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ORIGINAL ARTICLE

Soil contamination by heavy metals through irrigation with treated wastewater in a semi-arid area

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Abstract

In arid and semi-arid regions, the adoption of unconventional water sources, including treated domestic wastewater (TWW), as an alternative water resource for crop irrigation has gained widespread attention. Addressing the potential effects of utilizing TWW for irrigation on heavy metal (HM) soil contamination is crucial. Given the escalating apprehensions regarding contamination risks, this study aims to quantitatively analyze the impact of TWW irrigation on the accumulation of HM within soil. The HDRUS-1D model was employed to predict the concentration of HM in soil over a 30-year period. This modeling approach enabled the assessment of HM propagation within the profiles of two TWW irrigated soil types: Calcisol and Fluvisol in Sfax, Tunisia. The simulation results indicate that accumulation of HM increases with time, with the metals gradually penetrating deeper into the soil. In the case of Cr, Cu, and Ni, significant enrichment is observed primarily in the surface layer, while Zn and Fe exhibit enrichment across the entire soil profile. The Fluvisol soil type displays a higher accumulation of HM compared to the Calcisol, particularly in the deep sandy layers. Despite HM concentrations in TWW falling below Tunisian irrigation standards, continuous monitoring of metal accumulation in soil is imperative. The choice of utilizing TWW for irrigation must not only consider water quality but also account for the soil type and its propensity to accumulate heavy metals. Consequently, when considering TWW for irrigation, a comprehensive assessment encompassing water quality, soil characteristics, and potential HM accumulation should be undertaken.

Keywords Treated waste water · Irrigation · Heavy metal · Soil contamination · Modeling

Introduction

In the arid and semi-arid countries, the use of unconventional water resources such as treated waste water (TWW) for irrigation became a common practice to overcome drought and water scarcity. The main benefits of this practice are the contribution to reduce fresh water demand and the adding of significant quantities of nutrients (especially nitrogen, phosphorus and potassium) and organic matter to the soil. However, TWW contain heavy metals (HM) that may accumulate, as a result of long-term irrigation, in the ecosystem and particularly in the soil.

The effects of irrigation with TWW are felt on the soil, the unsaturated zone and in some cases the groundwater. Several authors (Rattan et al. 2005; Meng et al. 2016; Chaoua et al. 2019; Zeyad and Abdul Malik 2022; Dakouré et al. 2013) characterized the HM accumulation in soil and/or components of the ecosystem based on field and laboratory studies. Weissmannová and Pavlovsky (2017) gave a synthesis on indices of soil contamination by HM defined using relative concentration and various standard limits or geochemical background for the considered element.

HM contained in TWW are numerous; the most abundant (about some mg/l) are Fe, Zn, Cu and Pb. Other heavy metals (Mn, Al, Cr, As, Sn, Hg, Cd, Mo, Ni,) are present at the state of traces. Once consumed, HM can have major health effects and cause a variety of symptoms depending on their nature and amount (Angelakis et al.

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1999; Qadir et al. 2010; Khan et al. 2013; Sankhla and Kumar 2019; Alghobar and Suresha 2017).

Hence, the superficial unsaturated zone confronts with the contaminant accumulation (primarily HM) coming from TWW used in irrigation. In fact, during the last decades research on the unsaturated zone becomes increasingly as it has a major effect in the water movement, such as infiltration, moisture variation, groundwater refill and contamination, erosion and others. The contamination issued from these practices could have serious influences on the environment, the flora and fauna (Farhadkhani et al. 2018). Thus, the vadose zone is affected by the pollution from agriculture practice, industry or municipal activities, such as waste storage. The effect of the irrigation with TWW leads to the contamination by the pathogenic germs, pollution by HM and the possibility of groundwater salinization. The effects of TWW appears after some years of intensive irrigation (Bedbabis et al. 2015; Farhadkhani et al. 2018; Kass et al. 2005; Bouri et al. 2008; El Ayni et al 2011). However, a 15-year-long irrigation by TWW has limited the loss of micro nutrients, such as Mn and Zn. It has also supplied important amounts in macro nutrients (P and N) and enhanced the C and N turnover in the studied soil (Belaid et al. 2012).

The HM movement and/or accumulation in the soil system involve simultaneous processes, including water flow, chemical diffusion, hydrodynamic dispersion, and geochemical processes and reactions. Models, considering these various mechanisms, can be utilized to characterize inorganic and organic contaminants transport in the soil media taking into account the prevailing hydrologic and geochemical conditions (Mayer et al. 2002; Appelo and Postma 2005; Jacques et al 2008; Sámano et al. 2014; Yang et al. 2022). A great variety of numerical models, based on the coupled equation of flow and transport in porous media are used to simulate the transport of pollutants in general or the HM in soil in particular. After model calibration and validation through experimental and field data measurements, they are considered of first help to generate provisional scenario on the behavior of pollutants in soils.

