Soil, Site, and Management Factors Affecting Cadmium Concentrations in Cacao-Growing Soils

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Abstract: Soil contamination by potentially toxic trace elements (PTEs) such as Cadmium (Cd), is a major environmental concern because of its potential implications to human health. Cacao-based products have been identified as food sources with relatively high Cd contents. Here, we assessed Cd concentrations of cacao-growing soils in four major agricultural regions with contrasting climates in Peru, one of the main exporters of cacao products worldwide. At each study site ($n = 40$) a broad range of potential factors affecting Cd concentration in soils, i.e., site, soil and management, were evaluated. Concentrations of Cd ranged between 1.1–3.2 mg kg^{-1}. Mean values per region were below 2.7 mg kg^{-1}, usually established as upper-limit for non-polluted soils. Cadmium concentrations were significantly ($p < 0.001$) higher in sites at higher elevations and in a temperate, drier climate. Cadmium correlated positively with pH ($r = 0.57$; $p < 0.05$) and was higher ($p < 0.001$) in alluvial sediments and Leptosols. Management factors (cacao variety, cultivation year, management practices) and agroecology did not affect Cd concentrations directly. Overall, this study highlights the importance of considering a broad range of both natural and anthropogenic factors to evaluate Cd concentrations in cacao-growing soils and contribute to effective and sustainable cacao production by improving land management and planning.

Keywords: cacao plantation; cadmium; trace elements; soil quality; sustainable land management

1. Introduction

Soil degradation is presently one of the most serious environmental issues at the regional and global scales [1]. Anthropogenic disturbances such as intensive agriculture, deforestation, and industrial activities may result in high rates of land and soil degradation in the form of soil compaction, fertility loss and contamination, among others [2]. Soil contamination by potentially toxic trace elements (PTEs) such as Cadmium (Cd) is of great concern because of its implications to human health [3–6]. The Joint FAO/WHO Expert Committee on Food Additives (JECFA), and the World Health Organization

Global Environment Monitoring System-Food Contamination Monitoring and Assessment Programme (GEMS/Food), have identified cacao-based products as food sources that may contribute to the total human exposure to Cd [3,4]. Thus, in January 2019, the European Union Regulation (EU) No 488/2014 was enforced, setting threshold values for the presence of Cadmium in cacao powder and beans to maximum levels of 0.6 and 0.8 mg kg^{-1}, respectively [7,8].

Potentially toxic trace elements such as Cd are released into the environment in differing amounts because of natural processes and anthropogenic activities. Globally, values of Cd in unpolluted natural soils range from 0.01 to 2.7 mg kg^{-1} [9–11], while in managed soils the range extends up to 10.5 mg kg^{-1} [12]. Cadmium is naturally present in soils with concentrations varying across continents, countries, and soil types [13]. Important sources of natural Cd levels in soils include weathering of rocks, atmospheric deposition from forest fires, biogenic material, and volcanoes [14,15]. Generally, concentrations of Cd in soils are strongly related to their abundance in the parent rock. However, industrial or agricultural activities can discharge Cd into the soils, exceeding the capacity of the soil system and eventually resulting in plant uptake. Soils overlying volcanic rocks, such as those in South America, where cacao (*Theobroma cacao* L.) is cultivated [5,16–19], contain more Cd than the average concentration reported for natural soils [14,20]. High soil Cd levels of cacao plantations have largely been attributed to the weathering of parent rocks in South America [21], but how these levels are related to site characteristics, such as elevation and geology, has not yet been explored in detail.

Previous studies on cacao plantations in South America showed a strong relationship between Cd in beans and total soil Cd, and reported a considerable influence of soil pH on the increase of plant-available Cd [5,16,19,22]. Yet, there are few studies investigating the relationships between soil Cd and management and agroecological factors, and little attention has been paid to integrated geological and climatic attributes in the design and management of cacao plantations. Conversely, most of the studies conducted have focused on investigating Cd absorption by cacao trees [19,22]. Cadmium bioavailability in soils depends on several variables including total metal content, pH, soil organic matter (SOM), cation exchange capacity (CEC), and clay content [13,23]. Soil pH is particularly important as it controls the solubility and mobility of Cd in the soil (and therefore its availability for plants), with higher acidity increasing the mobility and solubility of this metal and thus, the risk of absorption by plants [24]. Short-term strategies that influence Cd absorption in plants have been investigated in recent years. While some studies have reported an ameliorant effect of organic wastes, phosphorus, zinc and zeolites on reducing the bioavailability of Cd to plants [25,26], much of this research has not been tested on cacao trees. Lime and biochar have shown to mitigate the Cd uptake by cacao trees [27]. However, the effect seems to be reduced under field conditions compared to in vitro, due to other site factor interactions including climate (i.e., precipitation; [6]). Blommaert [27] reported that the liming of the soil may mitigate the Cd uptake only if the subsoil layers are not rich in Cd. Appropriate mitigation strategies of Cd uptake on cacao trees are therefore needed, and additional studies are encouraged in the field rather than in controlled conditions [28]. In cacao plantations in South America, management factors such as the use of fertilizers, pesticides, irrigation, organic farming [29] and forestry, as well as plant biodiversity, have been related to the total Cd contents in cacao plantation soils [19] and cacao beans [22]. However, the understanding of soil and land management factors influencing the Cd contents in cacao beans is only partially resolved, so further investigation is needed to evaluate how a site's geological setting, climate and agroecological factors affects the levels of Cd in the soil [30].

