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Ahmed BARAKAT*[*1], Oumaima EL HARTI[1], Widad ENNAJI [1]

ENVIRONMENTAL POLLUTION FROM HEAVY METALS IN SOILS SURROUNDING THE ABANDONED MINE OF TANSRIFT (CENTRAL HIGH-ATLAS, MOROCCO)

SUMMARY

In this study, the heavy metal (HM) pollution status, possible sources, and potential ecological and health risk of heavy metals in soils around the Tansrift abandoned mine (Central High Atlas, Morocco) were investigated using the coefficient variation (CV), principal component analysis (PCA), enrichment factor (*EF*), geoaccumulation index (I_{geo}), Pollution Load Index (*PLI*), potential ecological risk index (*RI*), and total hazard index (*HI*). A total of 25 surface soil samples were collected and analyzed to determine the soil proprieties of magnetic susceptibility (MS), organic matter (OM) contents, texture, and the HM (Cd, Cr, Cu, Pb, and Zn) concentrations. The average metal concentrations of 7.74 (Cd), 64.19 (Cr), 333.59 (Cu), 29.95 (Pb), and 58.12 (Zn) mg/kg in soils are higher, except for Pb, than that of the local background and worldwide guidelines. The MS result showed that the soil magnetic material consisted of multidomain (MD), superparamagnetic (SP), and stable single domain (SSD) particles, which were derived from both natural and anthropogenic sources. Moreover, the statistical analysis indicated that Cu originated mainly from anthropogenic and lithologic sources such as agricultural practices, soil parent materials, and mineral dust and wastes from the Tansrift mine. In contrast, agricultural activities are the predominant origin for Cd, Cr, and Zn. *EF* values revealed a significant HM contamination of samples soils, which is in the order of Cd>Cu>Cr>Zn>Pb. The I_{geo} and *PLI* results attested that soils were significantly affected by Cd, Cr and Cu toxic metal and, to a lesser degree, by Pb and Zn. According to the *RI* levels (12.07-1540.37), 40% of sampling sites were at considerable to high ecological risk, mainly influenced by Cd and Cu. Based on the *HI* values, the non-carcinogenic risk of HMs in studied soils could be neglected for adults but not for children. These findings showed the adverse HM effects on the environmental

[1]Ahmed Barakat[*] (Corresponding author: a.barakat@usms.ma), Oumaima El Harti, Widad Ennaji. Geomatics, Georesources and Environment Laboratory, Faculty of Sciences and Techniques, Sultan Moulay Slimane University, MOROCCO.

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quality of soils and the need to implement protective management strategies in the study area.

Keywords: Tansrift abandoned mine, Mining and agricultural activities, Soil proprieties, HM concentrations, Pollution status, Ecological and health risks

INTRODUCTION

Heavy metals (HM) are known environmental pollutants because they are not readily degraded. Also, HM bioaccumulation via the food chain will promote human health hazards such as cancer and nervous system problems (Alomary & Belhadj, 2007; Madzin *et al*., 2015). Also, HMs with concentrations above the threshold levels can cause various effects on the microbiological balance of soils and consequently on their fertility (Barbieri, 2016). So, the properties of HMs in urban and agricultural soils have been gaining increasing attention over the last few decades. Numerous scientific studies have been focused on heavy metal pollution of soil worldwide. They reported that HMs mainly enter environments due to natural processes, anthropogenic activities, or both. Natural sources include weathering of soil and rock parents, while anthropogenic sources are mostly from industrial activities (Huang, 2021; Long *et al*., 2021; Yang *et al*., 2018), vehicle and domestic emissions (El Baghdadi *et al*., 2012; Hanfi *et al*., 2020; Roy *et al*., 2019), mining (Stefanowicz *et al*., 2020; Zhao *et al*., 2020), and agricultural production (Ennaji *et al*., 2020; Yan *et al*., 2020; Yang *et al*., 2018). Therefore understanding the source of HMs is essential to prevent pollution and formulate effective control strategies.

Mining activities are recognized as one of the greatest substantial causes of soil HM pollution affecting surrounding environmental compartments (Atibu *et al*., 2018; Sun *et al*., 2018; Wu *et al*., 2021). In recent decades, the environmental concerns produced by mining activities have attracted growing attention worldwide (Chun *et al*., 2021; Punia, 2021; Sun *et al*., 2018). Several studies have shown that mining activities polluted the surrounding area through inappropriate exploitation and uncontrolled dumping of the mine tailings (Abouian Jahromi *et al*., 2020; Sun *et al*., 2018; Wang *et al*., 2019). Due to its high geological complexity, mining activity in Morocco has enjoyed since its independence the multiplication of large projects concerning phosphates base and precious metals. Resources minerals are gradually extracted and exploited to meet the economic demands, but sometimes without appropriate management of mining tailings. Thereby, several uncontrolled mining areas having very significant concentrations of heavy metal (HM), including Pb, Zn, Cu, Cr, and Cd, have even now undesirable effects on the environmental aspects (El Azhari *et al*., 2017; Leila *et al*., 2021; Midhat *et al*., 2019; Nouri & Haddioui, 2015). These HM pollutants in impoverished heaps and tailings were generally very mobiles due to their smaller dimensions and reactivity linked to the chemical nature of sulfides and sulfates. So, if it is not managed correctly, it could become a pollution source of water, groundwater and soil (Azzeddine & El Hassan, 2020; Bouzekri *et al*., 2020; Moyé *et al*., 2017; Nassiri *et al*., 2021a; Zouhri *et al*., 2019). It can then cause potential risk to human health via direct inhalation, ingestion, and dermal contact absorption (De Miguel *et al*., 2007; Tang *et al*., 2021; Yang *et al*., 2018). Therefore, evaluating the potential environmental impact of Tansrift uncontrolled mining tailing allows residents and authorities to take protective measures.

In the light of the above, this study was conducted with the objectives to evaluate the HM (Cd, Cr, Cu, Pb, and Zn) contamination levels in soils surrounding the Tansrift abandoned mine using some environmental indices, to identify the possible sources of HMs using multivariate statistical methods, and to determine ecological risk level using the potential ecological and health risk indices. This assessment could also provide support and information for properly managing of the areas with a possible risk for residents and the general population.

MATERIAL AND METHODS

Study area

The study area is the Tansrift abandoned mine (32°12’1.72’’N, W6°17’59.09’’W) which closed in 1978. The Tansrift Village lies to the northeast of Ouaouizeght Town, Azilal Province, in the central High Atlas of Morocco (Fig. 1).

