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Heavy Metals in Agricultural Soils of Constanza, Jarabacoa, San José de Ocoa, Azua, Barahona and San Juan de la Maguana, Dominican Republic, 2022

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Abstract

The objective of this study was to establish a baseline for future studies that aim to determine possible contamination from human, agricultural and industrial activities. As well as the determination of the indices of environmental or geological contamination and enrichment factors of heavy metals Cr, Ni, Cu, Zn, As, Cd, Hg and Pb in agricultural soils of Constanza, Jarabacoa, Rancho Arriba and San José de Ocoa (SJO), municipalities located in the valleys of the Central mountain range of the Dominican Republic. The determination of the concentrations of heavy metals was carried out using the X-ray Fluorescence technique. Just like in Azua, San Juan de la Maguana (SJM) y Barahona in the southwest. Producer municipalities of vegetables, rice, beans, corn, melon, watermelon, tomato, banana, avocado, sugar cane and fodder for cattle. The concentration of 160 mg·kg^{-1} Probable Effect Level (PEL) of Cr according to the SQuiRTs table (USEPA-NOAA) for agricultural soils, were exceeded in 50% of the samples in SJM, SJO, Jarabacoa and Constanza; in Barahona and Azua by 20%. The PEL of 42.8 mg·kg^{-1} of the Ni was higher in more than 50% of the samples from SJM, Azua, Barahona and Jarabacoa; in SJO and Constanza at 35%. In the case of Cu with a PEL of 108 mg·kg^{-1} in SJO and Constanza, 5% of the samples exceeded its, in the other areas the concentrations were lower. Zn, As and Pb did not manage to exceed their respective PEL.

Keywords

Soil, Heavy Metals, Constanza, Azua, X-Ray Fluorescence

1. Introduction

Metals that have a density greater than the 5.0 gr·cm^{-3} are called heavy metals and those that are normally found in low concentrations in soils and the earth's crust are called trace metals (Delanoy et al., 2022). These are necessary in low concentrations as nutrients (López et al., 2010), but in high concentrations are toxic. Mercury, which is one of them, still does not know the benefits for human health, but the damage that it can cause, as well as lead if it is known (Poma, 2008). The way that humans, animals, and plants incorporate heavy metals into their body is through absorption, either through the digestive, respiratory, or contact routes (Freedman, 2018). In the case of plants, these enter through the leaves and roots, using it for their constitution and the production of fruits. These fruits are the source of food for animals and humans. High concentrations of heavy metals in the soil can be toxic for some plants (Aqeel Kamran et al., 2016) but it could also result in a particular species or organism bioaccumulates in them or their fruit (Barea-Sepúlveda et al., 2022; Vinodhini & Narayanan, 2008) and that later they are used as a source of food in the trophic chain, causing damage to health when ingested (Flores et al., 2019). Lead is a heavy metal that, according to a study, causes bone diseases and affects the brain (Turner & Lewis, 2018), decreasing brain analysis capacity, affecting the cardiovascular and nervous systems (Sanín et al., 1998). The presence of heavy metals in agricultural soils can come from native minerals in the region, use of fertilizers, insecticides, fungicides, contaminates water of domestic or industrial waste (Fonseca et al., 2011). In our case, due to the presence of Nickel and Chromium minerals in the study region (Lewis et al., 2006; Aiglsperger et al., 2015), high concentrations of these elements are to be expected in agricultural soils. With this study, we intend to establish a baseline of the heavy metals Cr, Ni, Cu, Zn, As and Pb in the agricultural soils of the Municipalities of San Juan de la Maguana, Azua, Barahona located to the southwest of the Central Mountain Range as well as Constanza, Jarabacoa and San José Ocoa located in valleys within the mountains (Butterlini, 1955). As no information was found on the concentrations of heavy metals in the agricultural soils studied, we cannot make a comparison to determine the index of environmental contamination (Al-Taani et al., 2021). In the same way, we cannot attribute the high concentrations of any heavy metal to agricultural, domestic, industrial or mining activities, since we did not know the concentrations of heavy metals in agricultural soils (Adagunodo et al., 2018), prior to this study. It can be seen that currently several countries have adopted values similar to those proposed by the Food and Agriculture Organization of the United Nations (FAO) in 1996 as a guide for the quality of agricultural soils (FAO, 2007) and for the United Nations Environment Program (UNEP) as is the Union Republic of Tanzania and others countries (UNEP, 2013). On the other hand, other countries, according to their reality, have not adopted these values as a guide (EU, 2015). If we do not have the baseline for heavy metals in our study area, it is not prudent to calculate an environmental or geological contamination index.

1.1. Geology

The Central Cordillera of Upper Cretaceous age is made up of volcanoclastic, sedimentary, volcanic, and subvolcanic rocks, and an igneous rock batholith. These rocks gave rise to the Quaternary sediments that cover the Constanza and Tireo depressions and a series of Quaternary volcanic manifestations in Valle Nuevo with rocks composed of minerals rich in iron, copper, chromium and nickel (Aiglsperger et al., 2015). Also in Jarabacoa is an Upper Jurassic and Upper Cretaceous volcano-plutonic complex (Lewis & Jimenez, 1991); and a set of Quaternary superficial terrigenous formations, as well as amphibolites characterized by flat-linear and frequently blastomylonitic textures. The San Juan valley formed during the Oligocene epoch of the Paleogene period of the Cenozoic era and the Pleistocene of the Quaternary period, by the depositions of alluvium composed of sand, sandstone, limestone, dolomite, shale fragments of igneous rocks, forming a composite soil of conglomerate fossiliferous salty clay and argillaceous sandstone, which were washed away by the Yaque del Sur river (Heubeck & Mann, 1991). In the Azua region there are Paleogene sediments with a turbiditic component; olistostromic. The Azua Basin is Neogene sedimentary of a shallow nature that evolved from the sea; an Oligocene alluvial fan system is found (Mann & Pierce, 2021). While in the Holocene, the soils of Barahona were formed by alluvial depositions of marly limestone and sandstone by the river Yaque of the Sur (Fernández-Fernández et al., 2003). In Baní carbonate and greyvakic sedimentary rocks, of Upper Cretaceous age, highly deformed, volcanic rocks to a lesser degree; south of the Central Mountain Range in San José de Ocoa, recent Quaternary alluvial fans (Dolan et al., 1991).