It is well established that regular agriculture, based on large-scale and monoculture cultivation, will often lead to significant degradation of ecosystem functionality, affecting processes such as pollination and nutrient cycling [31,32]. To better balance crop production and ecosystem functionality, several approaches have been suggested including integrating biodiversity within agricultural systems [33–35] and assessment of agroecosystem function [36]. Agricultural systems need to maintain functional relationships and proper balances among biotic and abiotic factors [37] that can enhance ecosystem services [38]. Thus, soil pH, soil organic matter and trace elements have been used as potential indicators

to evaluate agroecosystem functions [39]. Comprehensive studies on the agroecological parameters influencing cacao plantation sustainability are essential to better understand the functionality of agroecosystems [17,40], and the role of these factors on modulating trace elements such as Cd in the plantations [41].

Here, we assessed the Cd concentrations of cacao-growing soils in four major agricultural regions of Peru, one of the main exporters of cacao products worldwide. We aimed to: (i) assess Cd concentrations in soils in areas with contrasting climates; (ii) identify the main factors influencing Cd concentrations across these regions, including site (elevation, geology), soil (soil type, pH), management (cacao variety, year of cultivation, use of fertilizers) and agroecological factors; and (iii) discuss the implications of these results within the framework of current regulations for improving land management and planning in cacao cultivation in Peru and similar regions.

2. Materials and Methods

2.1. Study Area

This research was conducted in four regions of Peru with contrasting climatic characteristics (Figures 1 and 2; Table 1): Madre de Dios (MD), Ucayali (UC), San Martin (SM), and Amazonas (AM). These regions, selected because of their broadly recognized agronomical relevance in cacao cultivation [42], are located in the Amazon Basin which encompasses the most extended tropical forest in the world. The Amazon Basin is characterized by an exceptional heterogeneity of vegetation composition and floral biodiversity [43]. The tropical and subtropical dry broadleaf forests [44] are characterized by tree genera including *Bombax, Alseis, Centrolobium, Aspidorsperma, Clusia, Croton, Embothrium, Jaracandà,* and *Inga* [45]. Soils in the basin are mainly lateritic, alluvial, and partly flooded [42].

Figure 1. Location of the 40 study sites within the four study regions in Peru (Madre de Dios, Ucayali, San Martin, and Amazonas). Source: Google Earth.

Figure 2. Sampling sites in the four regions: (**A**) Madre de Dios, (**B**) Ucayali, (**C**) San Martin, and (**D**) Amazonas.

Table 1. Description of the study regions: Madre de Dios (MD), Ucayali (UC), San Martin (SM), and Amazonas (AM).

Region	Sites (n)	Climate [1]	[2] TAP (mm)	[3] MAT (°C)	[4] Ecoregion
AM	13	Temperate, warm summer (Cfb)	995.0	24.1	Napo moist forest/Iquitos varzea
MD	12	Tropical rainforest (Af)	1938.5	26.0	Southwest Amazon moist forests/Peruvian Yungas
SM	7	Temperate, cold summer (Cfc)	1188.0	25.0	Peruvian Yungas
UC	8	Tropical monsoon (Am)	1667.1	26.4	Southwest Amazon moist forests

[1] Koppen–Geiger climate classification for 1980–2016 [45]; [2] TAP: total annual precipitation (mm); [3] MAR: mean annual temperature (°C); Climate data (2012) from stations in Puerto Maldonado (MD), Pucallpa (UC), Bagua (AM), Tarapoto (SM) [46]; [4] Ecoregions according to Olson and Dinerstein (1998) [47].

2.2. Soil Sampling and Analysis

Across the four regions of this study, we selected 40 sites in cacao cultivation patches. Twelve, eight, thirteen and seven sites were sampled in the regions MD, UC, AM, and SM, respectively (Figure 1; Table 1, Table 2) between November and December 2014.

