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Heavy metals content in soils and cocoa tissues in Magdalena department Colombia: emphasis in cadmium*

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Abstract

Cocoa (*Theobroma cacao* L.) is a species originating from the humid tropics. Over 70,000 km^2 worldwide is dedicated to growing cocoa, and approximately 30% of its global production is concentrated in the equatorial regions of Central and South America, the Antilles and tropical Asia. The demand for cocoa is growing significantly, but the presence of cadmium (Cd) in the species is a potential problem limiting its commercialization. The present study determined the Cd, Ni, Pb and Cr content in two cocoa-producing regions in northern Colombia. Foliage (leaf, bean and shell) and soil were analyzed. The mean Ni, Pb and Cr content in the plant tissue did not exceed the maximum permissible limits set by the United States Environmental Protection Agency (EPA). The mean Cd content in the beans was 0.51 mg kg^{-1} in region 1 and 0.66 mg kg^{-1} in region 2, which are considered risky per reports from the European Union and restrict the product's export. These results serve as a reference for future research on cocoa.

Keywords
Cocoa bean; food safety; heavy metals; *Theobroma cacao* L.; soil pollutants

Ciencia y Tecnología

Contenido de metales pesados en suelos y tejidos de cacao en el departamento del Magdalena, Colombia: énfasis en cadmio

Resumen

El cacao (*Theobroma cacao* L.) es una especie originaria de los trópicos húmedos. Más de 70,000 km^2 en todo el mundo están dedicados a su cultivo y aproximadamente el 30% de su producción mundial se concentra en las regiones ecuatoriales de América Central y del Sur, las Antillas y Asia tropical. La demanda de cacao está creciendo significativamente, pero la presencia de cadmio (Cd) en la especie es un problema potencial que limita su comercialización. El presente estudio determinó el contenido de Cd, Ni, Pb y Cr en

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dos regiones productoras de cacao en el norte de Colombia. Se analizaron el tejido (hoja, frijol y cáscara) y el suelo. El contenido medio de Ni, Pb y Cr en el tejido vegetal no excedió los límites máximos permisibles establecidos por la Agencia de Protección Ambiental (EPA) de los Estados Unidos. El contenido medio de Cd en los frijoles fue de 0,51 mg kg^{-1} en la región 1 y 0,66 mg kg^{-1} en la región 2, que se consideran riesgosos según los informes de la Unión Europea y restringen la exportación del producto. Estos resultados sirven como referencia para futuras investigaciones sobre el cacao.

Palabras clave

Almendra de cacao; seguridad alimentaria; metales pesados; *Theobroma cacao L.*; contaminantes del suelo

Teor de metais pesados em solos e tecidos de cacau no departamento de Magdalena, Colômbia: ênfase no cádmio

Resumo

O cacau (*Theobroma cacao L.*) é uma espécie originária dos trópicos úmidos. Mais de 70.000 km^2 em todo o mundo são dedicados à sua cultura e aproximadamente 30% de sua produção global está concentrada nas regiões equatoriais da América Central e do Sul, Antilhas e Ásia tropical. A demanda por cacau está crescendo significativamente, mais a presença de cádmio (Cd) nas espécies é um problema potencial que limita sua comercialização. O presente estudo determinou o teor de Cd, Ni, Pb e Cr em duas regiões produtoras de cacau no norte da Colômbia. A folhagem (folha, feijão e casca) e o solo foram analisados. O teor médio de Ni, Pb e Cr no tecido vegetal não excedeu os limites máximos permitidos estabelecidos pela Agência de Proteção Ambiental dos Estados Unidos (EPA). O conteúdo médio de Cd nos grãos foi de 0,51 mg kg^{-1} na região 1 e 0,66 mg kg^{-1} na região 2, que são considerados de risco por relatórios da União Europeia e restringem a exportação do produto. Esses resultados servem como referência para futuras pesquisas sobre cacau.

Palavras-chave

Grãos de cacau; segurança alimentar; metais pesados; *Theobroma cacao L.*; poluentes do solo

1. Introduction

Cocoa (*Theobroma cacao L.*), a species native to the humid tropics of Central and South America, has more than 70,000 km^2 worldwide dedicated to its cultivation. Approximately 30% of its production is concentrated in Central and South America, the Antilles and the tropical zones of Asia (Kongor *et al.*, 2016; Ramírez Gil, 2016), with production of 47 million tons (ICCO, 2018). Cadmium (Cd) presence in cocoa fruit has been reported in Peru, Ecuador and Colombia, but only some of the territory is believed to be contaminated; thus, the environment must be analyzed to improve the availability and quality of this information. Cd presence in cocoa and its derivatives was reported at the sixth meeting of the Committee on Food Contaminants in 2012 and added to the list of contaminants and natural toxicants present in food (FAO/OMS, 2014). Therefore, starting in 2019, the European Union (EU) implements the standard and maximum permissible limits of Cd in cocoa and its derivatives.