1.2. Study Zone

The determination of heavy metals in agricultural soils was carried out in the valleys of Constanza, San José de Ocoa, and Jarabacoa located in the Central Mountain Range; Azua, San Juan de la Maguana and Barahona located to the south of the Central Mountain Range (**Figure 1**). The soils of the Constanza Valley are used to grow vegetables, among which are carrots, potatoes, lettuce, cabbage, celery, beets, garlic, onion, radish, among others, as well as fruits such as tomato, eggplant, tayota and chili peppers, largely supplying the national market and contributing to exports. In San José de Ocoa, the same fruits and vegetables as in Constanza, and avocado, are produced, although in smaller quantities. Jarabacoa currently its agricultural production is focused more on tayota and ornamental flowers. In Azua, the production of tomato, melon, watermelon and banana predominates. Barahona's production is basically sugar cane and plantain. In San Juan de la Maguana, called the granary of the south, beans, corn, rice, bananas, sweet potatoes, cassava and grass for cattle are produced.

Figure 1. Hispaniola Island. The box corresponds to the study area of the municipalities of Azua, Constanza, San Jose de Ocoa, Jarabacoa, San Juan de la Maguana and Barahona, Dominican Republic.

2. Materials and Methods

2.1. Sampling

Soil sampling was carried out by plots, referencing the sampling site with GPS. These points were taken randomly depending on the access facilities and the distribution of the crops. The distribution was very irregular because the agricultural lands were located around the municipalities, that is, they were not continuous plots in a given area. A plastic shovel was used to collect approximately 1.0 kg of soil, both at the surface level and at a depth of 30 cm. To take the sample at a depth of 30 centimeters, a hole was made with a stainless steel tool. Then they were packed in plastic bags to be transported.

2.2. Drying

The samples were dried, first at room temperature, and then to remove all humidity in an oven at a temperature between 30 - 100 degrees Celsius; for slow drying and to decrease the possibility that some elements will evaporate along with the water.

2.3. Sample Preparation

After drying, the samples were crushed in a mortar and then sieved at 75 microns. This sieve mesh was chosen because the texture of the soils in the region is composed of a significant amount of sand. About 3 g were taken, measured on a 0.1 mg precision balance. With this mass of soil, using a press (15 tons), a tablet with a diameter of 2.0 cm was made.

2.4. Methodology and Analysis

The tablet was taken and placed in an x-ray fluorescence spectrometer (Roca, & Bayon, 1981; Fernández-Ruiz, 2010) brand Skyray EDX36000B. The excitation voltage of the X-ray emitting source was 40 kV and 600 μA (Marguí et al., 2011). The spectrometer was previously calibrated using standard sediment and soil samples certified to ISO/IEC 17025 and ISO Guide 34 by Sigma-Aldrich (TraceCert; NIST, IAEA) and BAM-CRM (SRM1944, SRM2704, SRM1646a and IAEA356). The quality of the heavy metal determinations was verified using the certified materials BCR277 (Certified Reference Materials, European-CRM); SRM2710a, SRM2711a (National Institute Standard and Technology, USA-NIST). The values of the NOAA-SQuiRTs and USEPA guide, which are for marine sediments, fresh waters and soils, coincide with the Canadian agricultural sediments and soils guide (CCME-CEQGs, 2022). For this reason we compare the Probable Effect Level (PEL) of a metal with the limits of the guide for a healthy soil in relation to this metal, they coincide in both guides. They were also compared to (UNEP, 2013; FAO, 2007) data, for common agricultural soil concentrations.

3. Results and Discussion

3.1. Azua

In Azua, 50 samples were taken, the determined concentrations of Cr(10), Ni(26), Cu(0), Zn(2), As(0) and Pb(0) were higher than the Probable Effect Level (PEL) (**Figure 2**). Regarding the (UNEP/FAO) Level, 2008 Cr(38), Ni(41), Cu(12), Zn(1), As(0) and Pb(0) exceeded it (**Figure 2**). In relation to the reference level for CCME-CEQGs soils (2022), Cr(29), Ni(26), Cu(3), Zn(2), As(0) and Pb(0) exceeded it. The maximum reference values of the (UNEP/FAO) and CCME-CEQGs (2022) of Cr and Ni are very similars, according to **Table 1**; As for Cu, Zn, As and Pb, the guide levels are double, so fewer samples exceeded these values. Most of the concentrations determined in the samples were higher than the normal values in soil of the earth's crust (**Table 1**), as can be seen in the graphs (**Figure 2**). Only Ni was found with high values according to the PEL. The concentrations of the other metals, Cr, Cu, Zn, As and Pb, were below the PEL in