At each site, we selected a homogenous sampling area of 100 m^2 from which eight subsamples (300 to 500 g each) were randomly collected from the top layer (5 to 30 cm depth), excluding the upper 5 cm rich in litter, vegetation residues, and organic matter. Soil sub-samples were then thoroughly mixed in the field into a composite homogenous sample (*n* = 40). From each bulk sample, 300 g was taken and air dried. The samples were then posted to the soil laboratory Mac-Minoprio analisi e Certificazioni s.r.l., a Vertemate con Minoprio (CO, Italy), where the analyses of Cd and pH were performed.

The analysis of the total Cd in soil was performed using EPA-3015A [48], based on the microwave extraction. The method is designed to mimic extraction using conventional heating with nitric acid (HNO_3). After extraction, metals in solution from soil samples were determined by flame (direct aspiration) atomic absorption spectrophotometry (EPA 700 B:2007), dissolving 1 g of cadmium metal in 20 mL of 1:1 HNO_3 and diluting to 1 L with reagent water, with a detection limit at 0.2 mg/kg [49]. The soil pH was analyzed with a potentiometer (InLab® Expert Pro-ISM, Mettler Toledo, Australia), on a suspension of soil-water as in Davey and Conyers [50] at a rate of 10 g soil on 25 mL H_2O (1:2.5).

Table 2. Pearson's correlation with main relationships between tree coverage (TC, %), herb coverage (HC, %), years of cultivation (Year), pH, Cadmium mg kg^{-1} (Cd), AEF (agroecosystem function indicator) and elevation (m) ($n = 40$). Correlation coefficients (r) with * are significant at $p < 0.05$; ** at $p < 0.01$.

Variable	Tree	Herb	Year	pH	Cd	AEF	Elevation
TC	1.00	0.37 *	−0.13	−0.36 *	0.17	0.60 **	−0.10
HC		1.00	−0.34 *	−0.49 *	−0.24	0.40 *	−0.31 *
Year			1.00	0.56 **	0.15	−0.18	0.54 **
pH				1.00	0.57 **	−0.40 *	0.76 **
Cd					1.00	−0.1	0.40 *
AEF						1.00	−0.26
Elevation							1.00

2.3. Site Factors, Agroecological Parameters and Land Management

At each of the 40 study sites we assessed and recorded the following parameters to explore their relationships with Cd levels in the soil: site characteristics, i.e., coordinates (latitude, longitude), elevation, soil type and geological attributes, (Table S1); agroecological factors, i.e., cacao variety, year of plantation, use of fertilizers, tree and herbaceous coverage and agroecosystem functionality (AEF); and land management practices (Table S2). Coordinates and elevation were measured with a GPS 66 S (Garmin Ltd., Lenexa, KS, USA) in situ. Geological attributes and types for each location were obtained from the Geological map of Peru 1:1,000,000 [51] and soil types and associations from the Soil Map of Peru 1:1,000,000 [52], according to the FAO- World Reference Base for Soil Resources classification [53]. Sedimentary origin was obtained from the map of Peruvian Andes 1:100,000 [54].

The tree cover (% canopy coverage that shaded the parcel) and herbaceous cover (% of grasses and legumes distributed over the cultivation surface) were measured at each of the eight random soil sampling sites per plot, within an area of 5 × 5 m. We recorded the canopy coverage based on the number of plants under the cacao trees (estimated in percentage categories from 0 to 65%), and the estimated herbaceous coverage from 0%–100%. Agricultural management was classified as one of the following categories: "cut-deposit" (deposition of the organic matter obtained by grass cutting and pruning); "partial cut-deposit" (partial deposition of the organic matter obtained by grass cutting and pruning; "no cut" (absence of cuttings); and "compost" (organic matter fermented and deposited, including the epicarp of the cacao fruits). A qualitative parameter representative of the agroecosystem functionality (AEF) was assessed according to Norris [55], who integrated factors such as the landscape context, production intensity and tree biodiversity. Parameters captured in the AEF included habitat integrity (vicinity to the primary forest), tree species richness and pollination [56]. At each site, we assigned numerical scores as in Liebig et al. [35] with one point recorded per criteria satisfied as follows: (i) proximity (<0.5 km) to the tropical humid primary forest; (ii) presence of >2 tree species other than cacao within the plantation; and (iii) occurrence of native insects acting as ecological indicators, scoring a point when at least three individuals of each taxa were encountered (Table S3). Native insect occurrence was assessed through 40 min of observation on the flowering plants at each parcel along a 100 m transect. The native insects identified as functional pollinators were Euglossine bees (Apidae) and *Forcipomyia* flies (Ceratopogonidae). Euglossine bees have already been used for the study of

agroecological systems [57–59], are considered ecological indicators of neo-tropical forest, and are easy to assess by transect observations.

