Science of the Total Environment 649 (2019) 120–127

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Soil properties and agronomic factors affecting cadmium concentrations in cacao beans: A nationwide survey in Ecuador

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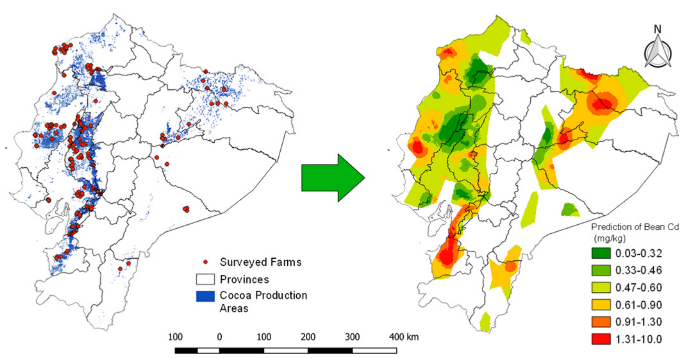
HIGHLIGHTS

- Spatial distribution of Cd concentrations in cacao beans is proposed for Ecuador.
- 45% of samples exceeded the 0.60 mg kg^{-1} (dry weight bases) threshold.
- Total soil Cd, pH and % organic carbon are the main factors affecting Cd concentration in beans.
- Soil to bean transfer factors suggest a high affinity of cacao plants for Cd uptake.
- Cd hotspots were identified in several cocoa producing regions of Ecuador.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 8 May 2018
Received in revised form 20 August 2018
Accepted 21 August 2018
Available online 23 August 2018

Editor: Charlotte Poschenrieder

Keywords:
Cadmium cocoa
Soil survey
Spatial distribution
Ecuadorian cacao

ABSTRACT

Recent cadmium (Cd) regulation in chocolate threatens the sustainability of cacao production in Southwest America. Cadmium contamination in cacao beans has not been assessed at a country level. A nationwide survey was conducted in Ecuador to identify the spatial distribution of Cd in cacao beans, as well as soil and agronomic factors involved. Paired soil and plant samples (pods and leaves) were collected at 560 locations. Information on agronomic practices was obtained through a prepared questionnaire for farmers. Total soil Cd averaged 0.44 mg kg^{-1} which is typical for young and non-polluted soils. Mean Cd concentration in peeled beans was 0.90 mg kg^{-1} and 45% of samples exceeded the 0.60 mg kg^{-1} threshold. Bean Cd hotspots were identified in some areas in seven provinces. Multivariate regression analysis showed that bean Cd concentrations increased with increasing total soil Cd and with decreasing soil pH, oxalate-extractable manganese (Mn_{ox}) and organic carbon (OC) ($R^2 = 0.65$), suggesting that Cd solubility in soil mainly affects Cd uptake. Bean Cd concentration decreased a factor of 1.4 as the age of the orchard increased from 4 to 40 years. Bean Cd concentration was inconsistently affected by genotype (CCN-51 *vs.* Nacional), pruning or application of fertilizers. It is concluded that the relatively larger bean Cd concentrations in Ecuador are related to the high Cd uptake capacity of the plants combined with their cultivation on young soils, instead of Cd depleted weathered soils. Mitigation strategies should consider the application of amendments to modify such soil properties to lower soil Cd availability. There is scope for genetic mitigation strategy to reduce bean Cd, but this needs to be properly investigated.

1. Introduction

Cacao bean, the seed of *Theobroma cacao* tree, is an important agricultural commodity for several South American countries (Brazil,

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https://doi.org/10.1016/j.scitotenv.2018.08.292
0048-9697/

Colombia, Ecuador, Peru, Venezuela) and Europe is one of its main destination markets. In Latin America, Ecuador is the major cacao producing country with a production of 260,000 Mg in 2015 (ANECACAO, 2016) which represents 35% of total exports from Latin American countries (ICCO, 2016). Ecuadorian cacao is renowned worldwide for its fine aroma and flavour (Beg et al., 2017). Despite the good reputation of Latin American cacao beans, elevated concentration of toxic trace metals, such as cadmium (Cd), have been reported in cacao beans and chocolate produced in this region (Abt et al., 2018; Arévalo-Gardini et al., 2017; Chavez et al., 2015; Gramlich et al., 2018). Multi-elemental fingerprinting analysis of cacao beans of different origin showed that the concentration of Cd in beans from South America was nearly three times higher than in those from Central America and East Africa and ten-fold above those of West Africa (Bertoldi et al., 2016). That analysis suggested that the widespread high Cd concentrations in South America as well as in Central America (see below) might be of geogenic origin, unrelated to point pollution. The European Union (EU) has defined maximum permissible levels for chocolate and other cacao derived products. This regulation, which will be implemented in January 1st 2019, has set threshold limits ranging from 0.10 to 0.80 mg Cd kg^{-1} dry matter, depending on the percentage of raw cacao present in the final product (The European Commission, 2014). There are no limits on Cd in cacao beans but thresholds of 0.6 and 0.8 mg Cd kg^{-1} have been used as values that allow the manufacturing of cacao products meeting the EU Cd limits (Arévalo-Gardini et al., 2017; Barraza et al., 2017; Chavez et al., 2015; Gramlich et al., 2018). Recent surveys conducted in cacao farms in Peru, Ecuador and Honduras have concluded that the high Cd concentrations occur in localized regions. Arévalo-Gardini et al. (2017) reported concentrations of Cd in cacao beans in Peru where the lowest Cd concentration occurred in Cuzco whereas the highest was found in Tumbes, near the Ecuadorian southern border. In southern Ecuador, cacao beans collected in 19 farms showed that 66% of the samples exceeded the 0.60 mg kg^{-1} threshold (Chavez et al., 2015) and a similar survey in 31 farms in six provinces of Ecuador found that 53% of samples surpassed a 0.80 mg kg^{-1} threshold (Barraza et al., 2017). A recent survey in 55 farms in Honduras reported an average Cd concentration in beans of 1.1 mg kg^{-1} (Gramlich et al., 2018). These surveys indicate that the upcoming EU regulation will affect a large fraction of the cacao production in the region of origin of this crop.

