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Ecological Risk Assessment for Heavy Metals in Agricultural Soils Surrounding Dumps, Huancayo Province, Peru

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ABSTRACT

Contamination generated by dumps is an environmental problem because the soils around the dumps are used for the cultivation of agricultural products and pastures, and could constitute a threat to human health. The contamination index and ecological risk potential for heavy metals in agricultural soils surrounding the solid waste dumps in Agua de las Vírgenes (AV) and El Eden (ED) in Huancayo were evaluated. The concentration of heavy metals in the soils was measured using an inductively coupled optical emission spectrophotometer. The average concentration of As, Cd, Cr, Pb, Cu, Fe and Zn in the soils was 25.77, 1.03, 6.09, 112.07, 48.52, 53733.33 and 349.10 mg/kg in AV and 14.35, 0.28, 2.85, 123.01, 57.47, 36137.50 and 414.31 mg/kg in ED respectively. The mean values of the contamination factor for As determined a very high soil contamination status at both sites. The order of mean Igeo values was As> Cd> Pb> Zn> Cu> Fe> Cr. The soils are strongly enriched with As and Cd. The highest risk index was for Cd, with a higher contribution to the overall potential risk index. The principal component analysis explained that 71% of the data set and the identified sources of heavy metals are the varied composition of wastes in general and crop irrigation with wastewater. In general, agricultural soils are characterized for a level of ecological risk that varies from high to very high. Among the measured heavy metals As, Cd and Pb pose significant health risks. Continuous monitoring of the level of contamination of soils affected by dump leachate and wastewater irrigation is suggested.

Keywords: soils; dumps; heavy metals; pollution; sources of contamination.

INTRODUCTION

The rapid increase of the world population parallel with urbanization, changes in cities, social life, accelerated economic growth, income level in urban areas, low efficiency of the waste collection system and waste treatment have contributed to a rapid increase in solid waste generation [Chen et al. 2020; Mavimbela et al. 2019]. Human activities are causing important geochemical transformations in nature, and unregulated waste dumping is causing serious pollution of the surrounding environment, resulting in intense contamination of soil, water and atmosphere [Borjac et al. 2019]. On the other hand, inadequate waste management and disposal affects the environment and human health [Odonkor et al. 2020]. With the growth of urbanization, anthropogenic activities bring large volumes of pollutants into the urban environment, causing serious heavy metal contamination of soils [Zhang et al. 2019]. These toxic elements from dumps are substances that are introduced into the soil, can alter the quality and function of the soil, causing degradation and alteration of the basic structure of the soil, causing damage to ecosystems and the health of the surrounding population [Agbeshie et al. 2020; Yu et al. 2020]. Heavy metals are one of the most dangerous pollutants in our natural environment due to their toxicity, long-term persistence, non-biodegradability and bioaccumulation capacity [Wu et al. 2018], and are considered hazardous to human health and the ecosystem [Jiang et al. 2017; Rai et al. 2019; Yang et al. 2018]. These elements in the geochemical background are found naturally in very low concentrations, whereas elevated concentrations are commonly associated with human activities [Islam et al. 2017; Krishna and Mohan 2016]. The presence of heavy metals in soil is commonly associated with geochemical and biological processes and is strongly influenced by anthropogenic activities such as industrial activities, waste disposal and agricultural practices [Benson 2006]. Soil contamination is most acute where landfills lack base liners, where the leachate collection system and leachate treatment are absent. Therefore, the movement of leachate is subject to various physical, chemical and biological processes and geological condition that eventually affect the concentration of contaminants in soils and groundwater [Fatta et al. 1999; Onwudike et al. 2017; Samadder et al. 2017]. Some wastes can contaminate soil, groundwater or even surface water by leaching and runoff during rainfall. Soil texture, pH and organic matter content influence the bioavailability of heavy metals [Alloway 2013; Li et al. 2019; Xiao et al. 2019], and soil contamination with toxic metals is a global problem because they do not degrade in the soil and cannot be permanently removed [Essien et al. 2019]. Solid waste contaminants affect the physicochemical properties of soil [Ali et al. 2014], and leachate transport depends on the characteristics of the soil profile, which controls the movement and storage of water and solutes [Mavimbela et al. 2019].

The use of contamination indices is a key tool for effective assessment of soil contamination with heavy metals and is of great importance for monitoring soil quality and ensuring its sustainability in the future [Kowalska et al. 2018], in addition, these contamination indices are tools to assess the potential ecological risk from heavy metals in soil, in order to prioritize pollution control studies [Chandrasekaran and Ravisankar 2019; Huang et al. 2019; Keshavarzi and Kumar 2019; Kumar et al. 2019), indicators that are used by various researchers in China, Pakistan, Bangladesh, Iran, India, Spain, Nigeria, Poland. Due to the rapid increase in population and the increase of waste products in Peru, 2014 generated a total of 7 497 482 t/year of municipal urban waste, of which 64% is household waste and 26% is non-household waste; and the rate of solid waste generation per day in Peru is approximately 18 thousand 870 tons of solid waste, and the Junín region contributes 479 tons; also Huancayo is among the 10 cities in the country that generate more solid waste [Ministry of the Environment 2016]. The per-capita generation of municipal solid waste in 2009 amounted to 0.380 kg/inhab/day for a population of 116,842 inhabitants, which indicates that the daily volume of solid waste exceeds the installed capacity for its adequate management in the municipalities [Ministry of the Environment 2014]. The size of the population and waste management determine the volume of municipal waste generation [Ministry of the Environment 2010].

Inadequate management and uncontrolled open dumping of municipal solid waste on land located in "El Eden" and "Agua de las Vírgenes", generated by the population of Huancayo and El Tambo, have caused environmental impacts on the soil, water, air, human settlements and the health of the population of the Yauris sector, El Eden and La Ribera neighbourhoods. Currently, the El Eden dump has implemented a closure plan, but the Agua de las Vírgenes dump does not yet have a closure plan. The level of contamination and degradation of the agricultural soils surrounding the dumps due to leachates is unknown. Consequently, the soil, water bodies, human settlements and agricultural crops near the dumps are highly vulnerable to contamination by toxic elements such as heavy metals. In this context, the accumulation of these toxic substances in the soil is characteristic of an open dump, and constitutes a risk to human and ecosystem health due to exposure to toxic contaminants, which not only threaten aquatic and soil ecosystems, but also contaminate the food chain. The objective of the research was to evaluate the level of contamination and the potential ecological risk of agricultural soils associated with heavy metals, and to identify the sources in the soils surrounding the "Agua de las Vírgenes" and "El Eden" dumps in Huancayo province, Peru.

