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Investigation of heavy metalloid pollutants in the south of Tehran using kriging method and HYDRUS model

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Abstract

Due to the high cost of the large-scale measurement of heavy metals, the use of statistical land models and techniques is one of the proper ways to study their distribution and level of pollution. The study area, is agricultural lands of south Tehran in Iran. Municipal wastewater is often used for irrigation agricultural lands under surface irrigation method. To study the distribution of heavy metals including copper, nickel, and lead, the ordinary kriging method in the GIS environment was used. In addition, one-dimensional HYDRUS modeling of water flow and heavy metals in the soil environment was simulated up to a depth of 50 cm for 210 days and the concentration of heavy metals in the depth was simulated. Distribution of lead element in soil surface with spherical model showed that its variation was in the range of 20–70 mg/kg. These values were 50–60 mg/kg for copper and 30 mg/kg for nickel. Investigation of heavy metal concentrations in soil profiles using the HYDRUS-1D model showed that the major accumulation of heavy metals occurred in the surface layer of soil at a depth of 0–15 cm that was higher than the permissible level.

Keywords: Distribution, Heavy metals, HYDRUS, Kriging method, Wastewater

Introduction

Due to the scarcity of safe water resources in arid and semi-arid regions of the world, the reusing of municipal and industrial wastewater for irrigation is one of the alternatives to agricultural activities. Despite providing part of the water needed for irrigation, it is one of the causes of soil and crops contamination (Abdioo et al. 2010,). Studies from wastewater-irrigated fields in Mexico, India and Pakistan clearly associate helminth-contaminated soils and wastewater to increased human infections (Ensink 2006). The early systems of land application were plagued with raw wastewater and pollutant overloading and inexperienced operation, resulting in grave environmental pollution. Bad odor and concern about transmission of diseases, improper application on land and discharge of raw wastewater to irrigation farms are also of great concern, leading to the reinvestigation of irrigation with wastewater. Investigation of heavy pollutants in agricultural soils in study area is one of main points.

Qadir et al. (2010) showed that irrigation with raw wastewater is expanding in some developing countries due to the mismatch between urban development and the infrastructure needed for wastewater treatment. The impact of irrigation with wastewater on the accumulation of heavy metals in soil and crops in the region of Marrakech in Morocco was investigated. The results revealed high-risk indices. Heavy metal-contaminated food crops are a high risk to the health of local people and animal populations (Sana Chaoua et al. 2019). The use of wastewater is usually associated with microbial contamination and heavy metals that can affect different parts of nature and therefore human health. Chen and Wang (2005) also stated that due to the transfer of heavy metals in nature and endangering human health, the problem of heavy

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metals was a limiting factor in the use of this resource (wastewater). Heavy metal pollution and its implications for human health and the environment have increased research in developing low cost and sustainable remediation technology (Shaikh A. Razzak a, b et al. 2022).

Heavy metal intoxication can damage proteins and enzymes due to their binding heavy metals to active sites and causing deformation.

The use of wastewater for irrigation has several benefits, including the use of nutrients in it and reducing its entry into nature. However, the use of wastewater is usually associated with microbial contamination and heavy metals, which can affect different parts of nature and therefore human health. *Escherichia coli* in wastewater-irrigated soil was about 2×10^6 (CFU $g - ^1$) and about 15 (CFU $g - ^1$) in vegetables' edible parts (leaf, bulb, tuber and fruit) while the mean total coliforms was about 1.4×10^6 and 55 (CFU $g - ^1$) in soil and vegetables' edible parts, respectively. For human health risk assessment, the estimated daily intake (EDI) and human health risk index (HRI) ranged from 0.01 to 8 (EDI and HRI > 1.0 associated with adverse health effects) (Yahia A. Othman,*, Amani Al-Assaf, Maher J. Tadros and Abeer Albalawneh. 2021). One of the important tools in environmental studies is GIS software. This tool has been widely used in soil studies, engineering, and environmental issues. The kriging method of this software is one of the important capabilities that can interpolate the desired variables based on their weight on the adjacent points.

One of the studies in this area is research that zoned the distribution of heavy metals including zinc and copper by conventional kriging and exponential modeling (Khodakarami et al. 2011). Rahimpour et al. (2014) modeled the spatial variations of heavy metals of copper, zinc, iron, and manganese in Harris County using conventional kriging methods and basic radial functions. Sistani et al. (2017) investigated heavy metal contamination around the Kerman Steel Industry. Their results showed that lead and cadmium concentrations increased under the influence of the steel industry. Borges et al. (2014) investigated the distribution and zoning of heavy metals using GIS in Brazil. They also investigated the status of heavy metal contamination in the water and soil resources of the study area. For spatial distribution of heavy metals in the middle Nile Delta in Egypt, contamination factor, pollution load index, and degree of contamination indices were used to assess the environmental risks of heavy metal contamination of the soils (Shokr et al. 2016). Altan et al. (2011) also distributed heavy metals including cadmium, chromium, copper, nickel, zinc, and lead, which were investigated using GIS interpolation techniques. In addition to the surface distribution of heavy metals, their accumulation in soil due to the use of wastewater for irrigation or fertilizers has also been reported in different studies (Khai N.M. et al. 2007). Results of their study showed that heavy metal-contaminated soils of selected villages in Zamfara State, Nigeria were in the order of Fe > Pb > Cr > Zn > Cd > Ni, with Pb and Cd having a concentration higher than permissible levels for soils and accounted for 98.64% of the total potential ecological risk (Sharhabil Musa et al. 2021).

To investigate the movement of heavy metals, different numerical models can be used to simulate their transport in unsaturated soil. One of these models is the one-dimensional HYDRUS (HYDRUS-1D) that has been used in various studies. This model is used to investigate the infiltration of water and pollutants into the soil as well as their one-dimensional transport within the soil with different boundary conditions. This model has also been used by various researchers to investigate heavy metal transport. Sayaad et al. (2008) investigated the transfer of heavy metals in soil using HYDRUS-1D under safflower and wheat cultivation. They concluded that the HYDRUS model was able to give a good estimate of the metal transfer process in the soil. Dao et al. (2014) simulated heavy metal transfer to the soil using HYDRUS-1D and concluded that this model was able to predict the heavy metal transfer in the soil to a permissible level. Also, in another study by Behbahaninia et al. (2014) to investigate heavy metal transport in the unsaturated soil environment, the capability of the HYDRUS-1D model to study the transfer and estimation of heavy metals of iron and zinc concentrations within soil was emphasized. Mohtar et al. (2018) showed regional and local factors contribute to the different types of air pollutant concentrations in urban environments.