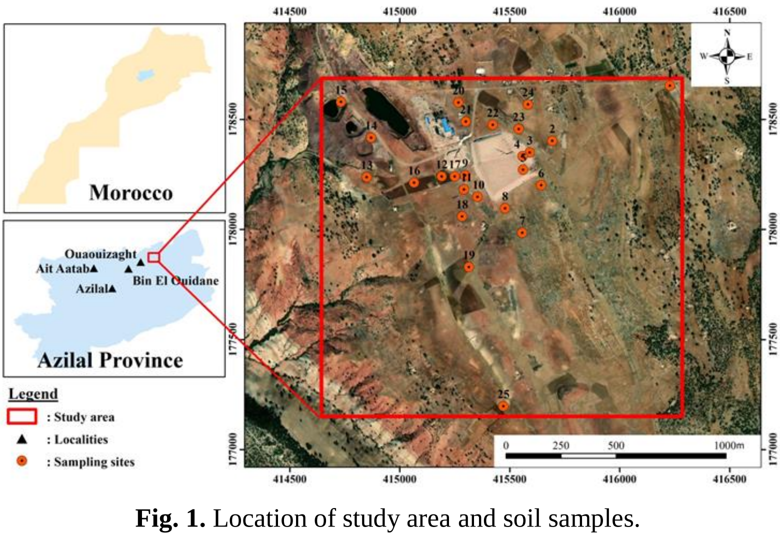

Fig. 1. Location of study area and soil samples.

During the mining years of the Tansrift abandoned mine, 650 000 tons of 1.5% Cu ore were surface mined from outcrops of copper-bearing sandstone. Since its closure, the tailings have been piled up on the mountainside with no protective measures. This area has an altitude of 1448 m and has a warm Mediterranean climate with dry summer. The long-term average annual temperature and precipitation are 19.2°C and 353.8 mm, respectively. With land formerly forest now agricultural and the steep slope of the area, the surface runoff and wind cause substantial surface erosion in the mining area. Fine tailings are easily transported by surface runoff and wind, entering the arable soils and streams of the mining area.

Geologically, the Tansrift area is located just south of the fault (Aghbala-Afourar accident) between the two synclinal structures of Ouaouizarth and Taguelft, having a Bajocian limestone frame and filled with Jurassic-Cretaceous Red Layers (Monbaron, 1988). The area is characterized by a sedimentary series of subsident platforms (transition to the High Atlas basin) comprising: (i) lower to middle Lias carbonate ensemble visible in the Jbel Ghnim to the northwest of the mine, (ii) alternations of red marl and limestone banks, (iii) Bajocian limestone bars visible at the Bin El Ouidane dam, (iv) Middle Jurassic to the Lower Cretaceous continental detrital ensemble of the "Red Layers", and (v) Aptian to the Eocene alternation of silts and red marls with dolomitic carbonate banks (Charrière *et al*., 2005; Haddoumi *et al*., 2010; Mojon *et al*., 2009). The Cu lenticular-shaped copper mineralization is hosted in the Jurassic-Cretaceous Red Layers as paleo-channels occupied mainly by sandstones and sometimes gray to yellowish arkoses (Ibouh *et al*., 2016). The Cu mineralization is mostly with chalcocite, chalcopyrite, pyrite, bornite, covellite, and rarely proustite. At the surface, it is primarily turned to supergene malachite and azurite.

Sampling and analysis

A study that focuses exclusively on HM contamination assessment of agricultural soil that is lacking in the Tansrift mine area. So, in the present study, the HM pollution status, possible sources, and potential ecological and health risk of heavy metals in soils around the Tansrift abandoned mine were investigated using various determining methods (physico-chemical analyses of soils, environmental indices, statistical analysis).

Samples of 25 surface tailings and agricultural soils (0–20 cm) were randomly collected in the study area in Marsh 2020 (Fig. 1). Three surface soil samples were also collected far from the Tansrift mine disturbance to establish the reference for soil HMs. The sampling site locations were obtained using a handheld global positioning system (GPS) (Fig. 1). The collected samples (0.5-1 kg) were stored in separate plastic self-sealing bags and marked and transported to the laboratory. The samples were dried, crushed, and sieved to less than 2 mm to remove coarse debris (Marguí *et al*., 2005). The samples are then used to measure the soil physical parameters of organic matter (OM), soil particle size (sand, silt, and clay), and magnetic susceptibility (MS). The OM content was determined by ignition at 550°C for 5 h in a muffle furnace and comparing pre- and post-ashing weights. The sand, silt, and clay contents were determined using the Robinson pipette method according to the NF X31–107 certified method (AFNOR, 2003). The soil texture is then predicted according to the USDA soil texture classes. The magnetic proprieties including the mass-specific low- and high-frequency magnetic susceptibilities (χlf and χhf at 470 and 4700 Hz) of sampled soils were measured by a Bartington Susceptibility Meter (Model

MS2B) with a dual-frequency sensor at the Georesources and environment laboratory of Sultan Moulay Slimane University., The frequency-dependent susceptibility (χfd) was thus calculated from the measured χlf and χhf (Dearing, 1996) by the application of Eq. (1) (Suresh *et al*., 2011):

$$\chi_{FD}\% = [(\chi_{LF} - \chi_{HF})/\chi_{LF}]x100 \tag{1}$$

The χfd is used to predict the presence of ultrafine superparamagnetic particles of ferrimagnetic components released by anthropogenic sources.

The concentrations of HMs (Cd, Cr, Cu, Zn, and Pb) and Fe in all soil samples were determined by applying the three-acid method (Allen, 1986). Tri-acid mixture (HNO3–HF–HCl) was added to the beaker containing soil sample and heated at 100-110°C until the solution becomes transparent. The resulting solution was finally maintained to 50 ml using deionized water and stored at room temperature before being placed in the Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) analyzer of a National Center for Scientific and Technical Research (CNRST) laboratory accredited ISO 9001 and ISO 17025. To ascertain the accuracy of the analysis and to ensure quality assurance and control (QA/QC), reagent blanks, duplicate tests and standard deviation of <5% were used for HM analyses (Essien *et al*., 2019). The detection limit for the studied element was 10-1 µg.l-1. The analysis of background samples has been carried out using the same approach.

All analysis data were statistically analyzed using IBM SPSS Statistics software. A Shapiro-Wilk test was used to assess the normality of data, which is an assumption to be met in data for statistical tests. Pearson correlation coefficients were determined to quantify the relationships among HM contents and magnetic properties. The coefficient of variation (CV) and the principal component analysis (PCA) were performed to identify the potential sources of HMs.

Assessment of heavy metal contamination

To assess the HM pollution of soils in the study area, environmental factors, namely enrichment factor (EF), geoaccumulation index (I_{geo}), pollution load index (PLI), and potential ecological risk index (RI), were employed as they generally are considered to be more efficient tools in environmental quality assessments (Barakat *et al*., 2012; Barakat *et al*., 2020; El Baghdadi *et al*., 2012; Ennaji *et al*., 2020; Hilali *et al*., 2020; Mirzaei *et al*., 2020; Oumenskou *et al*., 2018a; Yu *et al*., 2017).

