C until a jelly solution is obtained. Then 3.75 ml of 37% HCL and 1.25 ml of 65% HNO3 were added and the solution was reduced to a volume of 25 ml and analyzed by ICP-OES in the geochemical laboratory of the Southern Oilfields Company. Also, to verify the accuracy of the data results, repeat samples were also analyzed. Finally, the concentrations obtained for each metal were compared with standard values. Also, the geoaccumulation index, contamination factor and enrichment factor were determined for each of them.

RESULTS AND DISCUSSION

The concentration of the elements in the samples taken with the upper crust and control sample is shown in table (1).

Table 1: Comparison of metal concentrations with standard values in terms of (ppm)

Metal element	As	Ba	Cd	Co	Cr	Mn	Mo	Ni	Pb	Sb	V	Zn
Fresh mud	2.3	95	0.24	3	45	170	1.74	35	5	0.91	30	27
Inlet mud	5.2	124	0.25	7	83	299	1.72	59	6	1.08	51	41
Out flow mud	3.9	88	0.21	5	52	197	1.78	40	6	0.92	33	24
Waste 1	2.6	168	0.23	7	98	344	1.58	44	6	1.08	41	29
Waste 2	6.8	237	0.27	13	111	507	1.07	98	11	1.1	81	60
Waste 3	7.7	224	0.22	8	102	464	1.42	61	12	1.15	73	40
Waste ave.	5.7	209	0.24	9.5	103.5	438.5	1.35	67.5	9.5	1.11	65	43
Cuttings+mud	5.2	142	0.27	7	61	297	1.21	51	6	0.98	47	51
Soil around	13.2	522	0.28	9	90	532	1.22	64	15	1.21	61	56
Control soil	1.5	246	0.24	9.85	120	431.5	0.97	100	7	0.43	76	65
Crust ave.	1.5	668	0.11	20	100	950	1.4	80	14	0.3	160	75

As shown in table (1), the average values of the concentration of wastes relative to the Ag, As, Cd, Cr, Mo, Sb were higher than the control soil.

The concentrations of As, Cu, Mo in the fresh mud sample are higher than the control sample. Also, the concentration of Ag, As, and Cd in the samples is higher than the crust.

The concentration of As, Cd, Cu, Mo, Sb metals in the mud was higher than the control sample. Also, the concentration of Ag, As, Cd Mo, Sb in samples is higher than crust.

The concentrations of As, Mo, Sb metals in the outflow were higher than the control sample. Also, the concentration of Ag, As, Cd, Mo, Sb in samples is higher than crust.

The concentration of Ag, As, Ba, Cd, Cu, Mn, Mo, Sb metals in the soil around the rig is higher than the control sample. Also, the concentration of Ag, As, Cd, Pb, and Sb is higher than the crust.

The concentrations of As, Cd, Mo, Sb metals in cuttings and mud were higher than the control sample. Also, the concentration of Ag, As Cd, Sb in samples is higher than crust.

In general, all specimens were contaminated with Ag, As, Mo, Cd, due to the presence of these substances in the primary composition of the drilling mud as additives or impurities along with other materials. Sb was found in all specimens except for fresh muds, which probably has a reason for the origin of this metal from drilled formations.