Hydrus-1D model introduced by Šimůnek et al. (1998) is a software package for simulating water, heat and solute movement in one-dimensional variably saturated media.

In the present study, the essential goal is to recognize the effect of irrigation with TWW during periods of 4 and 15 years on HM movement in the vadose zone using the modeling of transport of aqueous solution in unsaturated medium. This work applies Hydrus-1D model to simulate the transport of Cr, Cu, Ni, Zn and Fe in two soils irrigated with TWW in Sfax, Tunisia as a case study.

Site description and methods

The study region is located on the Southern coast of Tunisia at 10 km to the south of the Sfax city (Fig. 1). It has an arid to semi-arid Mediterranean climate with an interannual pluviometry average of 220 mm/year. The Average temperature varies from 11 °C in January to 27 °C in August, with an annual mean of 19 °C. The morphology of the area is characterized by a fairly flat topography with an altitude varying from 12 to 30 m with a steady slope to the East. It consists of a vast alluvial plain.

The geology of the area has been described by Burollet (1956), Bouaziz (1995) and DGRE (2005). The outcropping geologic formations, composed of alternations of sandy clay and sand, belong to the Lower Pleistocene, Middle and Upper Pleistocene, and Actual (Fig. 1). The Lower Pleistocene is composed by red silts with calcareous concretions, overlaid by a limestone crust. The Middle and Upper Pleistocene are characterized by the development of a gypsum crust, ranging from 20 to 40 cm thick. This crust rests on a profile characterized by the expansion of red sandy-silty deposits, which cover vast expanses of cultivable fields. The Actual is represented by various deposits, including: grey silts forming the lower terraces of wadis and floodplain alluvial fans, eolian deposits filling small depressions and valley bottoms and alluvial deposits in watercourses composed of sands, gravels, and pebbles, occupying the beds of the main wadis.

The soil in the TWW irrigated area is mainly formed of two types: iso-humic soil or Calcisol and underdeveloped soil or Fluvisol (Belaid 2010).

The Calcisol with a depth of 150 cm is composed of 30 cm red ochre, loamy sandy texture, 20 cm of light beige, silty texture, 10 cm of calcareous crust and 90 cm of calcareous silty texture with nodules and calcareous pebbles.

The Fluvisol with a depth of 160 cm is formed of 50 cm: brown to grey, plastic clay texture, 40 cm of yellowish, sandy rich in fine sand, 20 cm of yellowish ochre, plastic clay to silty clay texture, 25 cm: of ochre, sandy rich in fine sand, 25 cm of beige, sandy-clay texture.

The water used for crop irrigation originates from the Sfax city waste water treatment plant with a capacity of 49,500 m^3/day. The Sewage passes through a mechanical screen, an aerated grit chamber and primary sedimentation containers. In the second treatment stage waste water passes through alternating anoxic/aerobic bioreactors for nitrogen and phosphorus removal and organic matter biodegradation.

The modeling of transport of an aqueous solution in unsaturated porous medium result from coupling its hydrodynamic movement and its physico-chemical interactions processes.

