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Application and Evaluation of Some Pollution Indices in Heavy Metal Contaminated Soil

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Abstract: Heavy metal and metalloid contamination significantly threaten the environment and human health. The use of soil environmental assessment indicators is important as they provide valuable information about soil health, quality, and potential risks to ecosystems. The purpose of this investigation is to determine the soil content of Cd, Cu, Fe, Mn, Ni, Pb, Zn, and Pb in an area exposed to various sources of pollution, as well as, assess soil heavy metal contamination using five of single pollution assessments, and three of integrated pollution assessments and compare the obtained data with risk assessment code (RAC). Results showed that concentrations (mg/kg) of Cd (3.32) and nickel (71.26) were above Threshold limits. While Cu, Fe, Mn, Pb and Zn contents, were below their corresponding threshold limits reported by the published threshold limits. Fractionation results showed that the general trend in almost all tested metals, on average, the order was residual (50.11 %)> followed by reducible (21.67%), carbonate (13.33%), organic (9.29%), and an exchangeable fraction (5.60%), however, the quantities varied. The calculated pollution assessments revealed that ; (CF) values showed that the soil was very highly polluted with Cd (8.1), moderately polluted with Cu (1.3), Mn (1.04), Ni (2.46), Pb (1.35), and Zn (1.05), while low polluted with Fe (0.36); Er index values indicated that the soil was considered low potential ecological risk, except for Cd which was categorized as highly potential ecological risk (242.93); EF index indicated that Cd and Ni were categorized as significant enrichment (22.74, and 6.9, respectively), while, (Mn, Pb, and Zn) were moderately enrichment, and Fe was minimal enrichment; Igeo index values revealed that only cd was categorized as moderately contaminated (2.43), Ni was slightly contaminated (0.71), while the rest of the metals were classified as uncontaminated (minus values); PIT index revealed that Cd categorized from (moderately to low polluted), while Ni was categorized low polluted, while (Cu, Fe, Pb, Mn, and Zn) were categorized as unpolluted. Also, the results of the integrated soil assessment indices were calculated; the results of the PLI index (1.45) revealed that the soil was considered polluted soil, the PERI index considered the soil moderate ecological risk, and the NCPI index classified the soil as slightly polluted. RAC indicated the soil high risk with Cd (30.04), and medium risk with (Cu (21.9), Mn (19.96), Ni (23.72), Pb (13.67), and Zn (16.14), while for Fe, the soil was lies in the low-risk category. Taking the RAC index as a reference result, the data revealed that among the tested single assessment indices (CF, EF) were consistent with RAC values, Also, the integrated indices (PLI, and PERI) indices, as well as NCPI when using threshold data of CSQG only were consistent with RAC data. So, it is clear that the indices that rely solely on the total or threshold level of pollutants may often be inaccurate and show several disagreements and make it challenging to compare soil quality data internationally or even locally, while soil evaluation using indices that depend on the speciation of pollutants in soil is more realistic.

Keywords: Soil Heavy Metals, fractionation of soil heavy metals, risk assessment code (RAC), Soil pollution indices.

1. Introduction

Agricultural soils are subject to high levels of pollution from different sources, naturally, through processes like the breakdown of parent rock materials and volcanic eruptions, or as a result of human influences **(Gao and Chen 2012; Ogunkunle and Fatoba 2013; Sayadi *et al*. 2015).** These pollutants persist in soil for long periods and can lead to soil function disorders, plant growth, and even pose a risk to human beings. So, the use of soil environmental assessment indicators is important as they provide valuable information about soil health, quality, and potential risks to ecosystems **(Bhairo *et al*, 2023; Mohamed, *et al*., 2014; Joanna, *et al*, 2018)**.

Among the most important used indices are; (1) single-element indices such as; Geo accumulation Index (Igeo) proposed by **Muller (1969)**; Pollution Index (PI), **Wu *et al*. (2015)**; Threshold Pollution Index (PIT), **Lu *et al*. (2009)**, and Contamination Factor (CF), **Hakanson (1980)**. And (2) Mult-element indices as; Pollution Load Index (PLI), **Liu *et al*. (2005)**; Nemerow Pollution Index (PINemerow), **Cheng *et al*. (2007)**; Degree of Contamination (Cdeg), **Hakanson (1980)**; Modified Contamination Factor (mCdeg), **Abrahim and Parker (2008)**.