2.4. Statistical Analysis

The soil, site, agroecological variables and management practices were tested for normality and variance homogeneity using the Shapiro–Wilk and Levene's tests. The differences for these variables between climatic regions, geology, and soil type were tested using one-way ANOVA, and comparisons between means were performed with the Tukey's honestly significant difference (HSD) test ($p < 0.05$). The analysed variables were log transformed or rank normalized to meet the assumptions for ANOVA (the presented data are non-transformed). Pearson's correlations were used to test the relationships between measured variables. Principal component analysis (PCA) was used to assess further relations and driving factors for Cd contents in soils. All analyses were performed with R statistical software version 3.1.2 R Core Team [60], and we used the packages agricolae, ggplot2, ggfortigy and viridis.

3. Results

Our results showed that Cd concentration in the cacao-cultivated soils ranged from 1.1 to 3.2 mg kg^{-1} and were significantly ($p < 0.001$) different across regions (Figure 3A). The mean concentrations per region were 1.57 ± 0.47 mg kg^{-1} (MD), 1.67 ± 0.14 mg kg^{-1} (UC), 1.92 ± 0.13 mg kg^{-1} (AM), and 2.46 ± 0.22 mg kg^{-1} (SM) (Figure 3A). The AM and SM regions, with temperate climate, low humidity, and high elevation (Table 1, Table S1), had consistently higher values.
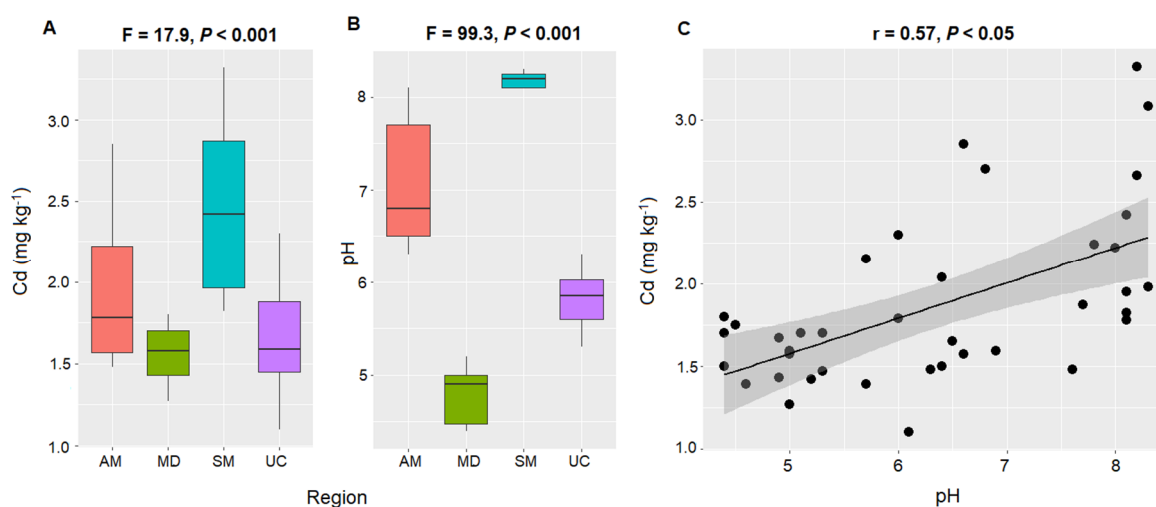

Figure 3. Boxplots of cadmium (Cd, mg kg^{-1}) in the soil (**A**) and soil pH (**B**) for each region ($n = 40$). Regions: Madre de Dios (MD), (UC) Ucayali, San Martin (SM) and Amazonas (AM). (**C**) Relationship between cadmium (Cd, mg kg^{-1}) in the soil and soil pH.