MATERIALS AND METHODS

Scope of the study

The Agua de las Vírgenes (AV) and El Eden (ED) dumps are located on the banks of the Mantaro River in the district and province

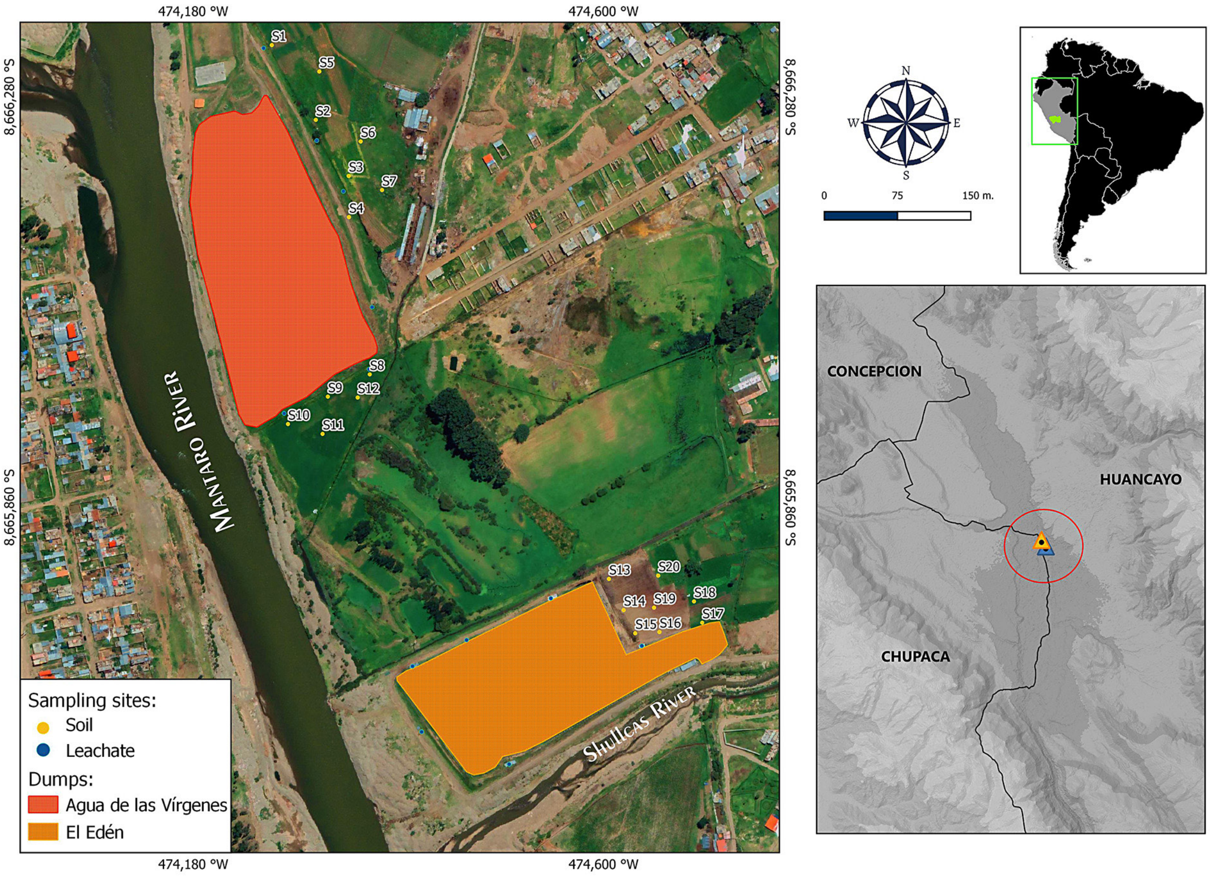

Figure 1. Location of agricultural soil sampling points at the Agua de las Vírgenes and El Eden dumps, Huancayo province

of Huancayo in the department of Junín (Figure 1), both managed by the Huancayo provincial municipality and the El Tambo district municipality. The dumps are located on the banks of the Mantaro River, within the alluvial plain approximately 100–200 m from the urban population. The area's climate is temperate sub-humid, with an average annual temperature of 11.5°C and annual rainfall of 649 mm. The rainy season is from January to March and the dry season is from June to August. The surface area of the dumps covers an area of approximately 3.95 ha (AV) and 2.89 ha (ED). The AV dump is a degraded area that currently does not have a closure plan; to the east there are three leachate wells, which during the rainy season overflow, affecting agricultural soils, and since 2015 solid household waste is no longer disposed of. The ED dump is located at the intersection of the Mantaro and Shullcas rivers and has six leachate wells. This dump finished operating in 2017. Both dumps do not have a lining at the bottom of the pit to prevent leachate to seep through and contaminate the underground water.

Soil sample collection

In areas with a history of agricultural activity surrounding the Agua de las Vírgenes and El Eden dumps, a total of 20 soil samples of 1 kg each were collected at a depth of 0–30 cm [Agbeshie et al. 2020; Martínez Mera et al. 2019; Nyiramigisha et al. 2021]. At each sampling point, the positioning coordinates were recorded with the help of GPS. Soil samples were collected with the help of a small stainless-steel shovel, previously removing materials such as pebbles, stones, organic and inorganic debris; they were then placed in airtight polyethylene bags, labelled and transported to the laboratory. Soil samples were dried at room temperature. The samples were crushed with a mechanical pulveriser and sieved using a 20-mesh nylon sieve, and stored in airtight plastic bags for the determination of As, Cd, Cr, Cu, Fe, Pb and Zn concentrations.

Sample preparation and analysis

For the quantification of As and selected heavy metals, 0.5 g of soil was weighed and transferred to plastic containers with lids. In each container, the soil samples were digested with 2.0 ml of nitric acid (HNO_3) and 6.0 ml of hydrochloric acid (HCl), which were placed in a block digester, the digestion temperature was adjusted to approximately 85°C for 90 minutes. After the time elapsed, the containers with the digested solution were removed, and once cooled, 25 ml of ultrapure water was added and allowed to stand for 12 hours to allow the undissolved material to precipitate. The samples were analysed using the inductively coupled optical emission spectrophotometer (ICP-OES, Agilent 700).

