Ecological and Human Health Risk Assessment of Heavy Metals in Municipal Sewage Sludge for Land Application

Laila A. Al-Khatib[1,*], Ahmed M. AlHanaktah[2], and Feras Y. Fraige[3]

[1]Environmental Engineering Department, Faculty of Engineering, Al-Hussein Bin Talal University, Ma'an, Jordan
[2]Laboratories and Quality Department, Aqaba Water Company, Aqaba, Jordan
[3]Mining & Minerals, Mechanical Engineering Department, Faculty of Engineering, Al-Hussein Bin Talal University, Ma'an, Jordan
Email: laila@ahu.edu.jo and lailaalkhatib2003@gmail.com (L.A.A.); ahmad112022@yahoo.com (A.M.A.); Ferasfraige@gmail.com (F.Y.F.)
*Corresponding author

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***Abstract*—Sewage sludge is an unavoidable waste product generated from wastewater treatment. In line with sustainable practice, it has been applied worldwide in land applications due to its nutrient value. However, heavy metals (HMs) in sludge have become a major limiting factor for this way of disposal. This study aimed to determine the potential ecological and health risks of HMs in sludge for land application. Samples were collected from five wastewater treatment plants (WWTPs) in southern Jordan. Results showed that the levels of heavy metals in sludge were below the limit threshold of EPA, EU, and Jordanian standards. However, the average potential ecological risk index (RI) was 462.15, indicating a high-risk level. Results of health risk analysis revealed that the dominant route of HMs exposure was ingestion. It was shown that all WWTPs have no potential health risk associated with the presence of non-carcinogenic HMs in the sludge. However, sludge sampled from two investigated plants had carcinogenic risk for children. This study indicated that relying solely on regulatory limit values is inadequate to determine the suitability of sewage sludge for agricultural use and land applications; instead, a comprehensive risk assessment should be carried out.**

***Keywords*—ecological risk assessment, health hazard assessment, heavy metal, land application, sewage sludge, wastewater treatment**

I. Introduction

Sewage sludge, a byproduct of wastewater treatment, poses disposal challenges due to its increasing volume and environmental impact. The main sludge disposal routes are incineration, sanitary landfill, or land application [1]. Due to its high content of Organic Matter (OM) and nutrients such as nitrogen and phosphorus which are requirements for many crops, sludge is used for land-based applications including structural soil improvement, soil buffer, and soil amendment. It was reported that applying sludge on land has been found to increase agricultural yield and improve soil qualities [2–4].

It was observed that there was a continual decline in the amount of organic matter present in the soil of some European countries in the Mediterranean region due to the combination of elevated summer temperatures and unsustainable farming methods [5]. As a result, and due to its substantial organic matter contents, 40% of sewage sludge is utilized as soil organic supplement [6]. In the United States about 60% of sewage sludge is utilized to improve soil while in China, about 48% of sewage sludge is used in agriculture [7]. In developing countries like Jordan, landfilling is the primary method of sludge disposal. However, due to sustainability concerns, reuse options such as land application have gained importance worldwide [8, 9]. Land application is the most effective and cost-efficient method for sewage sludge disposal, as it serves as a fertilizer or conditioner. This method demonstrates high efficiency and is economically and environmentally attractive [3, 10, 11]. It is particularly advantageous for developing countries, as it offers numerous benefits [12, 13].

Utilizing sewage sludge as a fertilizer to enrich the soil with nutrients and organic matter can have advantages, but it also has a potential risk due to the existence of pollutants such as heavy metals, organic compounds, and pathogens [6]. For example, when heavy metals in sewage sludge enter the soil through agricultural use, they can threaten the ecological environment and human health [7]. These metals can build up in the food chain, resulting in harmful consequences for both humans and the ecology [14]. Human health can be negatively impacted by being exposed to high levels of certain metals, leading to harmful non-carcinogenic (chronic) and carcinogenic disorders [15].

Therefore, it is essential to evaluate the toxicity and carcinogenic properties of sewage sludge to make informed decisions about its suitability for agricultural use [13]. To minimize risk from sludge land application, analyzing sludge for heavy metals is a critical step. While it is crucial to determine the metal concentration in sludge, this alone is insufficient for assessing its potential for land application. Therefore, it is necessary to conduct a thorough analysis of ecological and health risk assessment before considering land application. Both children and adults can be exposed to health risks, including both carcinogenic and non-carcinogenic, through inhalation, contact with the skin, and ingestion [9, 16].

The novelty of this work relies on conducting a thorough assessment of the potential ecological and health risks of heavy metals in sewage sludge. Studies concerning the human health consequences of applying sewage sludge in agriculture are rather rare in the literature [9, 13]. Moreover, there are only a limited number of research that evaluate the disparity in tolerance between adults and children when they are exposed to heavy metals by ingestion [17]. Furthermore, there is scarce research on the heavy metal composition of sludge produced by WWTPs in Jordan and its usage in agriculture [18, 19]. In addition, no prior research has been conducted on the ecological or health risk assessment of utilizing sludge in land applications in Jordan up to the author's knowledge. Thus, the uniqueness of this research relies on bridging the knowledge gaps of heavy metal profiles of WWTP sludges in Jordan, as well as the risk associated with their reuse.

The objectives of the current study were: (1) to measure heavy metal contents in sludge sampled from five WWTPs in southern Jordan; (2) to assess the potential ecological risks of heavy metals in sewage sludge; (3) to evaluate heavy metal exposure from sewage sludge and distinguish the difference in exposure of adults and children; and (4) to estimate carcinogenic and non-carcinogenic human health risks using ingestion, dermal contact and inhalation exposures to land-applied sewage sludge by children and adults.

II. Materials and Methods

A. Study Area

Five municipal wastewater treatment plants covering the south of Jordan were encompassed in this study, including Aqaba Mechanical Wastewater Treatment Plant, Wadi Mousa Wastewater Treatment Plant, Ma'an Wastewater Treatment Plant, Adnaniah-Mu'tah and Mazar Wastewater Treatment Plant, and Tafilah Wastewater Treatment Plant. Fig. 1 shows the WWTPs' geographic location. The details of WWTPs are presented in Table 1.

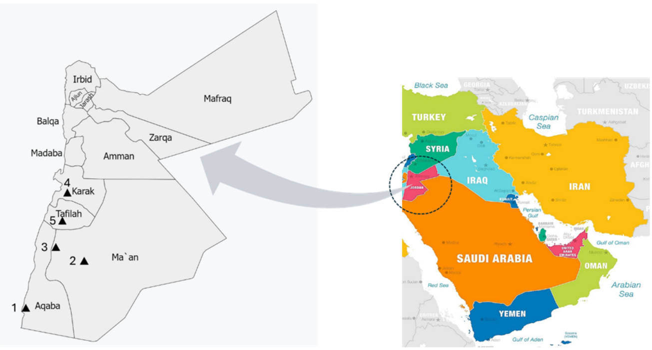

Fig. 1. The map presents the study area location and the investigated WWTPs (1 to 5).

Table 1. Wastewater treatment plants under investigation

Plant Name	Symbol	Type of Wastewater Treatment	Design capacity (m^3/d)
Aqaba Mechanical Wastewater Treatment Plant	WWTP1	Activated sludge	28000
Ma'an Wastewater Treatment Plant	WWTP2	Activated sludge	7000
Wadi Mousa Wastewater Treatment Plant	WWTP3	Activated Sludge	3400
Adnaniah, Mu'tah, and Mazar Wastewater Treatment Plant	WWTP4	Activated Sludge	7060
Tafilah Wastewater Treatment Plant	WWTP5	Activated sludge	5000

B. Sewage Sludge Sampling

The sewage sludge samples were collected from the different WWTPs located in the southern part of Jordan. Subsamples were collected from four different sites in each WWTP to obtain representative samples. Then they were combined and homogenized to form a single representative sample. The collected samples were kept in polypropylene containers and stored in a refrigerator at 4 °C until analysis.