The EF index is introduced to evaluate the anthropogenic HM pollution degree and is obtained by the application of Eq. (2) proposed by Selvaraj *et al*. (2004) (Table 1). C_n and C_{ref} represent the concentrations of given examined metal and given reference element, respectively. Fe was used in the present study as the reference element for geochemical normalization (Salati and Moore, 2010) because it is the major element included in this study. EF values are generally interpreted as follows:

$$EF = \begin{cases} < 1 = \text{minimal enrichment or no enrichment} \\ 1-3 = \text{ minor enrichment} \\ 3-5 = \text{ moderate enrichment} \\ 5-20 = \text{ significant enrichment} \\ 20-40 = \text{ very high enrichment} \\ > 40 = \text{ extremely high enrichment} \end{cases} \quad (3)$$

The I_{geo} index is employed to evaluate the anthropogenic heavy metal contamination in soils by comparing metal content in the sample with the geochemical background (Müller, 1979). I_{geo} is calculated as expressed in Eq. (4) (Table 1). Ci is the obtained metal concentration, Bi is the background concentration of analyzed metal, and 1.5 factor is used to attenuate lithogenic variations in background concentrations. According to the I_{geo} values, the soil pollution by HMs are classified as follows (Müller, 1979):

$$Igeo = \begin{cases} \leq 0 = \text{no contamination} \\ 0-1 = \text{ slight contamination} \\ 1-2 = \text{ moderate contamination} \\ 2-3 = \text{ moderate to severe contamination} \\ 3-4 = \text{ severe contamination} \\ 4-5 = \text{ severe to extreme contamination} \\ > 5 = \text{ exteme contamination} \end{cases} \quad (5)$$

The *PLI* index is proposed by Tomlinson *et al*. (1980) to assess the degree of HM contamination in samples by involving all elements concentrations. It was computed by applying Eqs. (6) and (7), where n is the number of determined heavy metals (Table 1). CF represents the contamination factor of the metals. The CF and PLI values are categorized according to respectively Hakanson (1980) and Zhang *et al*. (2018) as follows:

$$CF = \begin{cases} < 1 = \text{low contamination} \\ 1-3 = \text{ moderate contamination} \\ 3-6 = \text{ considerable contamination} \\ > 6 = \text{ very high contamination} \end{cases} \quad (8)$$

$$PLI = \begin{cases} < 1 = \text{unpolluted soil} \\ 1-2 = \text{ moderately polluted soil} \\ 2-3 = \text{ highly polluted soil} \\ > 3 = \text{ extremely polluted soil} \end{cases} \quad (9)$$

Description of the toxicity degree of HMs in the present study was carried out using the potential ecological risk index (RI) developed by Hakanson (1980). The RI index is calculated following Eqs. (10) and (11) as represented in Table 1:

E_r^i is the single potential ecological risk of HM i at the sampling site r, and T_i is the metal toxic factor for HM i. According to Hakanson (1980), the T_i values of Cd = 30; Cu = Pb = Ni = 5, Cr = 2, and Zn = 1 were used in this study. The E_r^n values and the RI index are generally grouped into the following levels (Hakanson, 1980):

$$E_r^n = \begin{cases} < 40 = \text{low risk} \\ 40 - 80 = \text{moderate risk} \\ 80 - 160 = \text{considerable risk} \\ 160 - 320 = \text{high risk} \\ \geq 320 = \text{very high risk} \end{cases} \quad (12)$$

$$RI = \begin{cases} < 150 = \text{low risk} \\ 150 - 300 = \text{moderate risk} \\ 300 - 600 = \text{considerable risk} \\ \geq 600 = \text{high risk} \end{cases} \quad (13)$$

The potential health risk by HMs, including non-carcinogenic effects on adults and children, was evaluated using a health risk quotient (HQ_i) and the total hazard index (HI). The two indices were calculated using Eqs. (14), (15) and (16) as given in Table 1.

Table 1. Pollution indices used in the present study.

Index name	pH	EC (µS/cm)
EF	$EF = (C_n/C_{ref})_{sample} \,/\, (C_n/C_{ref})_{background}$	Eq. (2) (Selvaraj *et al*. 2004)
Igeo	$I_{geo} = log_2\,(C_i/1.5B_i)$	Eq. (4) (Müller, 1979)
PLI	$CF = C_i/B_i$ $PLI = (CF_1 x CF_2 x \ldots. x CF_n)^{1/n}$	Eq. (6) (Müller, 1979) Eq. (7) (Tomlinson *et al*. 1980)
RI	$RI = \sum E_r^i$ $E_r^i = T_i x CF$	Eq. (10) (Müller, 1979) Eq. (11) (Hakanson 1980)
HI	$ADD_i = \frac{C_i x IR x EF x ED x CF}{BW x AT}$ $HQ_i = \frac{ADD}{RfD_i}$ $HI = \sum_{i=1}^{n} HQ_i$	Eq. (14) Eq. (15) Eq. (16)

ADD_i represents the average daily intake (mg/kg/day) of HMs through soil particle ingestion, Ci is the concentration of heavy metal i in soil (mg/kg), IR is the ingestion rate of soil/dust for children (200 mg/day) and adult (100 mg/day) (EPA, 2011), EF is the exposure frequency (350 days/year), ED is the exposure duration (6 years for children and 30 years for adults), BW is the body weight of the exposed individual (15 kg for children and 70 kg for adult), AT is the exposure period for non-carcinogenic effects (2190 days for children and 10950 days for adult), and CF is the conversion factor (10-6 kg/mg), and RfD representing the means reference oral dose, is 0.001, 0.003, 0.04, 0.0035, 0.3 and 0.7 mg.day/kg for Cd, Cr, Cu, Pb, Zn, and Fe, respectively (Leung *et al*., 2008). HQ and HI were classified into two levels: HQ or HI<1: no potential non-carcinogenic risk from HM accumulation, and HQ or HI> 1: significant non-carcinogenic health risk due to HMs.

RESULTS AND DISCUSSION

Physical parameters

The physical parameters and HM contents were determined at all soil samples around the Tansrift mine. The descriptive statistics of physical proprieties are presented in Table 2.

Table 2. Summary of descriptive statistics for soil physical properties.