Each sample was analysed in triplicate and the mean value was reported as mean ± SD as the final result, after properly calibrating the instrument using calibration blanks and calibration standard solutions of each metal to be analysed. The calibration curves were analysed based on their corresponding correlation coefficients (r^2) and evaluated at values greater than 0.9995. The standard precision of the analysis was less than 10% of the relative percentage deviation, indicating good reproducibility of the equipment and operating procedures. The percentage recovery of heavy metals ranged from 98.6% to 117. 8%. Likewise, the pH of the soil samples was determined by the potentiometer method (soil-water ratio 1:1), the organic matter (OM) content by wet combustion by the Walkey and Black method [Nelson and Sommers 1982] and the electrical conductivity per saturation extract of a soil [Corwin and Lesch 2005].

ASSESSMENT OF SOIL CONTAMINATION BY HEAVY METALS

Contamination factor

Contamination factor (CF) is an indicator of contamination from anthropogenic inputs associated with a single heavy metal [Hakanson 1980]. It evaluates the ratio between the content of each heavy metal in the soil with respect to the geochemical background value, calculated by equation 1:

$$CF = C_{soil}/C_{background} \quad (1)$$

where: C_{soil} is the concentration of each metal in the soil samples and $C_{background}$ is the geochemical background value of each metal [Hakanson 1980] categorized the contamination values into four classes: CF<1 (low contamination), 1<CF<3 (moderate contamination), 3<CF<6 (considerable contamination) and CF>6 (very high contamination).

Pollution load index

The pollution load index (PLI) evaluates the level of soil contamination by heavy metals, and determines the quality of soil [Tomlinson et al. 1980]. The PLI is defined as the nth root of the multiplications of the contamination factor of each metal, and was calculated by the equation 2:

$$\text{PLI} = (\text{CF1} \times \text{CF2} \times \text{CF3} \times \cdots \times \text{CF}n)^{1/n} \quad (2)$$

where: CF is the contamination factor and *n* is the number of heavy metals to be studied, Tomlinson proposed two kinds of PLI, when PLI>1, it means that there is contamination, if PLI<1 there is no metal contamination [Tomlinson et al. 1980].

Contamination degree

The CD is a tool to determine the level of soil contamination. The sum of the contamination factor (CFi) for all metals represents the degree of contamination (CD), which is expressed as the equation 3:

$$\text{CD} = \sum_{i=1}^{n} \text{CFi} \quad (3)$$

where: CFi is the contamination factor of metal i. CD was classified into four classes: CD<8 (low degree of contamination), 8≤CD<16 (moderate degree of contamination), 16≤CD<32 (considerable degree of contamination) and CD>32 (high degree of contamination) [Hakanson, 1980].

Geoaccumulation index

The geoaccumulation index (Igeo) quantifies the degree of contaminant load accumulated by anthropogenic or geogenic inputs in the soil and was determined by means of equation 4 suggested by Muller [Muller 1969]:

$$\text{Igeo} = \log_2(Cn/1.5Bn) \quad (4)$$

where: *Cn* is the heavy metal content in soil sample n and *Bn* is the geochemical background value of metal n in the corresponding soil. The constant 1.5 is the correction factor due to natural fluctuations of metals in the environment [Kamani et al.

2017]. The Igeo was classified into seven classes: uncontaminated (Igeo≤0), uncontaminated to moderately contaminated (0<Igeo≤1), moderately contaminated (1<Igeo≤2), moderately to heavily contaminated (2<Igeo≤3), heavily contaminated (3<Igeo≤4), heavily to extremely contaminated (4<Igeo≤ 5) and extremely contaminated (Igeo>5) [Muller 1979].

Enrichment factor

The enrichment factor (EF) is used to assess metals that originate primarily from human activities or natural sources, and to determine the degree of anthropogenic influence on heavy metal contamination of soil [Cai et al. 2019]. In this study, Fe was chosen as the normalization element to determine the FE values, since it is one of the widely used reference elements [Enuneku et al. 2017; Huang et al. 2019]. It was calculated using equation 5:

$$EF = \frac{\left(\text{Metal}/_{\text{Fe}}\right)\text{sample}}{\left(\text{Metal}/_{\text{Fe}}\right)\text{background}} \quad (5)$$

According to Sutherland [2000] EF classifies it into five categories: (EF< 2), mineral depletion or no enrichment; (2≤ EF<5), moderate enrichment; (5≤ EF<20), significant enrichment; (20≤EF<40), very high enrichment; (EF>40), extremely high enrichment.

EVALUATION OF THE POTENTIAL ECOLOGICAL RISK DUE TO HEAVY METALS

Ecological risk potential index

Ecological risk potential index (RI) represents the overall ecological risk of different heavy metals in soil, assesses the likely degree of contamination by trace metals taking into account the relative toxicity of the metals in general and the short and long term response of the environment [Benson et al. 2016; Hakanson 1980]. This risk index evaluates the harmful effect of heavy metals in soils, and was calculated through equations 6 and 7:

$$RI = \sum_{i=1}^{n} \text{ERI}i \quad (6)$$

$$\text{ERi} = \sum_{i=1}^{n} Trix\ \text{CFi} \quad (7)$$

Where Ri is the index of the ecological risk potential of an individual metal i, Tri represents the toxicological response factor of metal i, which reflects the level of toxicity and sensitivity of organisms to the metal and CFi is the contamination factor of metal i. The toxicological response factors for As, Cd, Pb, Cu, Cr, Fe and Zn are 10, 30, 5, 5, 2, 1 and 1 respectively [Bhatti et al. 2018; Hakanson 1980; Li et al. 2020; Mirzaei Aminiyan et al. 2018; Mirzaei et al. 2019]. The potential ecological risk factor associated with an individual metal (ERi) was categorized as: low risk (ERi<40), moderate risk (40≤ERi<80), considerable risk (80≤ERi<160), high risk (160≤ERi<320) and very high risk (ERi≥320). The ecological risk potential index (RI) was categorized as follows: Low risk (RI<150), moderate risk (150≤RI<300), high risk (300≤RI<600) and very high risk (RI≥600) [Enuneku et al. 2017; Hakanson 1980].

