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Heavy Metal Pollution Assessment in the Agricultural Soils of Bonao, Dominican Republic

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Abstract: Heavy metal content in agricultural soils potentially impacts the food chain and human health. The present study assessed the levels of heavy metals in topsoil samples collected within an agricultural region situated in Bonao, Dominican Republic. The Energy-Dispersive X-ray Fluorescence (EDXRF) technique was utilized to measure the concentrations of iron (Fe), manganese (Mn), chromium (Cr), copper (Cu), nickel (Ni), zinc (Zn), lead (Pb), and arsenic (As) in the samples. The assessment of soil pollution status and potential ecological risk (RI) involved the utilization of various soil pollution indices, such as the single pollution index (PI), integrated pollution index (IPI), and enrichment factor (EF). The average total concentrations of Fe, Mn, Cr, Cu, Ni, Zn, Pb, and As were 103,000, 2000, 347, 36, 92, 32, 9.6, and 4.2 mg·kg^{-1}, respectively. The results showed that the Mn, Ni, Cu, and As levels exceeded the Food and Agriculture Organization (FAO)'s recommended levels for healthy agricultural soils. The distribution pattern of each individual metal was different, indicating they had different sources of origin. The average pollution indices indicated low-to-moderate pollution, and the potential ecological risk obtained was low. This study emphasizes the need for soil management practices to mitigate heavy metal contamination for food safety and environmental health.

Keywords: soil pollution; heavy metals; Bonao; Dominican Republic

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1. Introduction

Soil is an essential resource for human survival and a fundamental component of terrestrial ecosystems. However, industrialization, rapid urbanization, and the excessive use of chemicals in soil have led to its contamination by heavy metals, which has turned into a significant environmental problem [1,2]. Heavy metals in agricultural soils and their interactions with rivers, plants, and food can have harmful effects on the environment and society by contaminating the food chain. Heavy metal exposure is also linked to serious health problems for humans and animals [3–5]. Heavy metal contamination can change soil composition, structure, and function; inhibit plant growth; and cause a decrease in crop yields [6–8]. These elements cannot be degraded or destroyed. Still, they can be dissolved by physical and chemical agents, leached, and distributed in ecosystems until they are incorporated into the trophic chain, mainly those that come from different anthropogenic sources, such as agricultural activities, mining, and wastewater [9–12]. Heavy metals have a direct correlation with soil pollution risks, plant toxicity, and adverse effects on natural resources and the environment [13,14]. These risks are based on factors such as the specific toxicity, persistence, bioaccumulation, and non-biodegradability of the metals [3,5,15,16].

Several research studies have been carried out in the Dominican Republic to investigate the heavy metal pollution of agricultural soils. Delanoy et al. found high levels of Ni and Cr in agricultural soils dedicated to rice and banana cultivation in the northwest and central regions [17,18]. Hernández et al. [19] and Pastor et al. [20] also reported high levels of heavy metals in various agroecosystems in the Pedernales province in the southern region of the country. However, there have been no systematic studies on the status of heavy metal pollution in agricultural soils in the Dominican Republic. This study aims to provide a baseline understanding of the heavy metal pollution status in cultivated soils for rice (*Oryza sativa* L.), which is one of the main agricultural crops in the country, with a total estimated consumption of 620,000 MT and a per-capita consumption of approximately 50 kg of rice per year [21]. The objective of this study is to determine the heavy metal concentration, spatial distribution, and pollution status of soils dedicated to rice production in Bonao, Dominican Republic. This information will help promote sustainable agriculture, establish soil management strategies for healthier food production, and minimize health and environmental risks.

2. Materials and Methods

2.1. Study Area

Bonao, situated in the Monseñor Noüel province of the central region of the Dominican Republic, was the studied area and is located between latitude 18°91′ north and longitude 70°39′ west, with a total geographical area of 5.4 Km^2. The weather is tropical humid, with an average annual temperature of 25.6 °C and an average annual rainfall of 2167 mm [22]. In this area, the main soil type is clayey derived from sedimentary deposits of the Yuna River, which are characterized by the presence of iron, nickel, and cobalt [17]. This area has been dedicated to intensive rice cultivation for over three decades and it is near to an active ferronickel mine and to a high-traffic highway. The area is irrigated with superficial water supplied from the Yuna River, the second most important river in the Dominican Republic. Figure 1 shows the studied area and soil collection sites.