The maximum standard limit for Cd in cocoa varies by norm and country. The EU legislated the maximum permissible levels in food products in regulation DO L 364 of 12/20/2006, p. 5, and established a permissible range between 0.2 and 0.5 mg kg^{-1} for Cd in powdered cocoa and chocolate products. García and Dorronsoro (2002) reported maximum levels in cocoa beans of 0.50 mg kg^{-1}, which would meet the EU and the international market's requirements.

Cd can bioaccumulate and persist but not biodegrade, and it is a precursor of various cancers, oxidative stress, inflammation, and tissue injury in humans (Das & Al-Naemi, 2019; Londoño-Franco, Londoño-Muñoz & Muñoz-García, 2016). In Japan, Itai-Itai syndrome, caused by consuming rice contaminated with Cd (Salama, 2018; Clemens, Aarts, Thomine & Verbruggen, 2013; Jiménez, 2015), revealed the risk of human contamination from cocoa and vegetable intake. Cd is distributed in the Earth's crust at a mean concentration of 0.1 mg kg^{-1}; in rocks, it does not exceed

0.3 mg kg^{-1}, and it has been found in limestone (Ramtahal *et al.* 2018; Wang *et al.* 2015). Cd contamination is associated with anthropogenic actions (Bravo & Benavides, 2020). Caviedes, Muñoz, Perdomo, Rodríguez, & Sandoval (2015), and Arévalo-Gardini *et al.* (2016) reported that irrigation waters from solid waste treatment systems are sources of Cd and other metals. Roberts (2014) reported that phosphorus fertilizers contain Cd; Alloway (2013) identified a close relationship between heavy metals and specific sources of fertilizers and pesticides and argued that this situation is exacerbated when species that absorb Cd are cultivated. On the other hand, high spatial variability in soil Cd contents is recognized, making it necessary to develop projects that seek their quantification, persistence, and speciation (Bravo & Benavides, 2020; Caviedes *et al.* 2015).

Cd is toxic to some species, and its specific entry mechanisms are unknown, although proteins are involved (Huamaní-Yupanqui, Huauya-Rojas, Mansilla-Minaya, Florida-Rofner, & Neira-Trujillo, 2012). Some species accumulate Cd, including barley (Hordeum vulgare), corn (Zea mays) (González & Lobo, 2013), alfalfa (Medicago sativa) (Lara-Viveros *et al.* 2015), rice (Oryza sativa) (Chaney, 2012), *T. cacao* L. (Ramtahal *et al.*, 2015), and some tree species (Callegario-Pereira, Dormelas-Rodrigues, Soares-dos Santos, do Nascimento-Guedes, & do Amaral-Sobrinho, 2012). In addition to generating risk to human and animal health, Cd contents can severely affect the agricultural export products such as cocoa, with demand in North American and European markets, where exists standard norms that prevent its reception if their content exceeds certain limits (García and Dorronosoro, 2002). The Cd accumulation process is of particular interest in cocoa farms in Colombia because there are producing areas with a high Cd content that could dangerously join the food chain and thus, affect human health. Therefore, it is of vital interest to know the soil Cd contents and determine if it moves to plant and, once inside it, establish if it is accumulating mainly in the organs destined for food (Lara-Viveros *et al.* 2015).

Therefore, areas are required where new cocoa plantations can be sown for export purposes. It is necessary to know the soil Cd contents and predict if it moves towards the plants and accumulates in the grain, and given the lack of information on whether cocoa (*T. cacao* L.) grown in northern Colombia absorbs Cd, we evaluated its presence in the soil, absorption, and accumulation in plant tissues.

2. Methodology

Location

This study was conducted in the department of Magdalena, Colombia, at the geographic coordinates 10°46'00" N and 74°8'00" W. The natural vegetation is classified as tropical dry forest, and the climate is warm and dry, with a mean annual temperature of 27°C and relative humidity of 82% (Piraneque, Aguirre, & Reis, 2018). Plant material and soil samples were collected in 20 georeferenced agricultural plots distributed in the Sierra Nevada of Santa Marta (zone 1) and the Banana Zone (zone 2) (Figure 1).

Soils characteristics

Soils in zone 1 were classified as Entic Haplustolls originating from granodiorite, quartz diorite, and granite. These are deep, well-drained soils with loam to sandy-loam texture; they are moderately acidic to neutral (pH 6.3 to 7.7) with moderate to high natural fertility due to low organic matter content and cation exchange capacity. These soils contain kaolinite (50%), micas (5-30%), montmorillonite (30-50%) and, vermiculite (5-30%) in the clay fraction and quartz (5-15%), feldspar (5-15%), and interstratified trace minerals in the sand fraction (Aguirre, Piraneque, & Vásquez, 2015; IGAC, 2009).

Soils in zone 2 were classified as Typic Ustorthens originating from gneiss and schists. These are relatively deep, well-drained soils with sandy texture; they are neutral to slightly acidic with moderate natural fertility due to medium organic matter content and high cation exchange capacity. These soils contain non crystalline material, kaolinite (40%), gypsite (30-50%) and, vermiculite (5-30%) in the clay fraction and quartz (22-26%), feldspar (36-38%), and hornblende (20-22%) in the sand fraction (Aguirre, Piraneque, & Vásquez, 2015; IGAC, 2009).

