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Original Research

Environmental Monitoring of Heavy-Metals Status and Human Health Risk Assessment in the Soil of Sahl El-Hessania Area, Egypt

Ahmed A. Abdelhafez[1]*, Mohamed H. H. Abbas[2], Tamer M. S. Attia[3]

[1]Soil Science Department, Faculty of Agriculture, Assiut University- The New Valley Branch, Egypt
[2]Faculty of Agriculture, Moshtohor, Benha University Qhalubia, Egypt
[3]Soils, Water and Environment Research Institute (SWERI), Agricultural Research Center (ARC), Giza- Egypt

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Abstract

Sahl El-Hussainia is an important reclamation project in Egypt located on El-Salam channel. Soils of these areas are irrigated with drainage water mixed with Nile water at a ratio of 1:1. North of Sahl El-Hussinia there exists Lake Manzalla, which receives large quantities of domestic, industrial, and agricultural discharges. The current study was aimed at measuring the contamination level of Sahl El-Hussainia soils with heavy metals and their effects on the grown plant contents of these contaminants. In addition, human risks resulting due to exposure to these heavy metals through different exposure routes were evaluated. To achieve these goals, soil and rice plant samples were collected from Khalid Ibn El-Waleed village, SahlEl-Hussainia, and analyzed for their contents of trace elements, i.e. Pb, Cd, Co, Ni, As, and Se. The results reveal that the soil of study suffers from moderate contamination with Pb, Co, Ni, and As to severe contamination with Cd and Se. Moreover, As and Pb were found at high concentrations in rice grains exceeding the permissible ones. The expected health risk assessments indicate that As, Se, and Pb can possess a health threat for children, whereas only As and Se can posses health problem for adults. Arsenic was found to contribute to more magnitude of cancer risks. Finally, bad management of Sahl El-Hussainia resulted in contamination of soil with trace elements. Accordingly, food obtained from the area of study might not be suitable for human consumption.

Keywords: Sahl El-Hussainia, heavy metals, contamination level, rice, risk assessment

Introduction

The Egyptian population has increased rapidly during the last decade from 70.2 million in 2002 to about 82.5 million in 2011 [1]. Such increases have caused growth in food demand [2]. Since 1980, the Egyptian government together with the private sector has moved towards further extensions in agricultural lands through a policy of land reclamation to increase the production of domestic food [3]. One of these projects is the reclamation of the areas surrounding El-Salam channel, known as Sahl El-Hussainia [4]. These areas are irrigated with drainage water mixed with Nile water at a ratio of 1:1 [5], in spite of the fact that the quality of irrigation water in El-Salam channel doesn't meet the standards of irrigation water [6]. Moreover, from time to time new areas are added to the El-Hussainia project through drying the wet lands of Lake Manzalla [4]. However, these soils are considered highly saline and not ideally suitable for crop production [7]. Thus, the farmers tend in many areas of Sahl El-Hussainia to establish fish farms during field cropping, particularly rice

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*e-mail: a.aziz@agrnv.au.edu.eg

Table 1. Physical and chemical properties of the studied soil.

pH	EC, dS m^{-1}	OM, %	Particle size distribution, %			Textural class
			Clay	Silt	Sand	
7.97±0.06	5.98±1.27	1.11±0.17	51.67±2.03	29.62±1.37	18.72±4.26	Clay

fields, and this dual utilization of the soil could increase the profit to the farmers to pay their debts [8]. Moreover, Lake Manzalla receives large quantities of untreated discharges, i.e. domestic, industrial, and agricultural [9, 10]. Therefore, the current research is aimed at monitoring the total and available contents of trace elements in the soil of Ibn El-Waleed village, Sahl El-Hussainia, which is considered among the new reclaimed lands in Egypt. Furthermore, distribution of these elements within the different parts of the plants grown therein was also investigated. Since soil contamination with trace elements can influence human health through different mechanisms, i.e., ingestion (either deliberate or involuntary), inhalation, and dermal absorption [11], human health aspects of the investigated trace elements therein will be under study.

Materials and Methods

Soil Sampling and Analysis

Surface soil samples (0-15 cm) were collected from eight different sites in Khalid Ibn El-Waleed village, Sahl El-Hessania, Port Said, Egypt, along the El-Salam irrigation channel. The collected samples were air dried and sieved to pass through a 2 mm sieve, and then used for further analysis. The particle size distribution was determined using the pipette method as described by Akoto et al. [12]. The soil reaction (pH) and electrical conductivity (EC) were determined in 1:1 soil to water suspensions and supernatant, respectively, using the method described by Jones [13]. Total organic carbon content was determined using Welkley-Black procedure [14]. Physical and chemical properties of the studied soil are shown in Table 1.