There is no doubt that different indices all have advantages, disadvantages, and uncertainties, therefore, the results of pollution assessment using one indicator will differ if another indicator were used. On the other hand, almost all of these indicators rely only on the total content or threshold levels of heavy metals as a tool to determine their potential impact on the environment. While the total concentrations of heavy metals do not provide adequate and insufficient information to evaluate their bioavailability or toxicity, in addition, the threshold level varies from one country to another depending on the environmental conditions in which the metal is present. Risk Assessment code (RAC) is an index that depends mainly on the laboratory results of sequential metal extraction procedures. RAC index formula is based on the calculation of the available and readily available fractions extracted sequentially by **Tessier *et al*. (1979)**, these fractions are softly bound to soil solid phases.

The current investigation aims to determine some of the general properties of heavy metal-contaminated soil and the content of Cd, Cu, Fe, Mn, Ni, Pb, Zn, and Pb metals. Additionally, assessing soil heavy metal contamination by using both single pollution indexes (Igeo, CF, and Er) as well as integrated (PLI, and PRI) pollution indices with a comparison with risk Assessment code (RAC).

2. Material and methods

2.1. Selected site

The selected site was Al-Kom Al-Ahmar, which is one of the villages belonging to the Osim Center in the Giza Governorate. It is located at 30° 6' 45" north and 31° 10' 25" east. This area is affected by various sources of pollution; urbanization, auto-exhausted, agricultural sources such as (heavy manuring, pesticides, and herbicides), as well as industrial emissions.

2.2. Soil Sampling

Twelve topsoil (0 -20 cm) agricultural soil samples were selected randomly from the area using stainless steel augers and debris and vegetation were removed by shovel, air-dried, ground, and sieved through a 2 mm sieve, and mixed thoroughly to make one composite soil sample, and stored away from light for the following analysis.

2.3. Soil analysis

2.3.1. General soil Characters

Soil pH, soil salinity (EC), soil texture, and calcium carbonate were determined according to **(Jackson, 1984)**. Soil organic matter (SOM) by modified Walkley and Black method **(USDA,1996)**.

2.3.2. Sequential fractionation of Soil heavy metals.

In triplicates, the studied pollutants in the soil were sequentially fractionated by the procedure of **Tessier *et al*. (1979)** into operationally five fractions. The extraction process steps and the resulting fractions are detailed in Table 1.

Table 1: The steps of the sequential extraction procedure used and the resulting fractions

Steps	**Fractions**	**Extraction process**
1	Exchangeable F.	1 g of soil sample, 8 ml 1M $MgCl_2$ (pH 7), 1 hr. shaking at room temp.
2	Carbonate F.	8 ml 1 M CH3COONa, at pH (5.0), 5 hr. shaking at room temp.
3	Oxides F.	20 ml 0.04M NH2OHÆHCl in 25% CH3COOH, pH (2.0), at 96 C, with 6 h, intermittent shaking
4	Organic F.	3 ml 0.02M HNO3, 30% H2O2 (adjusted to pH 2.0), water bath, 85 C, 5 h, 3.2 M CH_3COONH_4 in 20% (v/v) HNO_3, shaking for 30 min
5	Residual F.	Digested by 4 ml HNO3(70%) + 2ml HClO4(60%) + 5 ml HF (40%) to dryness

After each extraction, the mixture was centrifuged for 20 minutes, and filtered before the analysis of the tested metals

2.4. Assessment Indices of Soil Heavy Metals

To assess soil heavy metal contamination, two groups of Pollution Indices were applied; a) single pollution indices and; b) Integrated pollution indices.

a. Single pollution indices

- **Contamination Indices:** The contamination factor is determined using the method outlined by **Hakanson (1980)** as follows**;** **C= F Cm/ Cb**

Where; Cm is the metal content in the soil sample and Cb = is the reference value of the metal. In the current investigation, the data of the background content of the tested metal were taken from **Kabata-Pendias (2011)**. Following **Hakanson (1980)**, the soil is classified as follows: Cf ≤ 1 is low pollution; from ≤ 3 is moderate pollution; ≤ 6 is considerable pollution, and Cf > 6 is very high pollution.

- **Ecological risk assessment (Er):** The ecological risk factor is suggested by **Hakanson (1980)** to express the potential ecological risk of a given contaminant, and the calculated formula is;

$$Ei = CF \times Tr ;$$

Where; CF is the contamination factor, and Tr is the toxic response value of a given metal. Ti= Toxic response factors for Cd, Cr, Cu, Mn, Ni, Pb, Sb and Zn are 30, 2, 5, 1, 5, 5, 1, and 1, respectively.