Plant material, sampling and establishment

Different cocoa varieties are grown in small cultivated plots, but the product is recognized in the international market for its aroma and provides economic sustenance for indigenous people and farmers. The plots have the added value of proximity to the seaport, which is a determinant of cocoa export. Using a transect, 20 agricultural plots were selected with productive plants older than eight years from which cocoa leaves (healthy and functional from the central portion of the branches in the middle third of the crown) and fruits were collected. The plant material was transported to the Soil Laboratory of the University of Magdalena, were it was washed with distilled water and dried at room temperature; the beans were separated from the fruit or pod (shell), and the material was dried at 70°C for 48 hours, ground, packed and labeled.

Soil sampling and determinations

Soil samples (1 kg) were randomly collected for analysis from the 20 cocoa farms at two depths (0-10 and 10-20 cm) per

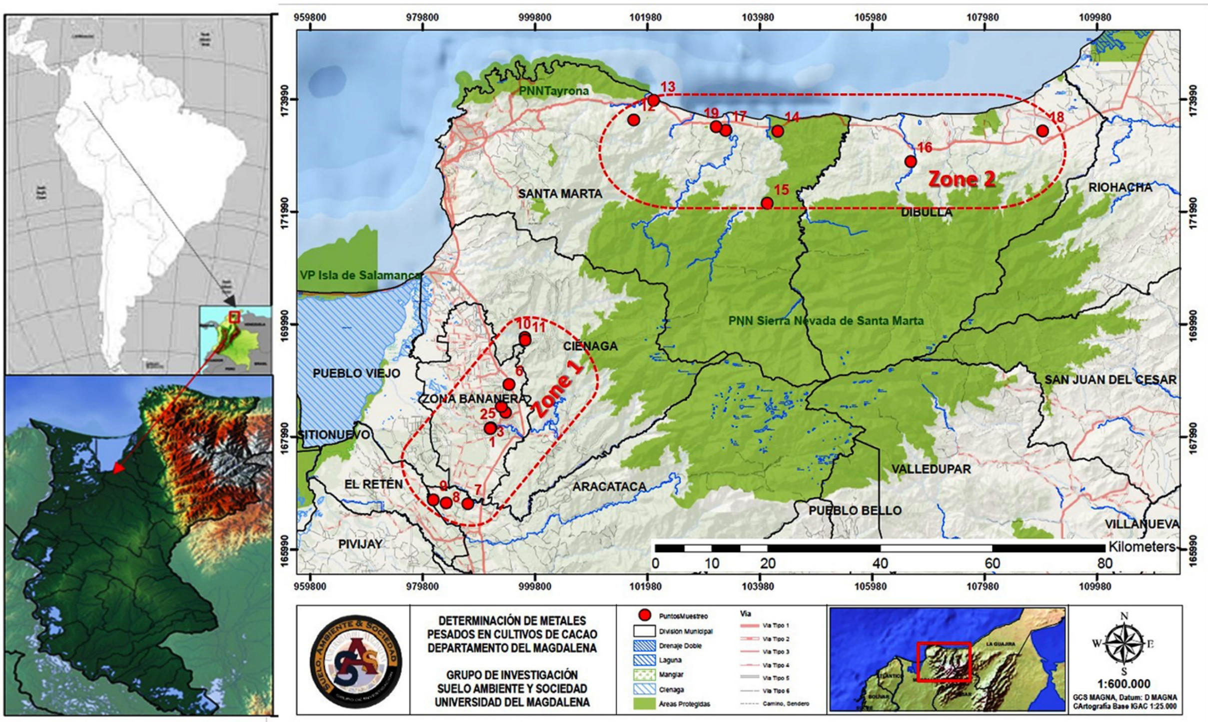

Figure 1. Location of the agricultural plots and sampling zones where Cd content in the cocoa tissues and soils cultivated with cocoa was identified.
Source: Authors

plot. The samples were air-dried, then, were sieved through a 2-mm mesh, packed in plastic bags, and tightly sealed (Figure 2).

The tissue and soil samples were taken to the laboratory of the International Center for Tropical Agriculture (CIAT) in Palmira for analysis, where the respective determinations were made per methods described in Table 1.

Statistical analysis

The parameters obtained from the chemical determinations (plant tissue and soil) were subjected to analysis of variance and Tukey's post hoc test ($p < 0.05$). The results that differed were subjected to Pearson's correlation analysis between the heavy metal content in the plant tissue and soil properties. Statistical analyses were conducted using R software v. 3.02 (R Development Core Team 2015, available at www.r-project.org).