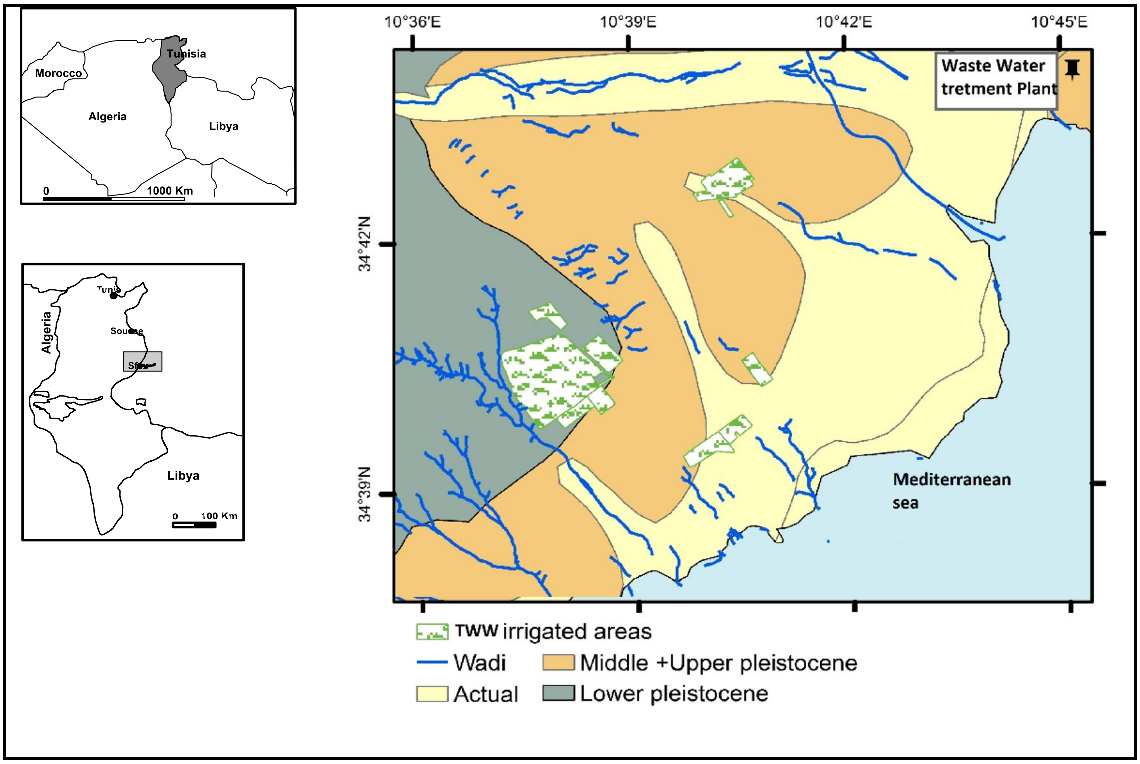

Fig. 1 Study area location and geologic outcrops map

The water flow and HM transport through the soil system can be modeled using the finite-element model HYDRUS-1D. This last is developed for simulating water, heat and non-conservative solute movement in one-dimensional variably saturated media.

For this investigation, HYDRUS 1D is selected because of its availability and the types of data it considers. The relevance of the use of the model Hydrus 1D for the estimate of the HM transfers in unsaturated mediums is recognized by several authors (Bragan et al. 1997; Gribb and Sewell 1998; Pang et al. 2000; Abbaspour et al. 2001; Abbasi et al. 2004; Jacques et al. 2008; Chu et al. 2014; Faisal Anwar and Thien 2015; Mirzaei et al. 2022) showed that the dynamics of the water and the transport of aqueous solutions in a soil is well-reproduced by the code HYDRUS.

The HYDRUS program numerically solves the Richards equation for variably saturated water flow and advection–dispersion equations for heat and solute transport. The following governing equation (Eq. 1) for flow and transport is solved numerically using the finite-element method:

$$\frac{\partial \theta C}{\partial t} + \frac{\partial \rho S}{\partial t} = \frac{\partial}{\partial x}\left(\theta D \frac{\partial C}{\partial x}\right) - \frac{\partial qC}{\partial x}, \tag{1}$$

where C is the contaminant concentration in the soil solution, S is the amount of contaminant fixed on soil particles, ρ is the soil bulk density 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 distance from the source point. A linear relationship between S and C is commonly considered as reconstituted by Freundlich adsorption model and may expressed by the following equation:

$$S = K_d C, \tag{2}$$

where K_d is the partition coefficient given by the ratio of sorbed/dissolved HM concentrations More details about the partition coefficient are given by Alisson and Allison (2005).

The hydraulic parameters of the unsaturated soil are defined using the Van Genuchten–Mualem of the following equation:

$$\theta(h) = \theta_r + \frac{\theta_s - \theta_r}{\left[1 + (\alpha h)^n\right]^{1-1/n}}. \tag{3}$$

In Eq. (3), $\theta(h)$ is the measured water content, θ_r is residual soil moisture, θ_s is saturated soil moisture, n and α are coefficients of the model, h is soil moisture potential.

The computer program ROSETTA (Schaap et al. 2001) was used to estimate the previous parameters as well as saturated and unsaturated hydraulic conductivity. The simulation of water and heavy metal transfer in soil up to a depth of 90 cm is undertaken for time periods of 4 and 15 years

Table 1 Mean HM content in TWW irrigation compared to Tunisian standard

HM (mg/l)	TWW	NT.106.03 (1989)
Cr	0.007–1.1	0.1
Cu	0.01–0.06	0.5
Zn	0.01–0.27	5
Ni	0.02–0.13	0.2
Fe	0.013–1.69	5

corresponding to the irrigation activity duration, respectively, in Calcisol and Fluvisol.

Results and discussion

Irrigation water quality

The main HM content of the TWW compared to Tunisian standard for irrigation with TWW is given in Table 1. The concentration of common HM in the TWW is found to be below the standard maximum permissible values expected for Cr exceeding these limits. The most abundant HM is the Fe followed by Cr. Even the TWW are in the standard fixed limits for HM in irrigation water, the use of such water may lead to soil contamination by adsorption/desorption and accumulation process particularly on clay minerals.

Heavy metals in irrigated soil

The average HM concentrations in irrigated and control soil samples for Calcisol and Fluvisol are given in Table 2 (Belaid 2010). The assessment of the concentration of these metals in all samples in Table 2 shows that concentrations for both irrigated or control soil decrease with depth and they are undetected for a depth of 90 cm in control soil.