The soil pH ranged from 4.4 to 8.3 across the study sites (Figure 3B) and correlated positively with Cd ($r = 0.57$; $p < 0.05$; Figure 3C). Pearson's correlation analyses also showed that elevation correlated positively with Cd ($r = 0.40$, $p < 0.05$) and pH ($r = 0.76$, $p < 0.01$) (Table 2, Figure 4). The year of plantation was not related to Cd, but significantly correlated with pH ($r = -0.56$, $p < 0.01$). We did not find a significant effect of management factors (cacao variety, use of fertilizers, or type of management) on the levels of Cd or pH in the soil.
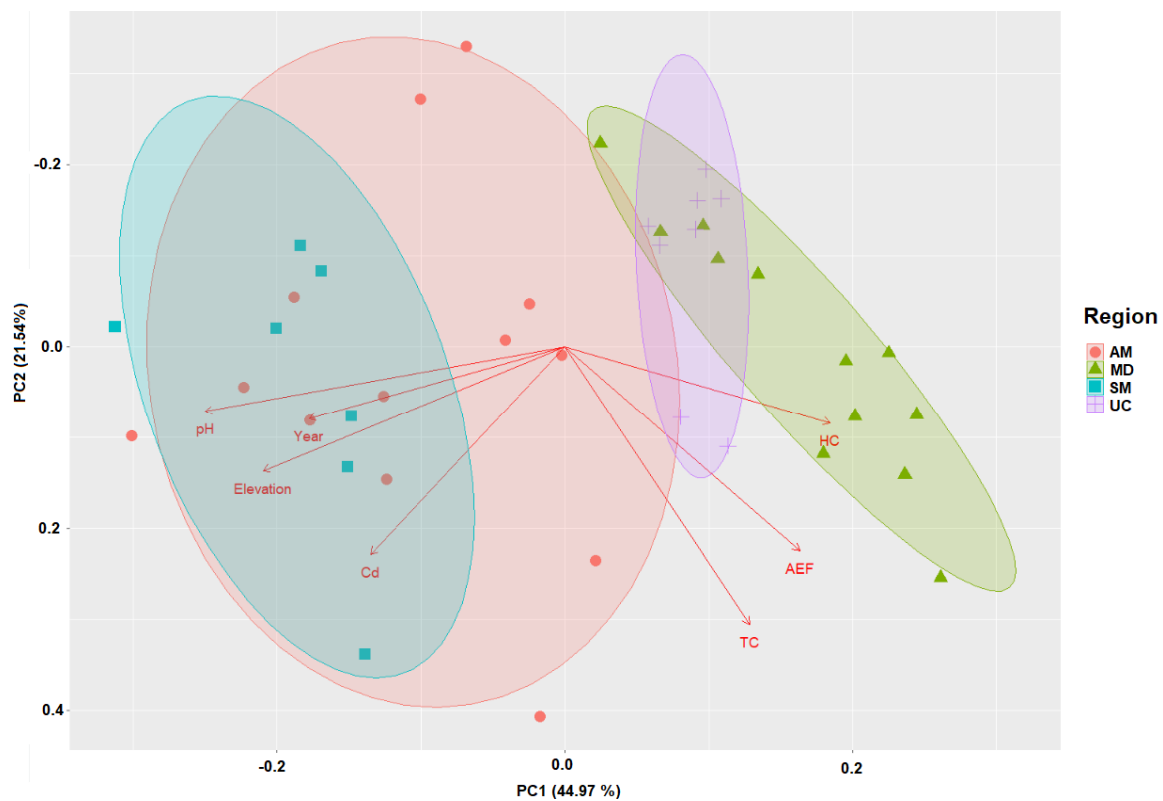
Figure 4. Biplot of the first and second axes obtained from the principal component analysis (95% confidence ellipses are shown) showing dependencies between site, soil and agroecological factors across regions. Points represent soil samples (*n* = 40). Cd = Cadmium; TC = tree coverage; HC = herbaceous coverage; Year = year of plantation; AEF = agroecosystem functionality. Regions: Amazonas (AM), Madre de Dios (MD), San Martin (SM) and Ucayali (UC).

In relation to the agroecological variables (AEF, tree and herb coverage), we did not find a clear effect on Cd levels, but a significant (and negative) correlation was found with pH, i.e., $r = -0.40$, -0.36 and -0.49; < 0.05, respectively (Table 2). The principal component analyses performed on all data (Figure 4) indicated that the first two axes could explain 66.51% of the variation between samples: 44.97% by axis 1 and 21.54% by axis 2. We found a clear discrimination between regions, with a strong cluster of MD and UC, where lower Cd values were reported. Although we did not find a correlation of Cd and agroecological factors, higher values for these parameters were reported in the abovementioned regions (MD and UC).