In general, the purpose of this study is to investigate the distribution of heavy metals in lands irrigated with wastewater in the south of Tehran using kriging interpolation in the GIS environment and to find areas with potential contamination of lead, copper, and nickel. Also, the purpose of this study was to investigate soil and agricultural products contamination and hence the extent to which people's health at risk.

The possibility of deep transfer of these pollutants and the deep penetration of these metals to the bottom layers of soil are evaluated using HYDRUS-1D software. Due to the method of surface irrigation (Basin) and soil saturation environment, similar to the infiltration of water in the plot, which is the dominant vertical component. To determine the concentration of heavy metals in the depth of heavy metal transfer in the vertical component is simulated by the Hydrus model. By interpolation of point concentrations of heavy metals on the soil surface, zoning has been done by kriging model.

It is expected that by examining the horizontal and depth distribution of these metals in the soils of the study area, comprehensive information on their distribution and concentration in the soil can be found.

Material and methods

The present study was conducted on lands south of Tehran (Fig. 1) that irrigated with municipal wastewater. The study area is located in approximate coordinates of 35° 30′ 35″ north latitude and 51° 26′ 29″ east longitude with an average elevation of 1050 m above sea level. The main soil in the study area is clay–loam with 1.1–5.3% organic matter. The area receives large amounts of municipal wastewater as well as surface runoff of Tehran's streets during the rainy seasons, which has always been a cause of concern for heavy metal pollution in the area. The major crops in these areas include vegetables and garden crops, which a surface irrigation method is often used for them. Therefore, as a result of using this irrigation source (raw wastewater), there is always a risk of soil and crops contamination and thus endangering human health.

Sampling and chemical analysis

After field visits of cultivated areas in the south of Tehran and sampling sites that were irrigated with raw wastewater, random sampling was performed. Thirty samples of surface soil at 0–15 cm depth were prepared from plots with approximate dimensions of 200 × 200 m. Date and time of sampling, was summer of 2016. In addition to soil samples, 30 samples were collected from the wastewater entering the study area in different times of during the 2016 crop season. After recording each sample's information including sample number, place of the sample with GPS, date and time of sampling, and area cultivation status, they were transferred to the laboratory for chemical analysis. In addition to soil samples, 30 samples were collected from the wastewater entering the study area in different sections and intervals. After the preparation of soil and wastewater samples, the concentrations of Pb, Ni and, Cu were determined by atomic absorption spectroscopy (AAS). Also, the amount of organic matter, acidity (PH), and electrical conductivity (EC) of the samples were measured in the laboratory.

Geostatistical analysis

To study the distribution of heavy metals including copper, nickel, and lead, the conventional kriging method in the GIS environment was used. This method uses a quantitative correlation between the measured points and then configures the space around the projected points based on the measured values. The computational function in the kriging method for estimating the desired values is given by Eq. 1:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [z(x_i) - z(x_i + h)]^2. \quad (1)$$

In this equation, the $Z(x)$ is the value of the i parameter at the point X_i and h is the distance between the pair of points and n is the number of pairs of measured points that are separated by h intervals. Estimated values using the above semivariogram are then fitted to a theoretical model such as circular, spherical, exponential, or Gaussian models. These models determine the spatial distribution as well as the parameters desired in the kriging

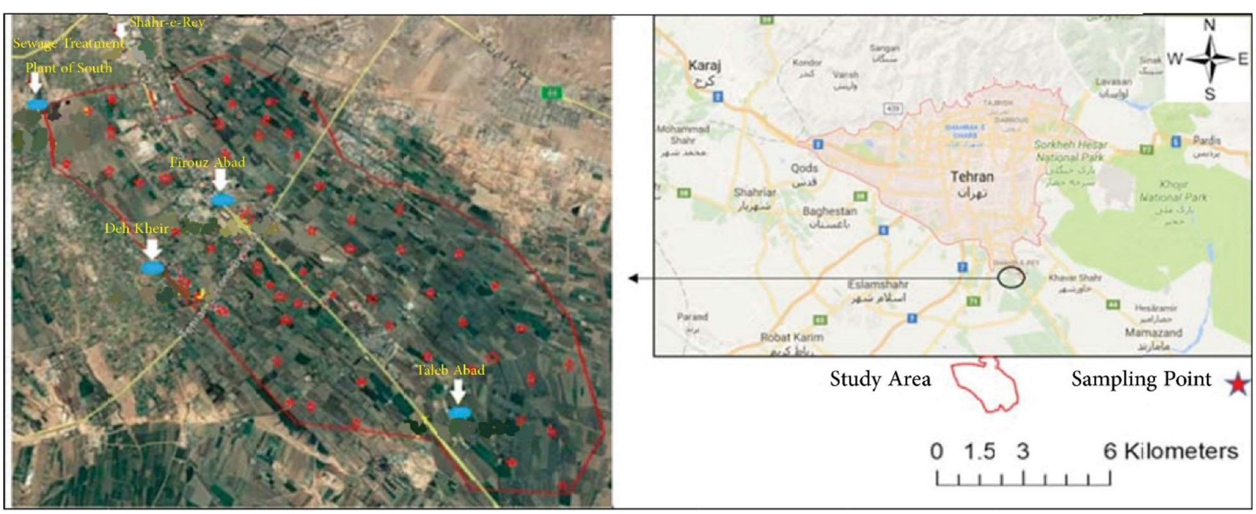

Fig. 1 The study area and sampling site

method. The kriging method uses the weighted average of the points to estimate the unknown value.

This method is given by Eq. 2:

$$z(x_0) = \sum_{i=1}^{n} \lambda_i z(x_i). \tag{2}$$

In this equation, the $Z(X_0)$ is the unknown value of the parameter desired at point X_0, and $z(Xi)$ is measured at point Xi and λ is weight.