2.2. Soil Sample Collection and Processing

A total of 30 surface soil samples at a depth of 0–30 cm were collected from rice-cultivated land. The soil sampling sites were randomly selected, ensuring an average distance of approximately 300 m between each site [23]. Three bottom soil samples were collected in the positions BG01-BG03 at 50 cm deep to determine the local background (Figure 1). In each position, a soil sample of about 1 kg was taken and placed in previously labeled plastic bags. A Global Positioning System (GPS) navigator was used to record the longitude and latitude of the sampling points.

In the laboratory, soil samples were air-dried for several days at ambient temperature (25–30 °C) and cleaned to remove any stones or plant roots. To analyze the soil properties, the samples were crushed using an agate mortar, passed through 2 mm nylon sieves, and stored in polyethylene jars at room temperature. For heavy metal analysis, the soil samples were further dried at 105 °C for 24 h, crushed using an agate mortar, passed through 0.2 mm nylon sieves, and stored in polyethylene jars at room temperature. Three grams of each soil sample was pressed at 15 tons into a pellet of 2.0 cm diameter and 2.0–3.0 mm height.

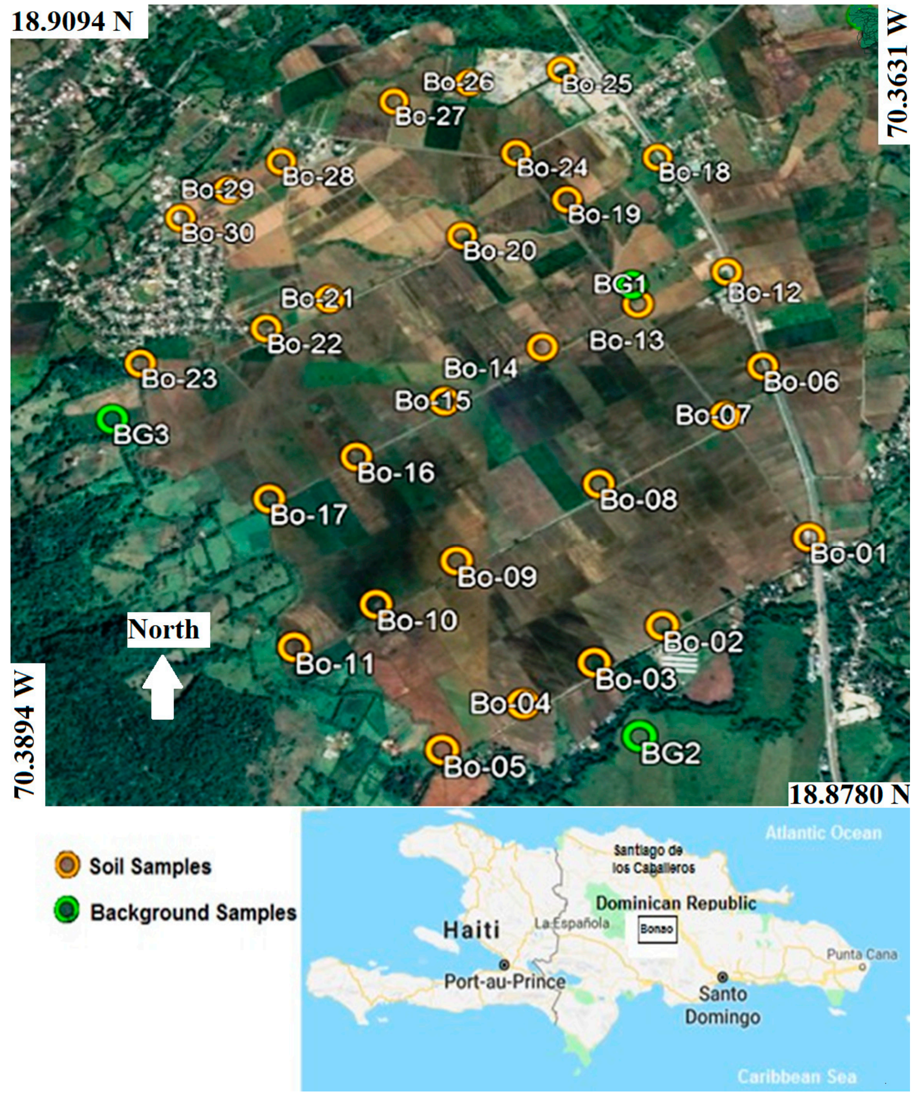

Figure 1. The locations of the soil sampling sites in Bonao, Dominican Republic.

2.3. Physicochemical Analysis in Soils

The pH was determined in a soil/water ratio of 1:2 using a glass electrode pH meter (Apera Instrument, LLC, Columbus, OH, USA, model MP511). The electrical conductivity (EC) was measured with a portable meter (Apera Instrument, LLC, model EC700). The organic matter (OM) content was determined following the Walkley and Black method [24]. The proportions of clay, silt, and sand particles in the soil were measured to determine soil texture, employing the hydrometer method [25]. The cation exchange capacity (CEC) of the mineral soils was measured in an extractable solution with 1 M of NH_4-acetate [26].

The total concentrations of Fe, Mn, Cr, Cu, Ni, Zn, Pb, and As were measured using the Energy-Dispersive X-ray Fluorescence (EDXRF) technique. This technique consists of a rapid, non-destructive, multi-elemental analysis with sensitivity in the range of a few ppm to percent [27]. EDXRF has been used to determine heavy metal concentrations in a variety of environmental samples [1,17,28,29]. The instrument used was a Skyray model EDXR-3600B X-ray spectrophotometer with a silicon detector at 45 degrees from the X-ray source (Ag). The excitation voltage of the X-ray-emitting source