3. Results

Heavy metals in plant tissues

The mean Cd, Ni, Pb, Cr, Ca, Mg and K content in the beans, shells and leaves of T. cacao grown in the study area are shown in Appendix 1. The beans from zone 1 had a mean Cd content of 0.51 mg kg^{-1}, whereas the beans from zone 2 had a mean of 0.66 mg kg^{-1}. These values exceeded the maximum permissible limit (0.5 mg kg^{-1}) reported by García and Dorronsoro (2002). The beans from plot 3 of zone 1 had a Cd content of 0.96 mg kg^{-1}, and those from plots 17, 11 and 15 of zone 2 had Cd contents of 1.25 mg kg^{-1}, 1.08 mg kg^{-1} and 0.86 mg kg^{-1}, respectively. These values are considered unsafe for direct consumption. In contrast, plots 6 and 16 had the lowest Cd contents, with 0.21 mg kg^{-1} and 0.15 mg kg^{-1}, respectively (Figure 3).

The mean Cd content in the leaves did not differ significantly across the analyzed samples, with means of 0.43 mg kg^{-1} in zone 1 and 0.41 mg kg^{-1} in zone 2. The highest Cd content was reported in plot 6, at 0.81 mg kg^{-1}, and in plot 11, at 1.24 mg kg^{-1}.

The Cd content in the shells was heterogeneous, but without significant differences. The mean values were 0.50 mg kg^{-1} in zone 1 and 0.40 mg kg^{-1} in zone 2. These values were lower than those reported by INIAP-PROMSA (2003) in Ecuador, where a mean of 1.08 mg kg^{-1} Cd was recorded in the shells.

The values reported for the shells and leaves generally did not exceed the permissible limits; however, more Cd had accumulated in the beans, with 0.51 and 0.67 mg kg^{-1} for

Figure 2. Cocoa pods and soil samples were collected in cocoa-producing areas in northern Colombia to determine the heavy metal content.
Source: Authors

Table 1.
Laboratory methods used to determine the heavy metal content in cocoa tissue samples from cocoa-producing areas in northern Colombia.

Variable	Method	Reference
pH	Potentiometric – soil: water ratio of 1:1	Jackson (2005)
Organic matter	Walkley and Black method	
Total Cd, Ni, Pd and Zn in the tissues	Regal water (HCl:HNO_3 -3:1), Quantification by atomic absorption spectrometry	NTC 3888, ISO 11466: 1964
Ca, Mg and K	Soils with pH ≤ 5.5 by 1 M ammonium acetate and soils with pH > 5.5 by 1 N KCl and determined by atomic absorption spectrophotometry	Anderson and Ingram (1993)
Total Cd Ni, Pb, Fe, Cr and Zn in the soils	Extraction with diethylenetriamine pentaacetate (DTPA) and quantification by atomic absorption EPA-3050B	US EPA (1996)

Source: Authors

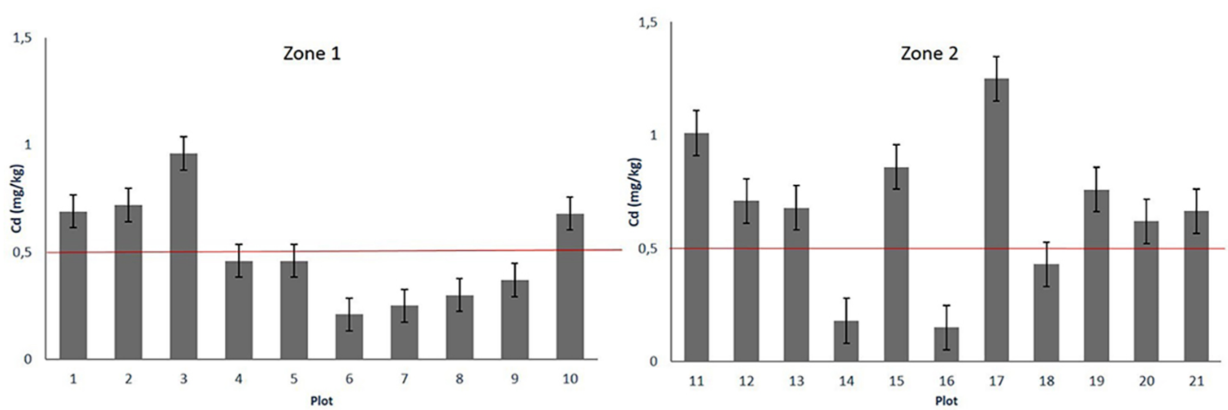

Figure 3. Total Cd content in cocoa beans from cocoa-producing areas.
Source: Authors
Legend: ___ Reference level from García and Dorronsoro (2002)

zones 1 and 2, respectively, demonstrating Cd mobility in the plant. Miranda *et al.* (2008) state that the Cd present in leaf tissue is linked to specific physiological stages and is involved in diverse metabolic pathways.