Statistical analysis

The data were processed using SPSS v23 statistical software and Microsoft Office Excel 2019 to analyse descriptive statistics of heavy metal concentration in the soil and pollution indices. The normality of the data was determined according to the modified Shapiro Wilks test. The Pearson correlation analysis and principal component analysis (PCA) were used to identify possible sources of toxic metal contamination in the soils surrounding the landfills. R studio software was used for Pearson correlation analysis, PCA and figures and tables.

RESULTS AND DISCUSSIONS

Concentration of heavy metals in leachate from dumps

The concentrations of As, Cd, Cr, Cu and Zn in the leachate from the AV dump did not exceed the national environmental quality standards (EQS) [Ministry of the Environment 2017b] and the Canadian Environmental Quality Guidelines of the Canadian Council of Ministers of the Environment (CEQG) [CCME 2021]. With the exception of Pb and Fe, which exceeded the Peruvian standard values of the Ministry of the Environment (0.05 and 5 mg/L), respectively. The concentrations of As and Cr in the ED dump leachate exceeded the national and Canadian standard (Table 1). The trend of the average concentration of heavy metals in the leachates from the AV dump

Table 1. Concentration of heavy metals (mg/L) in leachates from the Agua de las Vírgenes (AV) and El Eden (ED) dumps

Dumps	As	Cd	Cr	Cu	Pb	Zn	Fe
AV	0.083	0.003	0.036	0.081	0.124	0.726	5.434
ED	0.109	0.004	0.312	0.040	0.012	0.274	3.018
EQS	0.1	0.01	0.1	0.2	0.05	2.0	5.0
CEQGs	0.1	0.005	NA	NA	0.2	NA	5.0

EQS (Environmental Quality Standard); CEQGs (Canadian Environmental Quality Guidelines, Canadian Council of Ministers of the Environment); NA (Not available).

followed the following order Fe> Zn> Pb> Cu> As> Cr> Cd, while in the ED dump the decreasing trend was Fe> Zn> Cr> As> Cu> Pb> Cd.

Concentration of heavy metals in the agricultural soils surrounding dumps

The results of heavy metal concentration, pH, electrical conductivity (EC) and matter (OM) content in the agricultural soils surrounding the AV and ED dumps are presented in Table 2. The mean pH recorded at the different soil sampling points was 7.69 and 6.93, the mean EC recorded was 0.54 and 0.47 dS/m and the mean OM content was 6.52 and 6.72% in AV and ED respectively. The agricultural soils surrounding the dumps have neutral or near neutral to slightly basic pH, an EC below 2 dS/m, which indicates that the soils are slightly saline and do not affect crops, and a high OM content. No significant correlation (p>0.05) was observed between the chemical characteristics of the soil and the concentration of heavy metals in agricultural soils in both dumps. The distribution of heavy metal contents in agricultural soils varied widely. The mean concentrations of As (25.77±10.97 and 14.35±4.57 mg/kg), Cd (1.03±0.76 and 0.28±0.29 mg/kg), Pb (112.07±31.39 and 123.01± 40.19 mg/kg), Cu (48.52±14.05 and 57.47±23. 83 mg/kg), Fe (53733±5654.25 and 36137.63±5458.10) and Zn (349.11±142.31 and 414.31±105.65) in both dumps exceeded the reference values of the natural geochemical background. The trend of the mean concentration of heavy metals in the soils of both dumps followed the following order Fe> Zn> Pb> Cu> As> Cr> Cd. Higher values of As, Cd, Cr and Fe concentration in the soils were observed in the AV dump, while in the ED dump presented high values of Pb, Cu and Zn concentration. The coefficient of variation from large to small for the AV dump was Cd> Cr> As> Zn> Cu> Pb> Fe and for ED it was Cd>Cu> Pb> As>

Table 2. Concentration of heavy metals (mg/kg), pH, EC and OM in agricultural soils surrounding dumps

Dumps	Descriptive statistics	As	Cd	Pb	Cr	Cu	Fe	Zn	pH	CE (dS/m)	MO (%)
		(mg/kg)									
Agua de Vírgenes	Mean	25.77	1.03	112.07	6.09	48.52	53733.25	349.11	7.69	0.54	6.52
	DS	10.97	0.76	31.39	3.18	14.05	5654.25	142.31	0.15	0.09	0.46
	Minimum	11.31	0.22	71.54	2.66	25.20	46067.00	185.51	7.53	0.40	5.79
	Maximum	48.29	2.81	159.81	12.91	70.45	63933.00	549.10	7.93	0.71	7.52
	CV (%)	42.56	73.78	28.00	52.21	28.95	10.52	40.76	1.95	16.66	7.05
El Edén	Mean	14.35	0.28	123.01	2.85	57.47	36137.63	414.31	6.94	0.47	6.72
	DS	4.57	0.29	40.19	0.63	23.82	5458.10	105.65	0.30	0.06	1.43
	Minimum	6.58	0.10	68.43	2.08	20.55	29800.00	240.73	6.44	0.40	5.17
	Maximum	21.20	0.98	189.02	4.00	99.92	46033.00	550.86	7.23	0.56	9.24
	CV (%)	31.84	103.57	32.67	22.11	41.44	15.10	25.50	4.32	12.76	21.28
EQS		50	1.4	70	NA	NA	NA	NA			
UCC		1.5	0.1	20	35	25	35000	71			
CSQG-CCME		12	1.4	70	64	63	NA	200			

Note: EQSS – Environmental Quality Standard; UCC – Upper Continental Crust; CSQG-CCME – Canadian Soil Quality Guidelines Canadian – Council of Ministers of the Environment; NA – not available.

Zn> Cr> Fe. No significant correlation ($p > 0.05$) was observed between the concentration of heavy metals in the leachate and the concentration of heavy metals in agricultural soils in both dumps.

The concentrations of As, Cd, Cr, Cu and Fe in the agricultural soils surrounding the AV and ED dumps did not exceed the national environmental quality standards for soils (Ministry of the Environment, 2017a) and the Canadian Council of Ministers of the Environment (CSQG-CCME) [CCME 2007]. With the exception of Pb and Zn, which exceeded the Canadian Council of Ministers of the Environment soil quality guidelines (70 and 200 mg/kg respectively); and according to the Peruvian environmental quality standard, Pb would be the only element exceeding the recommended values. However, the average concentrations of As and Cd in both dumps exceeded more than 10 times the geochemical background reference values according to the Upper Continental Crust (UCC) [Taylor and Mclennan 1995], Pb and Zn exceeded more than five times, Cu and Fe exceeded between 1.5 and 2 times the reference values, which could indicate that the soils are highly contaminated with As, Cd, Pb and Zn, due to the influence of anthropogenic activities [Krishna and Mohan 2016] and the movement of leachates from the dump [Alam et al. 2020].