The following categories are used to describe the risk factor: Er<40, low potential ecological risk; 40≤Er<80, moderate potential ecological risk; 80≤Er<160, considerable potential ecological risk; 160≤Er<320, high potential ecological risk; and Er≥320, very high ecological risk.

- **Index of geo-accumulation**. The Index of geo-accumulation (Igeo) was established by **Muller (1969)**, for determining the levels of metal contamination in the soil as follows;

Igeo = log2 (Cm/1.5Cb)

There are seven classes for Igeo contamination as follows; Igeo ≤0: uncontaminated; Igeo ≤ 1: slightly contaminated; Igeo ≤ 2: moderately contaminated; Igeo ≤ 3: moderately to heavily contaminated; Igeo ≤ 4: heavily contaminated; Igeo ≤ 5: heavily to extremely contaminated; and Igeo > 5: extremely contaminated.

- **Threshold pollution index (PIT)**

PIT was proposed by Qingjie *et al*. (2008) and its formula is as follows;

PIT = Ci / CTL

Where; CTL is the toxic level of a particular metal, its classes are; PIT < 1 Unpolluted, 1 ≤ PIT ≤ 2 Low polluted,2 ≤ PIT ≤ 3 Moderate polluted, 3 ≤ PIT ≤ 5 Strong polluted, 5 ≤ PIT Very strong pollute

- **Enrichment factor (EF)**

The formula of the Enrichment factor is;

EF = (Ci/Cie)s / (Ci/Cie)Rs

Where; (Ci/Cie)s is the ratio between the content of the interested pollutant to immobile element (Al or Fe) in the sample, and (Ci/Cie)Rs is the ratio between the concentration of the interested metal to immobile element (Al or Fe) in the selected reference sample. The categories of enrichment are stated as : < 2 minimal enrichment; EF< 5 moderate enrichment; EF< 20 significant enrichment; ; EF< 40 very high enrichment; ; 40 ≤EF extremely high enrichment.

b. integrated pollution indices

- **Pollution load index (PLI) (Tomlinson *et al*., 1980), the formula is ;**

PLI = [CF1 x CF2 x CF3 x CF4 x----x CFn]1/n

Where n = the number of metals concerned; CFn = the calculated contamination factor of metal n. the (PLI) classes are; < 1 (unpolluted), 0 (Perfection), and > 1 (polluted)

- **The potential Ecological Risk Index (PERI).** This index is an integrated one that describes the degree of contamination by summation of the ecological risk indices of the heavy metals as follows; **PERI = ∑Er1 +Er2+Er3+…….**

Where Er is the Single Index of Ecological Risk of metal 1,2,3, … and its categories are; PERI < 150 low ecological risk; 150 ≤ PERI < 300 moderate ecological risk; 300 ≤ PERI < 600 considerable ecological risk; 600 ≤ PERI very high ecological risk.

- **Nemerow pollution index (PINemerow).** This index, (PINemerow) has been widely used to evaluate soil quality and is calculated as follows;

$$\mathbf{PINemerow = ((1/m\sum Pi)^2 + (Pi\ max)^2)/2)^{0.5}}$$

Where Pi is the single pollution index of heavy metal "i"; Pi max is the maximum value of the single pollution indices of all the heavy metals and "m" is the count of the heavy metal species. The quality of the soil environment was classified into 5 grades from; PINemerow < 0.7- clean; 0.7 ≤ PINemerow < 1.0 – warning limit; 1 ≤ PINemerow < 2 – slightly pollution; 2 ≤ PINemerow < 3- moderately pollution; 3 ≤ PINemerow heavy pollution.

C. Risk Assessment Code (RAC)

The percentage of easily extractable portions of a particular metal from soil could serve as a reliable indicator of its toxicity (Tessier *et al*., 1979). RAC is an index that depends mainly on the laboratory results of sequential metal extraction procedures **(Soon and Abboud, 1990)**, and its soil's evaluation based on the summation of the available and readily available heavy metal fractions extracted sequentially from the soil, as follows;

REC= (exchangeable fraction +carbonate fractions)/Sum of Five Fraction) * 100.