For analysis of the heavy metals content in soil, three soil samples of 0.50±0.04 g were taken from each site and placed in microwave Teflon vessels together with 9 mL of concentrated nitric acid (60%) and 3 mL of hydrofluoric acid. The microwave vessels were then sealed according to EPA method 3051 [15]. The vessels were placed in a microwave digestor (Mars-X, HP-500 plus, CEM Corporation) and the power was set at 1600 watts whereas the control temperature was kept at 180±5ºC until completion of soil digestion. The amount of available Pb, Cd, Co, Ni, As and Se in the soils were obtained by extracting with ammonium bicarbonate diethylene triamine penta acetic acid (ABDTPA). Specifically, 10 g of soil was shaken with 20 mL of AB-DTPA extraction reagent (0.005 M) for 2 h at 180 rpm [16]. The extracts were then passed through a Whatman No. 41 filter paper and transferred to 50 mL volumetric flasks, and then brought up to volume with deionized water.

Plant Sampling and Analysis

Ten whole rice plants were taken from the sampling spots of soils gently to avoid root's hair damage, washed

Table 2. Determination of Pb, Cd, Co, Ni, As, and Se (mg kg^{-1}) in standard reference materials.

Reference materials	Heavy metal	Found values (mg kg^{-1})	Certificate values (mg kg^{-1})	Recovery, %
Soil- GBW07429	Pb	39.4±3.5	38.0±2.0	97.0
	Cd	0.2±0.01	0.21±0.02	103.0
	Co	17.4±2.01	17.6±0.7	99.0
	Ni	39.1±4.71	41.0±1.0	95.0
	As	22.5±3.23	21.7±1.2	104.0
	Se	0.3±0.04	0.31±0.02	94.0
Plant- GBW07605	Pb	4.5±1.04	4.40±0.30	102.0
	Cd	0.1±0.01	0.057±0.01	93.0
	Co	0.2±0.03	0.18±0.02	106.0
	Ni	4.7±1.2	4.60±0.50	101.0
	As	0.3±0.02	0.28±0.04	96.0
	Se	0.1±0.01	0.072±0.0	93.0

with tap water, then deionized water, and separated into shoots, roots, and grains. The plant materials were oven dried at 70ºC for approximately 48 h, and then three portions of the dried plant samples (approximately 0.2 g each) were taken from each dried sample and placed in Teflon tubes together with 10 mL nitric acid (60%) and 2 mL hydrogen peroxide (30%). The vessels were left overnight and then digested at 180±5ºC using a microwave digestor (Mars-X, HP-500 plus, CEM Corporation) according to USEPA method 3052 [17]. The digests were then passed through a Whatman filter paper No. 41, and transferred to 50 mL volumetric flasks and then brought up to volume with deionized water.

Metal Determination and Quality Control

The concentrations of Pb, Cd, Co, Ni, As, and Se were measured in the digests of soil and plant samples by an inductively coupled plasma-mass spectrometer (ICP-mass, Agilent 7700, Japan). Reference materials of soil (GBW07429) and plants (GBW07605) were used to ensure high-quality results, and the recovery percentage of the studied elements ranged from 93% to 104% (Table 2), which ensure high precision for chemical analysis. Most of the chemicals used in this study were analytical grade, and mostly obtained from the Merck Company.

Statistical Analysis

All results were statistically analyzed using the SAS package (ver. 9.1). Means of four replicates for all chemical analysis were subjected to one way ANOVA. Tukey's honestly significant difference (HSD) studentized range test was applied for significant differences among means ($P<0.05$). Contamination factor (CF) and pollution load index (PLI) have been proved to be suitable tools for monitoring the levels of heavy metals in the environment [18-21]; therefore, CF and PLI were calculated to investigate the contamination level of the study area with heavy metals using the following equations:

$$CF = \frac{Cn}{Cr} \qquad \text{Eqs (1)}$$

...where Cn is the concentration of the investigated trace element in soil and Cr is the background level of the studied trace element. The level of soil contamination with the trace element could be interpreted as $CF<1$; indicating low contamination level; $1 \leq CF<3$; moderate contamination level; $3 \leq CF \leq 6$ high contamination; and $CF>6$; indicate severe contamination [22].

$$PLI = \sqrt{CF1 \times CF2 \times CF3 \times .... \times CFn} \qquad \text{Eqs (2)}$$

...where n, is the number of studied trace elements. The reference element used in this study was Al due to its high abundance in soil, and a lesser extent from other sources. The obtained results of PLI could be interpreted as: $PLI<1$; indicating the absence of contamination; $PLI=1$; indicating low contamination level; and $PLI>1$, refering to high contamination level and the probability of soil deterioration [23].