The contents of Cd in the soil were significantly different across geological substrates and soil types ($p < 0.001$) (Figure 5). Higher values of Cd were found in alluvial deposits (2.69 ± 0.24 mg kg^{-1}) and lower in detritus, sand, and gravel (1.55 ± 0.04 mg kg^{-1}) (Figure 4). Regarding the sediment origin, higher Cd concentrations were detected in sediments from the Cretaceous (2.10 ± 0.18 mg kg^{-1}) and paleo-neo age (2.14 ± 0.19 mg kg^{-1}) and lowest in those from the Quaternary (1.62 ± 0.06 mg kg^{-1}). Eutric Leptosol and Dystric Leptosol showed higher values for Cd (2.46 ± 0.22 mg kg^{-1} and 2.14 ± 0.19 mg kg^{-1}, respectively). The lowest concentrations were found in Dystric Gleysol (1.58 ± 0.10 mg kg^{-1}) (Figure 4).
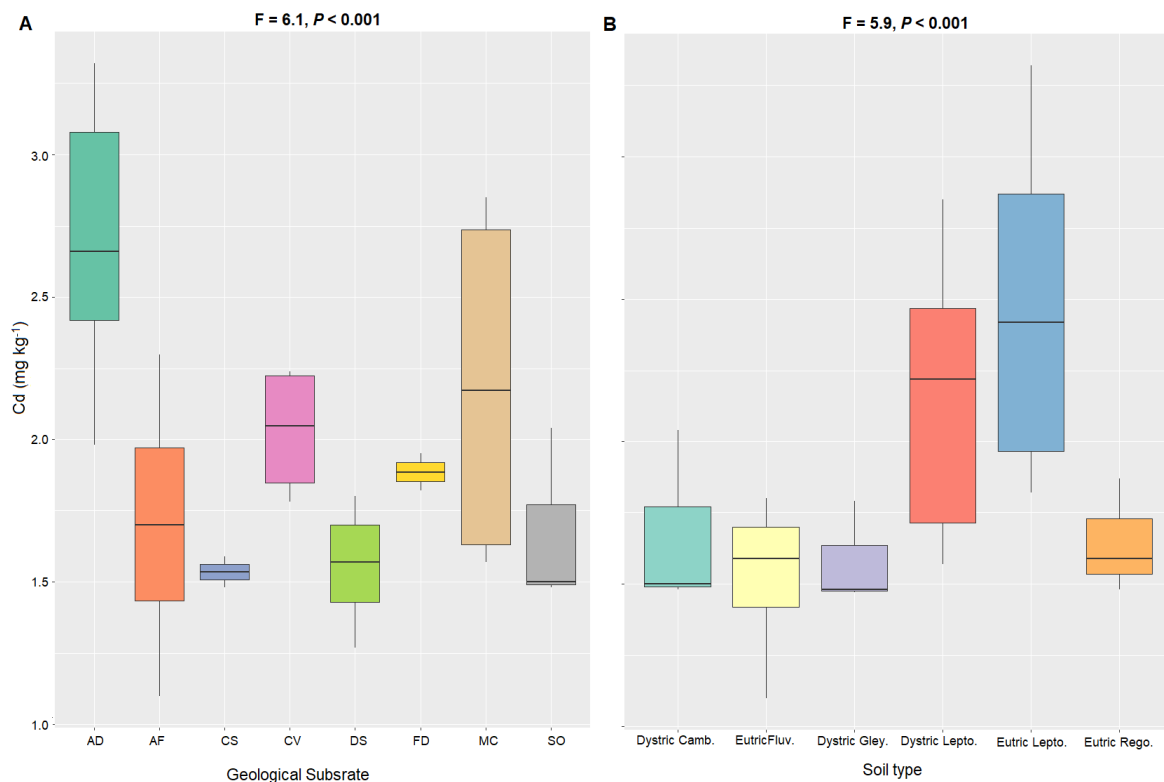
Figure 5. Boxplot of Cd (mg kg^{-1}) (n = 40) for each geological substrate (**A**): alluvial deposits (AD); alluvial, fluvial, colluvial deposits (AF); carbonaceous schists (CS); clastic and volcanic schists (CV); detritus, sand, and gravel (DS); fluvial deposits (FD); metamorphic complex, sandstones (MC); sandstones, outcrops with silty sand, clay and quartz (SO). Boxplot of Cd (mg kg^{-1}) (n = 40) for each soil association (**B**): Dystric Cambisol-Haplic Acrisol (Dystric Camb.); Eutric Fluvisol and Gleysol (Eutric Fluv.); Dystric Gleysol and Cambisol (Dystric Gley.); Dystric Leptosol-Cambisol-Regosol (Dystric Lepto.); Eutric Leptosol-Cambisol-Regosol (Eutric Lepto.); Eutric Regosol-Haplic Calcisol (Eutric Rego.)

4. Discussion

4.1. Cadmium Content in Soil of Cacao Plantations from Different Ecoregions

Overall, our results showed that Cd levels in soils from cacao plantations in Peru differed across regions with contrasting climatic characteristics, with most of the samples in this study (80%, Table 2) containing Cd concentrations below 2 mg kg^{-1}. Cadmium contents obtained here ranged between 1.1 and 3.2 mg kg^{-1} and the mean values per region were under the 2.7 mg kg^{-1} usually established as the upper limit for non-polluted soils [9–11]. These values were similar to those found in agricultural soils of cacao plantations in Ecuador [16], but higher than those reported by Arévalo-Gardini et al. [61] in cacao plantations from three productive areas of Peru. According to Arévalo-Gardini et al. [61], Cd in the soil was absent in the Huànuco region, while it was broadly present in other regions ranging from 0.01 ± 0.02 mg kg^{-1} (Cajamarca) to 0.53 ± 0.02 mg kg^{-1} (Piura).