RAC guidelines are, RAC: <1%: no risk, from 1:10 %: low risk; from 11: 30%: medium risk; From 31:50%: high risk; more than 50 very high risk.

3. Results and discussion

3.1. Soil Analytical Characteristics

Some of the soil properties of the tested soils are listed in Table 2. The soil has generally slightly alkaline reactions (pH = 8.28), high EC values (2.68 dS/m), low calcium carbonate content (1.63%), has measurable content of organic matter (2.53%). Meanwhile, the soil texture was characterized as sandy clay loam (clay, 29.20 %).

The total soil heavy metal contents of the tested soil and their threshold pollutant limits are given in Table 2. As shown from the Table the total contents of Zn, Cu, Fe, Mn, Ni, Pb, and Zn in (µg g-1) were; (3.32), (50.64), (13528.8), (506.18), (71.26), (36.50), and (73.48), respectively. It is clear from the results that total soil cadmium was higher than its corresponding threshold pollutant limits either compared by **World Health Organization (WHO), 1996; European Union (EUS), 2002**; or **Canadian Soil Quality Guidelines (CSQG), 2007** limits. In the same context, Ni content exceeded the pollutant limits set by **CSQG, 2007; WHO, 1996** but stayed below the threshold limits of **EUS, 2002**.

Meanwhile, the other pollutants (Cu, Fe, Mn, Pb, and Zn) were below the limits of the threshold used in this study.

So, according to the threshold limits used in this study, it could have been concluded that the tested soil was unpolluted with (Cu, Fe, Mn, Pb, and Zn) and polluted with (Cd and Ni). It is clear from the Table that the values of pollution vary greatly according to the threshold level used. These variations in threshold levels make it challenging to compare soil quality data internationally or even locally and this can complicate efforts to manage environmental issues. So, we need more efforts to harmonize standards and improve global regulations that can enhance consistency and effectiveness in managing soil quality.

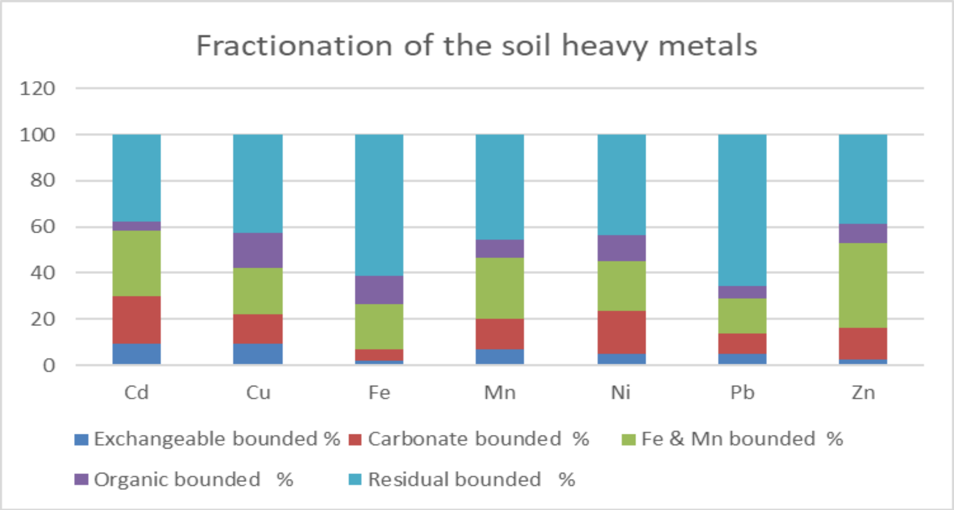
Table 1: General soil characters and heavy metal concentrations of the soil

Soil	pH	EC	CaCO3	OM	C. Sand	F. Sand	Silt	Clay
	8.284	2.684	1.63	2.53	10.4	23.97	36.43	29.20
Heavy Metal contents								
	Cd	**Cu**	**Fe**	**Mn**	**Ni**	**Pb**	**Zn**	
mg/kg	3.32	50.63	13528.8	506.18	71.26	36.5	73.48	
Threshold level								
EUS(2002)	3.00	140.0	50000.0	2000.0	75.00	300.00	300.0	
CSQG(2007)	1.40	63.00		2000.0	50.00	70.00	200.00	
WHO (1996)	3.00	100.0		2000.0	50.0	100.0	300.0	

Appr; (EUS), European Union; (CSQG), Canadian Soil Quality Guidelines; (WHO), World Health Organization.