Heavy metals in soils

Table 2 shows the soil analysis results, in which the pH ranged from 5.7 to 6.07, with no significant differences among sites or between zones. The Fe and Mn levels were optimal (10 to 50 mg kg^{-1}), and the Zn level was adequate in some sites but deficient in others. The Ca^{+2} content ranged from intermediate (3.0 cmol+ kg^{-1}) to high (≥6.0 cmol+ kg^{-1}), the Mg content was intermediate (1.5 to 1,7 cmol+ kg^{-1}), and K was deficient (< 0.2 cmol+ kg^{-1}). For this reason, the Ca/K and (Ca+Mg)/K cation ratios were analyzed, which confirmed that K deficiency may be induced. The percentage of organic matter was low. Regarding the total metal concentrations in the soil, no evidence of Cd, Pb, Ni or Cr contamination was found. Concentrations were not reported from some sites because the values were lower than the method's limit of quantification (< LOQ).

The mean heavy metal content in the analyzed soils did not exceed the maximum concentrations established by García and Dorronsoro (2002). In zone 1, the Cd concentration ranged from 0.19 mg kg^{-1} at 10 cm deep to 0.02 mg kg^{-1} at 20 cm deep. A similar pattern was observed in zone 2, consistent with the studies of Mite, Carrillo, & Durango (2010) and Gramlich *et al.* (2018), who reported surface Cd accumulation. Heavy metal adsorption is strongly conditioned by pH (and thus its solubility) and the organic matter content, clay, oxides and hydroxides in the soil (Olivares-Rieumont *et al.*, 2013).

Bioavailability is associated with soil concentration and plant species. Pearson correlation analysis between Cd, Pb, Ni and Cr content in the soil with foliar variables (bean, leaf and shell content) indicated an association between the heavy metal concentrations in the soil and foliar contents (Table 3).

The soil Cd content correlated positively with the plant Cd content as well as with Ca concentrations in the soil. However, it was inversely proportional to the plant's Ca content, while the plant's Cd content was inversely proportional to the Ca and K uptake.

Association between variables

The results showed that the soil Ni concentration was affected by that of soil Cr and is inversely proportional to

Table 2.
Results of the soil analysis by cocoa-producing zone in northern Colombia.

		ZONE			
		A		B	
Depth		1	2	1	2
pH	Un	5.78	5.81	6.07	5.77
Cox	%	1.16	0.72	1.06	0.66
MO	%	2.36	1.63	2.41	1.50
P	mg kg^{-1}	37.30	40.38	24.56	15.47
Ca	cmol kg^{-1}	6.00	5.53	5.58	4.74
Mg	cmol kg^{-1}	1.56	1.50	1.69	1.60
K	cmol kg^{-1}	0.10	0.09	0.21	0.18
Fe	mg kg^{-1}	21.62	29.07	18.37	17.81
Mn	mg kg^{-1}	16.07	14.78	28.60	22.25
Zn	mg kg^{-1}	1.95	2.21	3.20	1.38
Cd	mg kg^{-1}	0.02	0.00	0.00	0.00
Pb	mg kg^{-1}	0.57	0.47	6.80	2.78
Ni	mg kg^{-1}	18.15	14.98	11.60	20.13
Cr	mg kg^{-1}	39.97	31.80	20.60	29.80

Depth: 1 = 0–10 cm, 2 = 10–20 cm; Cd, Pb, Ni and Cr concentrations were determined by inductively coupled plasma mass spectrometry at the CIAT Laboratory.
Source: Authors

Table 3.
Pearson correlation between the Cd, Pb, Ni and Cr content available in the soil and their relationship to other nutrients.

Metal	Ca-p	K-p	Cd-p	Cr-p	Ca-s	Mg-s	K-s	Fe-s	Ni-s	Cr-s	pH
Pb-s						0.51	0.75	-0.39			
Cd-s	-0.4		0.3		0.34					0.02	
Cd-f	-0.29	-0.39									
Ni-s										0.412	-0.37
Ni-f	-0.59										
Cr-s				0.41							-0.37
Cr-f									0.62		

Significant correlation ($P < 0.05$); s = soil, p = plant tissue. s = soil, f = aerial organs
Source: Authors

the foliar Ca content as well as the pH. Although Cr poses no toxicological risk, it was positively correlated with the Ni content in the soil and is affected by the pH. Ni and Cr in the soil were inversely correlated with the pH, indicating that this parameter directly affects their solubility.

Figure 4 shows the association between the studied variables regarding the heavy metal concentrations in the cocoa crop in the cocoa-producing areas of northern Colombia. Larger balls indicate greater association, blue represents positive associations, and red represents inverse relationships, which confirm the previous analysis.

5. Discussion

Heavy metals in plant tissues

Chávez *et al.* (2015) reported Cd values of 0.02 to 3 mg kg^{-1} in cocoa beans in southern Ecuador. Gramlich *et al.* (2018) recorded Cd levels higher than 1.1 mg kg^{-1} in beans from Honduras. Huamaní-Yupanqui et al. (2012) reported a mean Cd content of 1.55 mg kg-^{1} in cocoa beans from the province of Leoncio Prado in Peru, and Martínez and Palacio (2010) found Cd content ranging from 4 to 7 mg kg^{-1} in San Vicente de Chucurí, Santander department, Colombia. These concentrations were higher than those found in the present study area.