The concentration values of As, Pb, Zn, Fe and Cu were higher than those reported by Alam et al. [2020] and Essien et al. [2019] in municipal waste, but similar in Cd content and lower in Cr content; these results could be attributed to the variation in the composition and decomposition of municipal solid waste, the displacement of leachate from the dump to the soil, the differential accumulation of heavy metals in a dump over a long period of time. Pb, Cd, Cu and As are toxic metal contaminants that, when present at high levels, cause metabolic disorders in most living systems, and prolonged exposure to these heavy metals can cause adverse health effects in humans [Singh et al. 2011]. High concentrations of these heavy metals can have dangerous effects on both the ecosystem and human health, where they are widespread through different forms of pollution [Borjac et al. 2019].

The Fe values found in this study exceed those reported by Agbeshie et al. [2020] and Alam et al. [2020], the high Fe content in the soil is attributed to the natural occurrence in the earth's crust, as well as to debris containing Fe [Agbeshie et al. 2020], and would be associated with the high organic matter content of organic wastes in the soil [Kabata-Pendias 2011]. However, Fe is an essential element for the functioning of the physiological activities of living organisms, but when exceeded it would be toxic.

Contamination of agricultural soils surrounding dumps

The level of contamination of the agricultural soils surrounding the dumps was categorized based on the calculations of the contamination factor (CF) (Figure 2A), the degree of contamination (CD) (Figure 2B) and the pollutant load (PLI). Soils near the AV dump have a very high level of contamination for As, Cd, Pb and Zn with 100%, 66.6%, 41.6% and 33.3% respectively, a considerable level for Pb with 58.33% of the sampling points, are moderately contaminated with respect to Cu and Fe and have a low level of contamination for Cr. The soils near the ED dump have a very high level of contamination for As and Pb with 87.5% and 50% respectively, 62.5% of the sampling points have considerable contamination for Zn, 87.5% and 75% have moderate contamination for Cd and Cu respectively, the presence of Cr and Fe are related to a low level of contamination with 100% and 62.5% of the sampling points, 58.33% of the sampling points in the soils surrounding the AV dump had a high degree of contamination, while 100% of the ED soils had a considerable degree of contamination. The contaminant load index (PLI) could indicate that the soils would be contaminated because the PLI was greater than 1.

The mean values of the geoaccumulation index (Igeo) of each metal in both dumps are presented in Figure 3(A). The Igeo results showed heterogeneous values in the soils of both dumps. In the agricultural soils of AV, 50% of the soil samples accumulated As with Igeo values between 3.1 and 4.4 classifying it as heavily contaminated ($3 < Igeo \leq 4$), 50% accumulated Cd and was classified as moderately to heavily contaminated ($2 < Igeo \leq 3$), 58.3% and 41.6% accumulated Pb and Zn classifying them as moderately contaminated ($1 < Igeo \leq 2$). The Igeo values for Cu (66.6%) and Fe (58.3%) indicated that the soils are not contaminated ($Igeo \leq 0$). While, in ED soils, 87.5% and 50% of soil sampling points accumulated As and Pb classifying them as moderately to heavily contaminated, 50% accumulated Cd and were classified as uncontaminated to

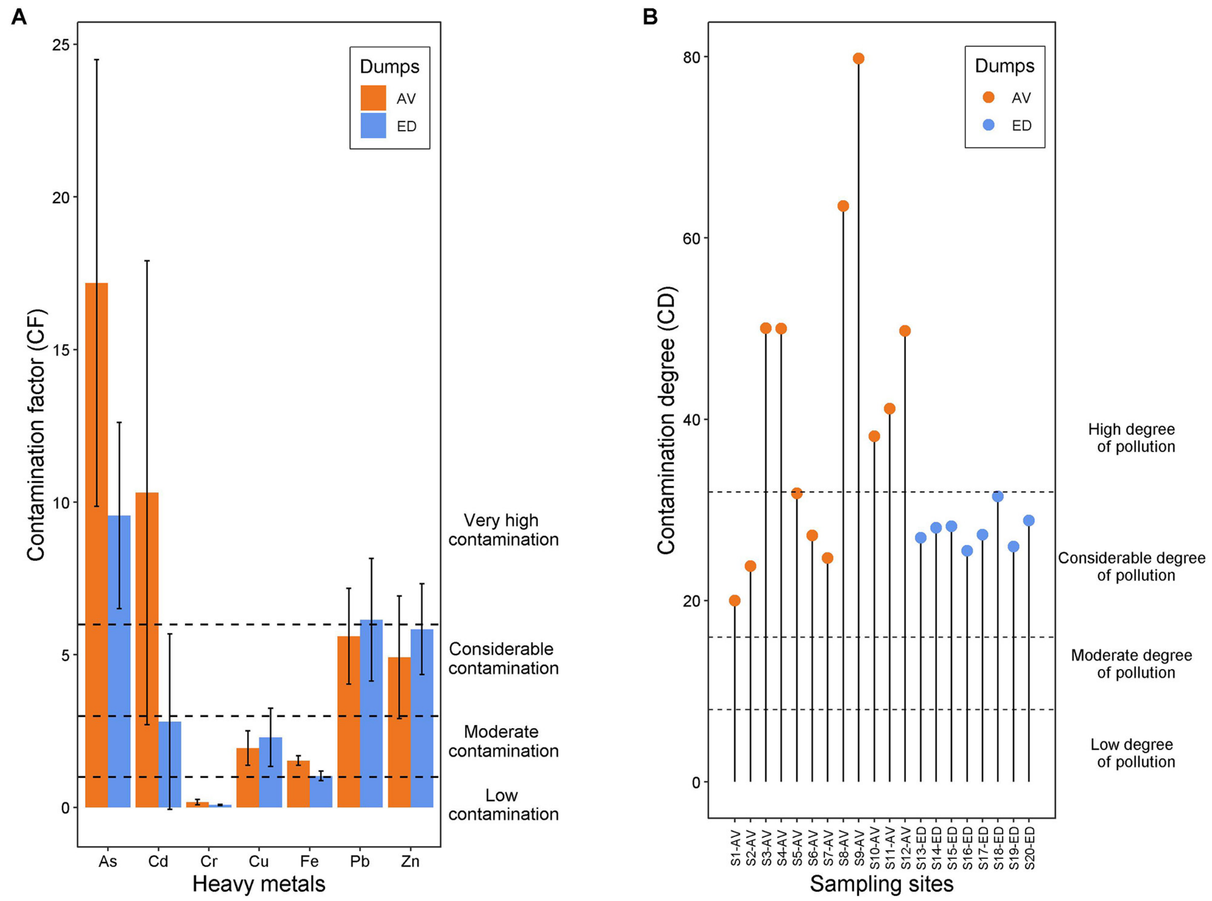

Figure 2. Contamination factor (A) and degree of contamination (B) of the soils surrounding dumps