The initial condition for using the values measured for interpolation by kriging is their normal distribution. For this purpose, using the logarithmic function, the distribution and data were normalized and then interpolated with different variograms. Also, before selecting the circular, spherical, exponential, and Gaussian models, their usability was evaluated and finally, the best model was selected for interpolation. For this purpose, statistical root mean square error indices (RMSE), Pearson correlation coefficient (R), mean absolute error (MAE), and MBE were calculated (Eqs. 3, 4, 5, 6) by using IBM SPSS statistics 23 software or in the Excel environment. The closer the value of R to 1 number in these relationships, the greater the correlation between observed and estimated data. And the closer the index to zero, the better the results of the model:

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n} (z^*(x_i) - z(x_i))^2}{n}}, \tag{3}$$

$$R = \frac{Cov(Z^*(x_i), Z(x_i))}{\delta(Z^*(x_i)).\delta(Z(x_i))}, \tag{4}$$

$$MAE = \frac{1}{n}\sum_{i=1}^{n} \left|z^*(x_i) - z(x_i)\right|, \tag{5}$$

$$MBE = \frac{1}{n}\sum_{i=1}^{n} \left[z^*(x_i) - z(x_i)\right]. \tag{6}$$

In these equations, the $Z^*(Xi)$ value is equal to the estimated value of the Z parameter by the model at point Xi and $Z(Xi)$ is the measured value of Z at point Xi and Cov, is data covariance and n is the number of samples.

In addition to the mentioned indices, the Nugget, Range, and Sill indices were also determined in the studied variograms.

The Range value is the distance after which the variogram value is fixed. It indicates physically that the pair of samples after this value is not spatially correlated. The Sill value is equal to the maximum variability between the sample pairs. In addition, the modeling of deep water flow and heavy metal transport in the soil environment was performed using a one-dimensional HYDRUS model.

In the one-dimensional HYDRUS, the flow of water is described using the Richards equation. Pollutant changes in the soil are calculated based on the transfer-diffusion equation (CDE) as follows:

$$\frac{\partial \theta C}{\partial t} + \frac{\partial \rho S}{\partial t} = \frac{\partial}{\partial X}\left(\theta D \frac{\partial c}{\partial X}\right) - \frac{\partial qc}{\partial X}. \tag{7}$$

In this equation, c is the contaminant concentration in the soil solution, S is the amount of contaminant absorbed, θ is the soil volumetric moisture, D is the diffusion coefficient, q is the transient flow value, t is the time, and X is the contaminant distance from the initial point.

The correlation between the heavy metals in the soil solution and the amount of adsorbed to the soil particles (S parameter in the above equation) is explained by Freundlich's adsorption model (Dao C.A. et al., 2014) which is given in Eq. 8:

$$Q_s = K_F C_e^{\beta}. \tag{8}$$

In this equation, Qs is equal to the amount of heavy metal absorbed, Ce is the concentration of heavy metals in soil solution, KF and β are also constant coefficients of Freundlich's equation. These coefficients can be estimated based on laboratory or based on earlier studies and then calibrated the model.

The HYDRUS model also uses the Van Genuchten–Mualem equation as follows to determine the hydraulic parameters of the soil:

$$\theta(h) = \begin{cases} \theta_r + \dfrac{\theta_s - \theta_r}{\left[1 + |\alpha h|^n\right]^m} & h < 0 \\ \theta_s & h \geq 0 \end{cases}. \tag{9}$$

In this equation, θr is the amount of residual soil moisture, θs is saturated soil moisture, m, n, and α are coefficients of the model, h is soil moisture potential, KS soil saturated hydraulic conductivity and Se is soil moisture effective content. Validation and Modification of the Van Genuchten Model for Eroded Black Soil in Northeastern China was used (Shuang Li et al. 2020).The results showed that the parameters in the VG model had significant difference under the two eroded soils of the saturated water content (θs), but the opposite was true for the residual water content (θr), the scale parameter (α) and the shape parameter (n). In addition, the θs and θr had no significant differences but the opposite was true for the α and n under the two input sets.

Simulation of water and heavy (Table 1) metal transfer in soil up to a depth of 50 cm soil for 210 days

Table 1 Soil physical parameters for model calibration

Θ_s	Θ_r	Alpha	*n*	K_S	Dispersivity
0.32	0.085	0.032	1.45	26	15.05

Table 2 Calibration results of the model based on various statistical indicators

Parameter tested	RMSE	Max ERR (%)	CRM	E	R^2
Soil moisture (%)	5.48	15.97	−0.02	0.62	0.52
Pb (ppm)	14.96	14.36	−0.03	0.53	0.59
Cu (ppm)	12.16	10.36	−0.05	0.63	0.78
Ni (ppm)	12.06	15.35	−0.06	0.55	0.62

(mid-November to mid-June) was carried out as the wheat-growing period (the dominant crop in the study area). The parameters *m*, *n*, *α*, and *Ks* were estimated inversely in the HYDRUS model. Inverse method for saturated water content (θs) was error, but the opposite was true for the residual water content (θr), the scale parameter (α) and the shape parameter (n).

Validation of the simulation of Hydrus model

Validation of the simulation of Hydrus model are presented in Tables 2 and 3.

The Feddes function was selected as the main function of water uptake by the plant, and its coefficients were selected from the default numbers in the model for the wheat crop. Boundary conditions and upstream initial values were considered for atmospheric water flow as well as irrigation water values.

Under these conditions, the height of soil water and rainfall amounts were considered equal to the depth of water required for irrigation, which was reduced by infiltration or evapotranspiration. Due to the low groundwater level and deep soil in the study area, downstream boundary conditions were considered as free flows. Soil moisture information for the depths of 0–15, 15–30, and 30–50 cm soil layers as input to the model were considered. In addition, the model boundary conditions for metal transport were also considered based on the initial concentration of heavy metal elements. In terms of climate and rainfall, most of the year's rainfall is from mid-autumn to mid-spring with an average of 400 mm. The warmest month of the year is August with an average of 36 degrees and the coldest month with an average minimum temperature of -1 degrees is related to February. The main soil of the study area is loamy clay with 1.1 to 3.5% of organic matter. The study area is plain with an average elevation of 1050 m above sea level and without mountains in its vicinity and has no river flow around. The area receives large amounts of municipal wastewater as well as surface runoff of Tehran's streets during the rainy seasons, which has always been a cause of concern for heavy metal pollution in the area.

Results

Laboratory analysis

The laboratory analysis obtained from the measurement of Cu, Pb, and Ni concentrations of soil samples of agricultural areas and raw wastewater imported into the study area is presented in Table 4.

A comparison of the concentration values of these metals in all samples in Table4 shows that Pb concentration was higher than copper and nickel.

Statistical results and selection of the best model

Statistical comparison of circular, spherical, exponential, and Gaussian models to determine the best variogram is presented in Table 5. The results showed that the exponential model with minimum RMSE, MAE, MBE, and maximum R has the best fit in drawing copper elements compared to other models were utilized as a variogram which used in heavy metal copper element zoning. This index was also used in the kriging method Pearson correlation coefficient indices (R) for evaluating models was used. This index (R) for nickel element, in spherical model is, 0.997 and for copper element, in exponential model , 0.704 and for lead, in spherical model, 0.85.