Nevertheless, the results of the Cd analysis for the cocoa beans of some plots exceeded the maximum value cited by García and Dorronsoro (2002) and therefore, the value allowed by Lanza, Churión, Liendo, & López (2016) and the EU. This finding suggests the need to further investigate Cd's origin to build a distribution map and monitor concentrations in the beans by zone.

The lowest leaf Cd content was observed in plot 17, at 0.20 mg kg^{-1}, but in this plot, the Cd content of the beans was relatively high, indicating Cd mobility in the plant. This finding agrees with the report by Caunii *et al.* (2015), who cautioned about Cd mobility in leaves and considered the maximum tolerable Cd content to be 0.5 mg kg^{-1}.

Finally, the values reported for the shells and leaves generally did not exceed the permissible limits; however, more Cd had accumulated in the beans, with 0.51 and 0.67 mg kg^{-1} for zones 1 and 2, respectively, demonstrating Cd mobility in the plant. Miranda *et al.* (2008) state that the Cd present in leaf tissue is linked to specific physiological stages and is involved in diverse metabolic pathways. Likewise, Pernía, De Sousa, Reyes, & Castrillo (2008) reported that Cd uptake by the roots reflects its concentration and availability in the soil.

Heavy metals in soils

Regarding the total metal concentrations in the soil, no evidence of Cd, Pb, Ni or Cr contamination was found. Concentrations were not reported from some sites because the values were lower than the method's limit of quantification (< LOQ). Thus, the mean heavy metal content in the analyzed soils did not exceed the maximum concentrations established by García and Dorronsoro (2002). In zone 1, the Cd concentration ranged from 0.19 mg kg^{-1} at 10 cm deep to 0.02 mg kg^{-1} at 20 cm deep. A similar pattern was observed in zone 2, consistent with the studies of Mite, Carrillo, & Durango (2010) and Gramlich *et al.* (2018), who reported surface Cd accumulation and that Heavy metal adsorption is strongly conditioned by pH (and thus its solubility) and the organic matter content, clay, oxides, and hydroxides in the soil (Olivares-Rieumont *et al.*, 2013).

Although Roberts (2014) reported that applied phosphorus fertilizers (phosphate rock) are a source of Cd contamination in agricultural land and constitute more than 50% of the Cd

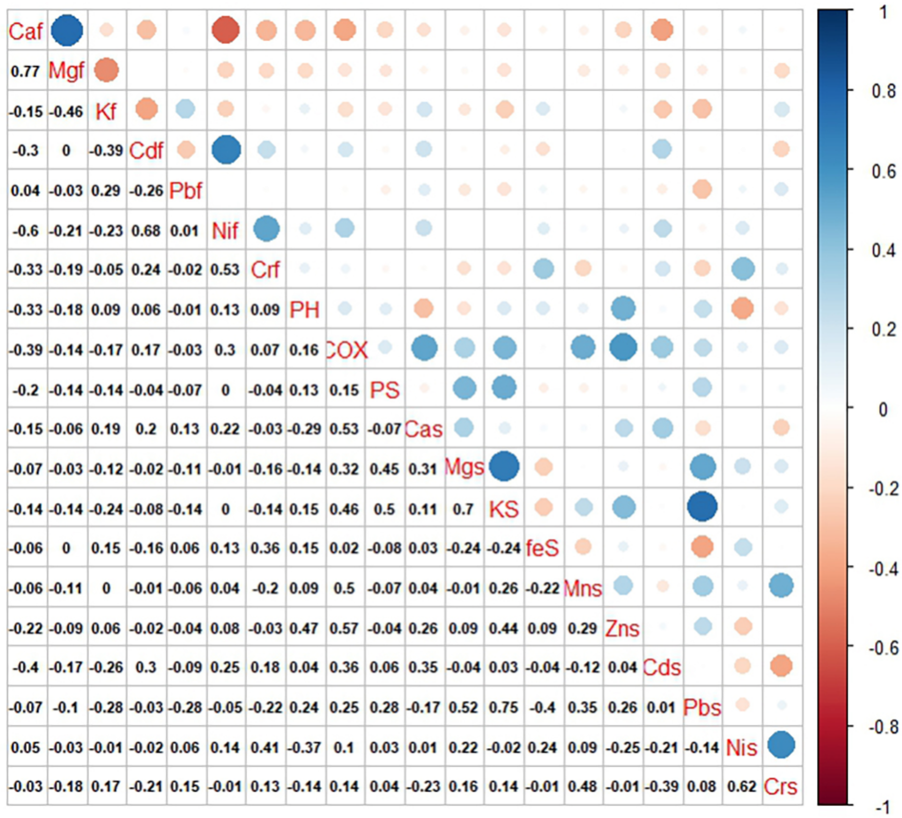

Figure 4. Associations between the variables measured in the soil and aerial organs of cocoa plants grown in northern Colombia. Blue circle = positive associations, and red circle = inverse relationships.
Source: Authors

that enters into the soil (this hypothesis is not tested here). However, given its extensive use, phosphate rock may be a possible contamination route for Cd in this area, further studies with more focus on fertilizers, are suggested in the north of Colombia.