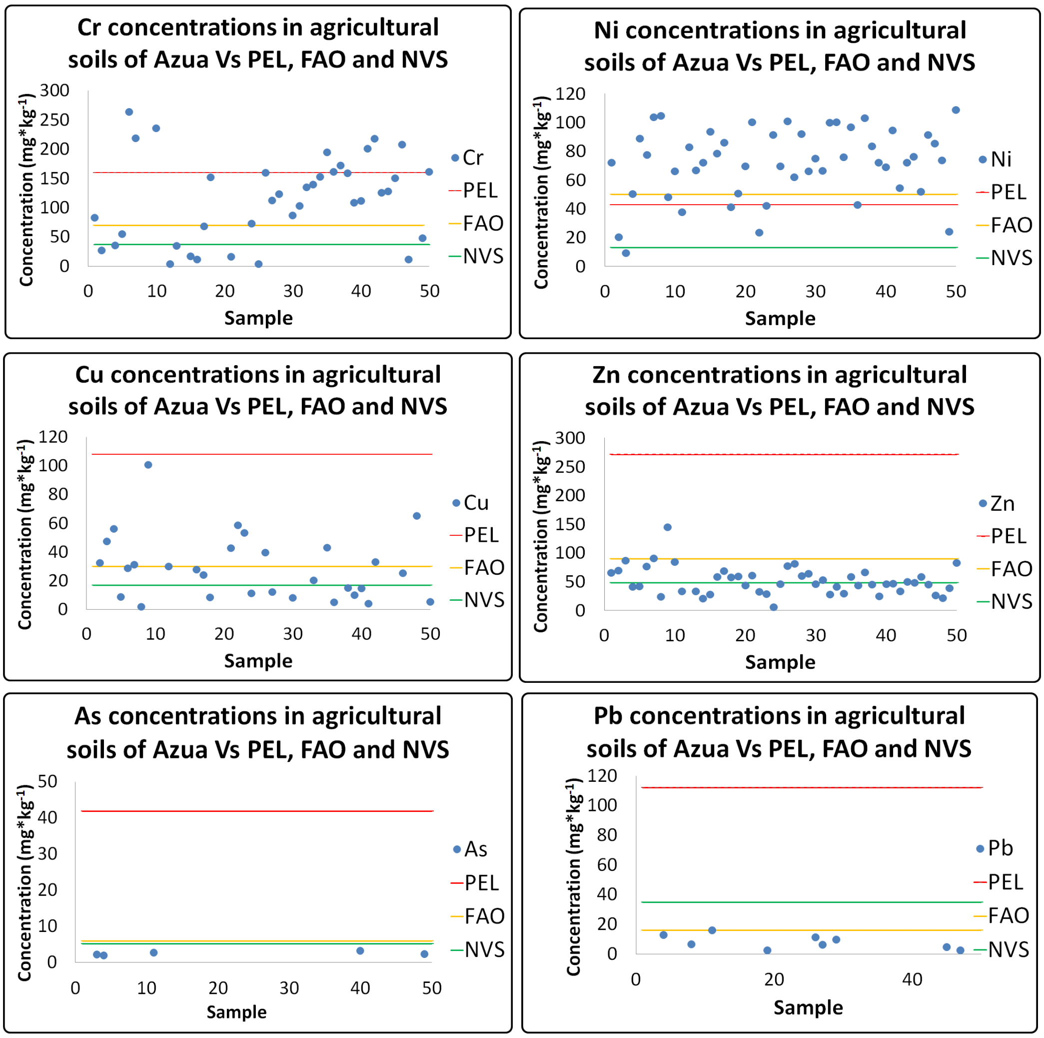

Figure 2. Scatter plot of the average values of heavy metals: Cr, Ni, Cu, Zn, As and Pb determined in the Azua region, Dominican Republic. Comparison with Probable Effect Levels (PELs), FAO reference levels for healthy soils and normal soil levels.

Table 1. Minimum, maximum and average values of heavy metals in agricultural soils of Azua, Barahona and San Juan de la Maguana, Dominican Republic.

Element	References				Azua (50)			Barahona (37)			San Juan de la Maguana (45)		
	mg·kg^{-1}												
	NVS	F[2]	S[3]	P[4]	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
Cr	37	70 - 100	64	160	0	264	89.4	9.5	300	152	81.2	629	292
Ni	13	50 - 100	45	42.8	9.4	109	71.7	12.1	133	84.4	2.1	299	87.7
Cu	17	30 - 100	63	108	2.1	101	28.9	3.7	88	39.4	13.8	94.3	64.9
Zn	48	90 - 200	250	271	5.9	145	51.2	16.7	105	60.1	69	188	130
As	5.2	6 - 5	12	41.2	2	3.3	2.5	2	3.2	2.4	3.8	7	4.8
Pb	16	35 - 60	70	112	2.7	16.1	8.1	3.8	19.9	9.8	17.6	61.2	36.3

NVS Normal values in soils (Freedman, 2018); F[2] Maximum levels according to the guide for agricultural land (FAO, 2007; UNEP, 2013); S[3] maximum levels for healthy agricultural soil according to Canadian guidance (CCME-CEQGs, 2022); P[4] Probable Effect Levels (PELs) to Human and Animal Health (NOAA-USEPA SQuiRTs).

most cases and the UNEP/FAO threshold. The sampled areas of the municipality of Azua are located near the Caribbean Sea. The soils have been formed by sediments that have been dragged from the southern side of the central mountain range (Díaz De Neira & Solé Pont, 2002), so it is expected that the mineralogical components of the soils are the same or similar to those found in the formations of the central mountain range. There is little mineralogical information about this sampled area, so the data obtained, we consider, are an important contribution in relation to its composition when we compare it with the concentrations found in agricultural soils. The high concentrations of Ni suggest minerals rich in this metal, also Cr and Cu but to a lesser extent if we compare it with the PEL and UNEP/FAO levels.

3.2. Barahona

In Barahona, 37 samples of agricultural soils were taken, the determined concentrations of the heavy metals Cr(17), Ni(35), Cu(0), Zn(0), As(0) and Pb(0) were higher than the PEL. On the other hand, Cr(33), Ni(34), Cu(25), Zn(2), As(0) and Pb(0) exceeded the (UNEP/FAO) reference level. Concentrations of Cr(35), Ni(35), Cu(6), Zn(2), As(0) and Pb(0) were higher than the CEQGs reference level (**Figure 3**). The samples in Barahona contained high concentrations according to the PEL and UNEP/FAO levels of Cr, Ni, and Cu. This suggests soils composed of minerals rich in these metals. There was no information on minerals that are composed of these heavy metals in the region.