was 40 kV and 600 μA. A calibration curve was prepared for each element using standard reference materials (SRMs). The curves obtained were evaluated using their determination coefficients (r^2), which were between 0.990 and 0.999. The Skyray program (Version RoHS4_1.147_110524_R, 2009, 20110524_R, Kunshan, China) provided by the manufacturer (Dallas, TX, 75238 USA) was used for processing the spectra to obtain the intensity of characteristic radiation for each element in the soil samples.

2.4. Data Analysis and Spatial Distribution

Descriptive statistics (minimum, maximum, mean, and standard deviation) were computed to represent the obtained dataset using the software package Excel 2023 (Microsoft, Redmond, WA, USA). A Pearson correlation analysis was run to determine the relationships between heavy metals and the examined soil properties. The heavy metal concentrations in paddy soils were mapped using ordinary Kriging in Grapher 8 for spatial distribution analysis.

2.5. Quality Control and Quality Assurance

To ensure the analysis quality, two soil standard reference materials (SRMs) were used to compare the certified value: SRM 2711a (Montana II soil, moderately elevated trace element concentrations) and SRM 2710a (Montana I soil, highly elevated trace element concentrations), from the National Institute of Standards & Technology, US Department of Commerce. The accuracy of the quantification was verified using the McFarrel criterion [30], in which an SR parameter is defined as:

$$SR = \frac{|C_x - C_w| + 2\sigma}{C_w} 100\% \tag{1}$$

where C_X is the experimentally determined concentration, C_W is the reported concentration for standard reference materials, and σ is the standard deviation of C_X.

Using this criterion, the comparison between the certified value and analytical data obtained via the proposed methods is classified into three categories: SR $\leq$ 25% is considered excellent; 25% $\leq$ SR $\leq$ 50% is considered acceptable; and for SR values > 50%, the method is considered unacceptable for quantification. All metals (Fe, Mn, Cr, Cu, Ni, Zn, Pb, and As) measured via EDXRF were "excellent" (SR $\leq$ 25%). The results of the analysis of five replicas of the SRM 2711a, the detection limits, and the quantification limits obtained via EDXRF can be found in Table S1.

2.6. Pollution Indices and Potential Ecological Risk Index

The soil pollution status and potential ecological risk (RI) were determined via different pollution indices: the single pollution index (PI), integrated pollution index (IPI), and enrichment factor (EF).