Regarding the limits for Cd concentrations in cacao-derived products imposed by current regulations, the scenario at the international level is varied, as each country adopts different approaches to determine the threshold of phytotoxicity of Cd in the soil [62–68]. The maximum value of Cd found in soils in this study was 3.2 mg kg^{-1} in a cultivated site from the San Martin region. This is above the reference values set by various countries such as Denmark, Italy, Germany, and China [9–16,68,69], but below the maximum limit of tolerance set by regulations of countries such as Finland, France, and Poland [66,67].

4.2. Factors Influencing Cadmium Concentrations in Cacao-Growing Soils

As reported by other studies [16,61], we found a strong correlation between Cd and pH in the soils analyzed here and higher Cd concentrations were evident in alkaline soils in the SM and AM regions. Fertilizers ("Guano de Isla", Sulfomag, dolomite and phosphatic rock) were applied only in the MD and SM regions. Regarding the management of the plantations, in most sites (93%) farmers applied "partial or total cut and deposit", which refers to the deposition of biological material resulting from the pruning of the trees. It has been previously reported that Cd accumulation in cacao-growing soils may depend also on the decomposition of litter and deposit from harvesting in the topsoil [70]. But, consistent with the results found by Argüello et al. [19] in cacao plantations in Ecuador, neither the type of management practice nor the use of fertilizers affected the levels of Cd in the soils in our study sites. The cacao clone CCN51 was cultivated in 33 of the 40 sites investigated; 38 of these sites were cultivated for ≤10 years, and 30 only for over six years. Similarly, these variables did not affect the Cd concentrations in the soil either. The lack of effect of the year of cultivation on Cd levels may be partly due to the predominance of young plantations in our study. Remarkably, age was positively related to pH, suggesting a likely modulation on Cd in the soil, but this effect should be investigated in a broader panel of plantations (i.e., in [22]). Soil pH was correlated with agroecological factors such as tree and herb coverage and the AEF, in agreement with Andersen et al. [71], who found that sites with a higher tree coverage proportion had more acidic soils, promoting the decrease of soil Cd concentrations in the topsoil. One of the components used here to assess the AEF was the occurrence of native insects. *Forcipomyia* flies are the pollinator of cacao (*Theobroma cacao)* and rely on these trees for their lifecycle, particularly in shady conditions [72]. Therefore, the canopy coverage is critical to support agroecological functions such as pollinator services.

Despite the lack of significant relationships with management and agroecological factors, the location of the cultivated sites in terms of climate and elevation was a determinant for the variation in Cd concentrations. Moreover, our results revealed the important influence of the geological nature, including substrate type, soil association and sediments' origin, on the soil Cd concentrations of the cacao-growing soils. According to their study in Honduras, Gramlich et al. [73] found that the variation of total Cd in the soil was strongly affected by the natural substrate. Similarly, Pérez-Sirvent et al. [74] found that Cd contents in soil were not linked to the soil use or characteristics when comparing natural sites to agricultural ones, but with mineralogical composition. Several studies have reported that Cd concentrations in soil under cacao plantations across the world are highest in alluvial soils originating from sedimentary material [13]. In South America, for example, Cd contents have largely been attributed to the weathering of parent rocks [21], which was evidenced here with the highest values of Cd in mountain soils subjected to intense erosion, e.g., Leptosols. Furthermore, we found that an ancient sedimentary origin (of the Cretaceous and Paleo-Neo age) was associated with highest contents of Cd in the soil (Table S1). Noteworthy, the variations in Cd were related to differences in the geological substrate, with highest values in alluvial deposits and associated clastic, volcanic, and metamorphic rock origins of the sub-Andean and Amazon belt [75].

As several authors have previously shown, Cd concentrations in soils can be ascribed to natural soil conditions as these are largely affected by Cd levels in the parent rock, local weathering conditions, or deposition [21]. In fact, as noted also by Bertoldi et al. [76], the much higher concentrations of Cd in cacao plantations (beans) found in South American countries than in Central America, and East and West Africa may be due to a geological nature rather than pollution.