C. Determination of Physicochemical Characteristics and Total Metal Concentrations of Sewage Sludge

The pH value of samples was determined using a calibrated digital pH meter (WTW PH, 7110, Germany). The content of dry matter, moisture content, and organic matter in sewage sludge were determined after drying at 105 °C and being ignited at 550 °C according to EPA Method 1684 [20].

The chemical analysis focused on the following heavy metals: Cd, Cr, Cu, Hg, Ni, Pb, As, Se, Mo, and Zn. These metals were selected because they are commonly found in sewage sludge and pose a potential risk due to their toxicity [21, 22]. In addition, they are regulated based on the requirements for sewage sludge intended for land application, which have been defined in the USEPA [23], EU [24], and Jordanian Standards [25] (see Table S1 in the supplementary material). The determination of the overall heavy metal concentrations involved the preparation of sludge samples. This included drying the samples in an oven at a temperature of 105 °C, followed by grinding and sieving them through a 0.2 mm sieve. The resulting mixture was then placed in plastic bags for further analysis. Then, samples were weighed and digested with nitric acid and hydrogen peroxide using a microwave digestion system based on the USEPA Method 3051B. The total heavy metal concentrations in the samples and extracts were determined using inductively coupled plasma optical spectrometry (ICP-OES, PerkinElmer Inc., USA). Mercury was analyzed using the cold vapor atomic absorption spectrometry technique. Each analysis was conducted three times, and average and standard deviation (SD) were calculated.

D. Assessment of the Level of Contamination and Ecological Risk

Pollution levels and ecological risks associated with sewage sludge are crucial considerations due to the potential environmental impact they may have. Various individual and complex indicators were employed to assess the pollution level and ecological risk associated with the application of sludge in agricultural soil amendment or use in land reclamation projects [13, 26].

1) Geoaccumulation index (I_{geo})

The I_{geo} aims to assess the level of contamination by heavy metals in sediments [22, 27]. Recently, the I_{geo} has been employed to evaluate the pollution degree of heavy metals in sewage sludge for agricultural use [28, 29]. This index was calculated based on the following equation [27]:

$$I_{geo} = \log_2\left(\frac{C}{1.5B}\right) \quad (1)$$

where C is the heavy metal content in sludge samples, B is the background heavy metal concentrations in the soil, and 1.5 is a correction factor for the background matrix due to lithospheric effects. In the current study the global average concentration of metals in shale by [30] was selected as the background value (Table S2 in the supplementary material). The classification of the value of the I_{geo} for the evaluation of contamination by HMs is shown in Table S3 in the supplementary material.

2) Potential ecological risk index (RI)

The RI is widely applied to evaluate heavy metal pollution [7, 31, 32]. RI assesses the ecological risk posed by heavy metals by considering their tendency to build up, their toxicity, and their interactions with the ecosystem [7]. It has been demonstrated that RI is a reliable, quick, and standard method for determining the pollution level of heavy metals and potential ecological risks [29]. In the present study, the RI was determined using the following formulas:

$$C_f = \frac{C}{C_n} \quad (2)$$

where, C_f is the single heavy metal pollution factor of the ith heavy metal, and C_n is the background values of the ith heavy metal.

$$E_r = T_r \times C_f \quad (3)$$

where E_r is the monomial potential ecological risk coefficient of the ith heavy metal; T_r is the heavy metal toxic response factor, according to Hakanson; the values for each heavy metal are Zn (1) < Cr (2) < Cu (5) = Pb (5) < As (10) < Cd (30) < Hg (40) [7].

The potential ecological risk index which is the sum of the monomial potential ecological risk coefficient factors of the contamination with heavy metals from sewage sludge was defined by the formula:

$$RI = \sum E_r \quad (4)$$

The classification of the value of E_r and RI for the evaluation of contamination by HMs is shown in Table S4 in the supplementary material.

E. Health Risk Assessment

Toxicity is a significant concern when utilizing sewage sludge for agricultural purposes [13]. In this study the US Environmental Protection Agency (USEPA) Part 503 Rule - Standards for the Use and Disposal of Sewage Sludge agricultural application regulation was applied for exposure assessment of heavy metal and risk characterization [23]. Due to differences in behavior and physiology between adults and children (including different responses to risks), they were treated differently when evaluating health risks.

1) Exposure assessment

The main pathways through which the person can be exposed to heavy metals contained in sewage sludge are accidental ingestion, inhalation, and dermal contact. The average daily dose for the individual exposure route can be determined by the following equations [33, 34]:

$$ADD_{ing} = C \times \frac{Ing\ R \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (5)$$

$$ADD_{inh} = C \times \frac{Inh\ R \times EF \times ED}{PEF \times BW \times AT} \quad (6)$$

$$ADD_{dermal} = C \times \frac{SA \times SL \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (7)$$

where, $\mathbf{ADD_{ing}}$, $\mathbf{ADD_{inh}}$, and $\mathbf{ADD_{dermal}}$ are the average daily doses (mg kg^{-1} d^{-1}) through ingestion, inhalation, and dermal contacts, respectively; **Ing R** is the ingestion rate of biosolid/sludge by children or adults which is 100 for adults and 200 mg d^{-1} for children [35]; **Inh R** is the rate of inhalation (7.6 and 20 m^3 d^{-1} for children and adults, respectively) [13, 36]; **EF** is the exposure frequency (d yr^{-1}) and it was assumed 350 d yr^{-1} [35]; **ED** is the exposure duration (year) assumed 30 years for adults and 6 years for children [35]; **SA** is the exposed skin area which is 2800 cm^2 for children and 5700 cm^2 for adults; **SL** is the skin adherence factor (0.2 and 0.07 mg cm^{-2} d^{-1} for children and adults, respectively) [13]; **ABS** is the dermal absorption factor (0.03 for As and 0.001 for the rest of the elements, unitless)[37]; **PEF** is the air particulate emission factor (m^3 kg^{-1}), 1.36 × 10^9 m^3 kg^{-1} [38]; **BW** the body weight (kg) which is 16 kg for children and 70 kg for adults [23]; **AT** is the averaging time, which is **ED** × 365 d for non-carcinogens and 70 years (lifetime) × 365 d for carcinogens [35]. The coefficient of 10^{-6} is the conversion factor of mg to kg.

2) Risk characterization

a) Non-carcinogenic risk assessment

To evaluate the combined non-carcinogenic impact of exposure to various heavy metals through different routes, the sum of the Hazard Quotient (HQ_{ij}) values for all individual heavy metals (i) via all specific routes (j) is expressed as the Hazard Index (HI). The equation used to determine this index was as follows [34, 39]:

$$HQ_{ij} = \frac{ADD_{ij}}{RfD_{ij}} \quad (8)$$

$$HI = \sum HQ_{ij} \quad (9)$$

where RfD_{ij} is the reference dose of individual metal (mg $kg^{-1} \cdot day^{-1}$) (Table S5 in the supplementary material).

In cases where the HI or HQ_{ij} value is more than 1, there can be non-carcinogenic effects [40].

b) Carcinogenic risk assessment

The probability of developing cancer as a result of a lifetime exposure to a potential carcinogen is known as the carcinogenic risk (CR) [41]. To determine the overall carcinogenic risk (TCR) effects of the HMs under study as well as the carcinogenic risk associated with each exposure pathway, the following equations were used [21]:

$$CR_{ij} = ADD_{ij} \times SF \quad (10)$$

$$TCR = \sum CR_{ij} \quad (11)$$

where, **SF** is the cancer slope factor (mg $kg^{-1} \cdot day^{-1}$) from individual heavy metal ingestion, inhalation, and skin contact (Table S5 in supplementary material). A **CR** of more than 1×10^{-4} is regarded as a potentially concerning cancer risk. The value of less than 1×10^{-6} can be disregarded as a cancer risk.