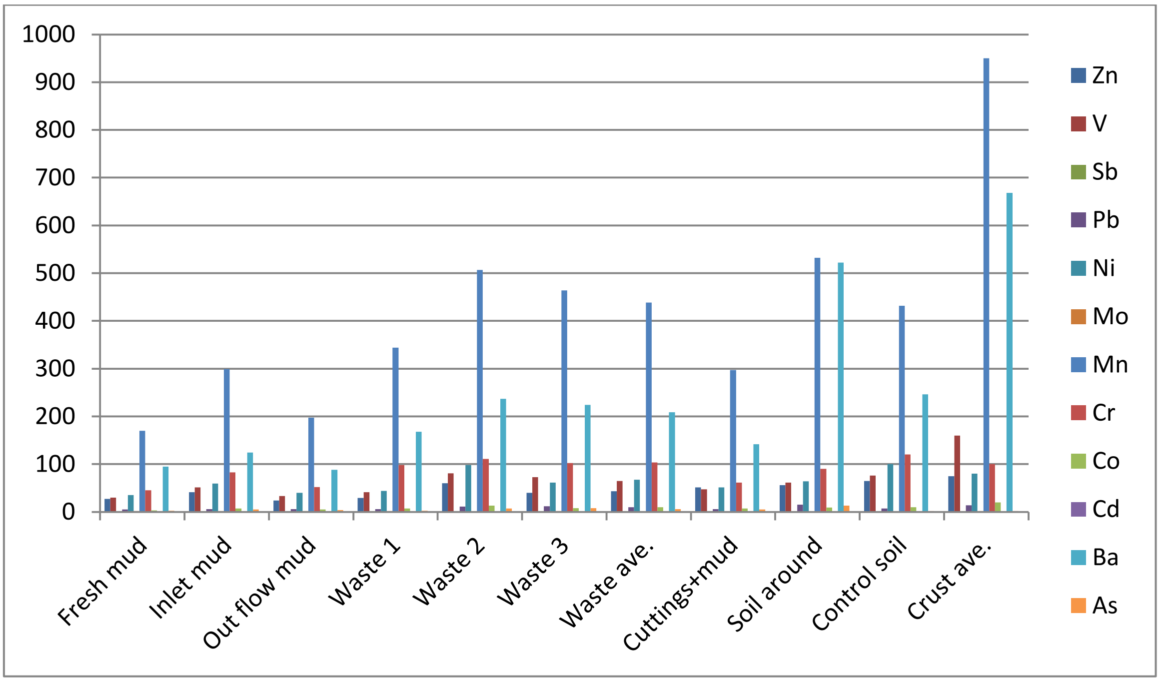

Chart 1: Comparison of metal concentrations with standard values in terms of (ppm)

One of the methods for assessing the risk of heavy metals is the calculation of the Enrichment Factor (EF) of a metal in sediment and soil. The enrichment coefficient is used to distinguish between human, natural or mixed resources (human and natural), as well as to determine the degree of contamination of heavy metals and to examine the effect of anthropogenicity (Hao et al., 2007; Meza-Figueroa et al. 2007). The enrichment factor can detect metals from human activities and cause natural factors, and identify the effects of human activities on subtraction between metals derived from human activities and natural origin, or could able to determine the source of complexity and the assessment of the degree of human effects (Hernandez et al., 2003). The enrichment factor is calculated from equation (1).

$$EF = \frac{(C_x/C_{ref})_{sample}}{(C_x/C_{ref})_{background}} \quad (1)$$

(EF): The enrichment factor of metals

$(C_x/C_{ref})_{sample}$: The concentration of the metal to the base metal concentration in the sample

$(C_x/C_{ref})_{background}$:The ratio of the desired metal concentration to the base metal concentration in the field

The base metal is usually a metal whose distribution in nature is not dependent on other metals, human activity has a small effect on its amount and therefore mainly effected by natural resources (Manganese, Lithium, Iron, Aluminum, Thallium). In some studies, heavy metal concentrations in non-contaminated areas are used as metal basic values. Other groups also use concentrations of heavy metals in the earth's crust for this purpose.

Table 2: Different groups and range of enrichment factor changes (EF)

Enrichment	EF
Low enrichment	EF< 2
Medium enrichment	2≤ EF< 5
High enrichment	5≤ EF <20
High enrichment	20≤ EF< 40
Extreme enrichment	EF≥ 40

In this study, aluminum was used as the base and crust values were used as background concentrations. Five different groups of EF values are defined for analyzing the values that are shown in Table (2) (Hernandez et al., 2003).

