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Article

Assessment of pollution with some heavy metals in agricultural soils near Qayyarah oil fields, southern of Mosul

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ABSTRACT

To assess soil contamination in Qayyarah sub-district of Nineveh Governorate with elements (Pb, Cr and Ni), the study area was divided into three areas depending on the distance of the land from the oil fields. The results showed an increase in the total concentration of heavy metals, Ni in all locations exceeded the maximum permissible limit of 50 $mg.kg^{-1}$ according to WHO (World Health Organization) and FAO (Food and Agriculture Organization), which ranged in concentration (61.15 - 221.4) $mg.kg^{-1}$. while the total soil content of pb ranged between (32.33 - 103.88) $mg.kg^{-1}$, also location (2) exceeded the maximum permissible limit in the soil of (100)$mg.kg^{-1}$.The total Cr content ranged between (17.76 - 71.08) $mg.kg^{-1}$. Enrichment factor for for pb, Cr and Ni ranged from (1-2.8)(0.8-2.7) and (1.8-4.2) respectively, in the surface soils affected by residues oil field when compared with the control soil C.S 16. It was found through the spatial distribution that the concentrations of lead were the highest in the soils of region A, which are located inside the oil fields, and they decrease as we move away from the polluted source, specifically in areas B and C, and this confirms the role of liquid, solid and gaseous oil residues that are thrown from those oil wells to those soils and agricultural lands. Which caused an increase in the total content of lead. As for chromium, it was found that it had a different soca from lead, and the highest concentrations appeared in the C region far from the oil fields with little variation from the A and B regions. The results of the spatial distribution of nickel also showed that it had a similar behavior to chromium Significant increases in concentration were shown in the soils of area C, which reinforces the assertion that the geological structure of the study area had an impact on the high values of nickel and chromium, particularly the areas far from the oil fields. The results of the correlation analysis showed a significant negative relationship of the studied heavy metals with sand and a positive correlation with clay, and the effect of this relationship was shown in Site 3, which is located within Zone A, which showed low concentrations of heavy metals due to the nature of its sandy texture, in addition to an important positive relationship between minerals Heavy, which confirms that it resulted from the same pollutant source.

Keywords: : heavy metals, Pollution, Soils

INTRODUCTION

Soil pollution refers to any chemical substance in soil that exceeds normal levels that causes adverse effects on plant growth as well as on animal and human health through its entry into the food chain and its impact on soil quality and the entire ecosystem [1], presence some pollutants may also lead to nutrient imbalances and soil acidification, which are important issues in many countries [2], therefore it is a chemical degradation causes partial or complete loss of soil functions. Soil productivity is affected as well as the organisms in polluted soil, when these pollutants exceed the

permitted levels, the soil becomes functionally dead, which adversely affecting the growth of plants in those soils [3], and soil is the main basin for those pollutants emitted by environment [4].
Due to the rapid economic development and severe industrial activities, pollutants are considered one of the most important human problems in the world, especially in recent years, which has high levels of pollutants are released, which led to their accumulation in the soil, which reached alarming levels [5], Therefore, some locations near the oil manufacturers have become contaminated by the distinctive activities of these businesses and their organic and inorganic pollutants, including heavy metals [6], Soil pollution with heavy metals is a serious global environmental issue and not less dangerous than water and air pollution because it harms the plant growth and adversely affects on soil microbial processes through its harmful effects on soil microorganisms such as bacteria, earthworms and other soil organisms that live in the soil [7], Whereas, high concentrations of heavy metals lead to a significant inhibition of soil respiration, because the severe pollution of heavy metals weakens soil microbial activity, which poses a serious threat to the function of the soil ecosystem [8], Also Soil contamination is directly related to the soil physical properties. Therefore In highly contaminated soils, surface hardening processes take place that increase of soil bulk density, degrade soil porosity, and significantly reduce water permeability [9], As a result of this adverse effect, many countries around the world have adopted national soil protection and pollution treatment regulations, and research on soil pollution assessment and treatment has increased [10].
Qayyarah is an Iraqi town located south of the city of Mosul and is characterized by the presence of many oil fields nearby as it contains large oil reserves, which led to attracting many international private investing companies to explore for oil, in addition to the presence of an oil refinery in it, and because of the lack of commitment of these companies operating in The field of the oil industry and the lack of respect for the environment, which led to significant pollution of the soil and air as a result of the release of liquid waste into the soil without treatment, in addition to the burning of fuel and the release of gases into the atmosphere, which led to the deterioration of the soil system in particular by affecting the crops grown near the oil fields in These agricultural lands and the ecosystem in general. Therefore, the current study aims to assess the pollution with lead, chromium and nickel elements in those lands close to the oil fields and compare them with lands and soils far from the source of pollution by relying on the enrichment factor and some global determinants of the World Health Organization and the International Food and Agriculture Organization.