F. Data Analysis

Statistical analyses were performed by Microsoft Office Excel 2019. The average, standard deviation, maximum, and minimum values of descriptive statistics were computed. Through the use of Pearson correlation analysis, the relationships between the concentrations of each heavy metal in sewage sludge were determined; this is a crucial indicator of the heavy metals' sources. This assists in identifying potential metal contamination sources and their distribution in sewage sludge [42]. Strong correlations between heavy metals are shown by correlation coefficients close to 1. In order to verify the sources and ascertain the association between specific heavy metals, hierarchical cluster analysis (HCA) was also employed. It is typically applied as a validation method to identify the origins of HM contamination. Using this method, HMs with comparable properties can be grouped together [43, 44]. The HCA was carried out with Python.

III. Results and Discussion

A. Sewage Sludge Physicochemical Characteristics

The physicochemical characteristics parameters of sludge from the five municipal Wastewater Treatment Plants are presented in Table 2. pH is a crucial factor in determining the mobility of metals in soil. Increasing pH values decrease the migration of heavy metals and enhance the content released [13, 45, 46]. The pH of sludge ranged from 6.9 to 7.9, which presents weak alkalinity. And is consistent with previous studies for sewage sludge land applications in other regions [13, 29, 45].

Table 2. Physiochemical characteristics of sludge samples

Parameter	WWTP1	WWTP2	WWTP3	WWTP4	WWTP5
pH	7.3	6.9	7.9	7.4	7.6
Organic matter (%)	61.20	49.80	46.70	44.30	53
Dry matter (%)	89.70	84.34	89.50	91.30	54.60
Moisture Content (%)	10.3	15.66	10.50	8.70	45.40

The moisture content of sewage sludge is 8.70%–45.40%. The Jordanian guidelines for disposing of sludge from municipal WWTPs state that the sludge used in agriculture must meet certain physical indicators [25]. The water content should be less than 10% for Class I, and less than 40% for Class II. Based on these requirements, it can be confirmed that the moisture content of sludge in each sewage treatment plant complied with the standard regulations for class II except WWTP5 (moisture content = 45.40%). While only WWTP1, WWTP3, and WWTP4 complied with the standard regulation for class I.

The organic matter content ranged from 44.30% to 61.20%. Determining the nutritional value of sewage sludge for plants in land application requires consideration of both organic matter and nutrient content [2]. These values exceeded the range typically found in organic soil, indicating that the samples were abundant in organic matter. This may be attributed to the composition of sewage sludge from WWTP, which mostly consists of residual organic materials, microbial biomass, and settled solids resulting from the treatment process. Recycling nutrients through land application promotes plant growth and improves soil properties [46].

B. Total Heavy Metal Concentrations

The heavy metal concentrations in sewage sludge from the five different WWTPs are presented in Table 3. According to the analysis, the concentration of zinc in sewage sludge was the highest, and it ranged from (130±60 to 770±110 mg/kg), followed by Cu (49±19 to 138±16.2 mg/kg). While Cd had the lowest concentration (1±0.1 to 5±0.5 mg/kg). Different previous studies also reported similar trends [47, 48]. According to Li *et al.*, [47], the concentrations of their studied heavy metals in sewage sludge were ordered as follows: Zn>Cu>Cr>Ni>Pb>As >Hg>Cd. The order for the sludge examined in the current study in terms of total heavy metal concentrations is generally the same. Also, slight differences in Pb, Mo, and Se concentrations can be observed, which could be due to the different characteristics of the sewage entering each plant. The findings of the present work represented by the lowest Cd concentration and not detecting As and Hg in all samples from the investigated WWTPs are significant, especially when sewage sludge is intended for agriculture uses as these heavy metals are the most toxic. In addition, it was found that Zn and Cu were the most predominant HMs, which are considered essential elements for higher plants [22, 48]. In summary, the concentrations of the investigated heavy metals were within the permissible limits of USEPA [23], EU [24], and Jordanian Standards [25] for sludge use in agricultural and land applications.

Table 3. Heavy metal content (mg/kg) in sludge samples (Average ± standard deviation)

Heavy metal	WWTP1	WWTP2	WWTP3	WWTP4	WWTP5
Cd	4±0.1	5±0.5	1±0.1	2±0.2	2±0.2
Cr	30±0.2	56±12.4	7±2.0	41±3	22±8
Cu	137±22	101±20	49±19	108±13	138±16.2
Mo	7±0.7	6±0.6	3±0.4	12±5.3	6±2
Ni	25±6	42±13	7±0.1	41±6	20±4.5
Se	11±0.6	-	-	25±1	11±0.7
Pb	11±3	5±2	8±3	10±1.1	8±2.3
Zn	770±110	715±141	130±60	570±122	680±180

The Pearson correlation coefficients (r) were used to analyze the relationships between different heavy metals and explain the interconnections among them (Fig. 2). This study found strong, positive connections between the following elements: Zn and Cd, Zn and Cr, Zn and Cu, Zn and Mo, Zn and Ni, Se and Mo, and Cr and Cd. Also, significant positive correlations were found between Ni and Cd, Ni and Cr, and Ni and Mo. The significant correlations revealed that the correlated metals may come from the same source.

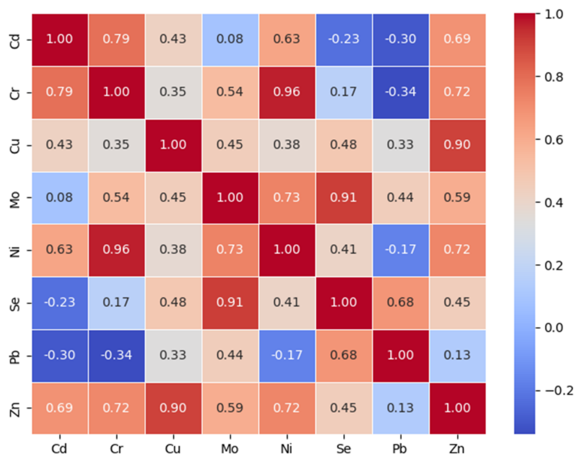

Fig. 2. Pearson correlation coefficients for heavy metal in sludge samples.

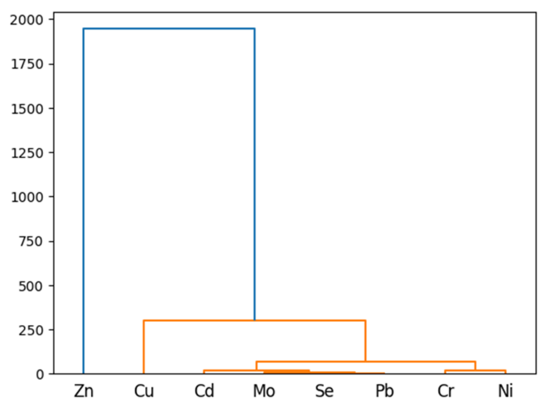

Fig. 3. Hierarchical cluster analysis of the heavy metals in sludge samples.

HCA was utilized to determine the origins of heavy metals present in sewage sludge. Fig. 3 shows the dendrograms based on the average heavy metal content of the samples. The dendrogram, constructed based on the heavy metal content, exhibited a single cluster consisting of one group and a singleton. Because of its considerable distance, Zn had no significant commonalities with the other elements. Cu, Cd, Pb, Mo, Se, Cr, and Ni were clustered into one group. The metal composition may indicate a range of human activities, including industrial and municipal discharges, agricultural practices, vehicle emissions, urban waste, and other sources [47].