Statistics	OM (%)	Grain size Sand (%)	Silt (%)	Clay (%)	χ_{fd} (%)	χ_{lf} 10^{-8} m^3 kg^{-}
Min	0.32	8.80	34.10	5.41	0.26	1.88
Max	9.36	31.60	52.00	14.95	12.38	463.65
Mean	4.06	17.23	48.47	10.52	5.09	111.82
SD	2.60	6.18	3.45	1.96	3.45	155.72
CV (%)	63.92	35.86	7.11	18.60	67.72	139.25

According to the USDA soil particle size classification, the texture of the soil samples was mostly silty sand indicating that soils of Tansrift are composed of coarse particles (Table 2). The sampled soil ranges of sand, silt and clay percentages were 8.80-31.60%, 34.10-52%, and 5.41-14.95%, respectively. Silt is the dominant facies in the sampled soils, followed by sand and clay in decreasing order. The samples collected near to the mining tailings showed sand to silty sand facies.

OM is a key factor inducing redox potential and biological processes in soils, including the control of the comportment of chemical species as HMs, through ion exchange adsorption, complexation or chelation reactions. In general, maximum adsorption of cationic HMs occurs at elevated OM levels. Then, OM is an important parameter for assessing HM pollution. The OM contents were in the range of 0.32 to 9.36%, with an average of 4.6%, which pertained to soils low in organic matter (Table 2).

As shown in Table 2, the MS values were in the ranges of 1.88×10^{-8}-463.65×10^{-8} m^3 kg^{-1} with a mean of 111.82×10^{-8} m^3 kg^{-1}, and 1.87×10^{-8}-456.82×10^{-8} m^3 kg^{-1} with a mean of 105.59×10^{-8} m^3 kg^{-1} for χ_{lf} and χ_{hf} frequency magnetic sensitivity, respectively. The slightly higher values of χ_{lf} than those of χ_{lf} revealed the presence of the superparamagnetic minerals (Dearing, 1996). Gautam *et al.* (2004) have described four categories of soils based on their MS values as follows: 'lowly magnetic' (MS<$10\times10^{-8}m^3kg^{-1}$), 'moderately magnetic' (MS $10-100\times10^{-8}m^3kg^{-1}$), and 'highly magnetic' (MS>$100\times10^{-8}m^3kg^{-1}$). According to Gautam *et al.* (2004) classification, the studied samples from the Tansrift area would be lowly, moderately, and highly magnetic, with the proportion of 15%, 55%, and 30%, respectively.

According to Hanesch and Scholger (2002), the MS values above $20\times10^{-8}m^3kg^{-1}$ are believed to be highly endangered by pollution. About 50% of the studied samples showing χ_{LF} values beyond this threshold value revealed anthropogenic stresses on the soil in the study area. Supported by CV coefficients (Table 2) below 139.25%, a tremendous spatial variation of MS might be explained by an enhancement in magnetic material contents derived from mining extraction.

Soils had χ_{fd} values ranging from 0.26% to 12.38%, with an average value of 5.09%. The χ_{fd} can be used to describe the magnetic grain variation in soils and to detect the possible presence of a superparamagnetic (SP) mineral fraction (Dearing, 1996). As predicted by the model of Dearing *et al*. (1996), χ_{fd} lower than 2%, between 2 and 10%, and greater than 10 % indicated a predominance of multidomain (MD) grains, a mixture of stable single domain (SSD) and superparamagnetic (SP) grains, and a predominance of SP gains, respectively. From this model, about 25% of the samples are dominated by coarse MD grains, 65% are a mixture of SP and SSD grains, 5% are dominated by SP grains, and 5% showed erroneous χ_{fd} values. The soil magnetic particles in the surface soil were commonly smaller like SP and PSD, as reported by Meena *et al*. (2011) and Poggere *et al*. (2018).

Regarding the χ_{fd} result, the studied samples are predominated by the three types of magnetic assemblages: MD, SP, and SSD, suggesting that MS values in studied soils might be attributed, in addition to pedogenic processes, to anthropogenic magnetic materials. The anthropogenic materials are suspected from the Tansrift mine environment as dust and erosion by water and wind of mining tailing. Also, the absence of significant correlations between χ_{lf} and χ_{fd} seems to agree with the χ_{lf} results, attesting a dominant portion of ferrimagnetic minerals in soils, and an anthropogenic influence, as mining tailings, on the MS enhancement in studied soils.

Heavy metal contents

The Cd, Cr, Cu, Pb, Zn, and Fe contents varied among sites, with values between 0.15 and 42.90 mg/kg, 16.55 and 215.03 mg/kg, 13.88 and 2444.45 mg/kg, 2.60 and 118.95 mg/kg, 12.98 and 144.68 mg/kg, and 952.25 and 4740.80 mg/kg, with a means of 7.74, 64.19, 333.59, 29.95, 58.12, 2273.92 mg/kg, respectively (Table 3). The HMs and soil physical properties were distributed differently between samples. The contents of Cd (100%), Cr (100%), Cu (67%), and Zn (68%) exceeded the local corresponding background values (Oumenskou *et al*. 2018b) in almost all sampling sites. They were also the two most abundant heavy HMs, with minimum concentrations exceeding the WHO and FAO guidelines (Chiroma *et al.*, 2014). Pb showed values exceeding the local background values and WHO guidelines in 9.14 times and 64.00, respectively. The Fe values remained below their corresponding background value, implying no enhancement in the iron contents of soils. The highest concentrations of HMs were recorded in samples closer to the mine wastes and in those located further away. This observation suggested that the studied soils received an important quantity of metallic pollutants directly associated with the mining activities.

To evaluate the degree of soil spatial variability, the coefficient of variation (CV) has been used, knowing that CV ≤ 15% indicates weak variability; 15% < CV < 100% indicates medium variability; CV ≥ 100% means substantial variability. From the CV results, Cr, Pb, Zn, and Fe were moderately spatially distributed, while Cd and Cu showed strong spatial variability. This disparity in the Cd and Cu concentrations can be attributed to the types and nature of metal sources. Also, their contents in surface soil have a very significant relative value compared to agricultural soils originating from other areas in the region (Barakat *et al.*, 2020; El Baghdadi *et al.*, 2012; Ennaji *et al.*, 2020; Hilali *et al.*, 2020; Oumenskou *et al.*, 2018a) (Table 3). They might be suspected of agricultural and industrial activities, urbanization, and wastewater reuse for crop irrigation.

Table 3. Basic statistics of HM concentrations (mg/kg) in studied soil samples (n=25) and in other studied farmlands of the region.