3.3. San Juan of the Maguana

In SJM 45 samples were taken, the determined concentrations of Cr(40), Ni(34), Cu(0), Zn(0), As(0) and Pb(0) were higher than the PEL (**Figure 4**). A total of Cr(45), Ni(34), Cu(44), Zn(41), As(0) and Pb(24) passed the (UNEP/FAO) reference level (**Figure 4**). The majority of the samples in San Juan de la Maguana had concentrations of Cr, Ni higher than the PEL level while for the UNEP/FAO threshold level Cr, Ni, Cu, Zn and Pb had higher concentrations. The soils of this municipality come from the southwest side of the central mountain range and borders Constanza where minerals composed of Ni, Cr, Cu, Zn and Pb are located, so their high concentrations could have their origin in the mountains of the Central Cordillera in the Duarte formation, which is contiguous to the Tireo formation found in Constanza and Jarabacoa (Lewis & Jimenez, 1991). Since we do not have a baseline, we cannot affirm that in this municipality many samples have been enriched with heavy metals, but we suspect that the use of agricultural inputs and domestic waste could have been enrichment factors.

3.4. Constanza

Of the 48 samples taken in Constanza the concentrations of Cr (26), Ni (20), Cu (9), Zn (0), As (0) and Pb (0) exceeded the PEL. From the (UNEP/FAO) reference level, Cr(45), Ni(20), Cu(35), Zn(36), As(0) and Pb(17) were higher (**Table 2**).

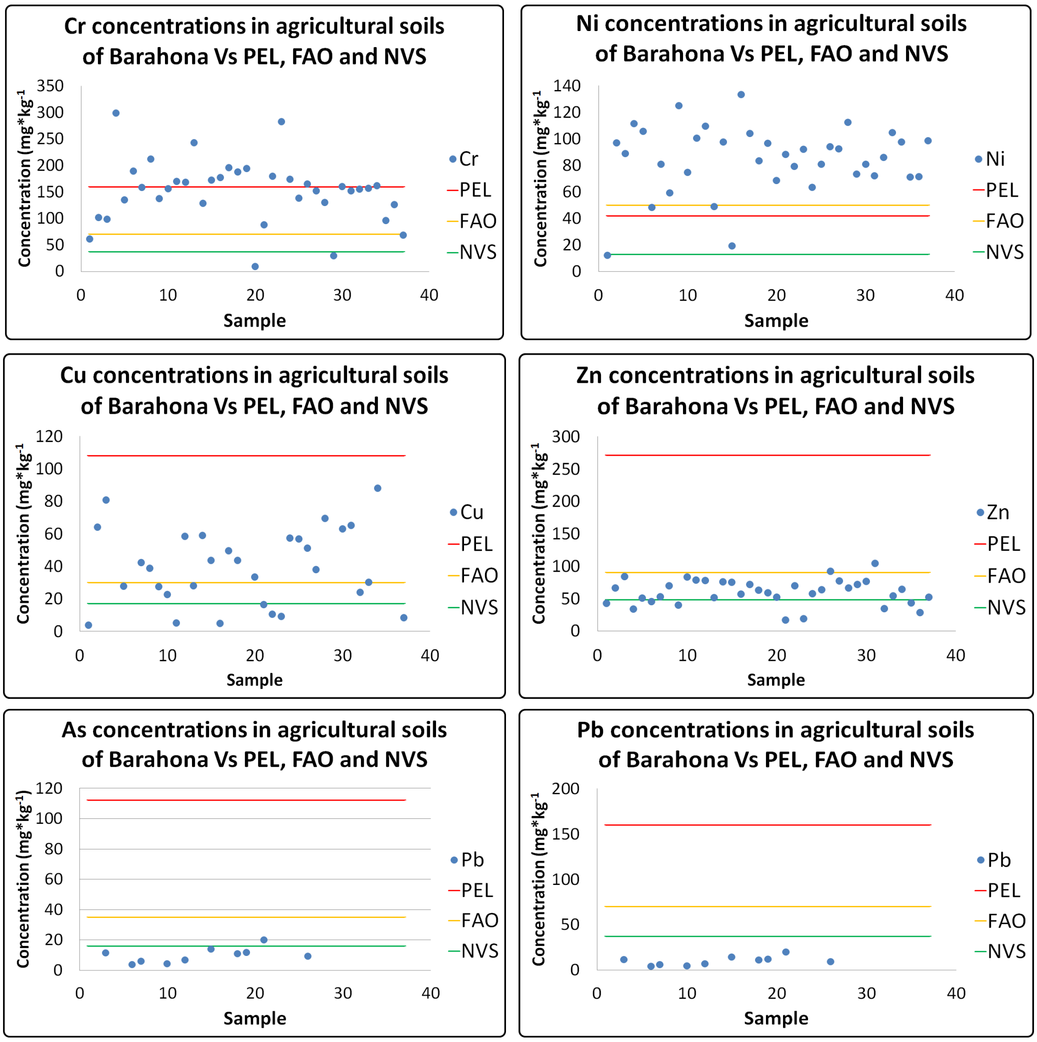

Figure 3. Scatter plot of the average values of heavy metals: Cr, Ni, Cu, Zn, As and Pb determined in the Barahona region, Dominican Republic. Comparison with Probable Effect Levels (PELs), FAO reference levels for healthy soils and normal soil levels.

Table 2. Minimum, maximum and average values of heavy metals in agricultural soils of San José de Ocoa, Jarabacoa and Constanza, Dominican Republic.