3.2. Heavy metal fractions of the soil

The fractionation of the tested heavy metals in the soil is presented in Fig 1.



On average, the data reveal that the tested pollutants were mostly abundant in the Inert fraction (50.11 %), followed by reducible (21.67%), carbonate (13.33%), organic (9.29%), and an exchangeable fraction (5.60%). The residual phase, which averaged (50.11%), largely emerged in the crystal lattice of the soil and is expected to be not available except under very extreme conditions. In the same context, the Iron-manganese oxide phase accounted (21.67%). This fraction is considered also as an inert pool for heavy metals. These oxides appear as a coating on mineral surfaces or as fine separated particles and they can involve in several soil reactions as precipitation, and adsorption process. Also, the Organic fraction which averaged organic (9.29%) is persistent in terrestrial ecosystems for a long period but under special conditions, it may be mobilized by the decomposition process **(Kennedy *et al*., 1997)**.

Only, the remaining two fractions (carbonate and exchangeable) are considered readily available pools for metals in soil. The carbonate fraction (av. 13.33%) is considered as a loosely bound fraction and is easily affected by changes in soil conditions such as PH and redox value of the soil **(Tessier *et al*., 1979)**. Also, the exchangeable fraction which accounts (av. 5.6%) consists of metal species found in soil solution as ions or either as free hydrated ions or as complexes with organic or inorganic ligands and exchangeable form which involves weakly adsorbed metals retained on the solid surface **(Tessier *et al*., 1979; El-Gendi *et al*, 2018)**. So, the risk assessment code (RAC) index which is based on the summation of the percentages of these two fractions could be more reliable and convenient in the evaluation of soil pollutants **(Dudley *et al*., 1991, El-Gendi *et al*, 2016)**.

3.3. Soil heavy metal contamination

Values of single pollution indices as well as integrated pollution indices and risk assessment are presented in Table (2). Calculating the indices of soil pollution requires selecting the appropriate background content of the concerned pollutant (GB). Background values are defined as the baseline or pre-industrial values. According to **Reimann and Garret (2005)**, geochemical background (GB) is the normal abundance of an element in barren earth material. **Matschullat *et al*. (2000)** offer another definition of GB as a relative measure to differentiate between natural and human-influenced element concentrations. **Xu, G., *et al*. (2015)** defined metal background concentrations as the natural concentration of elements in a virgin environment. At the same time, there are wide differences between the established GB data, due to many reasons such as the type of soil, environmental chemistry of the concerned metal, as well as the economic and social status. As a result, using different background values will certainly lead to discrepancies in the assessment of pollutant contamination. So, the issue of choosing the appropriate metal–background content is crucial in determining soil pollution indexes. In the study, the background levels of the tested heavy metals reported by **Kabata-Pendias (2011)** were taken as background concentration.

Table (3). Assessment Indices of the tested heavy metals in the soil

Index	**Single pollution indices**							**integrated pollution indices**		
	Cd	**Cu**	**Fe**	**Mn**	**Ni**	**Pb**	**Zn**	**PLI**	**PERI**	**NCPI**
CF	8.10	1.30	0.36	1.04	2.46	1.35	1.05	1.457		
Er	242.93	6.51		1.04	12.29	6.76	1.05		270.57	
EF	22.74	3.66	1.00	2.91	6.90	3.80	2.95			
Igeo	2.43	-0.20	-2.07	-0.53	0.71	-0.15	-0.51			
Pi according to (EUS) limit	1.11	0.36	0.27	0.25	0.95	0.12	0.24			0.85
Pi according to(CSQG, 2007)	2.37	0.80	0.26	0.25	1.43	0.52	0.37			1.78
Pi according to WHO,1996)	1.11	0.51	0.26	0.25	1.43	0.37	0.24			0.55
RAC (Risk assessment code)	30.04	21.9	0.09	19.96	23.72	13.67	16.14			

3.4. Evaluate soil heavy metal pollution using contamination indices

Based on values of the contamination factor (CF) index (Table 3), the tested soil was categorized as low polluted with Fe, while it was moderately polluted by (Cu, Mn, Ni, Pb, and Zn). Meanwhile, it was classified as very highly polluted by Cd.

Ecological risk factor (Er) index values (Table 3) indicate that all the tested metals (Cu, Fe, Mn, Ni, Pb, and Zn) were considerably low potential ecological while, Cd was classified as highly potential ecological risk. Regarding the Er of Fe, there is currently no published response value for it. Additionally, the wide variations in the published response values often result in unreliable and poorly documented results.