The solubility and availability of metals in soils is affected by the interactions between pH, cation exchange capacity (CEC), precipitation, complexation and chelation. In this regard, Silveira *et al.* (2003) reported that when metals adsorb to soil particles, their concentration in the solution is reduced; thus, the metals cannot be quantified therefore, soils with high CECs have more exchange sites available in the colloidal fraction for adsorption and possible immobilization. Likewise, Contreras, Herrera and Izquierdo (2005) identified that soil Ca is found both as free Ca and as carbonate and calcium phosphate; therefore, Cd can be controlled by alkalization and precipitated.

Further research is required to assess the risk of Cd soil contamination, and regular monitoring of heavy metals should be carried out to evaluate the existence of any potential threat to food safety. For this, it is necessary to advance in new detection techniques such as the use of two-dimensional electrical resistivity tomography described by Bravo and Benavides (2020) or application of organic amendments like biochar for reducing the soil Cd movement and Cd plant uptake reported by Cui *et al.* (2011), which promotes the soil quality, soil health, as well as food security.

Association between variables

Regarding the Ca and Cd association, Cui *et al.* (2011) and Solti *et al.* (2011) mentioned that high Ca concentrations in soils can reduce Cd uptake because ionic competition occurs between these elements, thus reducing their uptake. Likewise, Solti *et al.* (2011) reported that Cd entering the roots directly competes with Ca, K, Fe and Cu because the same transporter proteins carry out Cd absorption. Huamaní-Yupanqui *et al.* (2012) explained that for Cd to precipitate, it must have affinity for carbonates or phosphates, even if pH is slightly acidic.

The solubility and availability of metals in soils is affected by the interactions between pH, cation exchange capacity

(CEC), precipitation, complexation and chelation. Soils with high CECs have more exchange sites available in the colloidal fraction for adsorption and possible immobilization. Likewise, as the soil Ca is found either in free form or as carbonate and calcium phosphate, therefore, Cd can be controlled by alkalization and precipitated (Olivares-Rieumont *et al.*, 2013).

Carbonate presence in the study soils was not analyzed, but the study area's geological characteristics and proximity to the sea indicate that carbonate may have been present; thus, insoluble Cu, Zn and Cd compounds can form and be adsorbed by carbonates, and Ca and Cd in the soil may remain adhered and undetectable this is an essential issue for future research. So, the fact that soil Ni and Cr correlated inversely with pH, is consistent with that reported by Chávez *et al.* (2015), who reported that low redox potential increases heavy metal solubility, and with the Agency for Toxic Substances & Disease Registry (ATSRD, 2012), which maintains that Ni is immobile in alkaline soils and precipitates in the form of carbonates and insoluble phosphates. This is important to select new areas for the cocoa crop.

The foliar calcium content was associated with most of the variables studied and was inversely proportional to Cd, Ni, Pb and Cr uptake. If Ca uptake is increased, cocoa metal uptake may decrease. Although the calcium concentration in the soil is in the intermediate range, increasing it would be beneficial without neglecting the cationic relationships. Further research should focus on the application of soluble Ca effect on cocoa Cd uptake growing in northern Colombia.

6. Conclusions

The results of this study lead to the following conclusions regarding the heavy metals content in soils and cocoa tissues in northern Colombia:

- The cadmium concentration in the cocoa beans fluctuated between zones and plots, but the values all exceed the permissible limit for this element in cocoa beans (0.5 mg kg^{-1}), indicating that the grains are contaminated. However, the soil Cd concentration shows that soil is optimal for cocoa cultivation, which suggests that the Cd origin is anthropogenic and related to crop management. Still, the correlation with other plant nutrients suggests that soil carbonates or organic amendments can reduce Cd uptake.
- These results serve as a reference point for future research about heavy metals presence on cocoa, with an emphasis on the need for regular monitoring of soil Cd and plant Cd contents, to evaluate the risk for food safety. For this, currently, techniques with promising efficiencies are being developed to reduce energy and operating costs during the sampling stage, allowing for their implementation and sustainability.

Conflict of interest

The authors have no conflicts of interest to declare.

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Appendix

Appendix 1.
Heavy metal content in the cocoa beans, shells and leaves from cocoa-producing areas in northern Colombia.