Table 3. Parameters used to characterize the ADD and HI values.

Parameter	Description	Value		Unit
		Adult	Children	
C	Contamination level	–		mg kg^{-1}
IR	Ingestion rate per unit time			
	Soil^{a}	100	200	mg day^{-1}
	Rice^{c}	123.29	61.64	g day^{-1}
EF^{a}	Exposure frequency	180		days year^{-1}
ED^{a}	Exposure duration	30	6	years
BW^{a}	Body weight	70	15	kg
AT^{a}	Average time (non-cancer risk)	30 years × 365 days	6 years × 365 days	days
	Average time (cancer risk)	70×365		
SL^{b}	Skin adherence factor	0.7	0.2	mg cm^{-2} h^{-1}
SA^{b}	Exposure skin area	3300	2800	cm^{2}
ABS^{b}	Dermal absorption factor (As)	0.03		unit less
	For other elements	0.001		
InhR^{a}	Inhalation rate	20.0	7.60	m^{3} kg^{-1}
PEF^{a}	Particle emission factor	1.36E + 09		m^{3} kg^{-1}

ADD average daily dose; HI hazard index a) Data from the USEPA [62]; b) Data from Zheng et al. [63]; c) Data from the Ministry of Agriculture and Land Reclamation Economic Affairs Sector (EAS) [64]

Table 4. Toxicity parameters and the calculated HI values used to investigate the non-cancer and cancer risks.

		Element					
		Pb	Cd	Co	Ni	As	Se
RfD[a,b]	Ingestion	3.50E-03	1.00E-03	2.00E-02	1.40E-01	3.00E-04	5.00E-03
	Dermal	5.25E-04	1.00E-05	1.60E-02	5.60E-03	1.23E-04	5.00E-03
	Inhalation	–	5.71E-05	1.43E-04	–	v	–
SF[b]	Ingestion	–	–	–	–	1.50E+0.0	–
	Dermal	–	–	–	–	3.66E+0.0	–
	Inhalation	–	6.30E+0.0	9.80E+0.0	8.40E+1.0	1.51E+1.0	–
HI	Adult	0.4770	0.1242	0.0156	0.0108	**8.7475**	**1.0974**
	Children	**1.1084**	0.2751	0.0436	0.0263	**19.3229**	**2.4289**
CR	Ingestion	–	–	–	–	**3.92E-03**	–
	Dermal	–	–	–	–	1.33E-05	v
	Inhalation	–	3.20E-015	4.07E-014	8.38E-013	6.77E-015	–

RfD reference dose, SF slope factor, CR cancer risk a) Data from the US EPA US EPA [29, 65]; b) FERREIRA-BAPTISTA, L., DE MIGUEL [66]; Values of HI >1 indicate adverse health impacts, and are in bold; The values of CR exceed the exceeded the maximum acceptable risks of 1E-06 to 1E-04 as suggested by USEPA [29] are in bold.

Risk Assessment

Hazard evolution for people living in the study sites was assessed by calculating the average daily intake of heavy metals from soil and rice plants through different exposure pathways [24-27] according to the following equations:

$$\text{ADD soil and grain ingestion} = \frac{C \times Ing\,R \times EF \times ED}{BW \times AT}$$

Eqs (3)

$$\text{ADD inhalation} = \frac{C \times Inh\,R \times EF \times ED}{PEF \times RW \times AT}$$

Eqs (4)

$$\text{ADD dermal} = \frac{C \times SA \times SL \times ABS \times EF \times ED}{BW \times AT}$$

Eqs (5)

...where ADD is the average daily dose of heavy metals, C the metal concentration of media (soil or grains; $mgkg^{-1}$), IR the ingestion rate per unit time, ED the exposure duration, EF the exposure frequency, BW humans body weight, AT the average time, SL the skin adherence factor, SA the surface area of contact, ABS the absorption factor, InhR inhalation rate, and PEF the particle emission factor. The parameters in the ADD formulas are presented in Table 3 and the toxicity indices of the elements are presented in Table 4. The obtained ADD values were used to determine the hazard quotient (HQ) and the hazard index (HI), which are as follows.

$$\text{HQ} = \frac{ADD}{RfD}$$

Eqs (6)

$$\text{HI} = \sum HQs$$

Eqs (7)

...where RfD is the reference dose, which is defined as the maximum daily intake of a contaminant without deleterious health effect [28]. If ADD > RfD, there will be a possible deleterious health impact.