Table 3 Validation of results of the model based on various statistical indicators

Element	Parameter tested	RMSE	Max ERR (%)	CRM	*E*	R2
Pb	Soil moisture (%)	8.12	10.05	−0.10	0.66	0.62
	Concentration (ppm)	15.33	15.34	−0.09	0.68	0.66
CU	Soil moisture (%)	16.12	14.11	−0.08	0.79	0.55
	Concentration (ppm)	12.1	20.22	−0.11	0.65	0.7
Ni	Soil moisture (%)	18.11	23.14	−0.01	0.75	0.5
	Concentration (ppm)	20.21	19.34	−0.12	0.56	0.46

Table 4 Chemical analysis of heavy metal in soil samples

Parameter								
Sample	**Element**	**Number of samples**	**Minimum**	**Maximum**	**Average**	**Standard deviation**	**Standard error is about 95% confidence**	
Soil	Lead	30	17.1	79.9	38.79	13.71	2.5	**38.79 ± 5.31**
	Copper	30	17.6	65.5	29.65	8.91	1.63	**29.65 ± 3.56**
	Nickel	30	27.1	42.5	33.05	3.42	0.62	**33.05 ± 1.37**
	Organic matter	30	1.6	4.64	2.32	0.6	0.11	**2.32 ± 0.23**
Effluent	Lead	30	0.06	2.25	1.43	0.96	0.18	**1.43 ± 1.53**
	Copper	30	0.05	0.5	0.21	0.2	0.04	**0.21 ± 0.32**
	Nickel	30	0.06	0.1	0.08	0.02	0.01	**0.08 ± 0.03**
	Acidity	30	7.21	7.33	7.66	0.31	0.06	**7.66 ± 0.4**

Table 5 Comparing circular, spherical, Gaussian model in different variograms

Element	The variogram model	RMSE	MAE	MBE	*R*	Nugget	Partial sill	Range
Copper	Circular	23.733	22.522	−22.523	0.727	0.062	0.015	553.603
	Exponential*	5.714	3.486	−0.002	0.704	0.072	0.005	553.603
	Gaussian	6.649	4.131	−0.042	0.507	0.027	0.038	527.84
	Spherical	6.418	3.971	−0.031	0.561	0.075	0	553.621
Nickel	Circular	1.364	1.018	0.091	0.98	0.008	0.018	524.552
	Exponential	2.35	1.253	0.025	0.912	0.003	0.025	551.284
	Gaussian	1.83	1.361	−0.202	0.961	0	0.026	552.36
	Spherical*	0.565	0.424	−0.105	0.997	0.01	0.016	551.213
Lead	Circular	25.649	16.131	−0.442	0.565	0.044	0.052	533.84
	Exponential	31.536	29.239	2.314	0.598	0.011	0.056	527.84
	Gaussian	28.649	12.131	−0.142	0.523	0.017	0.033	529.84
	Spherical*	10.692	7.502	−0.358	0.85	0.091	0.011	385.49

The models were selected with regard to minimum RMSE, MAE, MBE, and maximum R from Table 5.

Selected models had maximum R.Coefficient of determination index was used in the calibration and validation stage of Hydrus model. In the calibration stage, this coefficient was obtained from 0.51 to 0.8 and in the validation stage, its value was obtained between 0.46 and 0.7. Kh. Khosravi*1, M.H. Nejad Roshan, A. Safari (2017) accomplished through comparing different methods including simple kriging and ordinary kriging, RBF, and IDW with powers 1 and 5 for interpolating groundwater depth and water table in SariNeka Plain for the years 2001, 2006 and 201 and reported mean absolute error (MAE) from -0.07 --0.15 and mean bias error (MBE) from 2.28-2.50.

The comparison of these indices for the nickel element showed that the spherical model was better as the desired variogram. The spherical model had the best fit for the copper element. Also, the comparison of other parameters showed that the selected variograms had minimum partial sill value, which means that the maximum variability between sample pairs was smaller.

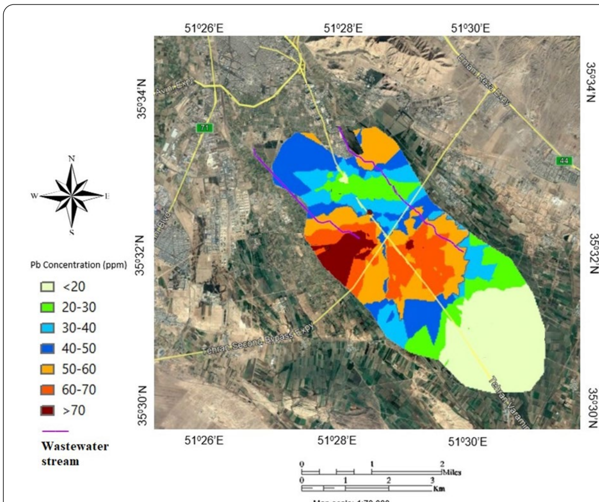

Fig. 2 Distribution heavy metal lead in south Tehran

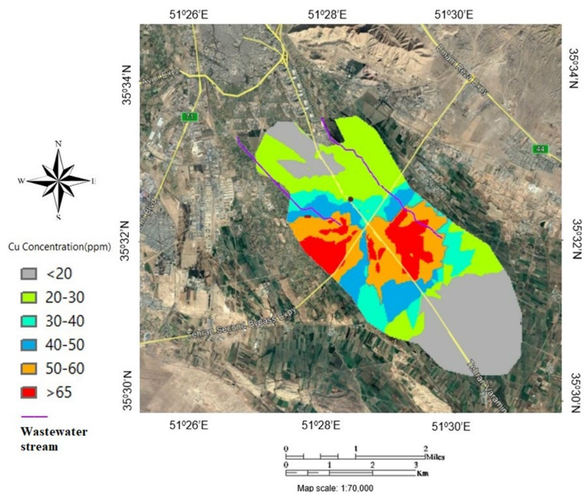

Fig. 3 Distribution heavy metal copper in south Tehran

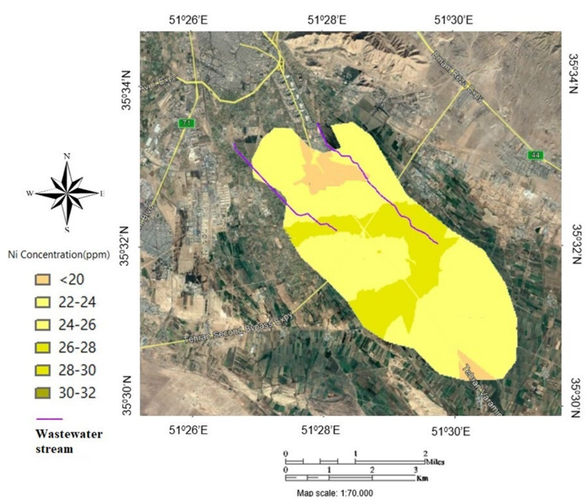

Fig. 4 Distribution heavy metal nickel in south Tehran

After selecting the most suitable variogram, Cu, Ni, and Pb zoning maps were prepared in the study area (Figs. 2, 3, 4).