Samples	**Concentrations**						
	Cd	Cr	Cu	Pb	Zn	Fe	
Min	0.15	16.55	13.88	2.60	12.98	952.25	
Max	42.90	215.03	2444.45	118.95	144.68	4740.80	
Mean	7.74	64.19	333.59	29.95	58.12	2273.92	
SD	8.33	42.71	620.73	28.53	30.40	864.86	
CV (%)	107.67	66.53	186.08	95.25	52.31	38.03	
Earth's soil	0.11	84	26	29	60	3.2	Taylor (1964)
Earth's crust	0.6	100	50	14	75	4.1	Taylor (1964)
FAO/WHO	3	100	100	100	300	2.78	Chiroma *et al.* (2014)
Local Background	0.85	25.21	31.40	32.54	43.76	1.21	Oumenskou *et al.* (2018b)
Urban soil of Beni Mellal City	0,76	44,58		183,12	127,45	30160	El Baghdadi *et al.* (2012)
Peri-urban soils of Beni-Mellal city	2.20	140.41	37.24	31.45	154.63	33700	Barakat *et al.* (2020)
Northeast area of Tadla plain	0.92	32.72	138.10	31.72	162.11	19249.27	Ennaji *et al.* (2020)
Beni Moussa irrigated perimeter	4.89	77.77	87.40	109.66	75.98	26248.18	Hilali *et al.* (2020)
Beni Amir irrigated perimeter	8.77	57.06	25.95	33.32	294.71	23891.57	Oumenskou *et al.* (2018b)

Analysis of HM sources

Before statistical analysis, the data distribution was evaluated using the Shapiro-Wilk test; and since the distribution was not normal, the data were log-transformed. The correlation between soil proprieties, magnetic parameters, and HM contents was analyzed to determine the origin of HMs in the study area. Table 4 presented the Pearson correlation coefficient (R^2) among all analyzed parameters and metal elements.

Table 4. Pearson correlation among HM concentrations and magnetic proprieties.

	Zn	**Cd**	**Cr**	**Cu**	**Pb**	**Fe**	χ_{FD} **(%)**	χ_{LF} **(10^{-8} m^3 kg^{-1})**
Zn	1							
Cd	**0.58**	1						
Cr	**0.85**	**0.78**	1					
Cu	0.05	0.10	0.04	1				
Pb	0.06	0.03	-0.04	**0.70**	1			
Fe	0.07	**-0.27**	-0.14	**0.35**	**0.33**	1		
χ_{FD} **(%)**	0.09	**0.30**	**0.20**	**0.20**	**0.35**	-0.06	1	
χ_{LF} **(10^{-8} m^3 kg^{-1})**	0.07	**0.22**	0.15	**-0.33**	0.04	**-0.23**	0.11	1

Positive and significant correlations were observed between sand and Cu and between clay and Fe. In contrast, sand with Fe and clay with Cu and Pb showed negative and significant correlations indicating that the contents of these metals were affected by sand and clay contents in the studied samples. The other metal elements showed very low correlation coefficients with sand, silt, and clay, oscillating between -0.02 and 0.16. Therefore, all the correlations mentioned above prevent us from concluding the relationships between soil grain size distribution and HM concentrations.

The highest positive correlation was also recorded among Cd, Cr, and Zn, implying that these cations were derived from similar potential sources. Significant positive correlations were observed between Cu, Pb, and Fe, revealing thus that these elements might have common sources. Moreover, Cd exhibited a negative correlation with Fe, reflecting that Cd and Pb share different sources.

Highly significant correlations between HM concentrations and PLI were observed, with the R^2 coefficient increased in an order of 0.31 (Cr)<0.33 (Cd)<0.35 (Zn)<0.42 (Fe)<0.80 (Pb)< 0.88 (Cu). The results attested that metal pollution in the study area was greatly affected by Cu exploitation, even though there might have been natural sources.

The χ_{lf} magnetic susceptibility showed a significant positive correlation with Cd and Cr and a significant negative correlation with Cu and Fe. Such correlations are so weaker between χ_{lf} and contents of Zn and Pb. Similar observations were made by Beckwith *et al*. (1986), which reported that the correlation between HMs and MS is lost when the sediment samples having low MS are added to the statistical analysis. This may explain why the correlation of

MS with some HMs in the present study is so low. Some previous works indicated that the correlation between MS and HM accumulation is complex and varies by assessed areas and pollutant sources (Lu *et al*., 2007; Schmidt *et al*., 2005). Schmidt *et al*. (2005) stated that a significant correlation between MS and a given HM occurs if only samples above a threshold value of 2 % more than the mean or 52% below the median of all samples.

The χ_{fd} values showed a significant positive correlation with Cd, Cr, Cu, and Pb, a weaker positive relation with Zn, and a significant negative correlation with Fe. From this, it is assumed that Cd, Cr, Cu, and Pb may be mainly added through mining tailings and dust at the studied area, while the concentration of Fe could be primarily linked to the parent material of soils. Fe is an omnipresent element in the environment, but its content in soils is generally low (<0.1%). Thus, there is often little direct connection between magnetic susceptibility and Fe content in soils (Maher, 1998). Liu *et al*. (2003b) suggested that the MS of soils formed in sedimentary rocks frequently increases with an increase in χ_{fd} frequency-dependent susceptibility. This explained the close relationships between χ_{fd} and HM contents in studied soils developed from sedimentary rocks. The heavy metals are due to natural particles present in the bedrock and ore wastes and dust brought into the topsoil environment.

Among soil-magnetic parameters, there are no significant positive correlations between χ_{lf} and χ_{fd} in studied soils, which did not exceed R^2 of 0.11. The absence of a significant positive correlation revealed that a magnetic enhancement might cause the MS variations due to mining pollution.

For more evaluation of sources of the HMs, PCA was performed on log-transformed data that are non-normal distributed. Based on the grouping of HMs using varimax rotation factor analysis, four components with eigenvalue >1 were calculated and described 79.72% of cumulative percentage for the soils (Table 5, Fig. 2). According to Liu *et al*. (2003a), the component loadings were classified as ‘strong’, ‘moderate’, and ‘weak’, corresponding to absolute loading values of 40.75, 0.75–0.50, 0.50–0.30, respectively.

PC1 that produced 25.08% of the total variance was strongly positively correlated with Cr, Zn, and Cd and weakly positively loaded with Cu. This finding suggested that these elements are highly associated with each other. The primary sources of these elements are probably the dust fallout and gaseous emissions from the Cu ore mining and processing. Cd and Co frequently occur as an accompanying mineral of Cu ores (Esmaeili *et al*., 2014; Yang *et al*., 2020). Besides, the application of agrochemicals like phosphate fertilizers and pesticides could be another source for the high content of these HMs. PC2 accounting 22.50% of variance showed moderate to strong positive factor loadings with OM and sand variables and moderate to strong negative loading with Fe and clay variables. Local normality of Fe content might be derived from the weathering of parent material and subsequent pedogenesis. This finding is consistent with the positive between Fe and clay that clays can also influence. PC3, explaining for 19.53% of the data variance, had strong positive loadings on Cu, Pb, and Fe. The result suggested that these HMs is derived from the same source.