MATERIALS AND METHODS

Study Area:

The study area is located 60 km southern of Mosul, which is represented by agricultural lands in Qayyarah district of Nineveh Governorate, which lines between longitudes (43°20'55.87"E-43°6'2.66"E) and latitudes (35°55'30.42"N - 35°34'36.56"N) (Figure 1), Where the study area was divided into three parts depending on the distance of the agricultural lands from the oil fields from all directions, part (A) represent lands close to the fields, which are located under the influence of solid, liquid and gaseous oil wast, at a distance of (400-800) m, (B) lands about (1500-2000) m away from center, and (C) area about (4000-5000) km from the oil fields. Because wind movement and direction play an important role in the distribution of pollutants, in addition to the comparison sample (C.S), which is located about 6000 m northeast of the oil fields, 16 surface soil samples included at a depth of 0-30 cm. All soil samples were dried upon collection and sieved through with a diameter of (2) mm, thus became ready for the following physical and chemical analyzes.

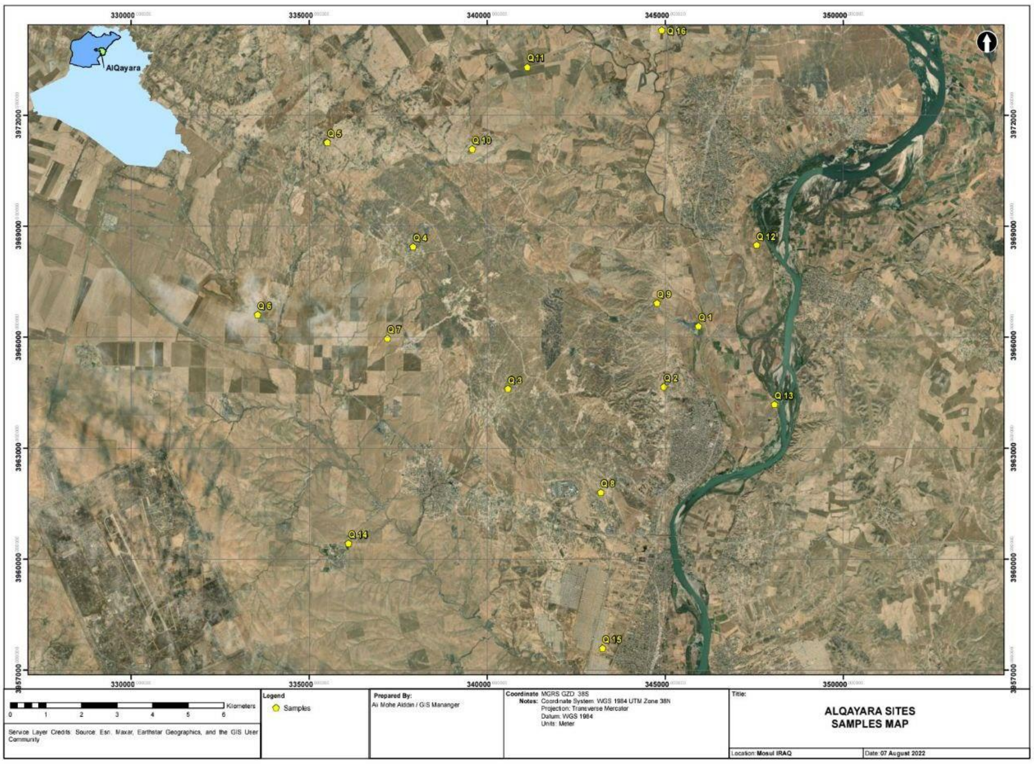


Figure 1. Study area in the Qayyarah district.