moderately contaminated. Cr and Fe presented values of *Igeo*≤ 0 categorizing them as uncontaminated soils, meaning that these two metals did not cause contamination. The mean values of the enrichment factor (EF) of the analysed metals with respect to natural background concentrations are presented in Figure 3(B). 100% of the sampling points showed significant enrichment for As (20≤EF<40) in the agricultural soils surrounding AV and ED, 100% and 62.5% showed moderate (5≤ EF<20) and significant (5≤ EF<20) enrichment for Pb respectively. Cr presented EF< 2 values indicating that there was no enrichment with this metal in both study sites. The mean EF was higher for As in the two sampling sites, and decreased in the following order As> Cd> Pb> Zn> Cu> Cr in VA soils; and in DE soils it was As> Zn=Pb> Cu> Cd> Cr.

The agricultural soils surrounding the AV and ED dumps according to the contamination factor (CF), degree of contamination (CD), geo-accumulation index (Igeo) and enrichment factor (EF) would be heavily contaminated, and would present a very high level of contamination for As and Cd in AV, and a high level of contamination for As and Pb in ED, which would indicate that the soils present significant enrichment for As followed by Cd, Pb, Zn and Cu, suggesting that soils in general are contaminated with respect to heavy metal concentration influenced by leachates and waste composition from dump and other human activities such as wastewater irrigation and past metallurgical activity.

Ecological risk potential

The results of the individual potential ecological risks (ERI) and total ecological risk (RI) of the agricultural soils are presented in Figures 4(A) and 4(B). The mean ERI values for the soils surrounding the two dumps decreased in the order of Cd (309.25)> As (171.78)> Pb (28.02)> Cu (9.70)> Zn (4. 92)> Fe (1.54)> Cr (0.35) at the AV dump, and As (95.66)> Cd (84.33)> Pb (30.75)> Cu (11.49)> Zn (5.84)> Fe (1.03)> Cr (0.16) at the ED dump. Cd and As imply a high ecological risk condition

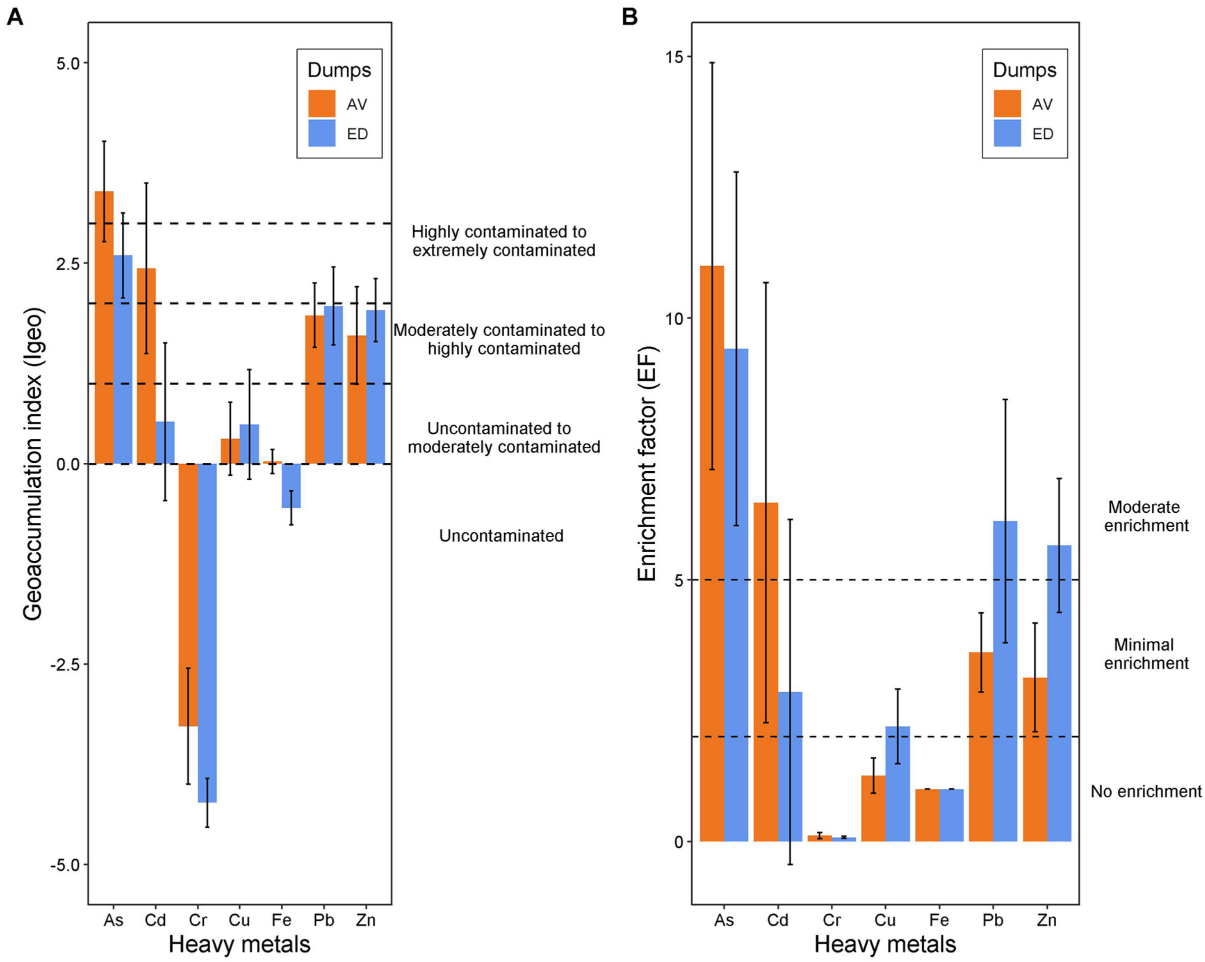

Figure 3. Geoaccumulation index (Igeo) (A) and enrichment factor (EF) (B) of the soils surrounding dumps

for soils near the AV dump and considerable risk for soils near the ED dump, while the ERI values for Pb and the rest of the selected heavy metals implied a low ecological risk. The maximum RI value was 1226.6 for the soil samples in AV and 403.5 for ED. Cadmium (59%) and arsenic (33%) were the elements with the highest contribution to the high-risk levels in the soils of AV.