Element	References				San José de Ocoa (39)			Jarabacoa (21)			Constanza (48)		
	mg·kg^{-1}												
	NVS	F²	S³	P⁴	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
Cr	37	70 - 100	64	160	3.6	881	337	9.9	549	257	34.8	888.1	249
Ni	13	50 - 100	45	42.8	2.9	899	218	10.3	356	149	15.1	589.3	115
Cu	17	30 - 100	63	108	7.1	228	57	4.2	60.3	23.2	8.5	296.9	84.1
Zn	48	90 - 200	250	271	3	260	95.5	2.5	249	97.7	3.8	244.2	113
As	5.2	6 - 5	12	41.2	2.7	6	4.1	2.8	8	5.9	2.4	6.8	4
Pb	16	35 - 60	70	112	2.1	64.4	36	3.2	76	35.4	2.7	132	39.8

NVS Normal values in soils (Freedman, 2018); F² Maximum levels according to the guide for agricultural land (FAO, 2007; UNEP, 2013); S³ maximum levels for healthy agricultural soil according to Canadian guidance (CCME-CEQGs, 2022); P⁴ Probable Effect Levels (PELs) to Human and Animal Health (NOAA-USEPA SQuiRTs).

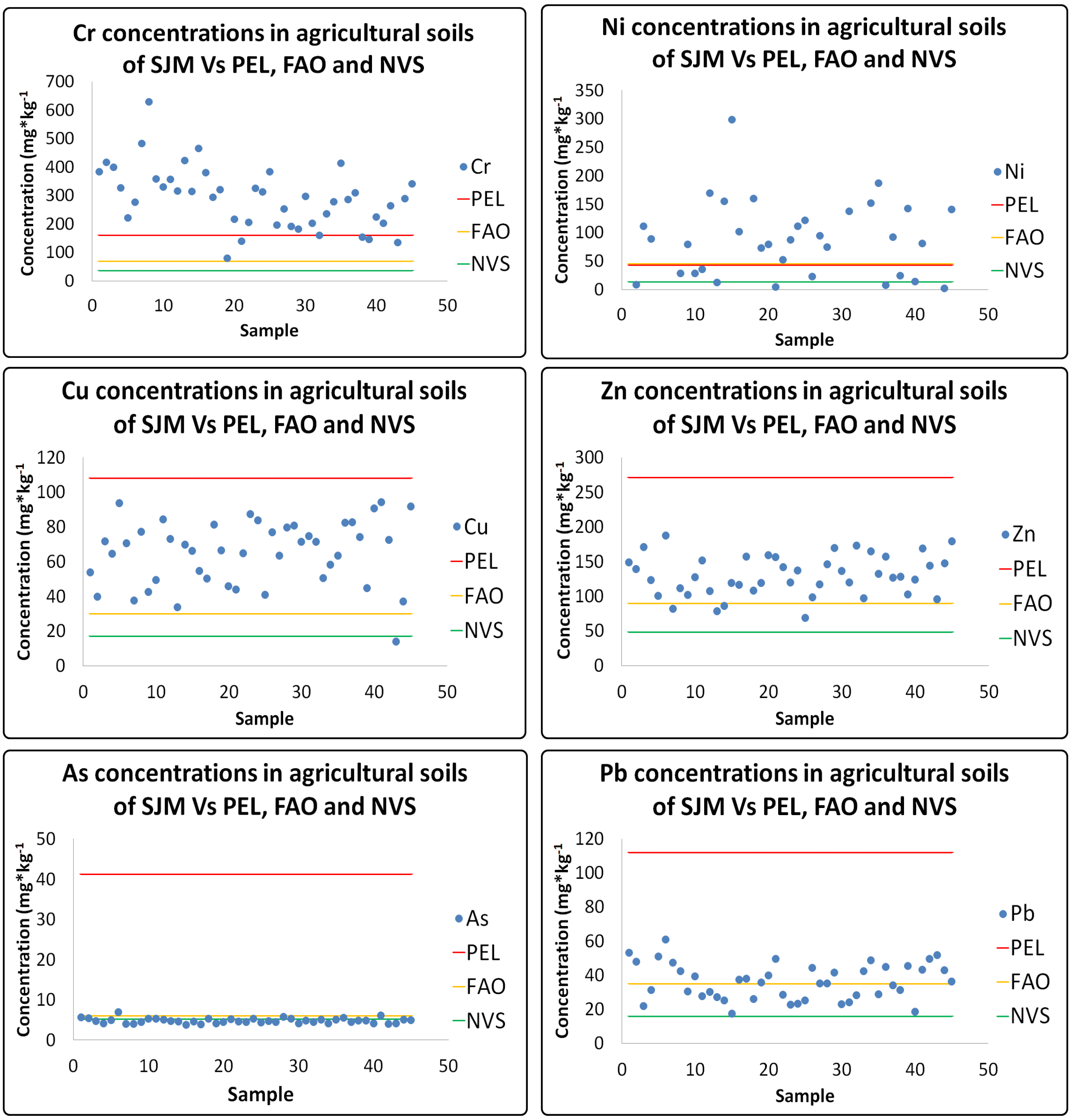

Figure 4. Scatter plot of the average values of heavy metals: Cr, Ni, Cu, Zn, As and Pb determined in the San Juan de la Maguana region, Dominican Republic. Comparison with Probable Effect Levels (PELs), FAO reference levels for healthy soils and normal levels.

The elevated concentrations of heavy metals determined in the agricultural soils of Constanza when compared with the PEL and UNEP/FAO are possibly due to the presence of minerals rich in Cr, Ni, Zn, Cu and Pb from the Tireo geological formation (Aiglsperger et al., 2015), especially Cr and Ni; we cannot blame agricultural, domestic and industrial activities since there are reasons why this is due to the mineralogy of the region (Figure 5). The minerals found in the Tireo formation are chalcopyrite, sphalerite, pyrite, chromite and laterite. Sphalerite is a mineral composed of Zn, Cu and Pb; laterite rich in Ni and Fe; chromite in Cr. Reasons why these soils have high concentrations of heavy metals such as Ni, Cu, Zn and Pb.