According to table (2), the EF number to close to 1, represents ground source. Also, metals with an enrichment factor of greater than 10 have significant amounts of enrichment and are mainly due to human and non-terrestrial resources (Hernandez et al., 2003); therefore, they will be hazardous to the environment and to the health of organisms. The results show the medium enrichment of metals, As, Pb, Ba, Mn, Mo, Cd, Co, which confirms the Anthropogenic content of these metals. Some of these metals are used to improve the efficiency of drilling mud as an additive or as impurities accompanying with other materials such as Bentonite and Barite, which are operated to prepare drilling mud from the main compounds. Because these metals have an effect on human health and most of them are carcinogens, they should be care seriously.

Hackenson used an index of total pollution for heavy metals based on whole data. This method is based on calculating the pollution factor for each pollutant. The contamination factor is calculated according to the following formula (Abrahim & Parket, 2008).

$$CF = \frac{C_m}{C_{ref}} \quad (2)$$

(CF): The contamination factor of metals

$(C)_m$: Concentration of the measured metal in the sample

$(C)_{ref}$: Base metal concentration (control sample)

Based on the enrichment factor, 5 contamination groups have been identified (Sutherland, 2000) which are presented in (Table 3).

Table 3. Different groups and range of CF changes

CF	**Contamination rate**
CF < 1	Low pollution
1 < CF <3	Medium pollution
3 < CF < 6	High pollution
CF > 6	Very high pollution

The Igeo was calculated according to formula (3) (Tang et al., 2013):

$$Igeo = Log_2 \left[{C_i}/{1.5C_{ri}} \right] \quad (3)$$

(Igeo): The geoaccumulation index
$(C)_i$: Concentration of metal i in the sample
$(C)_{ri}$: Geochemical background or reference value of the metal i

To describe the concentration of metals and the severity of contamination in the waste, drilling mud and soil of the area were calculated. Tables (4, 5 and 6) show the field index of element accumulation. The results show that according to the defined range of geoaccumulation index in 5 samples, the values showed above the normal range of the field. Some soils showed moderate to severe contamination with arsenic, barium, lead, copper and zinc. Also, some wastes showed moderate to severe contamination with barium, cadmium, copper and lead metals, and finally, only one sample of drilling mills showed moderate contamination with barite. The mean values of residue and drilling mud for all metals were in the non-contaminated range and the soil average was only relative to the element barium in the first and non-contaminated to moderately contaminated range and lead metal in the second and middle contaminated ranges. In general, the order of reduction of the land index of metal accumulation in the area of Bibi Hakimeh oil field is as follows: Pb> Ba> As> Cu> Zn> Cd>…

The calculated values of EF, CF and Igeo for the specimens studied are shown in tables (4, 5, 6).

Table 4. Calculation of pollution indices for waste mud

Metal element	**As**	**Ba**	**Cd**	**Co**	**Cr**	**Mn**	**Mo**	**Ni**	**Pb**	**Sb**	**V**	**Zn**
CF	3.8	0.85	1	0.94	0.86	1.01	1.39	0.67	1.38	2.58	0.85	0.66
EF	8.81	0.75	5.32	1.09	2.53	1.10	2.46	1.96	1.63	9.12	0.95	1.33
Igeo	-1.6	-2.4	-2.0	-1.5	-1.1	-2.3	-1	-1.5	-2.8	-0.6	-1.6	-2.3

The soil contamination with Ba, Cd, Mn, Mo, Pb, Sb was moderate and for Ar, was very high. The enrichment of Ba, Cr, Mo, Ni, Pb, Zn was moderate and for Ag, As, Cd is high.

Table 5. Calculation of pollution indices for the soil of the area

Metal element	**As**	**Ba**	**Cd**	**Co**	**Cr**	**Mn**	**Mo**	**Ni**	**Pb**	**Sb**	**V**	**Zn**
CF	8.8	2.12	1.16	0.91	0.75	1.23	1.25	0.64	2.14	2.81	0.8	0.86
EF	25.76	2.28	7.45	1.31	2.63	1.63	2.55	2.34	3.13	11.8	1.11	2.18
Igeo	-1.1	-1.6	1.4	-1.8	-1	-1.2	-1.3	-0.8	-1.7	-0.5	-0.07	-0.4

The fresh mud showed moderate contamination to As, Mo, Sb and enriched in moderate enrichment Cr and Ni and high enriched Ag, As, Cd, Mo, Sb in terms of EF.