2.6.1. Single Pollution Index (PI)

The single pollution index (PI) for each targeted heavy metal was calculated [31] as:

$$PI = C_i/C_b \tag{2}$$

where C_i is the concentration of the metal i in soil samples, while C_b is the average concentration of the metal in the local background. PI values are classified into three levels: low pollution (PI < 1), moderate pollution (1 $\leq$ PI < 3), and strong pollution (3 $\leq$ PI).

2.6.2. Integrated Pollution Index (IPI)

The integrated pollution index (IPI) is defined as the mean value of the PIs of the studied metals

$$IPI = \frac{1}{N}\sum_{i=1}^{N} PI_i \tag{3}$$

where N is the number of determined metals, based on the IPI value, and following the classification proposed by Wei and Yang [32], soils must be classified as slightly polluted soil (IPI $\leq$ 1); moderately polluted soil (1 < IPI $\leq$ 2); highly polluted soil (2 < IPI $\leq$ 5)); and extremely polluted soil (IPI > 5).

2.6.3. Enrichment Factor (EF)

The enrichment factor can be employed to distinguish between metals that come from human activities or natural sources [33]. Elements such as Fe, Mn, Al, Ti, and Sr are usually employed as reference elements in calculating the EF values [15,34,35]. In this study, Fe was taken as the reference element due to its relatively high concentration and stability in the Earth's crust. It was calculated as follows [36].

$$\mathrm{EF} = \frac{\left(\frac{M}{Fe}\right)_{sample}}{\left(\frac{M}{Fe}\right)_{background}} \tag{4}$$

where $(\mathrm{M/Fe})_{\mathrm{sample}}$ is the ratio of metal and Fe concentrations in the sample, and $(\mathrm{M/Fe})_{\mathrm{background}}$ is the ratio of metal and Fe concentrations in the background sample. EF values can be categorized into six levels: no enrichment ($0 < \mathrm{EF} < 1$), minimal enrichment ($\mathrm{EF} < 2$), moderate enrichment ($2 < \mathrm{EF} < 5$), significant enrichment ($5 < \mathrm{EF} < 20$), very high enrichment ($20 < \mathrm{EF} < 40$), and extremely high enrichment ($40 < \mathrm{EF}$).

2.6.4. Potential Ecological Risk Index (RI)

To calculate the potential ecological risk index (RI) of heavy metals, the following equation was used:

$$RI = \sum\nolimits_i^n E_r^i = \sum\nolimits_i^n T_r^i \times \frac{C_s^i}{C_n^i} \tag{5}$$

where E_r^i is the potential ecological risk factor of a single heavy metal, T_r^i is the toxic response factor, C_s^i is the measured concentration of the heavy metal, and C_n^i is the average concentration of the metal in the local background. The $\mathrm{T_r}^{\mathrm{i}}$ value is 2 for Cr, 5 for Cu, Pb, and Ni, 1 for Zn, and 10 for As [37]. The values of RI indicate the following: low ecological risk ($\mathrm{RI} < 150$), moderate ecological risk ($150 \leq \mathrm{RI} < 300$), considerable ecological risk ($300 \leq \mathrm{RI} < 600$), and very high ecological risk ($\mathrm{RI} \geq 600$) [31].

3. Results and Discussion

3.1. Physicochemical Properties in Soils

The physicochemical properties evaluated in the surface soil samples (0–30 cm) of Bonao, Dominican Republic, are presented in Table 1. The soil pH ranged between 4.8 and 6.7, with a mean value of 5.8, which indicates a slight acidity in the nature of the soil, which could favor the bioavailability of heavy metals [38]. The results of the pH observed in this study were lower than those reported by Zhang et al. [12] in soils used for rice cultivation in Southwest China. The electrical conductivity (EC) varied between 0.1 and 0.6 (mS/cm), which indicates that the salinity of the soil is low. The organic matter content (OM) ranged between 2.6% and 13.4%, with a mean value of 5.2%. The results of the present study are higher than those reported by Guo et al. [39] in soil used for rice cultivation in the Jin-Qu Basin of China. The cation exchange coefficient (CEC) varied between 3.8 and 21.2 meq/100 g, with a mean value of 12.1 meq/100 g. These results are lower than those obtained by Marrugo-Negrete et al. [34] in agricultural soils along the Sinu River Basin, Colombia. The soil texture has a content of silt, clay, and sand in the ranges of 16.7–41.3%, 8.2–50.2%, and 18.5–73.2%, respectively. In the study area, clayey soils are predominant.