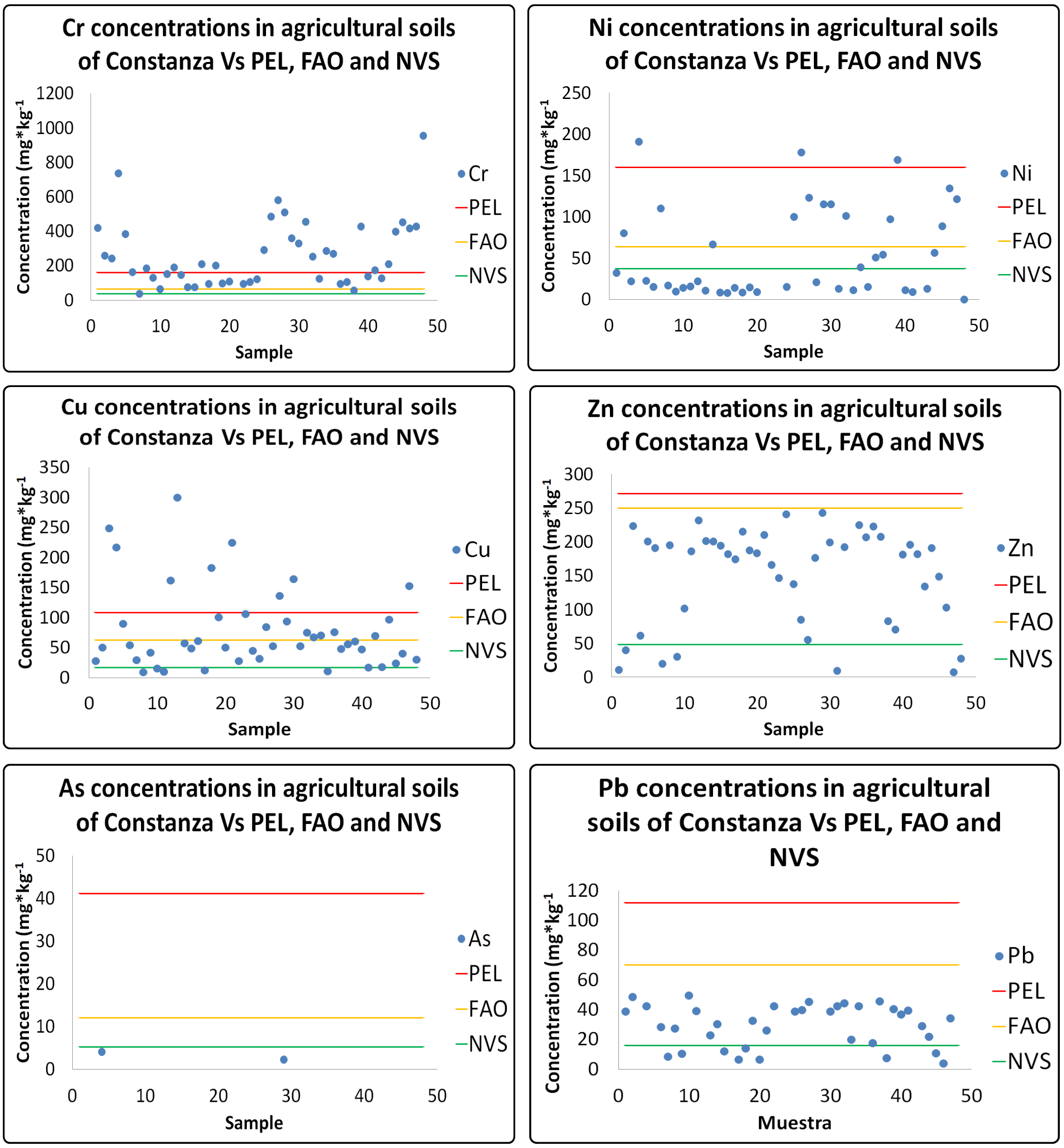

Figure 5. Scatter plot of the average values of heavy metals: Cr, Ni, Cu, Zn, As and Pb determined in the Constanza region, Dominican Republic. Comparison with Probable Effect Levels (PELs), FAO reference levels for healthy soils, and normal soil levels.

3.5. Jarabacoa

Much of the land used for agriculture in this municipality has been used for the construction of infrastructure. In the soils still suitable for agriculture, some 21 samples were taken in which the concentrations of Cr(12) and Ni(10) were higher than the PEL. They exceeded the reference level of the (UNEP/FAO) Cr(16), Ni(14), Cu(7), Zn(8), Pb(10) and As(2) (Figure 6). The same occurs in Jarabacoa as in Constanza in comparison with the PEL and UNEP/FAO (Table 2), as well as the presence of minerals rich in Cr, Ni, Zn, Cu and Pb from the Tireo geological formation (Aiglsperger et al., 2015), very marked Cr and Ni. The