Table 6: Contamination indices for fresh mud

Metal element	As	Ba	Cd	Co	Cr	Mn	Mo	Ni	Pb	Sb	V	Zn
CF	1.53	0.38	1	0.3	0.37	0.39	1.79	0.35	0.71	2.11	0.39	0.41
EF	7.77	0.72	11.05	0.76	2.28	0.9	6.29	2.21	1.81	15.37	0.95	1.82
Igeo	-3.9	-3.9	-2.4	-3	-1.2	-3.9	-2.3	-2.8	-3.5	-0.9	-2	-2.8

The Table (7) compares the concentrations of heavy metals in Bibi Hakimeh oil field with different standards and the contamination or non-contamination of the studied samples according to these standards, including the average concentration of elements in the earth's crust and the average global soil, soil background Localization and Comparison with European Soil Standards, California (Wanty et al., 2009) and Ireland (Zhang et al., 2008) and the United States (Burt et al., 2003). If the concentration of elements in the samples is close to the average, there is no risk in terms of contamination, but if it is higher than this amount, there is a risk of contamination and needs further investigation.

Table 7: Comparison of Bibi Hakimeh oil field average soil with world standards in terms of (ppm)

	As	Ba	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	V	Zn
Europe	6	65	0.14	7	22	12	382	0.62	14	15	33	48
Ireland	7.25	230.2	0.32	6.2	42.6	16.2	462	91.0	17.5	24.8	52.2	53.6
UK	-	121	0.7	9.8	39.3	18.1	577	-	22.6	40	-	82
Denmark	3.3	-	0.16	-	9.9	7	-	-	5	11.3	-	26.8
California	6	601	0.2	16	96	37	752	0.7	50	23	135	88
USA	-	-	0.16	7	24	17.3	372	-	18.3	10.1	-	56.5
M global	4.7	362	1.1	6.9	42	14	418	1.8	18	25	60	62
M crust	1.5	668	0.11	20	100	50	950	1.4	80	14	160	75
Soil witness	2.5	142	0.27	7	61	20	297	1.21	51	6	47	51
Area soil	22	1237	0.3	8.3	75	77	537	1.44	57.67	199	60	166

According to the tables of copper, lead and zinc in the soil average of Bibi Hakimeh oil field is higher than other parts of the world. Cadmium levels in the region are slightly higher than the global average, in England and Ireland. Cobalt is also slightly higher in Europe, Ireland, the United States, the world average, and soil. Chromium content is lower than the shell but higher than elsewhere. Manganese levels are lower than in the UK, California and crust and higher than elsewhere. Molybdenum is lower than the global and California average and higher than elsewhere. Nickel is just below average crust, but vanadium is lower than California soil in addition to crust. Chromium, cadmium, copper, lead and zinc are derived from drilling mud compounds. Nickel and molybdenum are high in both crude oil and oil and petroleum materials added to drilling mud. Probably due to the high amount of lead, part of it is related to drilling rigs and the fuel used in them.