Cancer risks were calculated for appropriate media and pathways; specifically, the cancer risks calculated through ingestion, inhalation and dermal exposure pathways for As. In addition, possible cancer risk due to exposure to Cd, Co, and Ni was calculated through the inhalation pathway. The average chronic daily intake ADD over 70 years and the slope factor SFs were used to calculate the possible numbers of cancer occurrence using the following linear equation:

$$\text{Cancer risk (CR)} = ADD \times SF$$

Eqs (8)

The total cancer risk for each metal was calculated for each exposure pathway, and compared with the maximum acceptable risk of 1E-06 to 1E-04 as suggested by the USEPA [29].

Results and Discussion

Total Contents of Trace Elements in Soil

Total concentrations of the studied trace elements in soil are illustrated in Fig. 1a. Comparing the concentrations of these trace elements in soil with the natural background levels of the earth's crust might give a more clear view of the levels of soil contamination that might occur

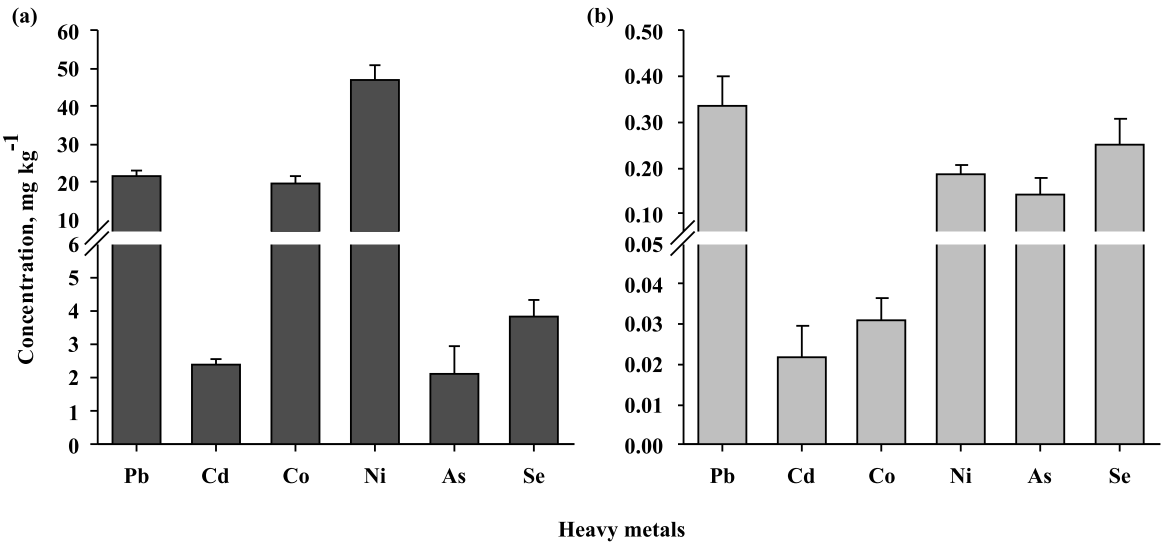

Fig. 1. Total (a) and AB-DTPA-extractable (b) contents of heavy metals (mg kg^{-1}) in soil.

within the area of study. The results show that the highest concentrations were recorded for Ni, Pb, and Co with average values of 46.91, 21.76, and 19.52 mg kg^{-1}, respectively. However, these concentrations are not much higher than the corresponding natural concentrations of the earth's crust; whereas the mean values of Cd, As, and Se exhibited far higher concentrations than the corresponding natural ones of the earth's crust, indicating considerable inputs of these metal ions from other sources than natural ones.