Table 5. Rotated principal component matrix and factor loadings for HMs in sampled soils.

	Principal component (PC)			
	1	2	3	4
Cr	**0.93**	0.10	0.02	-0.03
Zn	**0.90**	-0.14	0.07	-0.22
Cd	**0.87**	0.01	0.10	0.13
Pb	0.02	0.01	**0.86**	0.07
Cu	**0.23**	0.15	**0.81**	0.16
Fe	-0.07	**-0.48**	**0.67**	**-0.39**
OM	0.10	**0.56**	-0.15	0.01
Sand	-0.07	**0.91**	0.04	**-0.32**
Clay	0.10	**-0.91**	**-0.25**	-0.12
Silt	-0.08	-0.10	0.11	**0.95**
Eigenvalue	2.51	2.25	1.95	1.26
% of total variance	25.08	22.50	19.53	12.61
Cumulative variance (%)	25.08	47.58	67.11	79.72

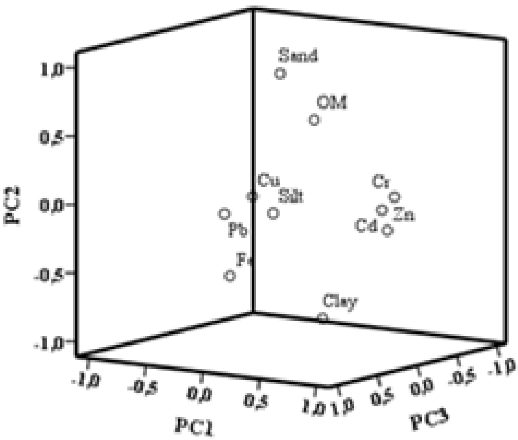

Fig. 2. Varimax-rotated components of the HMs in studied soils.

Since the contents of Pb and Fe in the study area do not exceed their contents in background soils, it can be inferred a major natural lithologic origin of the HMs of PC3. As the study area is located at and near the copper deposit and vicinity of the abandoned mine, the source of these elements could be lithogenic and related to ore extraction (waste minerals, tailings, and mine drainage). PC4, explaining for 12.61% of the data variance, mainly loaded silt and weakly negatively loaded sand and Fe, suggesting that it originated from weathering of the parent material of soils.

A cluster method is employed to assess the spatial variability and identify similarities among the sampling stations, and the obtained results are presented as a dendogram in Fig. 3. According to Fig. 3, all soil samples could be grouped in three clusters, with 14 samples in cluster 1, 3 samples in cluster 2, and 8 samples in cluster 3.

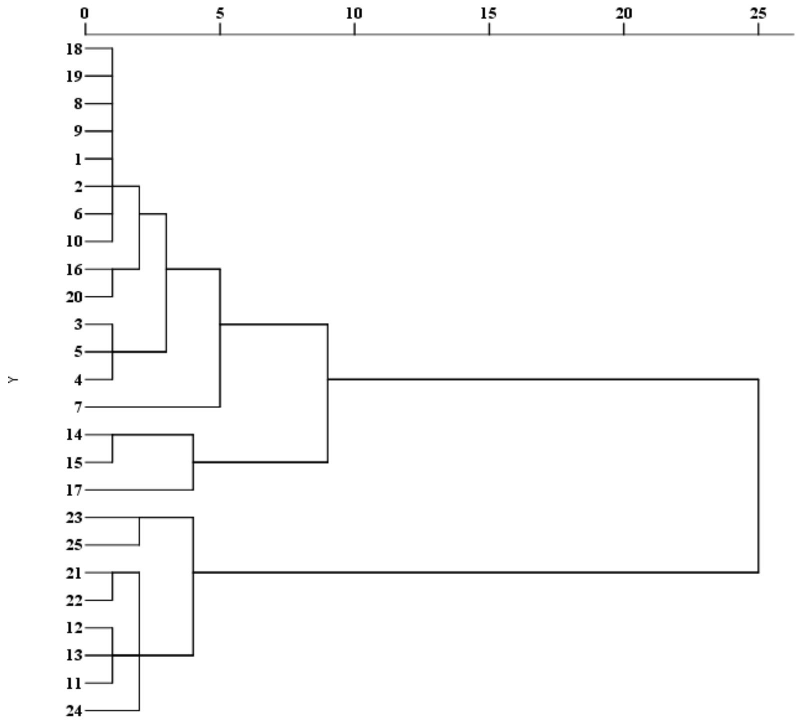

Fig. 3. Clustering tree of sampling sites based on the ward method.

The distribution patterns of soil samples based on their HM concentrations indicated spatial heterogeneity. Some highly polluted samples are located near or far away from abandoned mine wastes. This variation in the spatial distribution is probably controlled by the wind and water transport of the HM pollutants from the abandoned mine site.

HM contamination assessment

The effects of mining activities were further assessed using a few environmental, ecological, and health risk indices, namely *EF*, I_{geo}, *PLI*, *RI*, and *HI*. The results are presented in Tables 6, 7 and 8.

Table 6. Descriptive statistics of *EF*, I_{geo} and *PLI* values.

	EF					I_{geo}					***PLI***
	Cd	Cr	Cu	Pb	Zn	Cd	Cr	Cu	Pb	Zn	
Min	0.18	0.66	0.44	0.08	0.30	-3.09	-1.19	-1.76	-4.23	-2.34	0,39
Max	50.47	8.53	77.85	3.66	3.31	5.07	2.51	5.70	1.29	1.14	6,93
Mean	9.10	2.55	10.62	0.92	1.33	1.93	0.54	0.75	-1.20	-0.35	2,31
SD	9.80	1.69	19.77	0.88	0.69	1.64	0.78	2.35	1.25	0.74	1,58
CV (%)	107.67	66.53	186.08	95.25	52.31	84.90	144.23	311.75	-104.17	-210.81	68,49

Table 7. Statistical attributes of E_r^i and *RI* for ecological hazard of HMs in studied soil samples.

	E_r^i					***RI***
	Cd	Cr	Cu	Pb	Zn	
Min	5.29	1.31	2.21	0.40	0.30	12.07
Max	1514.12	17.06	389.24	18.28	3.31	1540.37
Mean	273.07	5.09	53.12	4.60	1.33	337.21
SD	294.01	3.39	98.84	4.38	0.69	322.96
CV (%)	107.67	66.53	186.08	95.25	52.31	95.77

Table 8. *HQ* and *HI* for non-carcinogenic risk of HMs in studied soil samples.