Soil Sampling and Analysis

Particle size distribution was determined using the hydrometer method [11], Soil pH was determined by a pH meter, Soil EC determined by using the (EC meter), Organic matter and Calcium carbonate determined according to the method by [12], Cations Exchange Capacity determined according to the method by [13].

Determine of Heavy Metals in soil

Soil samples were digested using royal water (acidic method) [14] to estimate the total amount of heavy lements (Pb, Ni, Cr) in soil. (2) gm of each soil sample was added to a conical flask with (15) ml of royal water consisting of 3:1 ratio of hydrochloric and nitric acids and left for 24 hours. Then heated for digestion to 120°C, after two to three hours of heating (when the color turns from brown to colorless), leave the mixture to cool, then used Whatman filter paper and filled in to 50 ml distilled water. Heavy metals concentrations in the extracts were estimated using a German-made Atomic Absorption Spectrophotometer, model Analytika jena 350A.

Enrichment Factor (EF)

Enrichment factor used to assess the level of soil pollution and seeks to know the natural or human inputs and their impact on the soil. The most common reference elements used in determin the EF value are Aluminum (Al), Tin (Sn), Iron (Fe), and Manganese (Mn) [15], In current study, (Fe) selected as a reference element because of its high concentration in study area soils, where Fe is stable in soil and is characterized by the absence of vertical movement within the soil. In addition, its concentration does not change in human terms [16]. Enrichment factor values were calculated according to the following equation [17]:

$$EF = (C_m/C_{Fe})\text{sample} / (C_m/C_{Fe})\text{Background}$$

where

(C_m sample)= Heavy metal Concentration in soil sample.
(C_{Fe} sample) = Total Iron concentration in the same sample.
(C_m Background) = Heavy metal Concentration in comparison soil.
(C_{Fe} Background) = Iron concentration in comparison soil.

Seven categories have been identified to classify the results of the enrichment factor as described by [18] as shown in table (1).

Table (1): Interpretation Levels of Enrichment factor

Enrichment Factor	Designation of Street Dust Quality
EF ≤ 1	No enrichment
EF ≤ 3< 1	Minor enrichment
EF ≤ 5 < 3	Moderate enrichment
EF ≤ 10 < 5	Moderately severe enrichment
EF ≤ 25 < 10	severe enrichment
EF ≤ 50 < 25	severe enrichment Very
EF > 50	Extremely severe enrichment

RESULTS

The Physicochemical Properties of Soil

It is evident from Table 2 that the proportion of separated (clay, sand and silt) was different for the soil of the study area, as it ranged between (224.5-387) g.kg^{-1} for clay, and the percentage of silt ranged between (125-460) g.kg^{-1}, while it was The sand ratio ranged between (212-603) g.kg^{-1}, and the electrical conductivity EC values ranged between (0.6-8.6) ds.m^{-1} . As for the pH values, it ranged between (7.38-7.92), and the results showed that the organic matter ranged between (6.87-20.63) g.kg^{-1}. The content of the soils of the study area of calcium carbonate ranged between (45-370) g.kg^{-1}. The results show that the cation exchange capacity of CEC was between (9.56-23.48) centimole (+).kg^{-1}.

Total Content of Heavy Metals

When heavy metals are found in large quantities in soil, this may be due to the accumulation of chemicals containing heavy metals that are discharged into the soil as a result of oil exploration and production activities [19].Table .3