Plot	Zone	Plant material	Ca	Mg	K	Cd	Pb	Ni	Cr
			g kg^{-1}			mg kg^{-1}			
1	1	Bean	1.33	3.73	15.42	0.69*	0.49	12.66	13.3
	1	Shell	11.15	4.10	24.5	0.38	0.26	2.99	0.67
	1	Leaf	19.79	6.86	15.5	0.43	2.12	4.03	1.09
2	1	Bean	1.76	4.41	19.71	0.72*	0.58	17.6	15.9
	1	Shell	10.37	4.35	30.02	0.23	7.46	5.00	1.98
	1	Leaf	17.07	5.37	19.45	0.35	1.20	2.56	0.75
3	1	Bean	1.86	4.19	20.20	0.96*	3.44	16.4	0.96
	1	Shell	6.18	3.30	29.13	0.15	0.87	5.13	1.38
	1	Leaf	14.15	5.34	26.54	0.25	4.61	5.46	1.93
4	1	Bean	0.79	4.11	19.82	0.46	1.86	6.26	1.29
	1	Shell	7.98	3.76	30.98	0.20	0.61	3.94	1.53
	1	Leaf	7.98	6.01	21.90	0.15	0.26	4.96	1.34
5	1	Bean	0.99	3.64	15.17	0.46*	1.62	6.06	1.21
	1	Shell	7.82	3.62	29.17	0.16	0.92	4.34	1.24
	1	Leaf	19.08	8.55	14.26	0.81	1.42	8.93	1.03
6	1	Bean	1.14	3.37	18.75	0.21	0.47	8.54	0.90
	1	Shell	3.04	2.7	27.76	0.55	0.32	4.91	1.87
	1	Leaf	12.4	6.94	21.57	0.73	2.6	6.33	1.33
7	1	Bean	1.58	4.26	15.23	0.25	0.98	7.87	0.92
	1	Shell	8.94	5.37	29.01	0.53	0.30	3.76	3.17
	1	Leaf	10.85	5.20	28.89	0.19	2.09	3.44	1.05
8	1	Bean	0.83	3.81	14.42	0.30	0.71	7.32	0.89
	1	Shell	6.72	3.07	39.33	0.12	0.22	2.26	2.13
	1	Leaf	18.66	7.35	14.31	0.27	2.87	2.95	2.04
9	1	Bean	0.69	3.47	14.92	0.37	0.28	8.08	2.53
	1	Shell	4.05	4.57	57.6	0.28	0.06	4.75	0.83
	1	Leaf	17.4	8.02	15.24	0.61	0.3	4.89	0.65
10	1	Bean	0.72	3.80	14.44	0.68*	0.48	6.77	2.48
	1	Shell	2.77	2.65	41.66	0.56	0.10	5.25	1.22
	2	Leaf	19.63	7.33	18.78	0.52	0.28	4.45	0.51
11	2	Bean	1.20	3.87	20.99	1.01	0.57	9.76	0.73
	2	Shell	15.53	7.98	19.75	0.24	0.28	2.10	0.39
	2	Leaf	17.57	5.91	13.85	1.24	0.68	5.78	1.60
12	2	Bean	0.96	4.04	16.95	0.71*	0.80	7.71	1.45
	2	Shell	11.00	4.62	29.35	0.22	0.55	4.22	2.86
	2	Leaf	13.10	5.82	15.56	0.19	0.54	3.13	1.71

Continued on next page

Plot	Zone	Plant material	Ca	Mg	K	Cd	Pb	Ni	Cr
				g kg^{-1}			mg kg^{-1}		
13	2	Bean	1.01	3.99	12.84	0.68*	1.13	15.5	0.95
	2	Shell	9.30	4.68	26.37	0.13	9.00	6.89	2.46
	2	Leaf	21.25	7.73	14.56	0.59	0.53	6.41	0.91
14	2	Bean	1.41	3.69	13.82	0.18	0.60	5.04	1.60
	2	Shell	9.08	3.22	23.93	0.87	0.72	5.99	1.32
	2	Leaf	10.69	5.65	24.44	0.40	0.51	3.08	1.38
15	2	Bean	1.08	4.00	16.68	0.86*	0.41	13.06	1.85
	2	Shell	18.00	6.28	16.80	0.74	0.75	4.40	0.45
	2	Leaf	18.35	7.47	14.54	0.15	0.59	4.34	0.90
16	2	Bean	1.46	4.76	18.62	0.15	0.38	16.02	1.06
	2	Shell	12.08	4.43	28.49	0.41	3.65	4.04	2.89
	2	Leaf	16.50	6.00	15.45	0.28	0.51	2.12	1.26
17	2	Bean	1.77	4.53	14.13	1.25	0.53	14.64	1.40
	2	Shell	15.36	4.47	21.66	0.33	0.32	2.69	0.63
	2	Leaf	14.6	6.38	12.61	0.20	0.43	2.37	0.78
18	2	Bean	2.27	3.91	14.48	0.43*	0.34	6.58	0.98
	2	Shell	14.92	3.95	17.86	0.24	0.67	4.65	1.13
	2	Leaf	15.42	7.23	13.78	0.35	0.52	2.39	0.61
19	2	Bean	3.36	3.79	16.85	0.76*	0.36	11.54	1.12
	2	Shell	16.07	4.18	19.25	0.52	0.67	2.14	1.24
	2	Leaf	10.23	4.87	20.7	0.35	0.50	8.73	0.78
20	2	Bean	1.66	4.84	20.59	0.62*	0.37	10.84	0.64
	2	Shell	1.20	3.94	21.04	0.37	0.70	2.98	0.97
	2	Leaf	15.53	5.91	14.78	0.32	0.67	4.82	0.61

Source: Authors