By comparing the concentration of HM in soil with each other, it was observed that the highest concentration is of Cr > Zn > Fe. The Cu and Ni concentrations are the lowest with a relatively uniform distribution in the soil samples. Furthermore, when comparing these values with the concentration of the same elements in the TWW, it was revealed that the elevated level in the soil samples might be attributed to its high concentration in the TWW used for irrigation. This suggests that the treatment of raw wastewater before agricultural use is crucial. This finding is in agreement with Sharma et al (2007) and Mirzaei et al. (2022) conclusions, suggesting a soil HM concentration related basically to TWW composition which is consistent with the present study.

Table 2 HM concentration contained in irrigated and control soils for Calcisol and Fluvisol

HM	Soil depth (cm)	HM (mg/cm^3) in irrigated Calcisol and (control)	HM (mg/cm^3) in irrigated Fluvisol and (control)
Cr	0–30	0.045 (0.03)	0.1 (0.077)
	30–60	0.038 (0.029)	0.1 (0.068)
	60–90	0.029 (–)	0.034 (0.019)
Cu	0–30	0.012 (0.0084)	0.022 (0.017)
	30–60	0.0097 (0.0077)	0.021 (0.014)
	60–90	0.0062 (–)	0.006(0.0037)
Ni	0–30	0.0123 (0.0058)	0.0336 (0.024)
	30–60	0.0066 (0.0042)	0.0308 (0.02)
	60–90	0.0046 (–)	0.0043 (0.003)
Zn	0–30	0.046 (0.026)	0.1 (0.077)
	30–60	0.044 (0.021)	0.1 (0.063)
	60–90	0.038 (–)	0.02 (0.015)
Fe	0–30	0.0203 (0.013)	0.058 (0.04)
	30–60	0.021 (0.012)	0.056 (0.035)
	60–90	0.015 (–)	0.017 (0.01)

The comparison of HM relatively concentration of irrigated soil to that of control (in brackets) soil show an enrichment throughout the soil profile. The measured concentration is used for model calibration and verification; however, the concentration in control samples is assigned to initial soil concentration.

Considering the relatively great organic matter in upper soil layers, it can be concluded that the accumulation of HM in this level can be associated with this particular factor. A similar result is found by Mirzaei et al. (2022) and Rattan et al. (2005), where they highlight the significant influence of soil organic matter in regulating the downward transfer of HM.

The Concentrations of all HM along the soil profile are the lowest for the Calcisol due to its more clayey texture and carbonate rich composition relatively to Fluvisol. These two facts are recognized by Zhao et al. (2009), considering the role of soil pH (related to carbonate level in soil) and clay minerals in HM transfer in soil.

Hydrodynamic and transport parameters

The considered Van Genuchten parameters (Van Genuchten 1980) for the various types of soil texture are determined using the module ROSETTA integrated in Hydrus 1D (Table 3).

The most important level of residual water content is recorded at the ground level for clayey texture; however, the lowest levels are for sandy texture. The α and n parameters depend on the form of water retention curve characteristic, which depends on granulometry, are more important for

Table 3 Hydrodynamic parameters as recognized by Rosetta modulus

Texture	θ_r (%)	θ_s (%)	α (cm^{-1})	n (cm)	Ks 10^{-4} (cm/s)
Sand	2.5	43	0.145	2.68	82.5
Silty sand	5.7	41	0.124	1.56	12.2
Silt	3.4	46	0.016	1.37	2.8
Clay	6.8	38	0.008	1.09	0.05

coarse texture (sand) and lowest for fine texture (clay). The hydraulic conductivity varied between 82.5 10^{-4} cm/s in the case of sandy texture and 0.05 10^{-4} cm/s for clay texture.

For the transport parameters, the soil bulk density is assumed between 1.45 and 1.68 for various soil texture as recognized from laboratory tests (Belaid 2010).

The initial dispersivity values are set based on literature, they varied from 5.5 to 17.3 cm for a sandy clayey soil (Gonçalves et al. 2002, 2006; Schoups et al. 2006) and may reach 40 cm for a sandy soil (Jury and Flühler 1992). During the model calibration, the dispersivity was adjusted to find the best fit between measured and calculated HM concentrations.

For the adsorption coefficient (Kd), the values for various heavy metals and soil textures are taken starting from the values suggested in literature and adjusted based on comparison between field and calculated concentrations.

The validation process relies on comparing simulated concentrations of HM with concentrations measured in the field. This validation approach is widely acknowledged, as the model's predictions of system behavior usually fall within acceptable limits (Anderson and Woessner 1991). Numerous cases of HYDRUS validation are showcased in studies by Šimůnek and de Vos (1999), Luo and Sophocleous (2010), Wang et al. (2010), and Zhao et al. (2009), as referenced by Šimůnek et al. (2012).

HM transport simulation in Calcisol

The HM transport simulations were carried for a time period of 15 years corresponding to the time span of irrigation using TWW. The simulated HM concentrations are performed for 90 cm of soil depth divided in three 30 cm thick layers.

For the water dynamics, the boundary conditions are set to a variable flow for the higher limit and a free drainage for the lower one.