C. Assessment of Pollution Level and Ecological Risk

1) Geo-accumulation Index results

Table 4. Results of geoaccumulation index (I_{geo}) for heavy metals

Heavy metal	I_{geo}				
	WWTP1	WWTP2	WWTP3	WWTP4	WWTP5
Cd	3.15	3.47	1.15	2.15	2.15
Cr	-2.17	-1.27	-4.27	-1.72	-2.62
Ni	-2.03	-1.28	-3.87	-1.31	-2.35
Pb	-1.45	-2.58	-1.91	-1.58	-1.91
Cu	1.02	0.58	-0.46	0.68	1.03
Mo	0.84	0.62	-0.38	1.62	0.62
Se	3.61	-	-	4.80	3.61
Zn	2.43	2.33	-0.13	2.23	2.25

The Geo-accumulation index for sewage sludge land application indicated varying levels of heavy metal contamination. Contamination by metals in sewage sludge for agricultural and land application purposes from the five WWTPs was classified between classes 0 and 6 (see Table S3). The results summarized in Table 4 showed that Cd, Se, and Zn contamination was the highest; Mo and Cu contamination was moderate. While the calculated index values showed that Ni, Pb, and Cr in all WWTPs were placed in the zero class (i.e., practically uncontaminated). Consequently, Cd, Se, and Zn were chosen as the primary control metals. The I_{geo} values for the metals in all five WWTPs exhibited a wide range, indicating the diverse characteristics of sewage sludge and pollution sources of metals.

2) Potential ecological risk assessment of heavy metals

The potential ecological risk of heavy metals in sewage sludge were evaluated using the Hakanson coefficient method. The average value of the related heavy metals was computed after the E_r value of each individual heavy metal in each sewage plant was estimated. Table 5 presents the results of the heavy metals evaluation conducted at each of the five WWTPs. The average value E_r of heavy metals follows the order: Cd (280) > Se (156.67) > Cu (11.84) > Zn (6.24) > Mo (2.62) > Pb (2.10) > Cr (0.69). Cd had the highest potential ecological risk, which belongs to extremely strong ecological risk, and the other metals had low ecological risk (E_r <30) except Se which has a very strong risk.

The comprehensive ecological risk RI values of municipal WWTPs ranged from 110.64 to 646.76 with an average value of 462.15 which belongs to the high-risk level. This indicated that in case the sludge is released into the environment without undergoing pretreatment, it would pose a threat to ecological safety. Moreover, Cd is the prime contributor to the high ecological risk associated with sludge.

Table 5. Potential ecological risk assessment results from heavy metals contained in sewage sludge used for land application

WWTP	E_r								RI	RI Description
	Cd	Cr	Cu	Mo	Ni	Se	Pb	Zn		
WWTP1	400.00	0.67	15.22	2.69	1.84	183.33	2.75	8.11	614.61	Very High
WWTP2	500.00	1.24	11.22	2.31	3.09	0.00	1.25	7.53	526.64	High
WWTP3	100.00	0.16	5.44	1.15	0.51	0.00	2.00	1.37	110.64	Low
WWTP4	200.00	0.91	12.00	4.62	3.01	416.67	2.50	7.05	646.76	Very High
WWTP5	200.00	0.49	15.33	2.31	1.47	183.33	2.00	7.16	412.09	High
Min	100.00	0.16	5.44	1.15	0.51	0.00	1.25	1.37	110.64	
Max	500.00	1.24	15.33	4.62	3.09	416.67	2.75	8.11	646.76	
Mean	280.00	0.69	11.84	2.62	1.99	156.67	2.10	6.24	462.15	
SD	146.97	0.37	3.60	1.13	0.97	153.70	0.51	2.46	193.68	
Slight Risk	0	100%	100%	100%	100%	40%	100%	100%	20%	Low Risk
Medium Risk	0	0	0	0	0	0	0	0	0	Moderate Risk
Strong Risk	20%	0	0	0	0	0	0	0	40%	High Risk
Very Strong Risk	40%	0	0	0	0	40%	0	0	40%	Very High Risk
Extremely Strong Risk	40%	0	0	0	0	20%	0	0		

where E_r is the monomial potential ecological risk coefficient and RI is the potential ecological risk index.

D. Health Risk Assessment

1) Exposure assessment results

Heavy metals present in sewage sludge can enter the human body via ingestion, inhalation, and dermal contact pathways. The values of the average daily exposure dose (ADD) of metals in sewage sludge samples from the different wastewater treatment plants are shown in Tables (S6 – S10). The results revealed that the most significant pathway of exposure was ingestion while inhalation was the least for both adults and children. This is similar to the findings of previous studies [22]. It was observed that Cu and Zn had the highest values of ADD for different routes of exposure to non-carcinogenic metals for adults and children. While the lower values were indicated to Cd and Pb. Overall, the magnitude of adult exposure rates was comparatively lower than that of children, indicating that children are more exposed to these metals. Consistent results were noted by [41] and [49].

2) Health risk characterization

Analytical assessment was conducted to determine the concentrations of Cd, Cr, Cu, Mo, Ni, Se, Pb, and Zn in various samples. The health impacts, both non-carcinogenic and carcinogenic, were then evaluated using risk values and hazard indices.

a) Non-carcinogenic health risk

To assess the non-carcinogenic health risks, the site-specific overall Hazard Index (HI) values for children and adults were calculated by summing up the exposure due to ingestion, inhalation, and dermal contact of all heavy metals, as shown in Fig. 4. Individual HQs for each heavy metal were also presented in Tables S11 – S15.

The study revealed that the average values of HQs for non-carcinogenic HMs can be ranked in the following sequence: ingestion > dermal contact > inhalation. This trend aligned with prior research findings [13, 21, 45].

The samples acquired from WWTP4 had the highest Hazard Index (HI) values, which represents the combined exposures to heavy metals through ingestion, inhalation, and dermal contact, for both children and adults. This may be due to the high concentration of Mo and Se observed in this plant. Also, the overall mean values of HI for children were higher than for adults for all analyzed samples from the five investigated WWTPs. In all instances, however, the HI and HQ values for the HMs under consideration did not surpass the threshold value of 1. This indicated that the presence of non-carcinogenic HMs in the dewatered sludges from the studied wastewater treatment plants does not pose any possible concern to human health if they are used for land applications and agricultural purposes.

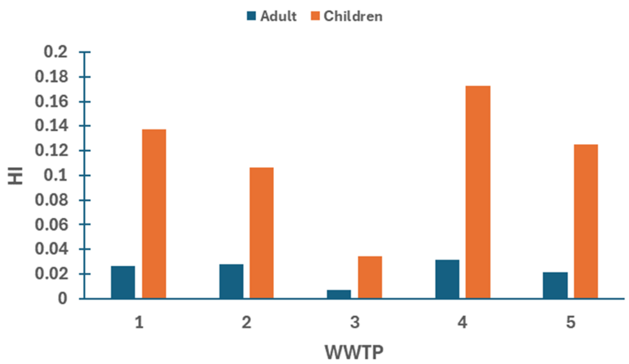

Fig. 4. Site-specific overall Hazard Index (HI) of accumulative exposure routes to heavy metals.

b) Carcinogenic health risk

The carcinogenic risk (CR) and total carcinogenic risk (TCR) for children and adults exposed to HMs in sludge samples are estimated and presented in Tables S11 – S15. The results showed that the carcinogenic effect varied according to the exposure pathways. Ingestion was the major contributor pathway of TCR for both children and adults. And TCR through inhalation was the least contributor. The average TCR for the exposure to HMs for all routes of exposure for all samples is shown in Fig. 5. The results revealed that the cancer risk for children is higher than that for adults. In addition, it is observed that the highest TCR was obtained for WWTP2.

The estimated carcinogenic risks for WWTP1, WWTP3, and WWTP5 indicated that there were no carcinogenic risks of exposure to HMs in the samples from these WWTPs. On the other hand, the estimated carcinogenic risks for WWTP2 and WWTP4 were also considered safe for adults through all routes of exposure and for children through inhalation and dermal contact. However, the threshold value of 1×10^{-4} has only been surpassed for the ingestion exposure of children in WWTP2 and WWTP4, with values of 1.35×10^{-4} and 1.06×10^{-4}, respectively. This indicated that sewage sludge from these plants may pose a cancer risk. Therefore, it is recommended to decrease the levels of HMs in sewage sludge prior to its utilization in agricultural and land applications in order to avoid the potential risks to health.