Evaluation of distribution of lead element showed that the highest concentration 70–50 mg/kg was found in central areas of the study that irrigated by raw wastewater. Concentrations of this element in soils often used from well water for irrigation are in the range of 30–40 or 40–50 mg/kg. Also, by increasing the distance from this area to the lands of Talibabad village (Fig. 2), the concentration of this element decreases and reaches about 20–30 mg/kg.

Similarly, the distribution of copper element showed that higher concentrations of this element were observed in the central areas of the study (the major consumer of wastewater for irrigation than elsewhere shown in Fig. 3) the concentration of this element is about 50–60 mg/kg. However, in the vicinity of the city of Rey, the concentration of this element reaches 20–30 or less than 20 mg/kg. In the surrounding areas of Talibabad village in Fig. 3 (as in the case of lead), a minimal distribution of copper was observed at concentrations less than 20 mg/kg.

The distribution of the nickel elements in Fig. 4 showed that the concentration of this element is in the range of 30 mg/kg. A study of the distribution of this element in the area shows a uniform distribution of it and, only in some central parts of the study area, the concentration of it slightly exceeds 30 mg/kg.

Investigation of heavy metal concentrations in soil profiles using the HYDRUS-1D model showed that the major accumulation of lead occurred in the surface layer of soil at a depth of 0–15 cm (Fig. 1). The simulated concentration of lead showed that the variations trend of this element versus depth of the soil is decreased so that at a depth of 15–30 cm is about 25 mg/kg and at a depth of 30 cm is below 15 mg/kg. Temporal changes in the transport of this element over 210 days from the soil surface to a depth of 50 cm (0–50) showed that the concentration of this element in the surface layer decreased from 45 mg/kg at the beginning of the period to 35 mg/kg at the end of the period. This decrease may be related to increased plant growth and so increased plant uptake.

The study of the concentration changes in the downstream layer also shows a similar trend during the growth period, as the concentration of Pb at the end of the period is reduced to less than 15 mg/kg at the beginning of the simulation period to less than 5 mg/kg at the end of the period.

The simulation of the deep transition of Cu and Ni also showed similar results, with the major accumulation of these two metals in the surface layer and with increasing depth, the concentration of these elements decreased rapidly (Fig. 5). The temporal variations of copper and nickel transfer in the upper boundary of the soil also show that although initially an increasing trend was observed, the amount of this element decreased over time and reached about 30 mg/kg at the end of the growth period. And at the lower boundary (the depth of 50 cm) the amount of element transport reached the small amount of about 2 mg/kg.

Discussion

The comparison of the mean concentration of heavy metals in soil with each other showed that the concentration of lead in these samples was higher than nickel and copper. On the other hand, comparing these values with the concentration of these elements in the wastewater