For HM transport the boundary conditions are fixed as a flow of concentration for the higher limit and a null concentration (zero concentration gradient) at the lower limit. This assumption is retained following the HM concentrations in control samples at 90 cm depth, where they found very weak and even null.

The HM content, simulated using HYDRUS-1D, are given in Table 4. An increase of HM concentration compared to the control soil is noticed. The most remarkable rise is detected for Zn concentration (0.05 mg/cm^3 at 30 cm depth).

Table 4 Comparison of measured and simulated HM concentrations (mg/cm^3) in Calcisol

HM	Depth (cm)	Simulated concentration	Measured concentration	Coefficient of determination R^2
Cr	0–30	0.0245	0.045	0.66
	30–60	0.028	0.038	
	60–90	0.0083	0.029	
Cu	0–30	0.0095	0.012	0.71
	30–60	0.011	0.0097	
	60–90	0.0026	0.0062	
Ni	0–30	0.008	0.0123	0.94
	30–60	0.0066	0.0066	
	60–90	0.0053	0.0046	
Zn	0–30	0.014	0.046	0.97
	30–60	0.016	0.044	
	60–90	0.021	0.038	
Fer	0–30	0.011	0.0203	0.99
	30–60	0.013	0.0213	
	60–90	0.0031	0.015	

The various HM have the same behavior throughout the soil profile with greater concentrations for Cr and the lowest for Ni.

The most notable augmentation for the various HM concentrations is located at 30 cm depth (the boundary between the silty sand and silt layers). A second-order peak of HM content is present at 60 cm depth, where a particularly high content of carbonate is encountered in soil at this level.

Results for simulated HM profiles showed that the highest concentrations are recorded at soil layer interface particularly at 30 cm and 60 cm depths (Table 4). At these locations, the layer boundary as recognized by Hansen et al. (2011), specific geochemical processes impact the transport of solutes in soil and lead to HM accumulation.

The measured and simulated HM contents are almost similar. The values of the simulated Cr, Fe and Zn content are slightly underestimated throughout the 90 cm profile, while the Cu and Ni contents are underestimated for the first thirty centimeters and overestimated for the last thirty centimeters and almost equal between these two intervals.

The coefficient of determination R^2 for comparison of measured and simulated concentrations varied from 0.66 to 0.99 and showed a high correlation level between simulated and measured concentrations for each HM. This indicated a good reliability of the model in reproducing the transfer of HM in Calcisol (Table 4).

HM transport simulation in Fluvisol

The Fluvisol irrigation with TWW began 9 years later than the Calcisol one. Hence, in the modeling, the calibration time period for this soil is set to 4 years.

A 90 cm soil profile of three horizons of 30 cm thick is assumed. The boundary conditions are the same to those of Calcisol modelling. The initial conditions for soil moisture and HM contents are, respectively, set to arbitrary values and measured soil concentration.

The hydrodynamic parameters are established using the ROSETTA modulus considering soil texture. However, transport ones are recognized based on literature and modified during model calibration. The retained values gave the best fit between measured and simulated HM concentrations.

The simulated and measured concentrations of the HM in Fluvisol are given in Table 5. The coefficient of determination R^2 is close to 1 for all HM indicating a high level of accuracy for the model to represent the transport process in Fluvisol. However, a slight underestimation of HM simulated concentrations is noted. The highest simulated HM concentrations are recorded in the clayey texture levels of the soil profile. The Cr and Zn have the greatest simulated concentrations which are concordant with the observed ones. The most important concentrations are encountered in the upper 60 cm of soil profile. The principal reason for this is that the transport of heavy metals in the soil are affected by numerous factors, such as organic matter, iron oxides, or clays contents which are higher in the surface layer (Rattan et al. 2005).

Table 5 Statistics of measured and simulated HM concentrations in Fluvisol

HM	Depth (cm)	Simulated	Measured	Coefficient of determination R^2
Cr	0–30	0.1	0.1	0.86
	30–60	0.067	0.1	
	60–90	0.012	0.034	
Cu	0–30	0.018	0.022	0.92
	30–60	0.0135	0.021	
	60–90	0.0048	0.006	
Ni	0–30	0.025	0.0336	0.88
	30–60	0.016	0.0308	
	60–90	0.0039	0.0043	
Zn	0–30	0.1	0.1	0.86
	30–60	0.0725	0.1	
	60–90	0.0265	0.026	
Fer	0–30	0.041	0.058	0.95
	30–60	0.032	0.056	
	60–90	0.0083	0.017	

Model exploitation

To assess the lasting effect of the irrigation with TWW, the calibrated model is used to simulate the HM transfer through a conservative scenario. Thus, same irrigation rates as the current one with a constant quality of TWW is considered. Provisional HM concentrations are calculated for time periods of 20 and 30 years in Calcisol and 8 and 10 years in Fluvisol.