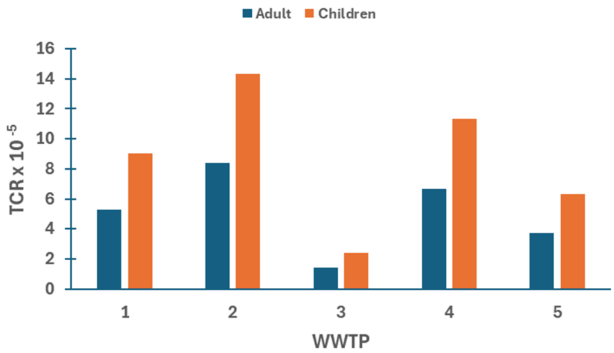

Fig. 5. Site-specific total carcinogenic risks (TCR) of accumulative exposure routes to heavy metals.

IV. CONCLUSIONS

Utilizing sewage sludge for reuse in agriculture is the most sustainable method to minimize environmental degradation and promote a circular economy. This study is the first comprehensive evaluation of the ecological and human health risks associated with heavy metals in sewage sludge derived from municipal wastewater treatment plants in Jordan. The results indicated that Zn and Cu exhibited the highest concentrations among the different sludge samples, whereas Cd demonstrated the lowest concentration. In addition, all heavy metals concentrations in sewage sludge samples were found within the standard limits stated by USEPA [23], EU [24], and Jordanian Standards [25]. However, the assessment of ecological risk index (RI) values of municipal sewage treatment plants varied from 110.64 to 646.76 with an average value of 462.15. RI indicated a high-risk level. On the other hand, the findings from the assessment of carcinogenic and non-carcinogenic risks revealed that ingestion constituted the primary route of exposure to HMs. The analysis of sludge samples from all WWTPs under consideration revealed that the presence of non-carcinogenic HMs does not pose a potential risk to human health. However, it has been suggested that children may be exposed to carcinogenic risk in the case of WWTP2 and WWTP4. This implies that sludge treatment is required before using it in land application.

The study limitations can be related to the design of human health risk assessments. Where the non-carcinogenic health risks were assessed for eight heavy metals, the carcinogenic were assessed only for four HMs due to the availability of valid health guidance values published by international agencies. Also, the intake of HMs from the soil by the plant is possible, however, this concern is not investigated in this study.

The results of this study indicated that it is not enough to determine HMs in the municipal sewage sludge to assess its suitability for potential reuse in land applications but, it is also crucial to assess the ecological and human health risks associated with the sludge to prevent environmental pollution and safeguard human health.

SUPPLEMENTARY MATERIAL

Table S1. Allowable concentration of sewage sludge samples according to USEPA [23], EU [24], and Jordanian Standards [25] for land application

Parameter	Allowable Concentration according to JS 1145 - 2016			EU	USEPA
	Class I	Class II	Class III		
As (mg/kg)	41	75	75	–	41
Cd (mg/kg)	40	40	85	20 – 40	39
Cr (mg/kg)	900	900	3000	–	1200
Cu (mg/kg)	1500	3000	4300	1000–1750	1500
Hg (mg/kg)	17	57	57	16–25	–
Mo (mg/kg)	75	75	75	–	–
Ni (mg/kg)	300	400	420	300–400	420
Se (mg/kg)	100	100	100	–	100
Pb (mg/kg)	300	840	840	750–1200	300
Zn (mg/kg)	2800	4000	7500	2500–4000	2800
Total Fecal Coliform Count (MPN/g)	1000	2000000	–	–	–
Salmonella (MPN/4g)	3	–	–	–	–
Helminth Eggs (Eggs/4g)	< 1	–	–	–	–
Enteroviruses (Unit/4g)	< 1	–	–	–	–
Moisture Content (%)	10	40			

Table S2. Global average concentration in shale background values [30]

Heavy metal	Average Shale (mg/kg)
As	13
Cd	0.3
Cr	90
Cu	45
Hg	0.4
Mo	2.6
Ni	68
Se	0.6
Pb	20
Zn	95

Table S3. Classifications for the geoaccumulation Index (I_{geo}) [29]

Class	Degree	Class quality
Not contaminated	0	$I_{geo} \leq 0$
Uncontaminated to moderately contaminated	1	$0 < I_{geo} \leq 1$
Moderately contaminated	2	$1 < I_{geo} \leq 2$
Moderate to heavily contaminated	3	$2 < I_{geo} \leq 3$
Heavily contaminated	4	$3 < I_{geo} \leq 4$
Very to extremely contaminated	5	$4 < I_{geo} \leq 5$
Extremely contaminated	6	$5 < I_{geo}$

Table S4. Classification for monomial potential ecological risk coefficient (E_r) and potential ecological risk index (RI) [12, 29].

E_r		RI	
Degree	Description	Degree	Description
Slight	$E_r < 30$	Low	RI<150
Medium	$30 \leq E_r < 60$	Moderate	$150 \leq$ RI <300
Strong	$60 \leq E_r < 120$	High	$300 \leq$ RI <600
Very strong	$120 \leq E_r < 240$	Very high	RI ≥ 600
Extremely strong	$E_r \geq 240$		

Table S5. Reference dose (RfD) and cancer slope factor (SF) of heavy metals via three exposure pathways [39]

Heavy metal	Reference dose (mg/kg day)			Cancer slope factor (mg/kg day)		
	RfD_{ing}	RfD_{inh}	RfD_{der}	SF_{ing}	SF_{inh}	SF_{der}
As	3.00×10^{-4}	–	1.23×10^{-4}	1.5	15.1	3.66
Cd	1.00×10^{-3}	1.00×10^{-3}	1.00×10^{-5}	6.30	6.30	–
Cr	3.00×10^{-3}	2.86×10^{-5}	6.00×10^{-5}	5.00×10^{-1}	42.0	20.0
Ni	2.00×10^{-2}	–	5.40×10^{-3}	1.7	8.40×10^{-1}	42.5
Pb	3.50×10^{-3}	3.52×10^{-3}	5.25×10^{-4}	8.50×10^{-3}	4.20×10^{-2}	8.50
Cu	4.00×10^{-2}	4.02×10^{-2}	1.20×10^{-2}	–	–	–
Hg	3.00×10^{-4}	8.57×10^{-5}	2.10×10^{-5}	–	–	–
Mo	5.00×10^{-3}	4.95×10^{-3}	1.90×10^{-3}	–	–	–
Se	5.00×10^{-3}	–	–	–	–	–
Zn	3.00×10^{-1}	3.00×10^{-1}	6.00×10^{-2}	–	–	–

Table S6. The average daily dose (ADD) of heavy metals from WWTP1

Life Range	Index	Cd	Cr	Ni	Pb	Cu	Mo	Se	Zn
Adults	ADD_{ing}	2.35×10^{-6}	1.76×10^{-5}	1.47×10^{-5}	6.46×10^{-6}	1.88×10^{-4}	9.59×10^{-6}	1.51×10^{-5}	1.05×10^{-3}
	ADD_{inh}	3.45×10^{-10}	2.59×10^{-9}	2.16×10^{-9}	9.50×10^{-10}	2.76×10^{-8}	1.41×10^{-9}	2.22×10^{-9}	1.55×10^{-7}
	ADD_{der}	9.37×10^{-9}	7.03×10^{-8}	5.86×10^{-8}	2.58×10^{-8}	7.49×10^{-7}	3.83×10^{-8}	6.01×10^{-8}	4.21×10^{-6}
	ADD	2.36×10^{-6}	1.77×10^{-5}	1.47×10^{-5}	6.48×10^{-6}	1.88×10^{-4}	9.63×10^{-6}	1.51×10^{-5}	1.06×10^{-3}
Children	ADD_{ing}	4.11×10^{-6}	3.08×10^{-5}	2.57×10^{-5}	1.13×10^{-5}	1.64×10^{-3}	8.39×10^{-5}	1.32×10^{-4}	9.23×10^{-3}
	ADD_{inh}	1.15×10^{-10}	8.61×10^{-10}	7.18×10^{-10}	3.16×10^{-10}	4.59×10^{-8}	2.34×10^{-9}	3.68×10^{-9}	2.58×10^{-7}
	ADD_{der}	1.15×10^{-8}	8.63×10^{-8}	7.19×10^{-8}	3.16×10^{-8}	4.60×10^{-6}	2.35×10^{-7}	3.69×10^{-7}	2.58×10^{-5}
	ADD	4.12×10^{-6}	3.09×10^{-5}	2.58×10^{-5}	1.13×10^{-5}	1.65×10^{-3}	8.41×10^{-5}	1.32×10^{-4}	9.26×10^{-3}

where, ADD_{ing}, ADD_{inh} and ADD_{dermal} are the average daily doses (mg kg^{-1} d^{-1}) through ingestion, inhalation, and dermal contacts, respectively.