It is evident from the results (Table 3) that the enrichment factor (EF) of Cd and Ni displayed significant enrichment, while Cu, Mn, Pb, and Zn were moderately enrichment, meanwhile, Fe displayed minimal enrichment. The low EF values indicate the pedogenic source of an element while the high EF values indicate anthropogenic origin. From the results, nearly, all the studied metals had considerable EF values indicating the anthropogenic origin of these metals.

In Table (3) almost all the Igeo values for Cu, Fe, Mn, Pb, and Zn are lower than zero, which indicates that the soil is uncontaminated by those metals, Whereas, Igeo for Ni (0.71) indicates that the soil is slightly contaminated by Ni, and it is moderately to heavily contaminated by Cd (Igeo 2.43).

The values of threshold pollution indices (PIT) as listed in Table (xxx), were calculated based on three of the published threshold limits **(WHO, 1996; EUS, 2002; and CSQG ,2007)**. The obtained results according to **(EUS,2002)** limit were nearly consistent with the results **WHO, 1996**) limits, in both PIT of Cd was considered as low polluted (1.11), PIT for (Cu, Fe, Mn, Pb, and Zn) were categorized as unpolluted. However, the difference between the two threshold limits was for PIT (Ni), which is categorized as low polluted and unpolluted in **EUS (2002)**, whereas in **WHO (1996)** threshold limit, respectively.

Using **CSQG (2007)** limits, the data shows that PIT of Cd (2.37) was moderately polluted, PIT of Ni (1.43), was low polluted, and PIT of the other tested pollutants (Cu, Fe, Mn, Pb, and Zn), were

From the previous results, it may be concluded that using different threshold limits will certainly lead to discrepancies in the assessment threshold pollution index. Some of the integrated pollution indices (PLI, PERI, and PINemerow) were also evaluated (Table 3). According to PLI value (1.45), the soil was polluted, while according to PERI, the soil was considered a moderate ecological risk (270.5). Concerning PIN index, the data showed that soil was considered as clean of pollution either taking threshold limits of **WHO (1996) or EUS (2002)**, in formula calculation, while the soil was categorized as slightly polluted when using **CSQG (2007)** limits, (PIN-less than 0.7). The results of the calculated risk assessment code (RAC) are also listed in Table (3) For (Fe), the soil was classified as low risk (RAC: 7.09), this outcome was consistent with only contamination factor (CF) value, However, these results were not consistent with all of the studied indices, except (CF, PLI, and PERI) indices, as well as NCPI when using threshold data of **CSQG (2007)**, only. The Cd, RAC (30.04), which is categorized as high-risk was consistent with nearly all of the studied indices, especially (CF, Er, EF, and PLI), and with less degree with (Igeo, PIT, and PERI) indices, as well as NCPI when using threshold data of **CSQG (2007)**, only. RAC values of (Cu, Mn, Ni, Pb, and Zn), were (21.9, 19.96, 23.72, 13.67, and 16.14, respectively) which are categorized as medium-risk were consistent with (CF, EF, PLI, and PERI) indices, as well as NCPI when using threshold data of **CSQG (2007)**, only.

4. Conclusion

The studied soil was alkaline, non-saline to slightly saline, had appreciable content of organic matter, and weakly calcareous soil. Regarding the textural characteristics, the soil is a clayey loam based on the USDA system. Concentrations of cadmium and nickel were above Threshold limits. While Cu, Fe, Mn, Pb, and Zn, were below their corresponding Threshold limits.

Fractionation results showed that the general trend in almost all tested metals the order was residual > followed by reducible, carbonate, organic, and an exchangeable fraction, with differences in quantity. Soil heavy metal assessment results revealed that among the tested single assessment indices (CF, EF) were consistent with RAC values, Also, the integrated indices (PLI, and PERI) indices, as well as NCPI when using threshold data of **CSQG (2007)**, only were consistent with RAC data.

The results of the employed risk assessment indices of the studied heavy metals showed that there are several disagreements between them. Therefore, relying solely on the total content or threshold levels of heavy metals as a tool to determine their potential impact on the environment may often be inaccurate, while the method of sequential extraction and estimation of the ARC index can be considered more realistic and represents the characteristics of the soil and environment under study.

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