The HM accumulation in the Calcisol as shown in Fig. 2 is greater in the soil layers of silty texture. We can notice that, as time increases, the quantity of metals propagates more and more in depth, where greater concentrations are recorded. For Cr, Cu, Ni and Fe the enrichment is mainly in deep layers of silty texture. Whereas for Zn the enrichment is of the same magnitude over the entire thickness of the soil studied (Fig. 2). This particularity for Zn is discussed by Streck and Richter (1997) for a soil irrigated with wastewater for 29 years and explained the phenomena by the slower attainment of Zn sorption equilibrium during irrigation. The study by Chu et al. (2014) reported that Zn have a higher mobility in comparison with other HM.

The particular effect of the layer's interface is also identified for the various HM concentrations along the soil profile. The clayey soil texture and carbonate rich horizon are distinguished by their greater concentrations as these components affects the sorption/desorption of soil minerals and soil pH.

After 30 years of irrigation in Calcisol, the peak concentration of HM reached remarkably high levels, surpassing the Tunisian standard for irrigation with TWW (NT.106.03 1989: Table 1). Specifically, the concentrations of Cr, Zn, Fe, Cu, and Ni were 80 ppm, 70 ppm, 36 ppm, 30 ppm, and 20 ppm, respectively. A similar trend was reported by Ben Fredj et al. (2014) following 12 year irrigation with TWW, the highest levels of Cr, Co, Ni, Pb, and Zn were detected at 20 and 40 cm horizons. Klay et al. (2010) studied the effect of a 15 year TWW irrigation on soil and noted that Cr, Zn, Cd, Fe, Cu and Pb have tendency to accumulate in surface horizons or more in depth depending on soil permeability.

These concentrations exceed the permissible values set for each HM. The elevated levels of these metals promote their potential absorption by plants, leading to adverse effects on plant health and growth. Furthermore, there is a risk of these metals being transferred through the food chain to plants and animals. Hence, Ben Fredj et al. (2014) investigated the stress and the estrogenic activity of TWW and irrigated soils in the same study area, with regard to their HM content, physicochemical properties, and the duration of the irrigation period and showed that longer irrigation period increased the accumulation of toxic compounds.

For the same assumptions of the Fluvisol model calibration, namely, keeping the same irrigation rate and the same quality of TWW. The prediction simulation periods are set