The results of Table 3 indicate that the concentrations of the total content of lead in the study soils ranged between (32.33-103.88) mg.kg^{-1}, and it is clear from Figure 1 that the highest values of lead were in the sites located within Zone A close to the source of pollution, specifically Sample 2 (Q2), then the concentrations start decreasing gradually as we move away from the oil fields in areas B and C until it reaches the lowest value of lead in the control soil C.S, while the results showed that the total chromium concentrations ranged between (19,56-71.08) mg.kg^{-1}, and Figure 2 shows that there is a discrepancy in the total chromium concentration of the studied sites between highs and lows within one region, as the results showed varying concentrations within region A, where the lowest value of chromium was recorded in sample 3 (Q3), then return to To rise in sites belonging to region B and C with a little difference from region A, as for nickel, the results of the total content indicated that it ranged between (54,2-221,4) mg.kg^{-1}, and it was shown by Figure 2 that the variation in the total content of nickel in the soil It was clear and the highest values were in the southeastern part of the study area within area A, specifically in sample 2, while in sample 3 It decreases suddenly, but in areas B and C it rises again significantly, specifically sample 12 and [13]. The results were less than what [20] found, who indicated an increase in total lead concentration in agricultural soils in Baghdad Governorate which is ranged between (56.73- 714.87) mg kg^{-1} and attributed this to the power stations impact and oil facilities such as Aldora Refinery, and higher than the results reached by [21] in agricultural soils close to industrial facilities as a result of the waste they pose and decrease the farther away from the source of pollution, as they noted an increase in lead concentrations, which ranged between (9.6 - 44.79)) mg kg^{-1}

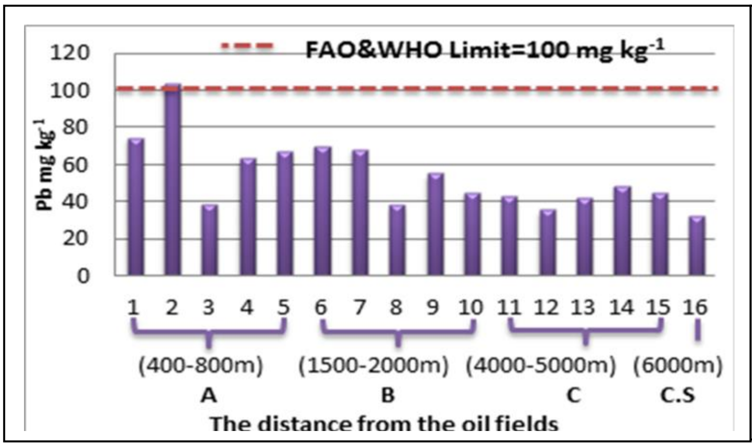
Table 2. physical and chemical properties of studied soil

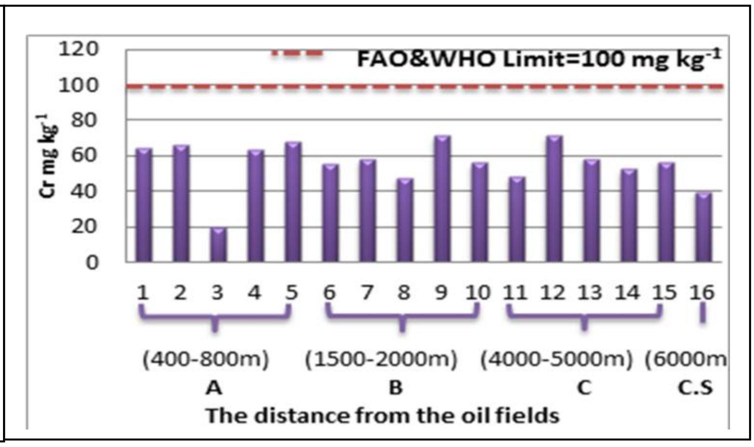
Zone	Nu	EC	pH	CEC	O.M	CaCO3	Sand	Clay	Silt	Texture
A	1	1.4	7.85	22.61	17.19	315	243	356	401	C.L
	2	2.5	7.69	23.48	15.06	290	250.5	357	392.5	C.L
	3	2.5	7.38	15.65	10.66	45	603	272	125	S.C.L
	4	1.2	7.54	20.87	16.28	225	228	362	410	C.L
	5	2.3	7.61	23.48	11.69	250	261.7	379.5	358.7	C.L
B	6	1.6	7.68	15.65	12.38	225	453	347	200	C.L
	7	0.7	7.69	20.00	12.38	240	361.7	254.5	383.7	L
	8	1.9	7.58	19.13	15.82	265	306	289	405	C.L

	9	2.7	7.83	19.45	15.87	270	265	382	353	C.L
	10	2.2	7.67	16.52	13.75	310	253	387	360	C.L
C	11	0.8	7.64	21.74	13.75	370	212	387	401	C.L
	12	1.1	7.59	18.26	20.63	155	335	240	425	L
	13	0.6	7.71	16.52	19.6	215	278	262	460	L
	14	8.6	7.84	19.13	10.31	200	214.2	349.5	436.2	C.L
	15	3.6	7.92	16.68	13.75	155	453	322	225	S.C.L
C.S	16	0.9	7.55	16.52	14.1	270	480	224.5	295.5	L