Contamination Factor as an Indicator for Soil Contaminated with Trace Elements

Table 5 reveals that the studied soil suffers from moderate contamination by Pb, Co, Ni, and As to severe contamination by Cd (CF=24.39) and Se (CFv=76.41). This might be attributed to the extensive applications of fertilizers containing traces of these contaminants. It is reported by Abdelhafez et al. [25] that excessive applications of mineral fertilizers, especially P-fertilizers, might account for contamination in soils in Egypt with Pb, Cd, Co, and Ni. Atafar et al. [30] almost got similar findings, where they found that the Cd, Pb, and As concentrations were increased in the cultivated soils due to mineral fertilizer applications. In addition, Abbas and Abdelhafez [31] highlighted the negative impact of pesticide manufacturing due to the presence of high As concentrations in the surrounding areas. Therefore, the unmanaged agricultural practices in terms of mineral fertilizer and pesticide applications might account for such contamination. Furthermore, the quality of irrigation water in El-Salam channel didn't satisfy the standards of irrigation water [6]. Moreover, the untreated water of Bahr El-Baqr Drainage discharges into El-Manzala Lake north of Sahl El-Hussinia [32]. Probably, there exists hydraulic continuity between the ground water of the area of study and the water in Lake Manzalla, at least during some periods of the year.

Table 5. Contamination factors (CFs) and pollution load index (PLI) of the studied heavy metals.

Pb	Cd	Co	Ni	As	Se	PLI
1.09	**24.39**	1.95	2.35	1.40	**76.41**	1.77

Values of CF < 1 indicate low contamination
Values of CF between 1 and 3 are underlined (moderate contamination)
Values of CF between 3 and 6 are in bold (considerable contamination)
Values of CF higher than 6 are underlined and bold (very high contamination)
PLI<1 no contamination
PLI=1 baseline levels of contaminants are present
PLI>1 high contamination

In general, the soil of study could be denoted to contaminated soil with trace elements (PLI = 1.77). Most of the previous studies considered total contents of trace elements a reliable indication for their toxicity; however, their available indices are more important than their total contents in determining their uptake and distribution in the different plant parts [31].

Available Contents of Trace Elements in Soil

Fig. 1b reveals that AB-DTPA-extractable Se and As were relatively high corresponding to their total content in the studied soil, whereas, the AB-DTPA extractable Pb, Cd, Co, and Ni were relatively low compared with their total content in soil. It is a well-known fact that for moisture, when it gets in contact with soil, microbial activity reduces the soil redox potential and thus affects the solubility and availability of trace elements in soil [33]. Under the waterlogged conditions of the paddy soils, the reduced soluble forms of As and Se are expected to be dominant, i.e. arsenite As^{3+} [34, 35] and selenite SeO$_3^{2-}$

Table 6. Total content (mg kg^{-1}) of the studied heavy metals in rice grains, shoots and roots.

Plant organ	Concentration, mg kg^{-1}					
	Pb	Cd	Co	Ni	As	Se
Rice grains	0.914^{b}	0.060^{ab}	0.158^{c}	0.774^{c}	1.465^{a}	3.074^{a}
Shoots	0.812^{b}	0.087^{b}	0.919^{b}	7.098^{b}	0.523^{b}	1.842^{ab}
Roots	1.534^{a}	0.409^{a}	3.944^{a}	9.489^{a}	0.797^{b}	1.426^{b}
MPL	0.200^{a}	0.100^{a}	-	-	1.000^{b}	-

Means with the same letter within a column are not significantly different at $p < 0.05$.
Maximum acceptable limits according to Das et al. and Commission Regulation of the European Union [49^{b},64^{a}].

[36, 37]. On the other hand, the alkaline conditions of the soil of study probably decreased the solubility and availability of Pb [38], Co [39,40], and Ni [41]. Cadmium precipitation is more likely to occur in the form of Cd under anoxic paddy conditions [42].

Distribution of Trace Metals in the Different Parts of the Plant Grown on the Study Area

The results shown in Table 6 reveal that the studied trace elements, i.e. Pb, Cd, Co, and Ni, recorded relatively higher concentrations in rice roots than in shoots and grains. Liu et al. (a and b) [43, 44] reported limited transfer of the absorbed Cd to rice grains. Also, Bhattacharyya et al. [45] recorded limited translocations for Co and Ni from rice shoots to grains. Although Pb content was relatively higher in roots than in shoots, its concentration in grain was much higher than the maximum permissible limits recommended by the European Union. It is reported that Pb precipitates mainly on root surfaces in the form of iron plaque [46]. However, the elevated concentrations of Pb have led to concurrent increases in Pb uptake and translocation to shoots [47]. These results agree with those obtained by Li et al. [48], who mentioned that feeding humans rice grains obtained from soils contaminated with Pb might possess high potential risk for health. On the other hand, high concentrations of As and Se were detected in rice grains. It was reported that the permissible limits of As in rice grain is 1.0 mg kg^{-1} [49]; moreover, the recorded concentrations even exceeded the 0.5±0.02 mg As kg^{-1}, which was recorded by Azizur Rahman et al. [50] for rice grain obtained from soil contaminated with 40 mg As kg^{-1}. However, in the current study the concentrations of As in grain were higher than those in roots and shoots. It is a well-known fact that As (mainly arsenite) has a relatively low mobility within rice plants [51, 52]; thus, we deduce that shoot uptake of As was probably more effective than root uptake under the paddy soil conditions. On the other hand, Se could transport via phloem and xylem [53], reaching concentrations in rice grains about 3 times higher than its concentration in shoots [54]. Riaz et al. [55] indicted that Se uptake could be stimulated by increasing As. Finally, the recorded high concentrations of As in rice grain may indicate potential health hazards in the food chain.