Considering the different toxicities of contaminants to humans, the ecological risk assessment (ERI) was adopted to comprehensively assess the potential ecological risk (ER) resulting from heavy metals. In general, high RI values for heavy metals were identified in VA, while moderate RI values were determined in ED soils. Cd was the main contributor to heavy metal contamination in soils surrounding the VA dump, coinciding with the results from Essien et al. [2019] and Tian et al. [2017], this would be explained by the organic and inorganic components of municipal waste in the area studied, and irrigation with wastewater. In ED soils, the greatest contribution to ecological risk would correspond to As followed by Cd. Cd is statistically associated with an increased risk of cancer; this element is nephrotoxic and can cause renal failure, and also participates in the process of bone demineralization [Cwieląg-Drabek et al. 2020].

Heavy metals in the agricultural soil around the dumps would be negatively affecting the quality of the agricultural soil, and eventually these heavy elements would be washed away by surface runoff and contaminate groundwater, thus increasing the adverse implications for ecosystems and human health [Yu et al., 2020]. The risk increases when farmers use the wastewater loaded with toxic metals for irrigation of their crops, and these toxic elements can be accumulated in plants and transferred to the food chain, therefore the consumption of meat and milk from animals that feed on pastures grown around the dump would be a high risk to the health of the population. The

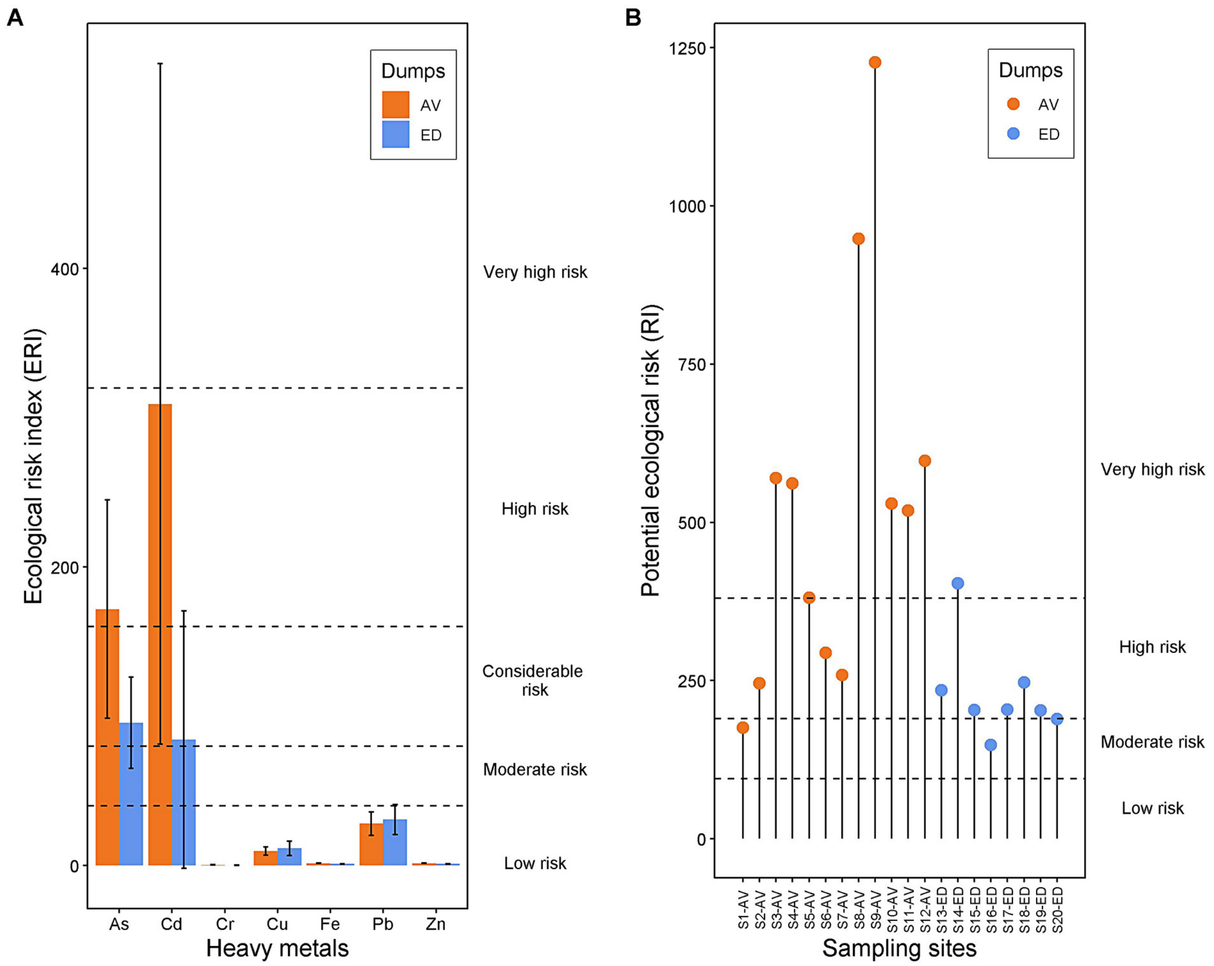

Figure 4. Individual ecological risk potential (A) and total ecological risk (B) of the soils surrounding dumps

results obtained require urgent attention due to the subsistence farming practices around the dump.