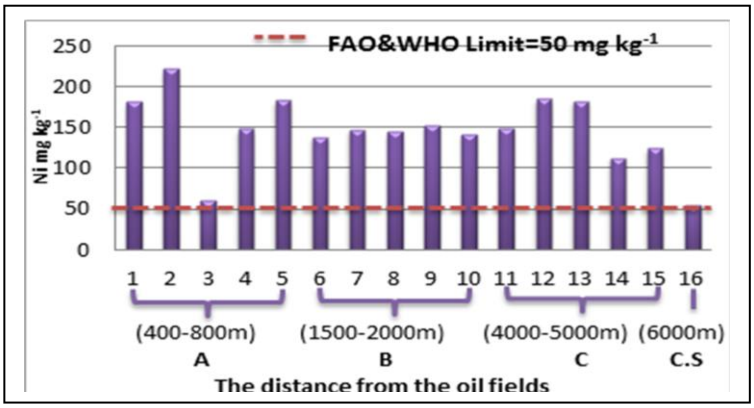
Table 3.Heavy metal analysis of study soils (mg.kg^{-1})

Zone	Nu	Site Name	Pb	Cr	Ni	Fe
A	1	Q1	74.15	64.56	181.00	3625
	2	Q2	103.90	65.96	221.40	3756
	3	Q3	38.44	19.56	61.15	1980
	4	Q4	68.23	63.22	148.20	3379
	5	Q5	67.35	67.59	183.50	3295
B	6	Q6	69.61	55.58	138.30	3295
	7	Q7	63.12	57.91	147.40	3412
	8	Q8	37.91	47.45	144.60	3357
	9	Q9	55.04	71.00	153.20	2191
	10	Q10	45.02	56.00	142.10	3370
C	11	Q11	42.58	47.71	148.70	3397
	12	Q12	35.60	71.08	185.40	3527
	13	Q13	41.46	58.11	182.20	2901
	14	Q14	48.21	52.24	112.10	3369
	15	Q15	44.33	56.23	124.00	3323
C.S	16	Background	32.33	39.37	54.20	3268





(a) **(b)**



(c)

Figure 2: (a) Total content of lead in the studied soils; (b) Total chromium content in the studied soils; (c) The total nickel content in the studied soils.

The enrichment factor was calculated from the concentration of heavy metals in 15 samples surrounding the oil fields of the study area, and the concentration of heavy metals in the sample 16 (comparison soil) was used as a standard concentration to assess the contamination with heavy metals for those soils [22].

Table 4. Higher, lower value and enrichment factor rate for heavy elements in the study soils.

Elements	Min	Max	Average	Enrichment
Pb	1	2.8	1.7	Minor enrichment
Cr	0.8	2.7	1.5	Minor enrichment
Ni	1.9	4.2	2.9	Minor enrichment

The results indicate that the values of the enrichment factor for heavy metal pollution are different for the study area soil, and the average values increase in the order Ni > Pb > Cr, where the values for lead ranged between (1 - 2.8) and fall within the range of little enrichment 1>EF>3, and ranged between (0.8 - 2.7) for chromium which also falls within the range of little enrichment 1>EF>3 except for site 3 it was low within the range of no enrichment EF < 1, while Nickel ranged between (1.9 - 4.2), where most of it falls within low enrichment range 1>EF>3 except for samples 1, 2, 5, 9, 12 and 13 that were within the moderate enrichment range 3>EF>5 according to 18, usually when the value of the enrichment factor is close to or less than 1, this reflect that the main source of heavy metals is from a natural source, but if the enrichment factor is greater than 1, this indicates that the main source from human activities [23]. Some opinions suggest that enrichment factor values of 1.5 or more are indicative of human influence, which were determined by [24]. According to this suggestion, the sources of heavy metals are considered human in most of the study's soil sites, and what confirms this is the spatial variation of the EF values which exceeded the critical limit mentioned by [24].