Table S7. The average daily dose (ADD) of heavy metals from WWTP2

Life Range	Index	Cd	Cr	Ni	Pb	Cu	Mo	Zn
Adults	ADD_{ing}	2.94×10^{-6}	3.29×10^{-5}	2.47×10^{-5}	2.94×10^{-6}	1.38×10^{-4}	8.22×10^{-6}	9.79×10^{-4}
	ADD_{inh}	4.32×10^{-10}	4.83×10^{-9}	3.63×10^{-9}	4.32×10^{-10}	2.03×10^{-8}	1.21×10^{-9}	1.44×10^{-7}
	ADD_{der}	1.17×10^{-8}	1.31×10^{-7}	9.84×10^{-8}	1.17×10^{-8}	5.52×10^{-7}	3.28×10^{-8}	3.91×10^{-6}
	ADD	2.95×10^{-6}	3.30×10^{-5}	2.48×10^{-5}	2.95×10^{-6}	1.39×10^{-4}	8.25×10^{-6}	9.84×10^{-4}
Children	ADD_{ing}	5.14×10^{-6}	5.75×10^{-5}	4.32×10^{-5}	5.14×10^{-6}	1.21×10^{-3}	7.19×10^{-5}	8.57×10^{-3}
	ADD_{inh}	1.44×10^{-10}	1.61×10^{-9}	1.21×10^{-9}	1.44×10^{-10}	3.38×10^{-8}	2.01×10^{-9}	2.39×10^{-7}
	ADD_{der}	1.44×10^{-8}	1.61×10^{-7}	1.21×10^{-7}	1.44×10^{-8}	3.39×10^{-6}	2.01×10^{-7}	2.40×10^{-5}
	ADD	5.15×10^{-6}	5.77×10^{-5}	4.33×10^{-5}	5.15×10^{-6}	1.21×10^{-3}	7.21×10^{-5}	8.59×10^{-3}

Table S8. The average daily dose (ADD) of heavy metals from WWTP3

Life Range	Index	Cd	Cr	Ni	Pb	Cu	Mo	Zn
Adults	ADD_{ing}	5.87×10^{-7}	4.11×10^{-6}	4.11×10^{-6}	4.70×10^{-6}	6.71×10^{-5}	4.11×10^{-6}	1.78×10^{-4}
	ADD_{inh}	8.63×10^{-11}	6.04×10^{-10}	6.04×10^{-10}	6.91×10^{-10}	9.87×10^{-9}	6.04×10^{-10}	2.62×10^{-8}
	ADD_{der}	2.34×10^{-9}	1.64×10^{-8}	1.64×10^{-8}	1.87×10^{-8}	2.68×10^{-7}	1.64×10^{-8}	7.11×10^{-7}
	ADD	5.90×10^{-7}	4.13×10^{-6}	4.13×10^{-6}	4.72×10^{-6}	6.74×10^{-5}	4.13×10^{-6}	1.79×10^{-4}
Children	ADD_{ing}	1.03×10^{-6}	7.19×10^{-6}	7.19×10^{-6}	8.22×10^{-6}	5.87×10^{-4}	3.60×10^{-5}	1.56×10^{-3}
	ADD_{inh}	2.87×10^{-11}	2.01×10^{-10}	2.01×10^{-10}	2.30×10^{-10}	1.64×10^{-8}	1.00×10^{-9}	4.35×10^{-8}
	ADD_{der}	2.88×10^{-9}	2.01×10^{-8}	2.01×10^{-8}	2.30×10^{-8}	1.64×10^{-6}	1.01×10^{-7}	4.36×10^{-6}
	ADD	1.03×10^{-6}	7.21×10^{-6}	7.21×10^{-6}	8.24×10^{-6}	5.89×10^{-4}	3.61×10^{-5}	1.56×10^{-3}

Table S9. The average daily dose (ADD) of heavy metals from WWTP4

Life Range	Index	Cd	Cr	Ni	Pb	Cu	Mo	Se	Zn
Adults	ADD_{ing}	1.17×10^{-6}	2.41×10^{-5}	2.41×10^{-5}	5.87×10^{-6}	1.48×10^{-4}	1.64×10^{-5}	3.42×10^{-5}	9.18×10^{-4}
	ADD_{inh}	1.73×10^{-10}	3.54×10^{-9}	3.54×10^{-9}	8.63×10^{-10}	2.18×10^{-8}	2.42×10^{-9}	5.04×10^{-9}	1.35×10^{-7}
	ADD_{der}	4.68×10^{-9}	9.60×10^{-8}	9.60×10^{-8}	2.34×10^{-8}	5.90×10^{-7}	6.56×10^{-8}	1.37×10^{-7}	3.66×10^{-6}
	ADD	1.18×10^{-6}	2.42×10^{-5}	2.42×10^{-5}	5.90×10^{-6}	1.49×10^{-4}	1.65×10^{-5}	3.44×10^{-5}	9.22×10^{-4}
Children	ADD_{ing}	2.05×10^{-6}	4.21×10^{-5}	4.21×10^{-5}	1.03×10^{-5}	1.29×10^{-3}	1.44×10^{-4}	3.00×10^{-4}	8.03×10^{-3}
	ADD_{inh}	5.74×10^{-11}	1.18×10^{-9}	1.18×10^{-9}	2.87×10^{-10}	3.62×10^{-8}	4.02×10^{-9}	8.37×10^{-9}	2.24×10^{-7}
	ADD_{der}	5.75×10^{-9}	1.18×10^{-7}	1.18×10^{-7}	2.88×10^{-8}	3.62×10^{-6}	4.03×10^{-7}	8.39×10^{-7}	2.25×10^{-5}
	ADD	2.06×10^{-6}	4.22×10^{-5}	4.22×10^{-5}	1.03×10^{-5}	1.30×10^{-3}	1.44×10^{-4}	3.01×10^{-4}	8.05×10^{-3}

Table S10. The average daily dose (ADD) of heavy metals from WWTP5

Life Range	Index	Cd	Cr	Ni	Pb	Cu	Mo	Se	Zn
Adults	ADD_{ing}	1.17×10^{-6}	1.29×10^{-5}	1.17×10^{-5}	4.70×10^{-6}	1.89×10^{-4}	8.22×10^{-6}	1.51×10^{-5}	9.32×10^{-4}
	ADD_{inh}	1.73×10^{-10}	1.90×10^{-9}	1.73×10^{-9}	6.91×10^{-10}	2.78×10^{-8}	1.21×10^{-9}	2.22×10^{-9}	1.37×10^{-7}
	ADD_{der}	4.68×10^{-9}	5.15×10^{-8}	4.68×10^{-8}	1.87×10^{-8}	7.54×10^{-7}	3.28×10^{-8}	6.01×10^{-8}	3.72×10^{-6}
	ADD	1.18×10^{-6}	1.30×10^{-5}	1.18×10^{-5}	4.72×10^{-6}	1.90×10^{-4}	8.25×10^{-6}	1.51×10^{-5}	9.35×10^{-4}
Children	ADD_{ing}	2.05×10^{-6}	2.26×10^{-5}	2.05×10^{-5}	8.22×10^{-6}	1.65×10^{-3}	7.19×10^{-5}	1.32×10^{-4}	8.15×10^{-3}
	ADD_{inh}	5.74×10^{-11}	6.32×10^{-10}	5.74×10^{-10}	2.30×10^{-10}	4.62×10^{-8}	2.01×10^{-9}	3.68×10^{-9}	2.28×10^{-7}
	ADD_{der}	5.75×10^{-9}	6.33×10^{-8}	5.75×10^{-8}	2.30×10^{-8}	4.63×10^{-6}	2.01×10^{-7}	3.69×10^{-7}	2.28×10^{-5}
	ADD	2.06×10^{-6}	2.27×10^{-5}	2.06×10^{-5}	8.24×10^{-6}	1.66×10^{-3}	7.21×10^{-5}	1.32×10^{-4}	8.17×10^{-3}