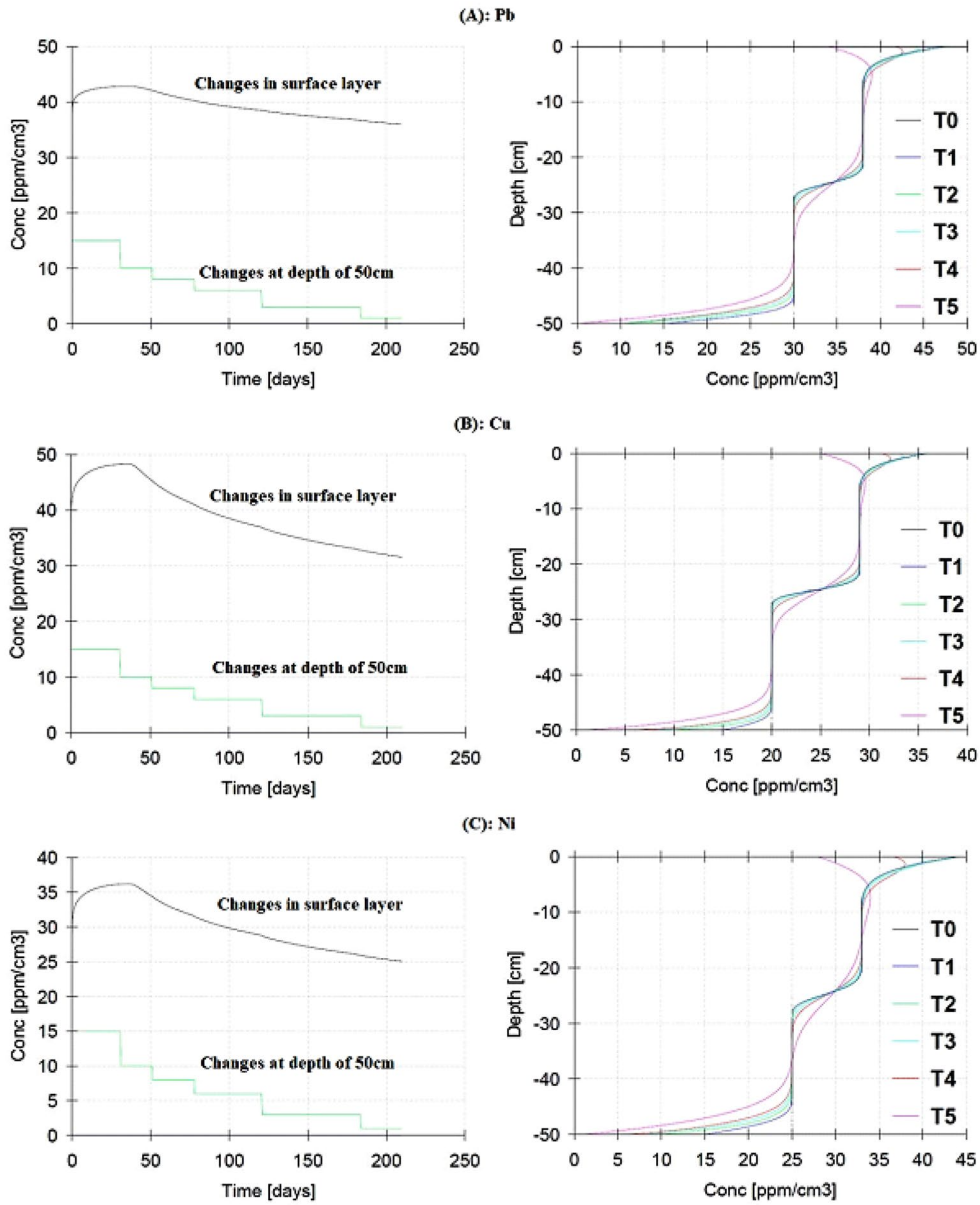

Fig. 5 Changes in the concentrations of copper, lead and nickel with depth (right) and time (left)

sample showed that the higher concentration of Pb in the soil samples may be related to its high concentration in the wastewater used for irrigation. This may indicate the importance of raw wastewater treatment before use for agricultural purposes.

The findings of Harati et al. (2010) also show a high concentration of lead in the study area, which is consistent with the results of this study.

The heavy metal pollution in the soil is very important. Because of the high costs of measurement methods and their low accuracy, the use of models is inevitable. In this regard, the use of conventional kriging based on different circular, spherical, exponential, and Gaussian models is one of the most common methods for investigating heavy element distribution. In this study, the spherical model for nickel and lead and exponential model for copper element allowed to study the dispersion and determination of contamination status of these elements. The results of this study are consistent with the study of Khaledan et al. (2017) as well as Rahimpour et al. (2014), who reported that the spherical model for the lead element and the exponential model for the copper element was the best fit. Toxic variogram analysis of these models showed a higher concentration of heavy metals in the central areas of the study. One of the reasons for the higher concentration of these elements in this area could be related to the repeated use of wastewater (the most important source of irrigation in this area). It should be explained that due to the low concentration of nickel in the wastewater of the inlet to the area and the uniform distribution of the concentration of nickel in the entire study area, there is no strong relationship between the use of nickel and its dispersion in this area. The results for the variations of this element are comparable to the results of the study by Fard Samiei et al. (2016), who reported a uniform concentration of the element in the study area. Another similar study is a study by Barzin et al. (2015), which investigated heavy metals in Hamadan province. Their research results also showed that lead element was affected by agricultural activities is at a high level of pollution.

It is important to determine the maximum permissible concentration of heavy metals in the soils of agricultural areas due to their potential absorption by the plant and its adverse effects on plant health and growth, as well as the possibility of their transmission through food cycles to plants and animals. Although an element such as copper is one of the necessary metals in the soil for plant growth, it is also found naturally in soil and is usually complex with organic matter, and rarely free or exchangeable. Due to the deficiency of this element as one of the micronutrients important for plant growth, there is a possible need for adding it to the soil. However, due to the low boundary between the amount required and the amount of poisoning in the soil, increasing its concentration in the soil may cause environmental pollution. Therefore, excessive entry of this heavy element by abnormal factors such as the use of agricultural fertilizers, pesticides, or entering wastewater into the soil, can be a potential contributor to pollution. Permissible amount of nickel in soil reported according to WHO standard maximum 50 mg/kg (Rathod et al. 2013). The study of heavy metals' levels in water, soil, and vegetables located in Tanzania, showed that the average daily intake for Pb (63 mg/person/day) was above the permissible maximum tolerable daily intake of 0.21 mg/person/day endorsed by WHO/FAO (David Sylvester Kacholi and Minati Sahu. 2018). And the heavy metals' levels in soil, water, and vegetables were in the order of Fe > Zn > Pb > Cu.

The maximum permissible values reported for lead, copper, and nickel in different countries (Table 6) show that their permissible values for different countries are significantly different.

Therefore, it is necessary to use a globally acceptable index for this purpose, including the World Health Organization (WHO) index. The maximum (Table 7) permissible levels in agricultural soils for lead, copper, and nickel are reported to be 60, 100, and 50 mg/kg, respectively, according to the WHO standard (Toth G. et al., 2016). Also, the permissible standard of agricultural soils in Iran has been introduced by the Environment Agency (Barzin M. et al., 2015) for these three elements are 75, 200, and 110 mg/kg, respectively. By comparing the concentrations of nickel and copper with the maximum permissible values based on WHO standards in agricultural soils, it can be said that there is no contamination of these two elements in the area.

The comparison of lead concentration with standard introduced by WHO as well as permissible value in agricultural soils of Iran shows that its concentration in the central areas of the study is due to repeated irrigation of this area by wastewater above WHO value and somewhat higher than the permissible level of WHO in agricultural soils of Iran.

It is necessary to explain that lead is one of the most important metals widely used in a variety of vehicles, electrical equipment, and buildings. Also, urban runoff transports pollutants from city vehicles and small industrial and domestic wastewaters to the irrigated areas and increases heavy metals. Therefore, considering the higher concentration of this element compared to nickel and copper in the wastewater entering this area, it seems important to control the concentration of this element. The results of HYDRUS model analysis of heavy metal concentrations in the soil profile showed that the accumulation of heavy metals in the soil surface layer was higher than the deeper layers. The comparison of the performance of this model with research by Behbahaninia et al. (2014) also shows that by providing enough information needed for model inputs, can accurately simulate element transfer. Another similar, comparable study is by Sayaad et al. (2008), which concluded that the HYDRUS model was able to simulate Cu and Pb transfer in the root environment.