Figure (3) shows that the highest values of lead were in samples close to the oil fields, specifically samples 1, 2, 3, 4, 5, 6, 7 and 9, which exceeded the critical limit (1.5). while for chromium the enrichment factor values in samples 1, 2, 4, 5, 9, 12 and 13, were greater or equal to (1.5), but nickel shows it appears that there is a significant increase in enrichment factor values, as the values of the enrichment factor in all the 24 study soil sites exceeded the critical value 1.5 , where it reached 4.2.

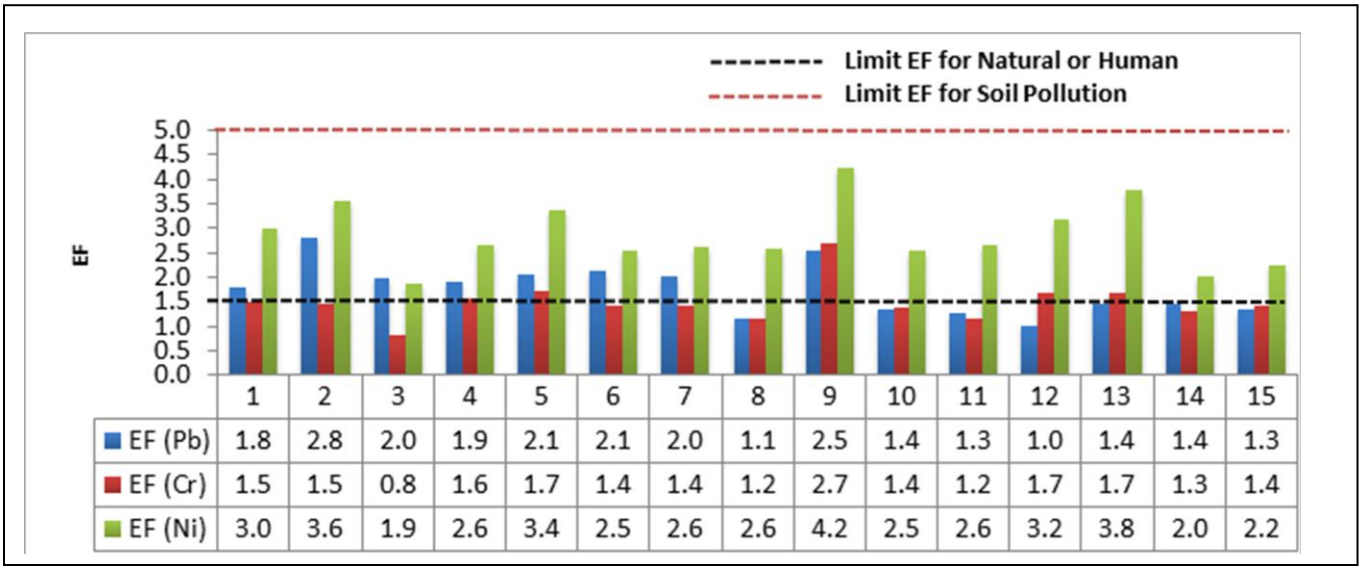


Figure 3. Enrichment factor value for lead, chromium and nickel.

Correlation Matrix

To describe the relationship between the total concentrations of heavy metals with the physical and chemical properties of the soil, it was done by Pearson correlation coefficient Table 4.

Table 5. Pearson correlation matrix between heavy metals and the chemical and physical properties of the soils of the study area.

	Clay	Sand	CEC	O.M	CaCO3	pH	EC	Pb	Cr	Ni
Clay	1									
Sand	-0.58*	1								
CEC	0.48	-0.67**	1							
O.M	-0.25	-0.30	0.05	1						
CaCO3	0.49	-0.67**	0.53*	0.13	1					
pH	0.36	-0.38	0.15	0.07	0.28	1				
EC	0.28	-0.13	-0.01	-0.49	-0.24	0.41	1			
Pb	0.48	-0.33	0.64**	-0.07	0.30	0.22	-----	1		
Cr	.0.36	-0.65**	0.50*	0.49*	0.38	0.53*	-0.06	0.49	1	
Ni	0.35	-0.67**	0.62**	0.52*	0.39	0.32	-0.20	0.60*	0.81**	1