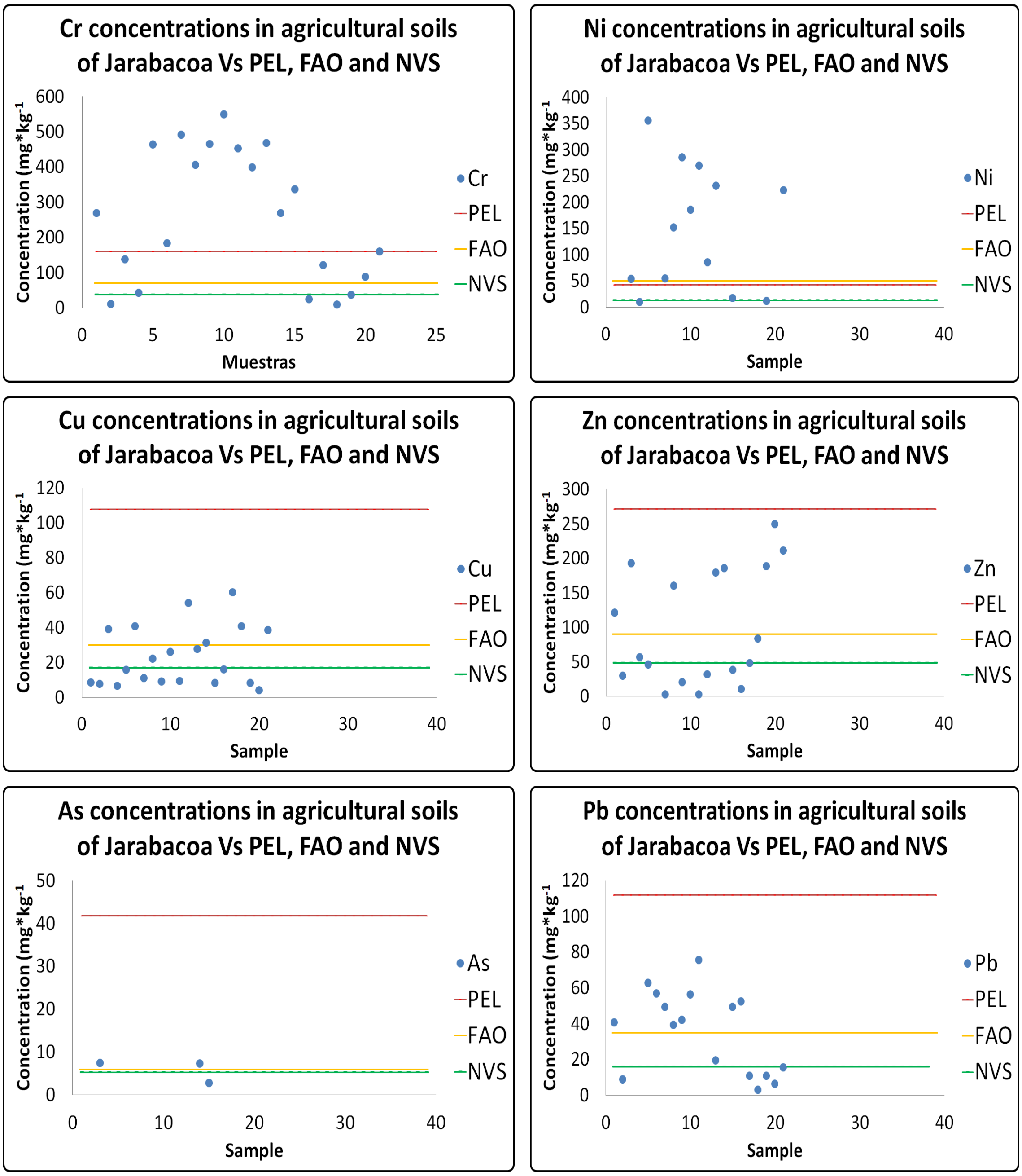

Figure 6. Scatter plot of the average values of heavy metals: Cr, Ni, Cu, Zn, As and Pb determined in the Jarabacoa region, Dominican Republic. Comparison with Probable Effect Levels (PELs), FAO reference levels for healthy soils, and normal soil levels.

geology as well as the mineralogy throughout the Central mountain range is similar, so it is expected that the soils will be very consistent in relation to the presence of heavy metals and their concentrations.

3.6. San Jose de Ocoa

Of the 39 samples of agricultural soils collected in San José de Ocoa, they presented Cr (27), Ni (23) and Cu (5) values that exceeded their respective PEL. The

(UNEP/FAO) reference level was exceeded by Cr (36), Ni (27), Cu (32), Zn (0), As (0) and Pb (15) (**Figure 7**). In both San José de Ocoa and Jarabacoa and Constanza, the high values compared to the PEL and UNEP/FAO are also due to the presence of minerals rich in Cr (**Table 2**), Ni, Zn, Cu and Pb from the Tireo geological formation (Aiglsperger et al., 2015), Cr and Ni are very marked; no to agricultural, domestic and industrial activities. There are three valleys located in the central mountain range with similar mineral soils from the Tireo geological formation.