Table 6 Heavy metal standard in agricultural soils in different countries

Countries	Heavy metal standard in agricultural soils		
Australia	300	600	100
Canada	200	100	150
China	80	60–40	200–50
Germany	1000	200	200
Tanzania	200	100	200
Netherlands	530	100	190
New Zealand	160	-	10,000

Table 7 Comparison of the verification results of this research with previous researches

Researchers and subject	Method	Results	Comparison
(Khodakarami et al., 2011)	Conventional kriging and exponential modeling	Zoned the distribution of heavy metals including zinc and copper	Trend results are consistent
Harati et al. (2010)	Simulation of Hydrus model	The findings show a high concentration of lead in the study area	Consistent with the results of this study
Khaledan et al. (2017) as well as Rahimpour et al. (2014)	Conventional kriging and exponential modeling	were reported that the spherical model for the lead element and the exponential model for the copper element was the best fit	Good agreement between them
Barzin M. et al., 2015	Ordinary kriging and exponential modeling	The permissible standard of agricultural soils in Iran has been introduced by the Environment Agency for these three elements are 75, 200, and 110 mg/kg, respectively	Consistent with the results of this study
WHO standards in agricultural soils	Presentation was standard	The concentrations of nickel and copper in study area, shows there is no contamination of these two elements in the area	Agricultural soils of study area for lead, copper, and nickel are less than the maximum permissible levels
Toth G. et al., (2016)	According to the WHO standard	The maximum permissible levels in agricultural soils for lead, copper, and nickel are reported to be 60, 100, and 50 mg/kg	Agricultural soils of study area for lead, copper, and nickel are less than the maximum permissible levels
Fard Samiei et al. (2016)	Remote sensing techniques	Was reported almost uniform concentration of the element nickel in the entire study area	Good agreement between them

The main reason for this is that the behavior of heavy metals depends on intermediate factors affecting the uptake of heavy metals in the soil such as organic matter, iron oxides, or clays which are higher in the surface layer (Rattan R.K. et al., 2005). For example, examining the relationship between the concentrations of heavy metals measured in the samples and the amount of organic matter in them showed that as the amount of soil organic matter increased, the concentration of heavy metals in the samples increased too (Fig. 2). Therefore, it can be said that one of the important factors is that the accumulation of heavy metals in the surface layers and its non-transfer to the lower layers are related to this parameter. The comparison of the findings of this study with the study by Dao et al. (2014) also indicates the important role of soil organic matter in controlling the transfer of heavy metals to the sublayers. In the present study, the total amount of organic matter was determined. However, the soil organic matter components of the study area have been reported in previous studies, including small (fresh) plant residues and small living soil organisms (15%), decomposing (active) (18%)) organic matter, and stable organic matter (humus) (68%).

Due to its high percentage of organic matter and clay, the 0–15 cm layer of soil tends to absorb heavy metals and delay their leaching to the lower layers. Decomposition of soil organic matter can release heavy metals and increase its concentration in soil solution. During the formation or accumulation of organic matter in the soil, the heavy metals can be absorbed by the soil, delaying their leaching. This illustrates the importance of soil organic matter in preventing heavy metal transport to the lower layers and ultimately to groundwater. In addition, the uptake of heavy metals into soil colloids, including clay minerals, is one of the factors that reduces the rate of ion transfer (8). In contrast, soil organisms' activity, plant root growth, and soil surface characteristics such as soil cracks in the dry seasons lead to preferential flow during irrigation. It can have a significant impact on the transmission of contamination to the lower layers of soil. The results also showed that the HYDRUS model was able to predict the values of the elements to a permissible level. The simulated values in the surface layer showed the highest and the lowest values in the lower layer, which were comparable to the measured values. The comparison of the performance of this model with research by Behbahaninia et al. (2014) also shows that by providing enough information needed for model inputs, one can accurately simulate element transfer. Another similar, comparable study is by Sayaad et al. (2008), which concluded that the HYDRUS model was able to simulate Cu and Pb transfer in the root environment.

Conclusion

Due to the high cost of the large-scale measurement of heavy metals, the use of statistical land models and techniques is one of the proper ways to study their distribution and level of pollution. Irrigation with raw and treated effluent for a long time in Tehran has caused polluted the study area. Showed that long-term use of effluents for irrigation in the Chinese city of Beijing has led to the accumulation of cadmium, chromium, copper and lead in the soil. Surface runoff in Tehran has also caused pollution in the study area. These runoffs including pollutant of the workshops and streets of the city.

In this study, conventional kriging in the GIS environment was used to analyze the values of heavy elements of lead, copper, and nickel in soils under wastewater irrigation in the south of Tehran. The findings showed that in general, the highest concentration of elements was in the central areas of the study, where the source of irrigation was mainly raw wastewater. Lead was found to be above the permissible level in the central areas of the study, which are often irrigated with wastewater, and more control studies are needed. In addition to the surface distribution of these elements, their deep transfer into the soil using one-dimensional HYDRUS software showed that the highest accumulation of elements occurred in the surface layer 0–15 cm. This is due to the presence of more organic matter, clay, iron, and manganese hydroxides as important factors in surface absorption in this layer, which indicates their importance in preventing the transfer of these elements to the deeper layers. In this study, due to the limitations of executive facilities, only the contamination of the three elements of lead, nickel, and copper was investigated, while the wastewater may contain more heavy metals. Therefore, considering this research shortcoming, conducting more studies to more comprehensively study heavy metals and zoning areas with excessive contamination limits can be very benefici.

In general, the objective of this study was to investigate the distribution of heavy metals in lands irrigated with wastewater in the south of Tehran using kriging interpolation in the GIS environment and to find areas with potential contamination of lead, copper, and nickel. Also, the purpose of this study was to investigate the risk of soil and agricultural products contamination and so the extent to which people's health is at risk. The work's hypotheses were that the horizontal distribution and depth distribution of heavy metals in large scale of the soils of the study area could be determined by using kriging interpolation in the GIS environment and HYDRUS-1D software. As can be seen, the conclusions are suitable response to the work's hypotheses and the objective of the research has been reached by preparing a large-scale map distribution of lead, nickel and copper heavy metals in the study area. Some of practical suggestions are as follows:

It is suggested to simulate heavy metal distribution for long periods after irrigation by wastewater in the study area. It is suggested to simulate chemical and microbial contamination in addition to heavy metals in the study area. It is suggested to simulate heavy metals by Hydrus-2D model under furrow irrigation method in the study area.