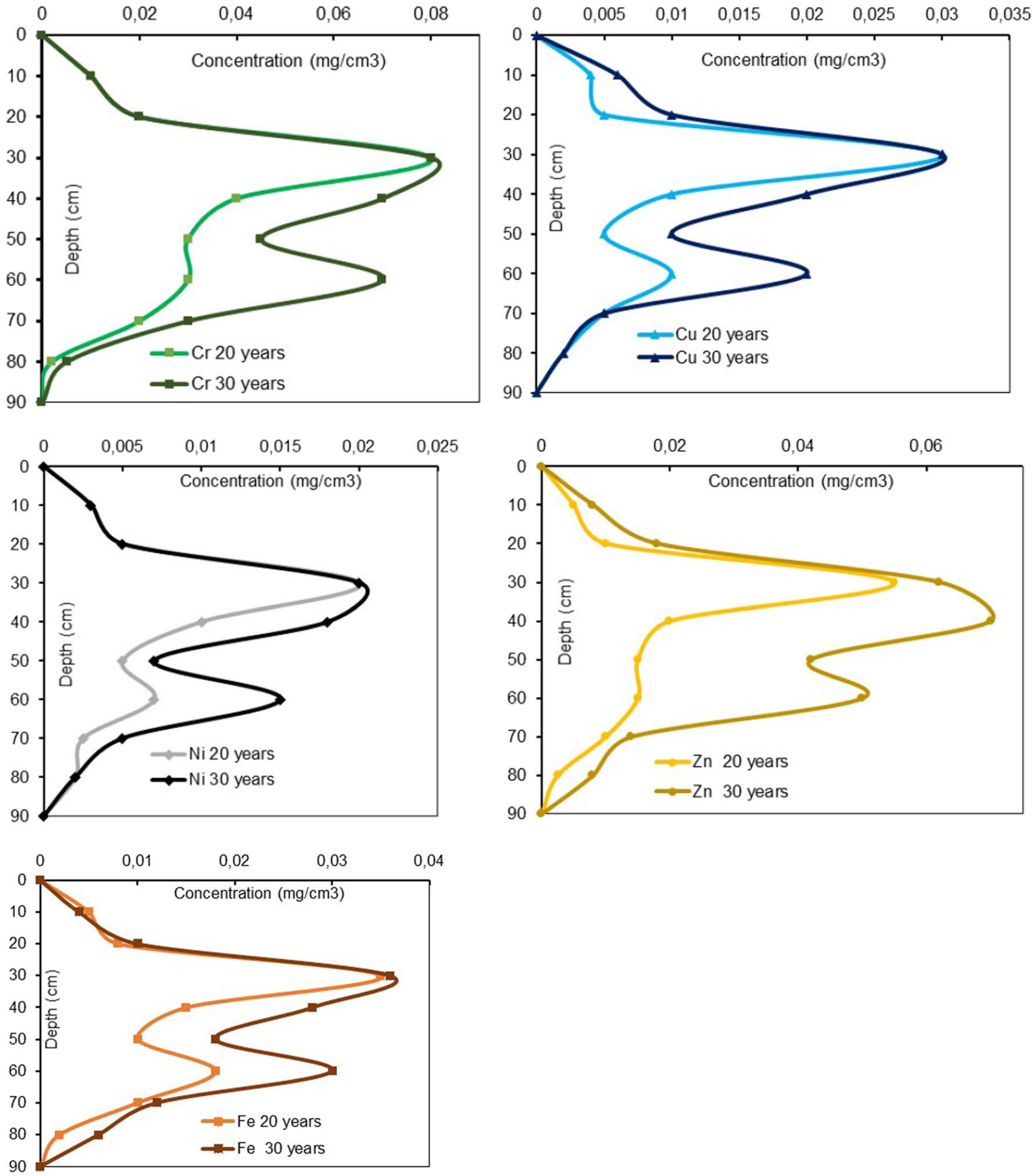

Fig. 2 HM simulated concentration for 20 and 30 years of irrigation in Calcisol

to 8 and 10 years. The evolution of the HM simulated concentration in Fluvisol as a function of time is illustrated in Fig. 3.

For all HM, the concentration in the topsoil gradually increases with respect to time because of the TWW flux resulted from irrigation. The evolution with depth is marked by an increase, where a first peck is reached at 30 cm depth. It than decrease till 50 cm, where the lowest concentrations are recorded. A second increase is recorded in depth with a peak at 60 cm and then decrease from the depth of 70 cm.

The Cr concentration does not increase dramatically with time at the surface. The enrichment is more obvious at deepest layers. The highest concentration is found at the level of the lower limit of the clayey layer. After 10 years of irrigation with TWW, the Cr concentration in the soil can reach 0.55 mg/cm^3. For the same period of irrigation with TWW, the amount of Cu in the clayey layers does not show a great evolution in the first thirty centimeters. However, the Cu concentration will be double relatively to that of 8 years in the deepest sandy horizon.