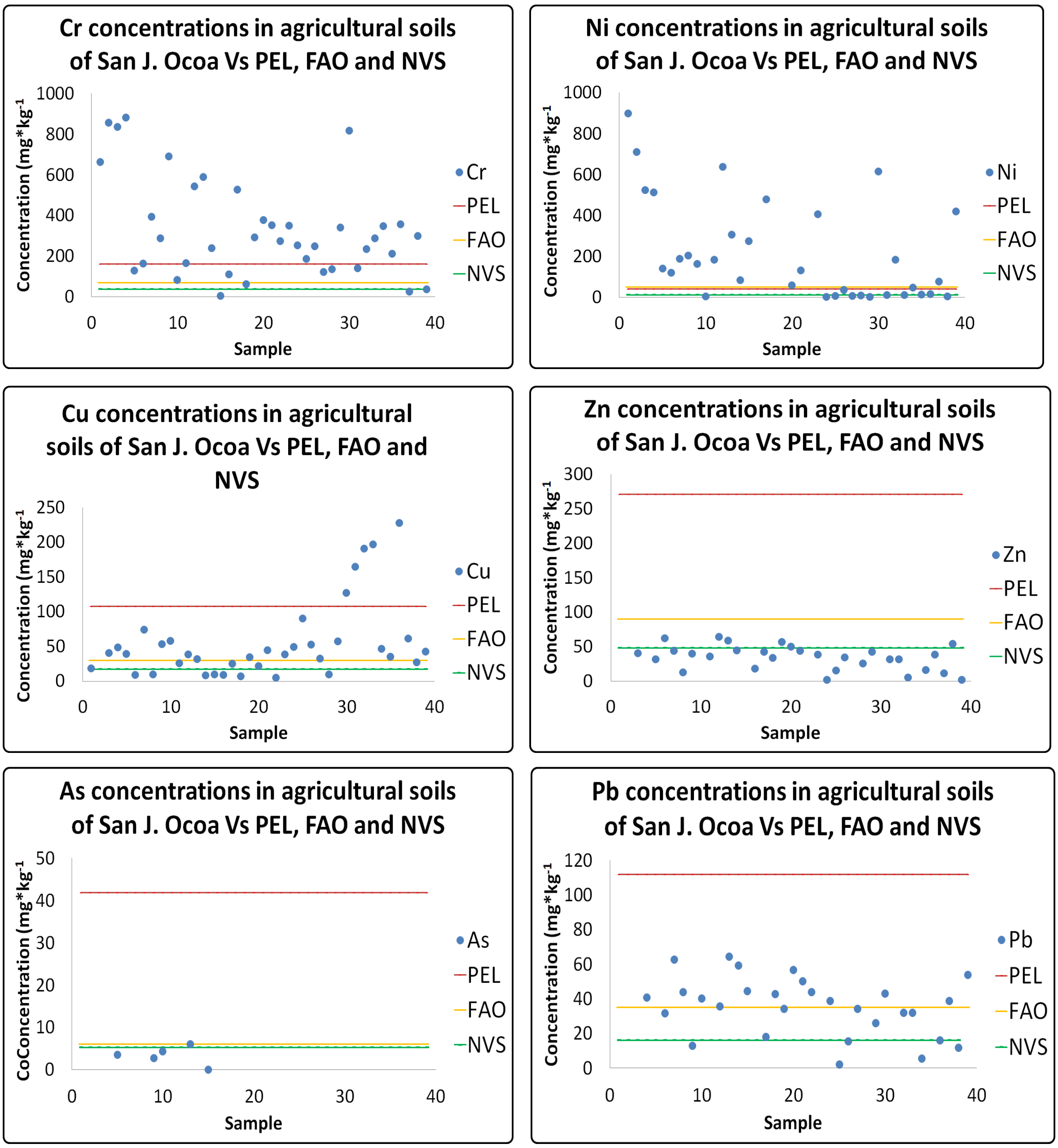

Figure 7. Scatter plot of the average values of heavy metals: Cr, Ni, Cu, Zn, As and Pb determined in the region of San José de Ocoa, Dominican Republic. Comparison with Probable Effect Levels (PELs), FAO reference levels for a healthy soil, and normal soil level.

The Cr concentrations in the soils of the Constanza, Jarabacoa and San José de Ocoa (SJO) valleys located in the Central Cordillera are generally above the probable effect level of 160 mg·kg^{-1} according to the table SQuiRTs, CEQGs cut-off level of 64 mg·kg^{-1} and (UNEP/FAO) reference level of 70 mg·kg^{-1}. In average value in SJO, Jarabacoa and Constanza were 337, 257 and 249 mg·kg^{-1} value, respectively. In the case of Ni, the same occurred in SJO, Jarabacoa and Constanza with 218, 149 and 115 mg·kg^{-1} respectively. The averages of the concentrations in the metals Cu, Zn, As and Pb were below their respective PEL. While the concentrations of Cu on average in SJO and Constanza were higher than the (UNEP/FAO) reference level. Also Zn values were similar to their (UNEP/FAO) reference level in all three regions. The As was found both below the PEL, CEQGs and FAO. Pb concentrations were similar to the (UNEP/FAO) value and below CEQGs. In the agricultural regions of the Southwest of Azua, Barahona and San Juan de la Maguana (SJM), in the latter the Cr on average was found above the PEL. While Ni concentrations, on average, were higher in these three regions than the PEL of 42.8 mg·kg^{-1} value. The Cu concentrations in Azua and Barahona were below the PEL of 108 mg·kg^{-1} and the CEQGs of 63 mg·kg^{-1} value; in SJM the Cu was similar to the CEQGs. The (UNEP/FAO) reference level of 30 mg·kg^{-1} value for Cu was exceeded in SJM and Barahona. The concentrations of Zn, As and Pb are lower than the PEL of 271 mg·kg^{-1} and the CEQGs of 250 mg·kg^{-1}. Zn in SJM exceeds the (UNEP/FAO) reference level, in Azua and Barahona they were below. In SJM the Pb concentrations are similar to the (UNEP/FAO) reference level and in Azua and Barahona they are below. Cr and Ni are the heavy metals that need more attention in the agricultural regions of the study.

4. Conclusion

The abundances of heavy metals Cr, Ni, Cu, Zn and Pb, especially Cr and Ni in the agricultural soils of the valleys studied come from the minerals of the rock formations of Tireo and Duarte, which through erosion and sedimentation processes have given rise to agricultural soils in the valleys of the Central Cordillera and Southwestern Dominican Republic. With this study we establish a baseline for future contamination and bioavailability studies. The crops currently being grown have not been reported to have suffered damage from the presence of these heavy metals in agricultural soils, nor have they bio accumulated large quantities of these. It is likely that these metals are not bioavailable in large quantities, so we suggest that bioavailability determinations be carried out to obtain information on the health of the soils in the region. The agricultural soils of San Juan de la Maguana, due to their abundance of the heavy metals studied and the possibility that these are being enriched by agricultural, domestic and industrial activities, should be given attention. The soils of Jarabacoa should be considered in the same way.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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