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Declarations

Competing interests
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References

Abadioo RC, Keraita B, Drechsel P, Dissanayake P, Maxwell AS (2010) Soil and crop contamination through wastewater irrigation and options for risk reduction in developing countries. Soil biology and agriculture in the tropics. Springer, Heidelberg, pp 275–297

Altan M, Ayyildiz Ö, Malkoç S, Yazici B, Koparal S (2011) Heavy metal distribution map in soil by using GIS techniques. J Environ Sci Eng 5(1):15–20

Barzin M, Kheirabadi H, Afyuni M (2015) An investigation into pollution of selected heavy metals of surface soils in Hamadan Province using pollution index. J Water Soil Sci 19(72):69–80

Behbahaninia A, Farahani M (2014) Investigation of leaching process heavy metals (Fe, Zn) in the soil un- der sewage sludge application by using hydrus-1D. J Biodivers Environ Sci (JBES) 5(4):35–41

Borges RC, Caldas VG, Filho F, Ferreira MM, Lapa CMF (2014) Use of GIS for the evaluation of heavy metal contamination in the Cunha Canal watershed and west of the Guanabara Bay, Rio de Janeiro. RJ Ma- Rine Pollution Bulletin 89:75–84

Chaoua S, Boussaa S, ElGharmali A, Boume A (2019) Impact of irrigation with wastewater on accumulation of heavy metals in soil and crops in the region of Marrakech in Morocco. J Saudi Soc Agric Sci 18(4):429–436

Dao CA, Phuong KM, Vy AP (2014) Application of Hydrus -1D model to simulate the transport of some selected heavy metals in paddy soil in Thanh Tri, Hanoi. VNU J Sci: Earth and Environ-Mental Sci 3(1):22–30

David Sylvester K, Minati S (2018) Levels and health risk assessment of heavy metals in soil, water, and vegetables of dares salaam, Tanzania. J Chem. https://doi.org/10.1155/2018/1402674
EL Shokr MS, Baroudy AA, Fullen MA, El-Beshbeshy TR, Ali RR, Elhalim A, Guerra AJT, Jorge MCO (2016) Mapping of heavy metal concentration in alluvial soils of the Middle Nile Delta of Egypt. J Environ Eng Landscape Management. 24(3):218–231
Harati M, TamadonRastegar M, Hariri N, Varavipour M. 2010. Effects of wastewater application for irrigation on heavy metals accumulation (case study south of Tehran, Iran). Papers of the 1st Iranian Fertilizer Challenge Congress.2010.Tehran.Iran .Half a Century of the Fertilizer Consumption. Iran (in Persian).
Jeroen H J Ensink. Wastewater quality and the risk of hookworm infection in Pakistani and Indian sewage farmers. Thesis for: PhD .Advisor: Brooker S, and Blumenthal U. 2006. DOI: https://doi.org/10.13140/2.1.1288.6723.
Khai NM, Ha PQ, Öborn I (2007) Nutrient flows in small scale per urban vegetable farming systems in South- east Asia—A case study in Hanoi. Agric, EcoSyst Environ 122(2):192–202
Khaledan S, Taghavi L, Paykanpour FP (2017) Investigation of spatial distribution of lead and cadmium using geostatistical techniques and GIS (case study: field's surrounding Mobarakeh steel complex). Iran J Health Environ 10(2):151–164
Khodakarami L, Soffianian A, TowfighE M, Mirghafari N (2011) Study of heavy metals concentration copper, zinc and arsenic soil using GIS and RS techniques (Case study: Kaboudarahang, Razan and Khonjin- Talkhab catchment in Hamedan province). J Appl RS GIS Tec Nat Res Sci 2(1):1–14
Khosravi Kh, Nejad Roshan MH, Safari A (2017) Assessment of geostatistical methods for determining distribution patterns of groundwater resources in Sari-Neka coastal plain. North Iran Environ Resour Res 5(2):2017
Li S, Xie Y, Xin Y, Liu G, Wang W, Gao X, Zhai J, Li J (2020) Validation and modification of the van Genuchten model for Eroded Black Soil in Northeastern China. Water 12(10):2678. https://doi.org/10.3390/w12102678
Mohtar AAA, Latif MT, Baharudin NH, Ahmad F, Chung JX (2018) Variation of major air pollutions in different seasonal conditions in an urban environment in Malaysia. GeoSci Lett. https://doi.org/10.1186/s40562-018-0122-y
Othman YA, Al-Assaf A, Tadros MJ, Albalawneh A (2021) Heavy metals and microbes accumulation in soil and food crops irrigated with wastewater and the potential human health risk: a metadata analysis. Water. https://doi.org/10.3390/w13233405
Qadir M, Wichelns D, Raschid-Sally L, McCornick PG, Drechsel P, Bahri, a., Minhas, P.S., (2010) The challenges of wastewater irrigation in developing countries. Agric Water Manag 97:561–568. https://doi.org/10.1016/j.agwat.2008.11.004
Rahimpour F, Abbaspour RA (2014) Zoning soil heavy metals pollution using Kriging and radial basis function methods (case study: Harris County). Sci-Entific Res Quart Geogr Data (SEPEHR) 23(91):56–67
Rathod PH, Rossiter DG, Noomen MF, van der Meer FD (2013) Proximal spectral sensing to monitor phytoremediation of metal-contaminated soils. Int J Phytoremediation 15:405–426. https://doi.org/10.1080/15226514.2012.702805
Rattan RK, Datta SP, Chhonkar PK, Suribabu K, Singh AK (2005) Long-term impact of irrigation with sewage effluents on heavy metal content in soils, crops and groundwater a case study. Agr Ecosyst Environ 109(3–4):310–322
Razzakab SA, Faruquea MO, Alsheikhd Z, Alsheikhmohamad L, Alkuroud D, Alfayez A, Hossaine MM (2022) A comprehensive review on conventional and biological-driven heavy metals removal from industrial wastewater. Environ Adv. https://doi.org/10.1016/j.envadv.2022.100168
Samiei Fard R, Keshavarzi A, Etesami H, Rostami nia M, Rahmani A. 2016. Monitoring the accumulation of Cd, As, Ni and Pb in soil using Landsat 8 images. 5th National Conference of Sustainable Agriculture and Natural Resources. Iran.
Sayaad GA, Mousavi SF, Abbaspour K, Afyouni M (2008) Simulating the transport of Cd, Cu, Pb and Zn in a non-disturbed calcareous soil under wheat and canola cultivation using Hydrus-1D model. Agric Sci Iran 39(1):187–200
Sistani N, Moeinaddini M, Khorasani N, Hamidian AH, Ali-Taleshi MS (2017s) Heavy metal pollution in soils nearby Kerman steel industry: metal richness and degree of contamination assessment. Iran J Health Environ 10(1):75–86
Toth G, Hermann T, da Silva MR, Montanarella L (2016) Heavy metals in agricultural soils of the European Union with implications for food safety. Environ Int 88:299–309
Yahaya SM, Abubakar F, Abdu N (2021) Ecological risk assessment of heavy metal-contaminated soils of selected villages in Zamfara State, Nigeria. Springer, Heidelberg. SN Applied Science ASPRINGER NATURE journal. SN Applied Sciences (2021) 3:168. https://doi.org/10.1007/s42452-021-04175-6.

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