4.3. Implications for Land Management

Overall, our study highlights the importance of including a broad range of potential factors when assessing concentrations of Cd in cacao-growing soils. These assessments can have substantial implications for adequate management of cacao plantations. Several directives, such as the European Union cadmium (Cd) regulation enforced in January 2019, are threatening the sustainability of plantations in several South American countries. In Peru, cacao plantations have major social relevance,

being the sixth most important crop in the number of farmers (138,000) that cultivate an area covering 199,000 ha [77]. Peru is one of the top producers of special and aromatic cacao and the second largest producer of organic cacao globally [78]. With a total cacao bean production of 121,825 tons in 2017, Peru exported more than $235 million of cacao and derivatives, 50% of it directed to the European market [79]. The majority of cacao varieties (i.e., forastero) grown in the world (95%) are native to the Amazon Basin [80,81], and that produced in Peru and other South America countries has been considered suitable for fine chocolate manufacture [82]. Although we did not measure Cd contents in the cacao beans or plants at our study sites, multiple studies have directly associated Cd contents in soils with those found in the cacao beans. It is therefore critical to assess the factors influencing these concentrations in soils.

Some of the strategies for neutralizing PTEs are the use of living organisms such as plants, e.g., phytoremediation or microorganisms [83], or selecting cacao cultivars that accumulate less Cd [84]. Proposed mitigation solutions also include the improvement of the post-harvesting processes, increasing the nutrient status of plants and using genotypes that are naturally low accumulators of Cd [21]. However, other ecological processes, such as pollination functionality and the integration of cacao plantations with native vegetation, could be relevant and indirectly affect Cd concentrations in soils, as evidenced here. Although over the years Cd levels in soils have been reported to progressively increase in relationship with anthropogenic processes (such as intensive agricultural practices), natural factors and processes are also critical for assessing areas at risk [41]. Some of the solutions to reduce the levels of Cd in soil proposed in previous studies include avoiding high-risk areas for establishing plantations [21]. Our study suggests that areas with lower amounts of rainfall and higher elevation that are intensively subjected to erosion, e.g., mainly including Leptosols, can encompass soils with higher Cd concentrations. This is linked to the natural sources of soil Cd in these areas from, for example, the rock weathering processes [18,21].

The widespread high Cd concentration in cacao plantations in South America have been attributed to a geogenic origin (unrelated to point pollution), as the Cd concentrations of cacao beans in these areas have been reported to be three times higher than those from Central America and East Africa, and ten-fold more than those of West Africa [19,27]. It is important to highlight that, as reflected in this research, most studies have shown a large variation across sites with particular areas exhibiting higher levels or "hotspots" [19,21]. Given this level of heterogeneity among sites (even within farmer fields), field trials across different environments and land management types are required in order to design cost-effective solutions and reduce the risk of accumulation of Cd in soils and plants where cacao is cultivated.

5. Conclusions

The results in this study showed a large variability in the Cd concentration of cacao-cultivated soils that ranged from 1.1 to 3.2 mg kg^{-1}. These concentrations were positively related to pH. The location of the cultivated sites in terms of climate and elevation was also a determinant for the variation in Cd concentrations. Soil Cd also reflected the geological nature of the sites, with soils of an alluvial origin having the highest Cd content. Higher concentrations were also found at higher altitudes and in regions with a temperate climate and lower humidity (Amazonas and San Martin). We did not find a significant relationship between Cd concentrations and land management and agroecological factors, but soil pH was largely influenced by environmental and management factors such as year of plantation. These results highlight the importance of considering a broad range of both natural and anthropogenic factors to assess Cd concentrations in cacao-growing soils. Additionally, it is important to evaluate the features of cultivation sites, in addition to land management factors, for effective and sustainable cacao production in Peru.

geological substrate according to Geological map of Peru [53] soil types extracted from Mapa de Suelos de Peru [54] and sediment origin according to Pfiffner and Gonzales [56]; Soil classification according to the WRB (2015) [55]. Table S2. Site number and parameters recorded in the cultivation sites. Regions: Madre de Dios (MD), Ucayali (UC), San Martin (SM) and Amazonas (AM). N: site number; TC: tree coverage (%); HC: herbaceous coverage (%); Year: year of the plantation; Cd: cadmium concentration (mg kg^{-1}); Management of cultivation: cut and deposit (CD), partial cut and deposit (PCD), compost (C); no cut (NC); + indicates the use of fertilizers (including Guano de Isla', Sulfomag, Dolomite and Phosphatic rock). Table S3. Site number and AEF: agro-ecosystem function indicator based on the scores assigned for each of the criteria satisfied: (a) proximity to the primary forest (<0.5 km); (b) presence of >2 tree species other than cacao within the plantation; (c) occurrence of native insects acting as ecological indicators, scoring a point when at least three individuals of each taxa (Euglossine bees (Apidae) and Forcipomyia flies (Ceratopogonidae).

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