Table 1. Selected physicochemical properties in agricultural soils of Bonao, Dominican Republic.

Soil Parameter		**Min**	**Max**	**Mean ± STD ***
pH (1:2)		4.8	6.7	5.8 ± 0.4
Electrical conductivity (mS/cm)		0.1	0.6	0.2 ± 0.1
Organic matter (%)		2.6	13.4	5.2 ± 1.9
Cation exchange capacity (meq/100 g)		3.8	21.2	12.1 ± 4.5
Soil texture	% Silt	16.7	41.3	30.4 ± 5.9
	% Clay	8.2	50.2	35.4 ± 8.5
	% Sand	18.5	73.2	33.6 ± 10.3
Texture class	Clay			

* STD, standard deviation.

3.2. Heavy Metal Concentration in Soils

Table 2 summarizes the descriptive statistics related to the total heavy metal concentrations in the agricultural soil samples and local background values. The mean heavy metal contents in the surface soil were 102,577, 2040, 347, 36, 92, 32, 10, and 4.2 mg.kg^{-1} for Fe, Mn, Cr, Cu, Ni, Zn, Pb, and As, respectively. The average levels of Fe, Ni, Cu, As, and Pb in the topsoil were higher than the natural background values. This suggests that the amount of these elements may have increased due to human activities such as mining, insecticide use, fertilizer applications, and transportation [6,40]. Because of the lack of official guidelines for healthy concentrations of metals in agricultural soils in the Dominican Republic, the mean concentrations of the different heavy metals were compared to the maximum levels of heavy metals for healthy agricultural soils adopted by the FAO [41] and the normal range of concentrations of heavy metals in agricultural soils introduced by Kabata-Pendias [42]. The mean concentrations of Zn and Pb are lower than the maximum values given by FAO and are within the normal range of concentrations given by Kabata-Pendias [42]. In contrast, the mean concentrations for Mn, Ni, and Cr are higher than the maximum values given by FAO. The mean concentration of As in the study area is higher than the normal range of concentration for agricultural soil given by Kabata-Pendias [42], similar to that reported by Delanoy et al. [17] in two different agricultural areas of the Dominican Republic, and lower than the mean concentration reported by Zhang et al. [43] in soils used for rice cultivation in Southwest China. The total heavy metal concentrations for all sampling points can be found in Tables S3 and S4.

Table 2. Descriptive statistics of the total heavy metal concentrations and local background values in agricultural soils of Bonao (mg·kg^{-1}).

Heavy Metal	**Min**	**Max**	**Mean ± STD ***	**Local Background Value ± STD ***	**FAO [a]**	**Kabata Pendias [b]**
Fe	57,800	157,700	102,577 ± 25,569	71,000 ± 60,200	-	-
Mn	400	5300	2040 ± 1014	1900 ± 500	<0.01	-
Cr	121	843	347 ± 164	354 ± 22	70	50–200
Cu	8	185	36 ± 44	23 ± 10	30	60–150
Ni	2	332	92 ± 93	58 ± 2	50	20–60
Zn	2	121	32 ± 30	35 ± 18	90	1–300
Pb	<0.1	59	10 ± 17	12 ± 9	35	20–300
As	3.6	5.0	4.2 ± 0.4	<0.1	-	1.5–3

* STD, standard deviation. [a]: maximum heavy metal concentration values for healthy agricultural soil, according to the Food and Agriculture Organization (FAO) (Rodríguez Eugenio et al., 2019 [41]). [b]: normal range of concentrations of heavy metals found in agricultural soil (Kabata-Pendias, 2010 [42]).