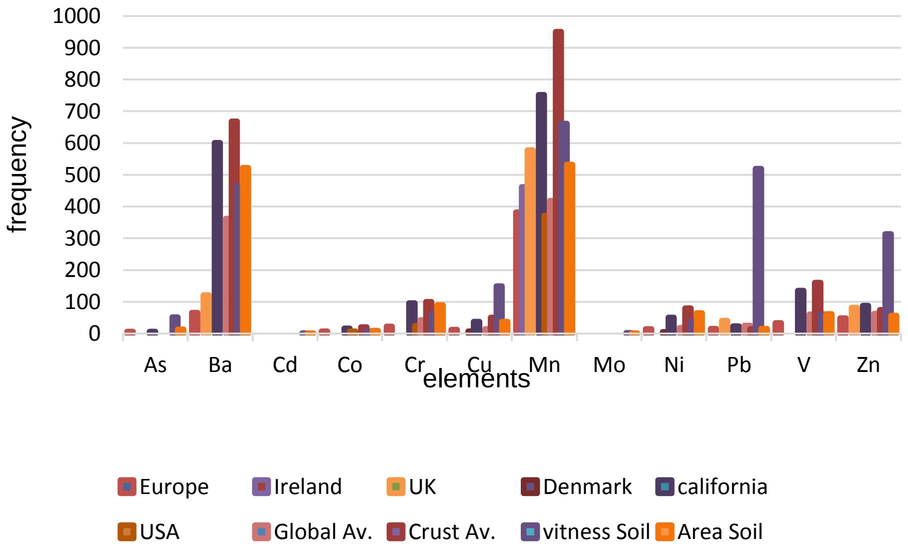

Chart 2. Comparison of soil in the study area with standards and other countries in the world

CONCLUSION

The high amounts of Arsenic, Lead, Barium, Cadmium, Molybdenum, Antimony and Silver in samples show the contamination of these elements in comparison to standards and the area. The calculation of enrichment also confirms the human source for these metals.

Pollutants of these ponds can spread to soil, weather, or penetrate into the underground. Also, by filling these ponds with rainfall, contaminants are scattered around the environment and lead to environmental contamination. Given that drilling operations are carried out in different geological areas in land and offshore, disposal of waste and in waste disposal ponds should be managed correctly in accordance with the laws and regulations.

Comparison between the concentration of heavy metals in Bibi Hakimeh oil field has been done with different standards and the contamination or non-contamination of the studied samples according to these standards, including the average concentration of elements in the earth's crust and the average global soil, local soil background and comparison with the standard. California (Wanty et al., 2009), Ireland (Zhang et al., 2008) and the United States (Burt et al., 2003). If the concentration of elements in the samples is close to the average, there is no risk in terms of contamination, but if it is higher than this amount, there is a risk of contamination and needs further investigation.

In India, Kotoky et al. Chemically decomposed heavy metals around the Assam oil facility. The concentrations of heavy metals and their relationship with the amount of organic matter in both wet and dry seasons were determined and it was found that the amount of lead in carbonate formations is higher than other metals. Also, the concentration of other heavy elements is four times higher than the local average of the region (Kotoky et al., 2003).

Concentration of heavy metals in the topsoil of an oil field was determined by Iwegbue et al. in Nigeria and compared with the local average. The result did not show much contamination, but since this area is close to agricultural lands, fishing ponds and surface water sources, their amounts should be controlled (Iwegbue et al., 2006). Malika Ghazi et al., Assessed the impact of the oil drilling system in Algeria, Heavy metals were measured and their effects on groundwater, soil and humans were investigated. The best treatment and freezing in place of waste was proposed (Ghazi et al., 2011).

According to Table (3-4), the amounts of copper, lead and zinc in the average soil of Bibi

Hakimeh oil field are higher than other parts of the world. Cadmium levels in the region are slightly higher than the global average, in England and Ireland. Cobalt is also slightly higher in Europe, Ireland, the United States, the world average, and soil. Chromium content is lower than the shell but higher than elsewhere. Manganese levels are lower than in the UK, California and crust and higher than elsewhere. Molybdenum is lower than the global and California average and higher than elsewhere. Nickel is just below average crust, but vanadium is lower than California soil in addition to crust. Chromium, cadmium, copper, lead and zinc are derived from drilling mud compounds. Nickel and molybdenum are high in both crude oil and oil and petroleum materials added to drilling mud. Probably due to the high amount of lead, part of it is related to drilling rigs and the fuel used in them.

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CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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