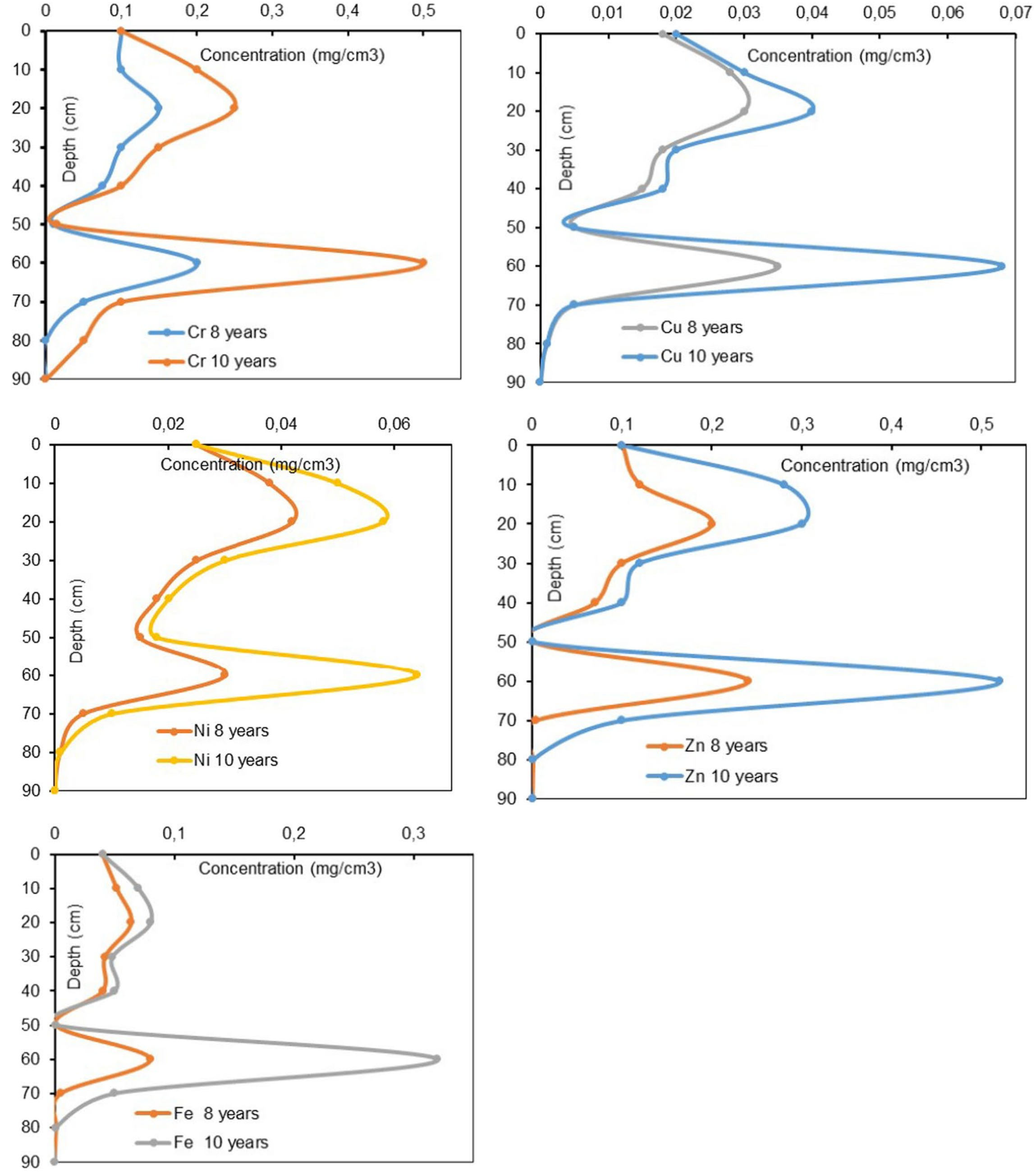

Fig. 3 HM simulated concentration for 8 and 10 years of irrigation in Fluvisol

The increase in Ni concentrations is almost the same for the different layers of the Fluvisol. The peak Ni concentration after 10 years of irrigation will reaches an average of 0.062 mg/cm^3.

The simulation of the concentration of Zn in Fluvisol shows the most enrichment for the deep soil layers. A comparable HM simulated profiles was found by Dos Santos et al. (2013) for Zn and Pb movement modeling in soil for a period of 50 years. Khaskhoussy et al. (2015) studied a TWW irrigated Tunisian clay loam soil and found equivalent result for HM behavior with depth particularly for Zn, Cu, Ni.

Following 10 years of irrigation in Fulvisol, the concentration of heavy metals reached elevated levels, surpassing the allowable thresholds outlined in the Tunisian standard for irrigation with TWW. More specifically, (Cr registered a concentration of 50 ppm, Zn reached 52 ppm, Fe measured 32 ppm, Cu amounted to 68 ppm, and Ni recorded 64 ppm. These facts indicate a substantial contamination level and pose a significant risk to the various components of the ecosystem and particularly to plants irrigated with TWW as revealed by Charfi (1995).

Conclusion

Even though, difficulties in modeling HM transport in the unsaturated soil media, their provisional concentrations were calculated and compared to measured ones for a relatively long period of irrigation with TWW. The modeling software in the unsaturated zone HYDRUS 1D was used to monitor the concentration of Cr, Cu, Ni, Zn and Fe in the different soil horizons to a depth of 90 cm for Calcisol and Fluvisol types. The model calibration is checked based on the coefficient of determination R^2 for measured and calculated HM concentrations and found close to one.

The prediction of HM concentrations in the Calcisol over 30 years shows that, over time, the quantity of metals propagates more and more in depth. The enrichment is more remarkable in the surface layer in the case of Cr, Cu and Ni and it affects the entire soil profile in the case of Zn and Fe.

In the case of Fluvisol, a prediction up to 10 years is undertaken. The enrichment is located in the deep sandy layers for all HM. We note the dominance of Cr and Zn with concentrations of 0.55 mg/cm^3 0.51 mg/cm^3 after 10 years of irrigation with TWW.

A greater enrichment of the quantity of metals in Fluvisol than in Calcisol is observed. The choice of irrigation by TWW will not considers only the water quality but also on the type of soil irrigated and its ability to accumulate metals. We noted in this work, that despite the duration of irrigation by TWW is lower than that applied to Calcisol, the accumulation of HM is greater in the case of Fluvisol.

Despite the TWW initially meeting the permissible threshold concentrations, the HM concentration exceeds the standard limits after several years of irrigation, primarily due to their accumulation in the soil. Consequently, excessive irrigation with TWW has the potential to contribute to pollution. To mitigate this issue, it is recommended to alternate continuous irrigation with water containing lower HM levels, such as third-stage TWW, to prevent further accumulation of heavy metals.

While the approach presented here effectively identified the accumulation and transport of heavy metals (HMs) by calibrating soil parameters using field data, it is important to acknowledge certain limitations. First, the model does not incorporate pH, which really influences the mobility and accumulation of HMs in soil. Second, the assumption of soil properties as constant throughout the simulations neglects potential variations over extended time spans, which is especially inaccurate. To enhance the modeling's representation of the real-world situation, future research should tackle these limitations head-on.

Authors' contributions All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by MA, SB, ZIA and MZ. The first draft of the manuscript was written by MA and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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Availability of data and materials The data sets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

Code availability The Hydrus 1D is free to download on https://www.pc-progress.com.

Declarations

Conflict of interest The authors declare that they have no conflict of interest.

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