3.3. Spatial Distribution of Heavy Metals

The spatial distribution of the concentration of Cr, Cu, Ni, Pb, Zn, and As is shown in Figure 2. The figures were generated via interpolation, using the ordinary Kriging method. This method represents the default linear variogram quite effectively for performing interpolations with many types of data sets [44]. The distribution pattern of each individual metal is different, indicating they have different sources of origin. The spatial variation observed in the distribution of individual heavy metals is specific to each element. High concentrations of Ni are observed in the central region of the study area, while there are higher Cu levels in the southwest, which could be associated with mining activities. The distribution patterns show high levels of Cr in the northeast, Zn in the northwest, and nearly uniform levels of As across the study area, possibly attributed to insecticide usage and fertilizer applications. Additionally, the distribution pattern of Pb shows higher concentrations in the east, likely influenced by transportation activities.

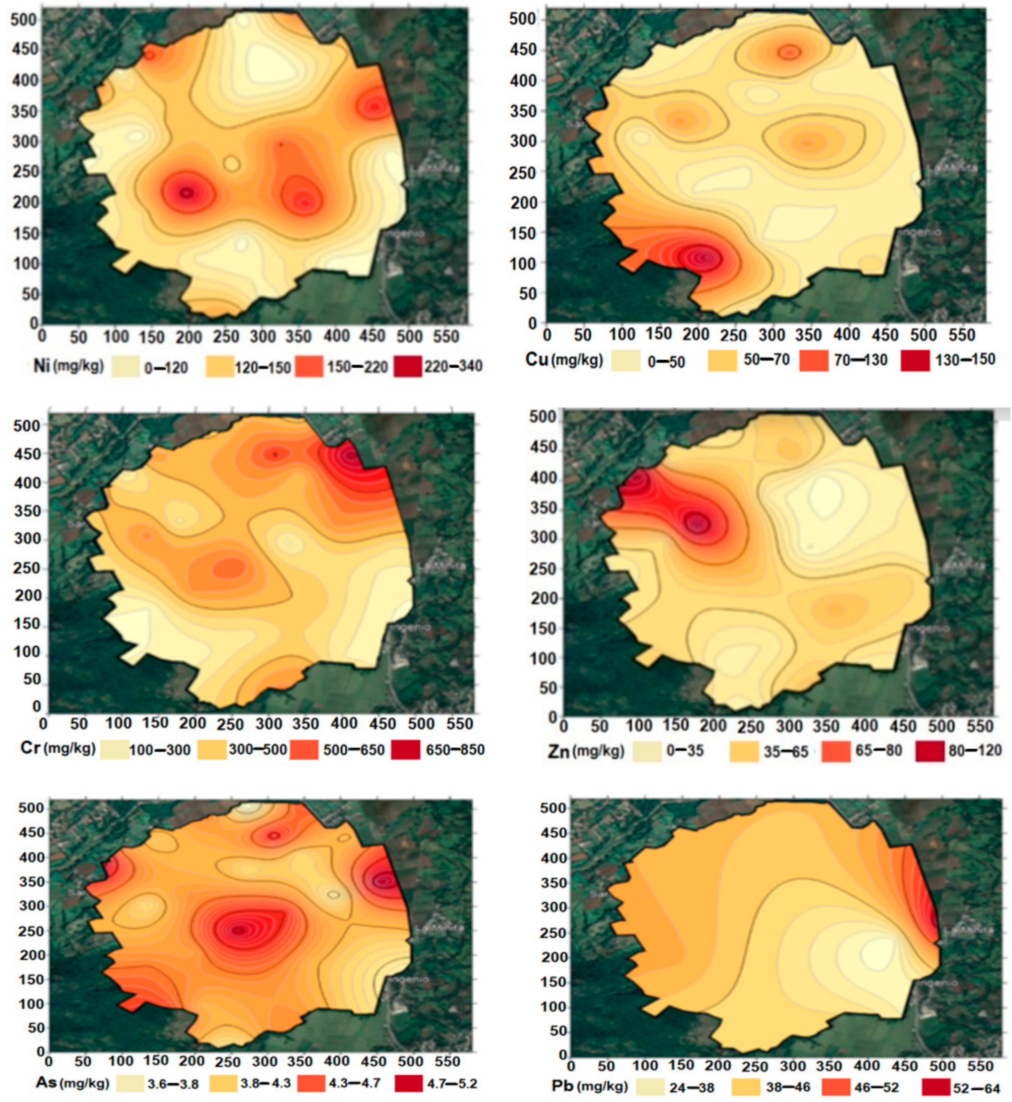

Figure 2. Spatial distribution of Cr, Cu, Ni, Zn, Pb, and As concentrations determined in the agricultural soil of Bonao, Dominican Republic.