****. Correlation is significant at the 0.01 level (2-tailed).**
***. Correlation is significant at the 0.05 level (2-tailed).**

The results showed a significant positive correlation between lead and CEC (0.64**), and a weak positive correlation with clay (0.47), CaCO3 (0.30) and pH (0,22), with a negative correlation of lead with sand (-0.33) and organic matter (-0.07), while chromium showed a significant positive correlation with CEC (0,50*), organic matter (0.49*), pH (0,53*), and a weak positive correlation with clay (0.36) and CaCO3 (0.38), while the correlation of chromium with sand was significant negative (-0.65**), and the results showed a significant positive correlation of nickel with CEC (0,62**) and organic matter (0,52*). And a weak positive correlation with clay (0.35), CaCO3 (0.39) and pH (0.32), while the correlation of nickel with sand was an significant negative correlation (-0.67**).

The relationship between heavy metals was evaluated in order to determine the common source of these elements in the study area, and it was found that there were significant positive correlations between heavy metals, as lead showed a significant positive correlation with nickel (0.60*), and the relationship between chromium and nickel showed a significant positive correlation also (0,81**), but it was a weak positive between lead and chromium (0,49).

DISCUSSION

Discussion the Chemecal Properties of Soil

It is clear from these results that most of the soils are normal soils whose salinity was (less than less than 4) dSm -1, with an increase in electrical conductivity in sample 14 observations whose conductivity values exceeded 4 dSm $^{-1}$, they appear as saline for plates [25]. This is a result of several factors, the most important of which are low rainfall, high surface evaporation, weathering of salt rocks, saline irrigation, and poor agricultural practices [26], indicate that the soils of the study area were of a neutral to slightly alkaline reaction degree. The reason for the high degree of soil interaction is due to the nature of the soil formation conditions in terms of climate and the origin material rich in calcium carbonate, as the presence of carbonates affects strongly on the degree of soil interaction, as carbonate soil is characterized by having a pH greater than 7 [27]. The organic matter it is clear that there is a discrepancy in the values of the organic matter between the soils, and in general it appears that there is a decrease in the values of the organic matter in all soils, due to the nature of the dry climatic conditions, with the exposure of these Areas of lower rainfall and higher temperatures result in less input of organic matter and nutrients from external sources [28]. The reason for the high values of calcium carbonate in most of the sites may be attributed to the fact that the nature of the origin material for the soils of the study area is mainly rich in calcium carbonate, which is also due to the geological formations of these The area, which was rich in these carbonate minerals, and the lack of rainfall It works on the accumulation of calcium carbonate in the sub-surface horizons [29]. The cation exchange capacity (CEC) is one of the important properties of soil and a basic criterion for its quality and the ability to remove environmental pollution, which affects the adsorption of heavy elements such as copper, zinc and lead [30].

Heavy Metals Content and Soil Pollution.

the highest Lead concentrations were in the sites located within the (A) area close to the source of pollution, where it was highest in Sample 2 (Q2), The reason for the high lead concentration due to its proximity to the oil fields and resulting from the solid, liquid and gaseous wastes that have contaminated these soils and agricultural land near oil wells, and the alkaline pH depositing lead in the arable soil layers in carbonate and phosphates form at pH greater than 6.5. [31] indicate that lead is significantly associated with organic matter and metal oxides at 26 % from the total amount.

The results show that there is a discrepancy in the total concentration of lead for all sites in the study area, as we notice a significant decrease in lead values for some samples in (A) part, and despite their proximity to the oil fields, specifically sample (Q3), which is located to the west of the oil fields compared to the rest of samples located at the same distance as the polluting source, This is due to its low content of clay and organic matter, which catch and fixation lead and the rest of the heavy metals. On the other hand, it is noted a high content of sand, as the clay soil retains a high amount of heavy metals compared to sandy soil [32]. The results obtained that all samples are con-sidered to have a high concentration of lead, but they are within the permitted limits in the soil, except sample Q2, which exceeded the maximum limit of (100) mg kg-1 according to the WHO and the FAO [33].