	HQ					***HO***
	Cd	Cr	Cu	Pb	Zn	
Child						
Min	1.92E-03	7.36E-02	4.63E-03	9.90E-03	5.77E-03	1.36E-01
Max	5.48E-01	9.56E-01	8.15E-01	4.53E-01	6.43E-02	1.78E+00
Mean	9.89E-02	2.85E-01	1.11E-01	1.14E-01	2.58E-02	6.35E-01
SD	1.07E-01	1.90E-01	2.07E-01	1.09E-01	1.35E-02	4.22E-01
CV (%)	1.08E+02	6.65E+01	1.86E+02	9.52E+01	5.23E+01	6.64E+01
Adult						
Min	2.05E-04	7.56E-03	4.75E-04	1.02E-03	5.92E-04	1.40E-02
Max	5.88E-02	9.82E-02	8.37E-02	4.66E-02	6.61E-03	1.84E-01
Mean	1.06E-02	2.93E-02	1.14E-02	1.17E-02	2.65E-03	6.57E-02
SD	1.14E-02	1.95E-02	2.13E-02	1.12E-02	1.39E-03	4.37E-02
CV (%)	1.08E+02	6.65E+01	1.86E+02	9.52E+01	5.23E+01	6.65E+01

The *EF* values of Cd varied between 1.48 and 643.51, indicative of significant contamination (20% of all samples) and very to extremely high contamination (72% of all samples) of sampled soils. The *EF* of Cr with values of 4.96-108.75 involved that 84% and 8% of all soil samples showed, respectively, significant enrichment and very- to extremely high enrichment. *EF* for Cu oscillated between 2.60 to 382.66 in soil samples. It indicated moderate, significant, and extremely high enrichment of Cu for 20%, 40%, and 28% of all samples. *EF* for Pb with values of 0.64-21.83 indicated that the sampled soils are minor, moderate, and significant enriched in Pb in 36%, 32%, and 28% of sampling sites. *EF* for Zn ranging between 2.49 and 39.08 revealed that 20% and 64% of all samples exhibited moderate and significant enrichment, respectively. The area surrounding the Tansrift mine tailings recovered high contamination that was in decreasing order Cd>Cu>Cr>Zn>Pb. Regarding *EF* results, it is significantly apparent that anthropogenic factors significantly impacted the sampled soils.

The results of the I_{geo} values (Table 6, Fig. 4) indicate that the studied soils were not polluted, slightly, moderately, moderately to severely polluted, severely polluted by Cd in 8%, 12%, 28%, 28%, and 20% of sites, respectively. The I_{geo} values of Cr classified the studied samples as not polluted (20% of all sites), slightly (60% of all sites), and moderately (12% of all sites). Moreover, the I_{geo} of Cu indicated that 56% and 44% of soil samples were respectively unpolluted and slightly to extremely polluted. Concerning I_{geo} of Pb, and Zn, the studied samples were mainly classified as unpolluted to slightly polluted. From the I_{geo} results, we acknowledge that the study area was greatly affected by Cd, Cr, and Cu and, to a lesser degree, by Pb and Zn, which might be mainly associated with mining. However, the naturally occurring source should not be neglected.

PLI used to evaluate the degree of HM pollution in the study area ranged from 0.39 to 6.93, with a mean of 2.31 (Table 6). Only 12% were not polluted among the entire soil samples, while 88% were polluted. As given in Fig. 5, the degree of pollution included moderately polluted, highly polluted, and extremely polluted, with the respective proportion being 48%, 16%, and 24% of analyzed samples. These *PLI* results combined with the I_{geo} results attested that HM pollution in the study area was affected by human activities.

The results of ecological indices of HMs in soils of the Tansrift are presented in Table 7. The results showed that the mean monomial potential ecological risk (E_r^i) was in the following order: Cd>Cu>Cr>Pb>Zn. The highest E_r^i of Cd was in a range of 5.29 and 1514.12, indicating significant risks of this metal. Cu had E_r^i varying between 2.21 and 389.24, indicating relatively high risks. E_r^i of Cr, Pb, and Zn were less than 18.3, suggesting quite low risks. The *RI* of the HMs in the studied soils ranged between 12.07 and 1540.37 (Table 7). According to the RI values, the studied soil samples are lowly, moderately, considerably, and highly contaminated, with the proportion being 24%, 36%, 24%, and 16%, respectively. Used for soil contamination health risk assessment, *HQ* values estimated for each element and the comprehensive non-carcinogenic risk index (*HI*), are given in Table 8. The *HQ* values of HMs for adult and children decreased in the Cr>Pb>Cu>Cd>Zn sequence. The *HI* values for adult were lower than 1, indicating no non-carcinogenic risk for the adult population, whereas, for children, four samples (S7, S10, S23, and S25) showed *HI* values exceeding 1. This finding indicated that the non-carcinogenic risk of HMs in the study area could be neglected for adult, but it could not be ignored for children health.

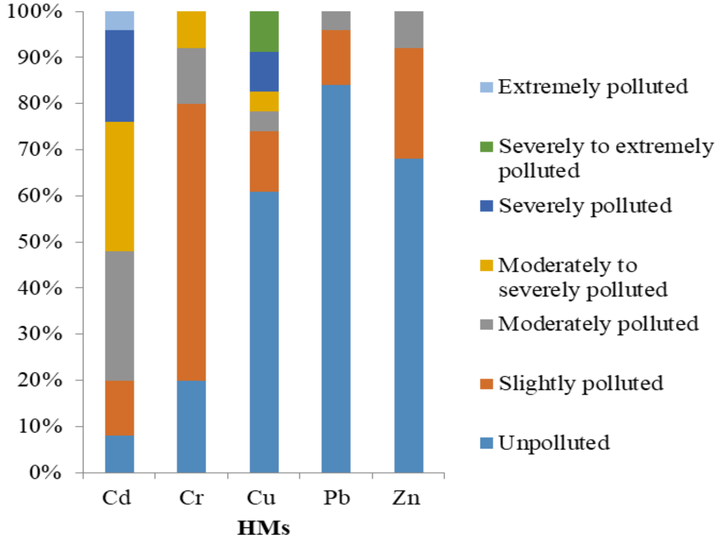

Fig. 4. HM pollution levels of the studied soils based on I_{geo} values.