3.4. Assessment of Heavy Metal Pollution in Soils and Potential Ecological Risk Index

The single pollution index (PI), the enrichment factor (EF), and the potential ecological risk index (RI) of the study area are presented in Table 3. The PI values calculated for Mn, Ni, Cu, and As indicated moderate contamination ($1 \leq \text{PI} < 3$), while Cr, Zn, and

Pb showed low contamination (PI < 1). The mean values of the EF for Cr, Mn, Ni, Cu, and Zn showed minimal enrichment (EF < 2), while Pb and As showed no enrichment (0 < EF < 1). Based on the obtained IPI value (1.1) and following the classification introduced by Wei and Yang [32], the studied soils can be classified as slightly polluted. However, the mean values of RI for all determined heavy metals indicated that the ecological risk is low (RI < 150). The average value of the integrated potential ecological risk index for the surface soils in the study area calculated as the sum of RI values of each individual heavy metal was 27.8, indicating an overall minimal ecological risk. The findings of the present study showed that the intensity of heavy metal pollution in the surface soils is low to moderate (Tables S5–S7).

Table 3. Pollution index (PI), enrichment factor (EF), and potential ecological risk index (RI) of heavy metals in soils of Bonao, Dominican Republic.

Heavy Metal	**Pollution Index (PI)**			**Enrichment Factor (EF)**			**Potential Ecological Risk Index (RI)**		
	Min	**Max**	**Mean ± STD ***	**Min**	**Max**	**Mean ± STD ***	**Min**	**Max**	**Mean ± STD ***
Mn	0.2	2.8	1.1 ± 0.5	0.3	2.3	1.1 ± 0.5	0.2	2.8	1.1 ± 0.5
Cr	0.3	2.4	1.0 ± 0.5	0.3	2.8	1.1 ± 0.6	0.7	4.8	2.0 ± 0.9
Cu	0.3	8.2	1.6 ± 1.9	0.3	11.0	2.0 ± 2.6	0.3	8.2	1.6 ± 2.0
Ni	0.03	5.6	1.6 ± 1.6	0.03	7.6	1.9 ± 2.0	0.2	28.0	7.8 ± 7.8
Zn	0.1	3.5	0.9 ± 0.8	0.05	5.8	1.1 ± 1.1	0.1	3.5	0.9 ± 0.9
Pb	0.0	4.8	0.8 ± 1.4	0.05	5.6	0.7 ± 1.5	0.0	24.0	3.9 ± 7.0
As	0.90	1.25	1.06 ± 0.09	0.04	5.7	0.8 ± 1.4	9.0	12.5	10.6 ± 0.9

* STD, standard deviation.

3.5. The Correlation of Soil Heavy Metals and Physicochemical Properties

Table 4 displays the correlation coefficients between physicochemical properties and heavy metal concentrations in the studied soils. As-Fe had a significant correlation at the 0.01 level, while As-Pb and Zn-Fe showed a correlation at the 0.05 level, indicating that the mentioned heavy metals must be associated with the same pollution sources. In the current study, a significant correlation between OM and Cu was observed, similar to that reported by Dragovic et al. [45], indicating that Cu was the preferred metal associated with the soil organic fraction. Additionally, a significant correlation between EC and Mn was observed, which could suggest a significant influence on the overall total content of this metal in analyzed soils. However, no significant correlations between physicochemical properties and the other heavy metals were observed.