Table S11. Human health risk assessment from heavy metals in the sludge from WWTP1

Non-carcinogenic	Adult			Children		
	HQ_{ing}	HQ_{inh}	HQ_{der}	HQ_{ing}	HQ_{inh}	HQ_{der}
Cd	2.35×10^{-3}	3.45×10^{-7}	9.37×10^{-4}	4.11×10^{-3}	1.15×10^{-7}	1.15×10^{-3}
Cr	5.87×10^{-3}	9.06×10^{-5}	1.17×10^{-3}	1.03×10^{-2}	3.01×10^{-5}	1.44×10^{-3}
Ni	7.34×10^{-4}	-	1.08×10^{-5}	1.28×10^{-3}	-	1.33×10^{-5}
Pb	1.85×10^{-3}	2.70×10^{-7}	4.91×10^{-5}	3.23×10^{-3}	8.97×10^{-8}	6.03×10^{-5}
Cu	4.69×10^{-3}	6.87×10^{-7}	6.24×10^{-5}	4.11×10^{-2}	1.14×10^{-6}	3.83×10^{-4}
Mo	1.92×10^{-3}	2.85×10^{-7}	2.01×10^{-5}	1.68×10^{-2}	4.74×10^{-7}	1.24×10^{-4}
Se	3.01×10^{-3}	-	-	2.64×10^{-2}	-	-
Zn	3.52×10^{-3}	5.17×10^{-7}	7.01×10^{-5}	3.08×10^{-2}	8.60×10^{-7}	4.31×10^{-4}
HI	HI_{ing}	HI_{inh}	HI_{der}	HI_{ing}	HI_{inh}	HI_{der}
	2.39×10^{-2}	9.27×10^{-5}	2.32×10^{-3}	1.34×10^{-1}	3.28×10^{-5}	3.60×10^{-3}
Carcinogenic	CR_{ing}	CR_{inh}	CR_{der}	CR_{ing}	CR_{inh}	CR_{der}
Cd	1.48×10^{-5}	2.18×10^{-9}	-	2.59×10^{-5}	7.23×10^{-10}	-
Cr	8.81×10^{-6}	1.09×10^{-7}	1.41×10^{-6}	1.54×10^{-5}	3.62×10^{-8}	1.73×10^{-6}
Ni	2.50×10^{-5}	1.81×10^{-9}	2.49×10^{-6}	4.37×10^{-5}	6.03×10^{-10}	3.06×10^{-6}
Pb	5.49×10^{-8}	3.99×10^{-11}	2.19×10^{-7}	9.61×10^{-8}	1.33×10^{-11}	2.69×10^{-7}
TRC	TCR_{ing}	TCR_{inh}	TCR_{der}	TCR_{ing}	TCR_{inh}	TCR_{der}
	4.86×10^{-5}	1.13×10^{-7}	4.11×10^{-6}	8.51×10^{-5}	3.75×10^{-8}	5.05×10^{-6}

where, HQ_{ij} is the Hazard Quotient, and HI is the Hazard Inde× for non-carcinogenic risk. While CR is the carcinogenic risk and TCR is the total cancer risk.

Table S12. Human health risk assessment from heavy metals in the sludge from WWTP2

Non-carcinogenic	Adult			Children		
	HQ_{ing}	HQ_{inh}	HQ_{der}	HQ_{ing}	HQ_{inh}	HQ_{der}
Cd	2.94×10^{-3}	4.32×10^{-7}	1.17×10^{-3}	5.14×10^{-3}	1.44×10^{-7}	1.44×10^{-3}
Cr	1.10×10^{-2}	1.69×10^{-4}	2.19×10^{-3}	1.92×10^{-2}	5.62×10^{-5}	2.68×10^{-3}
Ni	1.23×10^{-3}	-	1.82×10^{-5}	2.16×10^{-3}	-	2.24×10^{-5}
Pb	8.39×10^{-4}	1.23×10^{-7}	2.23×10^{-5}	1.47×10^{-3}	4.08×10^{-8}	2.74×10^{-5}
Cu	3.46×10^{-3}	5.06×10^{-7}	4.60×10^{-5}	3.03×10^{-2}	8.41×10^{-7}	2.82×10^{-4}
Mo	1.64×10^{-3}	2.44×10^{-7}	1.73×10^{-5}	1.44×10^{-2}	4.06×10^{-7}	1.06×10^{-4}
Zn	3.26×10^{-3}	4.80×10^{-7}	6.51×10^{-5}	2.86×10^{-2}	7.98×10^{-7}	4.00×10^{-4}
HI	HI_{ing}	HI_{inh}	HI_{der}	HI_{ing}	HI_{inh}	HI_{der}
	2.43×10^{-2}	1.71×10^{-4}	3.53×10^{-3}	1.01×10^{-1}	5.84×10^{-5}	4.96×10^{-3}
Carcinogenic	CR_{ing}	CR_{inh}	CR_{der}	CR_{ing}	CR_{inh}	CR_{der}
Cd	1.85×10^{-5}	2.72×10^{-9}	-	3.24×10^{-5}	9.04×10^{-10}	-
Cr	1.64×10^{-5}	2.03×10^{-7}	2.62×10^{-6}	2.88×10^{-5}	6.75×10^{-8}	3.22×10^{-6}
Ni	4.19×10^{-5}	3.05×10^{-9}	4.18×10^{-6}	7.34×10^{-5}	1.01×10^{-9}	5.13×10^{-6}
Pb	2.50×10^{-8}	1.81×10^{-11}	9.96×10^{-8}	4.37×10^{-8}	6.03×10^{-12}	1.22×10^{-7}
TRC	TCR_{ing}	TCR_{inh}	TCR_{der}	TCR_{ing}	TCR_{inh}	TCR_{der}
	7.69×10^{-5}	2.09×10^{-7}	6.90×10^{-6}	1.35×10^{-4}	6.94×10^{-8}	8.48×10^{-6}

Table S13. Human health risk assessment from heavy metals in the sludge from WWTP3.