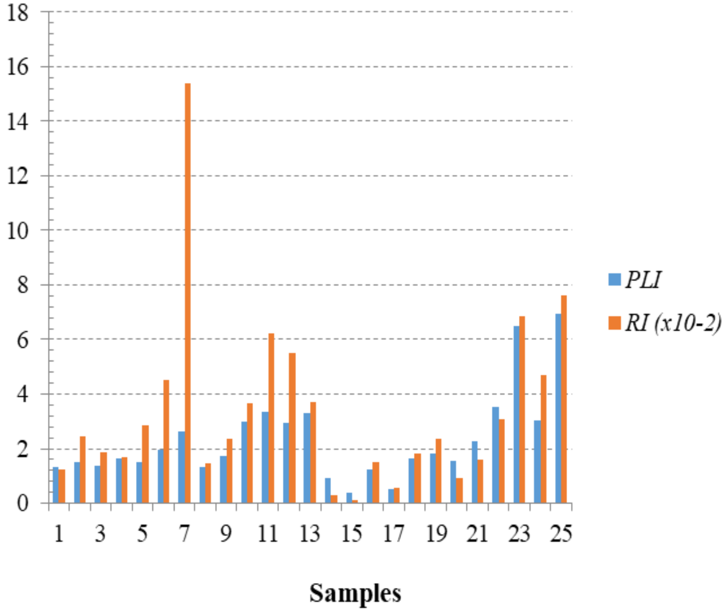

Fig. 5. *PLI* and *HI* values in soils near the Tansrift mine, Morocco.

Overall the obtained results in this study showed that the natural environment surrounding the Tansrift abandoned mine in the Azilal Province

(Morocco) is contaminated by HMs (Cd, Zn, Cr, Cu, and Pb). The HMs sources in question are closely connected to copper mine and the heaps of waste rock remaining after ore mining that are sources of HM from weathering and dusting.

Surface soil contents of Cd (100%), Cr (100%), Cu (67%), and Zn (68%) at the Tansrift area has a very high relative value compared to the corresponding average background values. The analyzed HMs were found to be the most polluting heavy metals in soils in Morocco. Reviews showed that metallic materials derived from different sources, including weathering of parent materials, agricultural activities, industrial activities, traffic, and mining activities (Barakat *et al*., 2019; Bouzekri *et al*., 2019; El Azhari *et al*., 2017; El Baghdadi *et al*., 2012; Ennaji *et al*., 2020; Hilali *et al*., 2020; Khafouri *et al*., 2021; Oumenskou *et al*., 2018b). In the study area, high concentrations of HMs added to high spatial variation indicated anthropogenic inputs for heavy metals. These high average concentrations are higher than those reported in agricultural soil samples of Tadla plain in central Morocco reported by Barakat *et al*. (2019), Ennaji *et al*. (2020) and Oumenskou *et al*. (2018b). Some previous case studies in Morocco focused on certain HMs in soils adjacent to the ore mines reported that the ore processing and the mineral residues affected the metallic mineral contents in the surrounding environments (El Amari *et al*., 2014; El Hamiani *et al*., 2015; Khafouri *et al*., 2021; Midhat *et al*., 2019). They reported that threats of HM pollution of the surrounding ecosystems of the abandoned Pb mines are strictly due to water and wind erosion that moved from the abandoned waste particles enriched in some HMs. Tuo *et al*. (2014) reported that the wind and water erosion of the surface soil produced a decrease in the content of fine particles and an increase in coarse particle content. This outcome is consistent with the coarse granulometry of the tailings evaluated in the field, suggesting that the tailings in the study area could be mobilized to the nearby environment under erosion. It is also supported by the fact that some soil samples located away from the mine site showed significant HM concentrations, and the magnetic properties result subsequently confirmed it. The type of magnetic material in the samples soils contained an admixture of MD, SP, and SSD grains, which is suspected of a combination of lithogenic and anthropogenic magnetic particles. Previous studies reported that the size of topsoil magnetic particles derived from anthropogenic is significantly coarser (Hu *et al*., 2007; Poggere *et al*., 2018).

Moreover, the lower correlation coefficients between χlf and HMs implied that HM contents in the soils are not controlled only by magnetic grains but can be affected by fertilization (Hu *et al*., 2007). Also, the PCA analysis finding attested that HMS in studied soils originated from different sources (lithogenic and anthropogenic) and confirmed the MS findings. The results of Igeo, PLI, and RI indices, calculated from spatially different soil samples, were also indicative of pollution driven by HMs in the Tansrift area. Cd, Cr, and Cu are considered the major contributors to contamination and toxicity in the investigated area. Many previous studies have been realized at other mines in Morocco and also reported abnormal accumulations of HMs accompanied by significant toxicity in soils and watercourse sediments, which often act simultaneously as a carrier and as secondary sources of pollutants in both the soil-plant and aquatic systems (Boularbah *et al*., 2006; Bouzekri *et al*., 2019; El Azhari *et al*., 2017; El Khalil *et al*., 2008; Khafouri *et al*., 2021; Nassiri *et al*., 2021b). In all of this, findings of the present investigation led to interesting conclusions on HM concentrations and their environmental toxicity in soils surrounding the Tansrift abandoned mine. Thus, based upon these conclusions, more attention should be paid to HM contamination of soils in the investigated area.

In summary, combining the analyses, the study area should be managed prior, and the protection of children should be prioritized. Therefore, the identification of priority control components from our study can help policymakers to form more effective exposure reduction and management measures. We also recommend implementing protective measures such as informing the local population about the health consequences and exposures.

CONCLUSIONS

The present study investigated the contents, contamination, and ecological and health risks of HMs in farmlands surrounding the abandoned mine of Tansrift (Central High Atlas, Morocco). The results showed that the HM concentrations followed the order of Cu>Cr>Zn>Pb>Cd, and the contents of Cd (100%), Cr (100%), Cu (67%), and Zn (68%) exceeded the local corresponding background values. Multivariate statistics suggested that Cu had anthropogenic and lithologic origins as agricultural practices, soil parent materials, and mineral dust and wastes from the Tansrift mine, while agricultural activities mainly governed cd, Cr, and Zn. Using cluster analysis, the polluted samples are heterogeneously spatially distributed, conforming that different sources contributed to the accumulation of HMs in soils. As regards *EF* results, significant contamination by HMs was recovered in decreasing order Cd>Cu>Cr>Zn>Pb. The I_{geo} revealed that the study area was greatly affected by Cd, Cr, and Cu toxic metal and, to a lesser degree, by Pb and Zn. According to I_{geo} values, the study area appeared more affected by Cd, Cr, and Cu toxic metal and, to a lesser degree, by Pb and Zn. The *PLI* values (0.39-6.93) used to evaluate the degree of HM pollution in the study area ranging between 0.39 and 6.93 showed that 88% of the soil samples were polluted. The potential ecological risk of HMs order was Cd>Cu>Cr>Pb>Zn. The criteria of ecological risk on Cd and Cu were significant risk, and the other metals were low risk. The *HI* values indicated that the non-carcinogenic risk of HMs in studied soils could be neglected for adult, but not for children.

To end, this study indicated that special actions could be carried out at the study area to reduce the inputs of HMs in agricultural lands. Furthermore, the study demonstrated that the adopted methodology combining soil proprieties' measurements, multivariate statistical analysis, and environmental indices could be a reliable tool to evaluate the HM pollution status and identify their origins and ecological impacts.

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