Finally, it is important to highlight that characterizing agricultural soils is essential for the development of sustainable agriculture in the Dominican Republic. The cultivation of rice is necessary for feeding the Dominican people, and the experience of this work can be used as a starting point to obtain the standard levels of heavy metals in rice soils and develop protocols that allow improving the quality of the soils and the agricultural sustainability of the country.

Table 4. Pearson correlation coefficients between metal elements and physicochemical properties.

	Fe	**Mn**	**Cr**	**Cu**	**Ni**	**Zn**	**Pb**	**As**	**EC**	**% Sand**	**% Silt**	**% Clay**	**pH**	**CEC**	**%OM**
Fe	1														
Mn	0.35	1													
Cr	0.05	−0.01	1												
Cu	−0.27	−0.03	−0.25	1											
Ni	−0.33	−0.08	0.17	−0.12	1										
Zn	−0.37 *	−0.22	−0.09	0.12	0.04	1									
Pb	−0.08	−0.07	0.45	−0.18	0.48	−0.19	1								
As	−0.53 **	−0.33	0.11	0.11	0.31	0.29	0.78 *	1							
EC	0.34	0.49 **	−0.10	−0.04	−0.05	−0.31	−0.67	−0.33	1						
% Sand	−0.11	0.16	−0.16	−0.09	0.25	−0.31	−0.67	−0.33	0.08	1					
% Silt	−0.29	−0.45 *	0.02	0.32	0.04	−0.31	−0.67	−0.33	−0.42 *	−0.55 **	1				
% Clay	0.34	0.11	0.18	−0.11	−0.33	−0.31	−0.67	−0.33	0.19	−0.82 **	−0.03	1			
pH	−0.10	0.07	−0.38 *	0.25	−0.24	−0.31	−0.67	−0.33	−0.05	−0.21	0.43 *	−0.04	1		
CEC	−0.03	0.02	−0.34	0.23	−0.30	−0.31	−0.67	−0.33	0.41 *	−0.02	0.13	−0.06	0.41 *	1	
% OM	−0.12	0.07	−0.03	0.52 *	0.05	−0.31	−0.67	−0.33	−0.38 *	0.1	0.01	−0.12	−0.38 *	0.15	1

**, correlation is significant at the 0.01 level. *, correlation is significant at the 0.05 level.

4. Conclusions

The results of this study revealed the presence of Fe, Mn, Cr, Ni, Cu, Zn, Pb, and As in rice-cultivated soils in Bonao, Dominican Republic. The average concentrations are in the order of Fe > Mn > Cr > Ni > Cu > Zn > Pb > As. The distribution pattern of each individual metal is different, indicating they probably have different sources of origin. The results of the pollution indices show moderate pollution by Mn, Ni, Cu, and As in the study area. Using the local background as a reference value, the potential ecological risk assessment indicated a low-risk status. This research will contribute to establishing a baseline of heavy metal levels in rice-cultivated soils in Dominican Republic. The results of this study can be used to develop soil management strategies that ensure food security, promote safer agricultural practices, and produce healthier food. For future studies, we recommend using a more sensitive analytical technique for the determination of cadmium and mercury concentrations.

Supplementary Materials: The following supporting information can be downloaded at: https://www.mdpi.com/article/10.3390/su152316510/s1, Table S1. XRF analysis of the standard reference material (SRM 2711a), %SR values (n = 5), and detection limits. Table S2. Geographic location and physicochemical parameters of Bonao, Dominican Republic. Table S3. Total heavy metal concentrations in surface soils of Bonao, Dominican Republic. Table S4. Descriptive statistics of pH, % Organic Matter (OM), and the heavy metal concentrations of local background samples in soils of Bonao, Dominican Republic (n = 3). Table S5. Single Pollution Index (PI) in surface soils of Bonao, Dominican Republic. Table S6. Enrichment Factor in surface soils of Bonao, Dominican Republic. Table S7. Potential Ecological Index (RI) in surface soils of Bonao, Dominican Republic.

Author Contributions: N.M.A.T., the design of the study and writing of the manuscript; R.D., validation and formal analysis; R.M.H., software; O.D.R., analysis and interpretation of the data; D.R.A., review; L.B., writing—review and editing; L.B., project administration. All authors have read and agreed to the published version of the manuscript.

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