Non-carcinogenic	Adult			Children		
	HQ_{ing}	HQ_{inh}	HQ_{der}	HQ_{ing}	HQ_{inh}	HQ_{der}
Cd	5.87×10^{-4}	8.63×10^{-8}	2.34×10^{-4}	1.03×10^{-3}	2.87×10^{-8}	2.88×10^{-4}
Cr	1.37×10^{-3}	2.11×10^{-5}	2.73×10^{-4}	2.40×10^{-3}	7.03×10^{-6}	3.36×10^{-4}
Ni	2.05×10^{-4}	-	3.04×10^{-6}	3.60×10^{-4}	-	3.73×10^{-6}
Pb	1.34×10^{-3}	1.96×10^{-7}	3.57×10^{-5}	2.35×10^{-3}	6.52×10^{-8}	4.38×10^{-5}
Cu	1.68×10^{-3}	2.46×10^{-7}	2.23×10^{-5}	1.47×10^{-2}	4.08×10^{-7}	1.37×10^{-4}
Mo	8.22×10^{-4}	1.22×10^{-7}	8.63×10^{-6}	7.19×10^{-3}	2.03×10^{-7}	5.30×10^{-5}
Zn	5.94×10^{-4}	8.73×10^{-8}	1.18×10^{-5}	5.19×10^{-3}	1.45×10^{-7}	7.27×10^{-5}
HI	HI_{ing}	HI_{inh}	HI_{der}	HI_{ing}	HI_{inh}	HI_{der}
	6.60×10^{-3}	2.19×10^{-5}	5.89×10^{-4}	3.32×10^{-2}	7.88×10^{-6}	9.34×10^{-4}
Carcinogenic	CR_{ing}	CR_{inh}	CR_{der}	CR_{ing}	CR_{inh}	CR_{der}
Cd	3.70×10^{-6}	5.44×10^{-10}	-	6.47×10^{-6}	1.81×10^{-10}	-
Cr	2.05×10^{-6}	2.54×10^{-8}	3.28×10^{-7}	3.60×10^{-6}	8.44×10^{-9}	4.03×10^{-7}
Ni	6.99×10^{-6}	5.08×10^{-10}	6.97×10^{-7}	1.22×10^{-5}	1.69×10^{-10}	8.56×10^{-7}
Pb	3.99×10^{-8}	2.90×10^{-11}	1.59×10^{-7}	6.99×10^{-8}	9.65×10^{-12}	1.96×10^{-7}
TRC	TCR_{ing}	TCR_{inh}	TCR_{der}	TCR_{ing}	TCR_{inh}	TCR_{der}
	1.28×10^{-5}	2.65×10^{-8}	1.18×10^{-6}	2.24×10^{-5}	8.80×10^{-9}	1.45×10^{-6}

Table S14. Human health risk assessment from heavy metals in the sludge from WWTP4.

Non-carcinogenic	Adult			Children		
	HQ_{ing}	HQ_{inh}	HQ_{der}	HQ_{ing}	HQ_{inh}	HQ_{der}
Cd	1.17×10^{-3}	1.73×10^{-7}	4.68×10^{-4}	2.05×10^{-3}	5.74×10^{-8}	5.75×10^{-4}
Cr	8.02×10^{-3}	1.24×10^{-4}	1.60×10^{-3}	1.40×10^{-2}	4.12×10^{-5}	1.97×10^{-3}
Ni	1.20×10^{-3}	-	1.78×10^{-5}	2.11×10^{-3}	-	2.18×10^{-5}
Pb	1.68×10^{-3}	2.45×10^{-7}	4.46×10^{-5}	2.94×10^{-3}	8.16×10^{-8}	5.48×10^{-5}
Cu	3.70×10^{-3}	5.41×10^{-7}	4.92×10^{-5}	3.24×10^{-2}	9.00×10^{-7}	3.02×10^{-4}
Mo	3.29×10^{-3}	4.88×10^{-7}	3.45×10^{-5}	2.88×10^{-2}	8.12×10^{-7}	2.12×10^{-4}
Se	6.85×10^{-3}	-	-	5.99×10^{-2}	-	-
Zn	3.06×10^{-3}	4.50×10^{-7}	6.10×10^{-5}	2.68×10^{-2}	7.48×10^{-7}	3.75×10^{-4}
HI	HI_{ing}	HI_{inh}	HI_{der}	HI_{ing}	HI_{inh}	HI_{der}
	2.90×10^{-2}	1.26×10^{-4}	2.28×10^{-3}	1.69×10^{-1}	4.38×10^{-5}	3.51×10^{-3}
Carcinogenic	CR_{ing}	CR_{inh}	CR_{der}	CR_{ing}	CR_{inh}	CR_{der}
Cd	7.40×10^{-6}	1.09×10^{-9}	-	1.29×10^{-5}	3.62×10^{-10}	-
Cr	1.20×10^{-5}	1.49×10^{-7}	1.92×10^{-6}	2.11×10^{-5}	4.94×10^{-8}	2.36×10^{-6}
Ni	4.09×10^{-5}	2.97×10^{-9}	4.08×10^{-6}	7.16×10^{-5}	9.89×10^{-10}	5.01×10^{-6}
Pb	4.99×10^{-8}	3.63×10^{-11}	1.99×10^{-7}	8.73×10^{-8}	1.21×10^{-11}	2.45×10^{-7}
TRC	TCR_{ing}	TCR_{inh}	TCR_{der}	TCR_{ing}	TCR_{inh}	TCR_{der}
	6.04×10^{-5}	1.53×10^{-7}	6.20×10^{-6}	1.06×10^{-4}	5.08×10^{-8}	7.62×10^{-6}

Table S15. Human health risk assessment from heavy metals in the sludge from WWTP5

Non-carcinogenic	Adult			Children		
	HQ_{ing}	**HQ_{inh}**	**HQ_{der}**	**HQ_{ing}**	**HQ_{inh}**	**HQ_{der}**
Cd	1.17×10^{-3}	1.73×10^{-7}	4.68×10^{-4}	2.05×10^{-3}	5.74×10^{-8}	5.75×10^{-4}
Cr	4.31×10^{-3}	6.64×10^{-5}	8.59×10^{-4}	7.53×10^{-3}	2.21×10^{-5}	1.05×10^{-3}
Ni	5.87×10^{-4}	-	8.68×10^{-6}	1.03×10^{-3}	-	1.07×10^{-5}
Pb	1.34×10^{-3}	1.96×10^{-7}	3.57×10^{-5}	2.35×10^{-3}	6.52×10^{-8}	4.38×10^{-5}
Cu	4.73×10^{-3}	6.92×10^{-7}	6.29×10^{-5}	4.14×10^{-2}	1.15×10^{-6}	3.86×10^{-4}
Mo	1.64×10^{-3}	2.44×10^{-7}	1.73×10^{-5}	1.44×10^{-2}	4.06×10^{-7}	1.06×10^{-4}
Se	3.01×10^{-3}	-	-	2.64×10^{-2}	-	-
Zn	3.11×10^{-3}	4.57×10^{-7}	6.19×10^{-5}	2.72×10^{-2}	7.59×10^{-7}	3.80×10^{-4}
HI	**HI_{ing}**	**HI_{inh}**	**HI_{der}**	**HI_{ing}**	**HI_{inh}**	**HI_{der}**
	1.99×10^{-2}	6.82×10^{-5}	1.51×10^{-3}	1.22×10^{-1}	2.45×10^{-5}	2.56×10^{-3}
Carcinogenic	**CR_{ing}**	**CR_{inh}**	**CR_{der}**	**CR_{ing}**	**CR_{inh}**	**CR_{der}**
Cd	7.40×10^{-6}	1.09×10^{-9}	-	1.29×10^{-5}	3.62×10^{-10}	-
Cr	6.46×10^{-6}	7.98×10^{-8}	1.03×10^{-6}	1.13×10^{-5}	2.65×10^{-8}	1.27×10^{-6}
Ni	2.00×10^{-5}	1.45×10^{-9}	1.99×10^{-6}	3.49×10^{-5}	4.82×10^{-10}	2.45×10^{-6}
Pb	3.99×10^{-8}	2.90×10^{-11}	1.59×10^{-7}	6.99×10^{-8}	9.65×10^{-12}	1.96×10^{-7}
TRC	**TCR_{ing}**	**TCR_{inh}**	**TCR_{der}**	**TCR_{ing}**	**TCR_{inh}**	**TCR_{der}**
	3.39×10^{-5}	8.23×10^{-8}	3.18×10^{-6}	5.92×10^{-5}	2.74×10^{-8}	3.91×10^{-6}

Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

Laila A. Al-Khatib; Designed the study, analyzed the data, and wrote the paper., Ahmad M. AlHanaktah; conducted the experimental work, Feras Y. Fraige; aided in analyzing the results and worked on the manuscript. All authors had approved the final version.

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