Human Health Aspects of the Investigated Trace Elements in the Studied Site

The hazard index (HI) was used by Abdelhafez et al. [26] as a measurement for the implications of ingesting grain and soil beside the dermal contact for health risk assessments. There is no doubt that the contact pathways vary between adults and children [56]. Thus, the hazard indexes of the studied trace elements should be considered separately for these two stages. The calculated values of (HI) for children exceeded 1 for As, Se, and Pb. On the other hand, the calculated hazard indexes for adults exceeded one only for As and Se. Such results indicate that serious health problems might occur for children and adults when feeding on rice grains or products obtained from this area. The calculated CF of Cd in soil was extremely high ($CF_{Cd}=24.39$), although no HI_{Cd} could be expected for adults or children feeding on the rice grains obtained from the studied area (Table 4). Accordingly, soil-Cd in Sahl El-Hussainia could be referred to as a hidden or potential contaminant. Similar results obtained by Jung and Thornton [57] reported that no health threats were detected due to feeding on rice grains obtained from paddy soil of relatively high concentrations of Cd and Pb. Also, Liu et al. [58] reported that paddy soil contaminated with Cr and Ni didn't possess health risks through rice feeding. Table 4 reveals that cancer risk CR exceeded the maximum acceptable risk of 1E-04 suggested by USEPA [29], and the recorded values ranged between 3.20E-015 and 3.92E-03 for Cd (inhalation route) and As (ingestion route), respectively. It is worth mentioning that As is considered a major source for cancer risk in the studied sites. The highest cancer risk was recorded for As through the ingestion pathway (3.92E-03) followed by As through the dermal contact (1.33E-05), pathway. However, cancer risk due to exposure to Cd, Co, and Ni was very low and didn't exceed the tolerable risk of 1E10-4. The results obtained of non-cancer and cancer risks are in agreement with those of statistical evaluation of soil contamination. The obtained results showed that humans are subject to potential risks due to exposure to heavy metals in Khalid Ibn Waleed village, Sahl El-Hussinia, Port-Said, Egypt through different exposure routes.

Conclusion

Levels of total Cd and Se in the soil of Khalid Ibn Waleed village, Sahl El-Hussinia, Port-Said, Egypt were relatively high, whereas those of Pb, Co, Ni, and As were moderate. The contamination of soil with heavy metals may be attributed to the seepage of water from Manzala Lake north of Sahl El-Hussinia, besides the contamination that probably occurs from using chemical fertilizers and pesticides. Accordingly, the concentrations of the investigated trace elements increased in the investigating soil. Under the conditions of the paddy soil, the reductions of As and Se were more likely to occur, thus increasing their availability and uptake by rice plants. It is thought that arsenite influx took place through aquaporins as H_3AsO_3 [59, 60]. Likewise, selenite influx took place through aquaporins in the form of H_2SeO_3 [61]. As and Se uptake by plant shoots was probably effective under the paddy soil conditions. Thus As and Se recorded relatively higher concentrations in grains than in roots, even As concentrations in rice grains exceeded the maximum permissible levels of 1 $\mu g\ g^{-1}$according to Das et al. [49]. On the other hand, reductions in the mobility and uptake of Cd might occur in the paddy soil; however, the HI_{Cd} indicates potential risk for children. Thus the rice grains obtained from this area might pose serious health aspects for children and adults. Arsenic was found to contribute to a greater magnitude of cancer risks. We concluded that continuous monitoring of heavy metals in the agricultural soils, of Khalid Ibn Waleed village, Sahl El-Hussinia, Port-Said; Egypt is necessary to ensure their safe use.

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