The result Chromium there was a variation in the in the study area, as the highest concentration appeared in (C) Area, which is 3 km away from the oil fields, specifically sample 12. which is explained by the wind speed and direction, which works to transfer pollutants and their chromium from the source of pollution to further areas and then are deposited on the surface of the soil in those locations, as weather conditions play an important role in the distribution of heavy metals in the air [34]. Or, its due to the original mater, since soil chromium is inherited from the mother rocks, as the chromium content increases in soils formed from mafic rocks and stone sediments [35]. [36] noted that the increase of chromium in Iraqi soils is the result of the transfer of calcareous deposits resulting from weathering processes from north and northeastern Iraq.While the lowest concentration of total chromium upper in sample (3) despite its proximity to the source of pollution, due to the low per-centage of clay and the prevalence of sand, as sandy soils have a weak ability to adsorb heavy elements [37].According to [33], the results of the total chromium content in all sites are considered within the permitted limits in soil, which is less than 100 mg kg-1, which are approved by standard regulatory bodies such as (WHO) and (FAO), which do not cause risks to Soil and plant system.

The spatial distribution of high nickel concentrations may be associated with oil combustion and agricultural activities such as phosphate fertilizers [38], as the variation in the total nickel content was clear and the highest values were in the southeastern part of the study area, where the highest concentrations in the sample 2 (Qayyarah 39), due to their proximitly to the oil fields and solid, liquid and gaseous wastes posed by these oil wells, in addition to their high content of clay. While the other far locations specifically (Q12 and Q13), which are located in the same direction, were also highly concentration due to the predominance of northwest winds in the region which transfer these pollutants and the elements they contain and deposit them in those sites. Ni can be released into the atmosphere through industrial activities which accumulates on the surface after precipi-tation reactions [39]. It is clear that all sites exceeded the maximum allowable limit for nickel com-ponent set by the World Health Organization and the Food and Agriculture Organization of (50) mg kg-1, which poses a threat to the soil, plants and humans system [40].

The Nickei it has been found that there is a noticeable increase in the total content of nickel in most locations compared to the rest of the heavy metals due to the high ability of nickel to ad-sorption on The surfaces of clay minerals, as clays has a high cation exchange capacity and therefore have a greater ability to stabilize heavy metals [38], Montmorillonite is the dominant min-eral in arid and semi-arid region. It may also be attributed to the increase in sediments and frag-ments transported from the northern and northeastern regions, which contain pyroxene, olefin and chlorite mineral rich in nickel.

These high values of lead, chromium and nickel in the above-mentioned areas are a clear confir-mation of human activity, which means that the enrichment of these elements results from indus-trial inputs such as liquid oil residues that flow from those oil wells or emissions of pollutants into the air, This is due to the burning operations of the oil wells that enriched the soils of the region, especially the nearby ones, with heavy elements as a result of smoke and ash emitted from it and the mineral and organic pollutants it contain, which eventually reach the soils of the neighboring lands either through sedimentation or by rain.

It appears in the rest of the locations that the values of the enrichment factor for lead and chromium decreased below 1.5 until it reached the lowest values of chromium, which is 0.8 in sample 3 (Fig.

1), due to the total chromium concentration at that point was slighty due to the nature of the sandy texture poor in clay minerals,

which It facilitates the movement and washing of heavy elements and thus reduces their amounts in soil, as well as the calculation of the enrichment factor index with the total iron concentration, whenever the Iron content was high thei is decrease in enrichment factor 21.

The close positive correlations between heavy metals, which emerged through Pearson correlation analysis, indicate that heavy metals may be produced from the same polluting source, which in our study represents oil fields [41], where the wind works to transfer gaseous emissions containing these heavy metals to agricultural lands. Near the oil fields, which led to an increase in the total content in the soil, and the geological composition of the area may contribute to an increase in the total concentration of nickel and chromium elements, especially in areas far from pollution sources (oil fields) [42].

CONCLUSIONS

The soil of the region was contaminated with Nickel, as it exceeded the maximum al-lowable limit according to FAO and WHO.The Lead and Chromium concentrations in soils was within the permitted extent, except sample 2 (Qiyarat 39), where Lead exceeded the upper limit allowed in the soil.The values of the enrichment factor explain that the enrichment of these heavy metals are results from industrial inputs such as oil residues and emissions of pollutants into the air, which are deposited on agricultural lands adjacent to the polluted source.

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