Sources of heavy metal contamination

Correlation analysis was used to measure the degree of association between heavy metal concentrations in soil [Borjac et al. 2019]. In Figure 5A, significant positive correlations ($p < 0.000$) are observed between As-Cd and Pb-Zn, which would indicate that they have similar possible sources. Likewise, high and positive correlations were observed for Fe concentration with As and Cd at a significant level ($p<0.001$), and a moderate and significant correlation between Cr-Fe and Cu-Zn ($p< 0.03$). Principal component analysis (PCA) is another multivariate analysis method that was applied for the identification of heavy metal sources in soil samples by applying varimax rotation with Kaiser normalization. The results of Kaiser-Meyer-Olkin (KMO = 0.579) and Bartlett's test of sphericity ($p < 0.00$) indicated that heavy metal concentrations in soils were suitable for PCA. According to eigenvalues greater than 1.0, principal component 1 (PC1) and principal component 2 (PC2) accounted for all heavy metals and explained 71.12% of the total variance. PC1 explained 41.38% of the total variance and is positively loaded with As (0.87), Cd (0.83), Pb (0.54), Cu (0.44), Fe (0.70) and Zn (0.69), which would indicate that they have the same sources of contamination. PC2 contains a high Cr load (0.76) as the only metal and has 29.74% as percent explained variance (Figure 5B).

The high correlation between As-Cd and Pb-Zn indicates that they probably originate from common sources and possess mutual dependence and identical behaviour during transport from the source to the impacted sites [Bastami et al. 2014], it could also suggest that the concentrations of these elements could be attributed to anthropogenic influence, such as the displacement of leachates

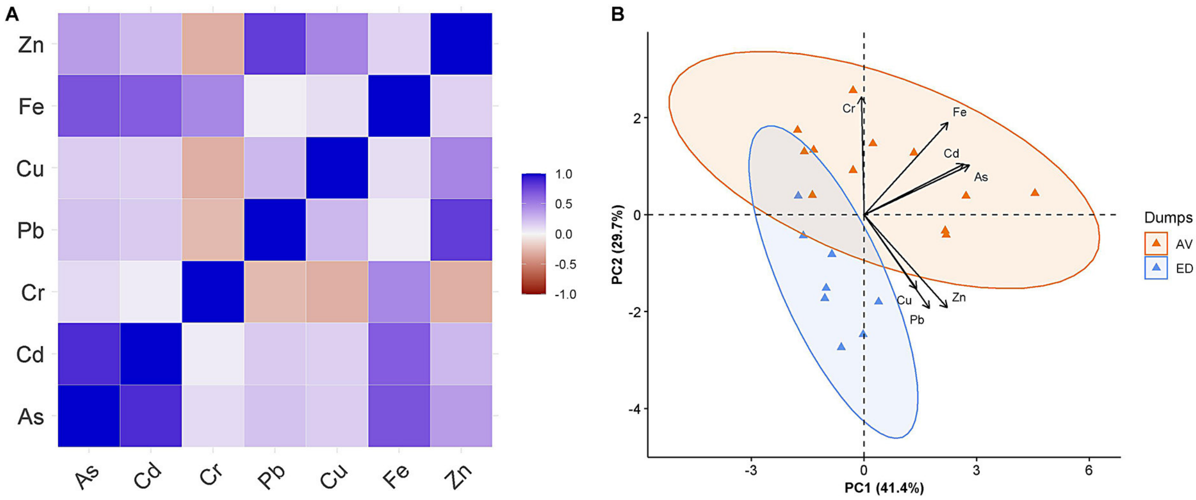

Figure 5. Pearson correlation coefficient values for heavy metal concentration in soil (A). Biplot of the principal component analysis (PCA) for heavy metals present in the soils surrounding dumps (B)

to the soil during the rainy season from dumps and other similar sources, especially at the AV dump.

According to the results of the principal component analysis (PCA), the PC1 component is dominated by As, Cd, Fe and Zn, with moderate contributions of Pb and Cu, which could be associated with anthropogenic activities and the presence of dumps. The main sources contributing to this factor are the differential composition of wastes in general (organic, inorganic, hospital, electrical and electronic, and industrial wastes), including galvanized metal wastes, obsolete metals, plastics, glass, electrical and electronic items, etc., coinciding with what was reported by Ogundele et al. [2020]. Potential sources of Pb, Cd and Zn in soils would derive from e-waste, as these in dumps release metals into the soil [Han et al. 2019; Olafisoye et al. 2013]. The sources of As and Cd would derive from industrial and agricultural waste [Essien et al. 2019], in addition, the use of domestic wastewater used by farmers to irrigate their crops in times of low water levels was detected, which would be contributing to the contribution of these two elements. On the other hand, the probable components of municipal waste that could release Cd are kitchen utensils, galvanized metals, cable sheathing [Ogundele et al., 2020]. In addition, the presence of Pb, Zn and Cu would also be related to the paralyzed metallurgical activity in Yauris, in charge of processing polymetallic minerals such as galena source of Pb, sphalerite source of Zn and chalcopyrite source of Cu, whose environmental liabilities were originated by the effluents of the different operations; furthermore, the presence of Cu would also be related to electronic waste and metal smelting [Yang et al. 2019]. The source of Fe in soils would be associated with natural sources, the high organic matter content of organic wastes in the soil and its high solubility [Kabata-Pendias 2011]. PC2 is characterized by a high Cr load in soils, the main source would come from stainless steel and chrome metal wastes, paints, varnishes, enamels, dyes, etc., according to Kabata-Pendias [2011] the main source of Cr contamination comes from dyes and leather tanning. Therefore, at toxic levels, metals are generally harmful to biological systems, including humans [Norouzirad et al. 2018].

CONCLUSIONS

The trend of heavy metal concentration in the agricultural soils surrounding the AV and ED dumps was Fe> Zn> Pb> Cu> As> Cr> Cd. The contamination characteristics (contamination factor, degree of contamination and contaminant load index) of the soil samples ranged from low contamination to very high condition. The order of the mean Igeo values was As> Cd> Pb> Zn> Cu> Fe> Cr. The soils around the dumps are enriched with As and Cd. Among the heavy metals analysed, Cd showed the highest risk index value and with a higher contribution to the overall potential risk index. The PCA explained approximately 71% of the data set, and the identified sources of heavy metals are the varied composition of waste in general (organic, inorganic, hospital, electronic, industrial) and irrigation of crops with wastewater. The findings are important and show that the agricultural soils surrounding the dumps have high levels of contamination by As, Cd and Pb. Continuous monitoring of the level of contamination of soils affected by leachates from dumps and irrigation with wastewater is suggested. On the other hand, it is urgent to determine the concentration of hazardous toxic metals in urban and domestic wastewater used to irrigate pastures and agricultural crops near the dumps.

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