Yet, few studies have presented information explaining the relationships between soil and agronomic factors and the Cd concentrations in cacao beans (Arévalo-Gardini et al., 2017; Chavez et al., 2015; Gramlich et al., 2018). This relationship is well established for other crops where Cd uptake in plants is enhanced at low soil pH, low soil organic matter content and high salinity. All these factors have been related to increased Cd mobility in soils. In addition, low soil zinc (Zn) availability increases Cd uptake due to Zn-Cd interaction during uptake and translocation (Mitra, 2015). Gramlich et al. (2018) found that Cd in the cacao bean was best predicted by the diffusive gradient in thin film (DGT) available soil Cd ($R^2 = 0.53$). The authors reported DGT as a tool to measure Cd diffusive fluxes (mobility) in cacao-growing soils. Agricultural practices and genetic factors could also influence Cd concentrations in foodstuffs. Gramlich et al. (2017) investigated leaf and bean Cd concentrations in relation to production systems (monoculture *vs.* agroforestry), management (organic *vs.* conventional), genotypes and soil properties. They found only weak effects of production systems, management and genotype, with a small (<factor 2) genotype effect on bean Cd in the monoculture and lower leaf Cd in agroforestry systems, explained by species competition effects (Gramlich et al., 2017).

Here, a large-scale survey on Cd concentrations in Ecuadorian cacao areas was set up for three purposes: (i) to identify the cacao production areas in Ecuador potentially affected by the new EU regulation, including a stringent quality assurance (QA) protocol which was not published in a large fraction of the existing cacao bean Cd surveys in South-America; (ii) to assess the spatial variability of Cd in cacao beans and to employ geostatistical tools to construct a map at a country level which can be further used to establish mitigation strategies in the regions of concern (*i.e.* where bean Cd exceeds 0.6 mg kg^{-1} dry weight); (iii) to determine the relationship between soil and agronomic factors controlling Cd accumulation in cacao beans as weak and inconsistent data have been presented in previous studies. The latter may be related to the limited sample size that reduces the statistical power, *e.g.* the maximal number of samples in a similar study was 110 (55 farms, 2 trees per location) from the studies cited above.

This nation-wide survey comprises 560 paired soil-plant samples that were collected at 159 farms throughout the cacao production areas in Ecuador. The sampling strategy was set up to measure spatial variability and to cover major cacao production areas allowing mapping of bean Cd concentrations at national scale. Soil properties and elemental composition of beans and leaves (as a proxy for nutrient status) were measured. Agronomic and edaphic factors were recorded, and all data were analyzed with multivariate approaches.

2. Materials and methods

2.1. Study area and sampling strategy

In Ecuador, cacao is mainly grown in the Coastal plain and Amazonia regions, which are characterized by an equatorial humid and tropical savannah climate. Cacao is widely cultivated in 21 provinces of Ecuador, but a large percentage of the production is concentrated in the Coastal region. For this study, farms were surveyed in 15 provinces which represent 97% (INEC, 2016) of the total production area (Fig. 1). The average annual rainfall in the sampled provinces ranged from 250 to 5500 mm y^{-1} (MAG, 2002) and the altitude of surveyed farms varied from 5 to 1000 m above sea level (Table S1). The survey was conducted from July to November 2017 in 159 cacao farms. In total, 560 paired samples of soil, leaves and cacao pods (beans) were collected; each single tree being one of the 560 observations. The surveyed farms were selected based on the following criteria: production area (≥1 ha), availability of mature pods, distance between the fields (>4 km) and accessibility for sampling. The number of soil-plant paired samples collected in each province was determined according to its overall production area (Table S1). On each farm, 2–18 cacao trees were randomly selected depending on the size of the field, *i.e.* three trees sampled for every five hectares. The coordinates were recorded using a GPS (Garmin® eTrex 30). The soil classification (U.S. soil classification system (Soil Survey Staff, 2003)) was derived from a digital soil map of Ecuador with 1:25000 resolution (Instituto Espacial Ecuatoriano, 2017).

A total of 15 to 20 middle-aged leaves and 2 to 3 mature cacao pods were collected from each tree. In addition, a composite soil sample was collected by combining 6 to 8 equidistant topsoil samples (0–15 cm depth) that were taken at 70 cm from the main trunk. The 0–15 cm topsoils were collected as this corresponds to the layer where most active roots are located. Soil samples were homogenized in a plastic bucket and a subsample of 0.5 kg was stored in Zip-lock® bags. During the field survey, a short questionnaire was prepared to obtain information from the farmers regarding agronomic management practices (*e.g.* application of fertilizers, pesticides, organic amendments) (Fig. S1), age of the orchard, genotype, irrigation, conventional or organic farming and agroforestry or monoculture system, the latter factor determined as the mixture of cacao trees with other crops such as fruits (citrus, banana, plantain, among others) or timber trees.

2.2. Sample preparation

Soils were air-dried for 72 h and sieved through a 2-mm stainless steel mesh. Leaves were washed with deionized water to remove adhering particles and thereafter oven-dried at 65 °C for 48 h. Dried leaves were ground using a kitchen blender and passed through a 0.16-mm