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# Soil cadmium uptake by cocoa in Honduras

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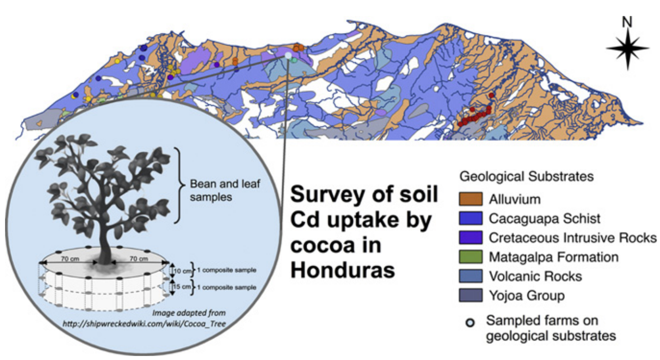
## HIGHLIGHTS

- Bean Cd exceeded European standards in some areas, although soils were uncontaminated.
- DGT-available soil Cd ($Cd_{DGT}$) best predicted bean and leaf Cd.
- Cadmium concentrations in cocoa beans were highest on alluvial substrates.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Cadmium (Cd) is a trace metal without essential biological functions that is toxic to plants, animals and humans at low concentrations. It occurs naturally in soils, but inputs from anthropogenic sources have increased soil Cd contents worldwide. Cadmium uptake by cocoa (*Theobroma cacao* L.) has recently attracted attention, after the European Union (EU) decided to bring into force values for maximum Cd concentrations in cocoa products that would be exceeded by current products of various provenances from Latin America. In order to identify factors governing Cd uptake by cocoa, we carried out a survey on 55 cocoa farms in Honduras in which we determined Cd concentrations in cocoa leaves, pod husks and beans and analysed their relationships to a variety of surrounding soil and site factors. Averaging $2.6 \pm 0.4$ mg kg$^{-1}$, the concentrations of Cd were higher in the leaves than in the beans. With an average of $1.1 \pm 0.2$ mg kg$^{-1}$, the bean Cd concentrations still exceeded the proposed EU limit, however. The bean Cd showed large differences between geological substrates, even though regional variations in 'total' soil Cd were comparably small and the average concentration was in the range of uncontaminated soils ($0.25 \pm 0.02$ mg kg$^{-1}$). As we found no influence of fertilizer application or vicinity to industrial sites, we conclude that the differences in soil Cd between sites were due to natural variation. Of all factors included here, DGT-available soil Cd was the best predictor of bean Cd ($R^2 = 0.5$). When DGT was not considered, bean Cd was best predicted by 'total' soil Cd, pH and geology. The highest bean Cd concentrations were found on alluvial substrates.



## 1. Introduction

Cadmium (Cd) is a trace metal without essential biological functions that is toxic to plants, animals and humans at low concentrations. The primary natural source of Cd in soils is the weathering of parent material

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(Adriano, 2001). The mobility of Cd in soils is high compared to other heavy metals, and it is taken up readily by plants, even though it has no essential biological functions (McLaughlin and Singh, 1999). While soil Cd can be naturally enriched to concentrations of 2 mg $kg^{-1}$ and more, especially in soils developed on alluvial sediments or on andesite and other volcanic rocks (Fauziah et al., 2001; Manton, 2013), anthropogenic inputs originating from sources such as mining and smelting, the plastic and microelectronics industry, and rock-based fertilizers are the primary cause of elevated Cd contents in many soils (He and Singh, 1994; Ross, 1994).

Cadmium uptake by cocoa plants (*Theobroma cacao* L.) has recently attracted increasing public attention, after rather high concentrations of Cd and lead (Pb) were found in cocoa products (Manton, 2013). Many chocolate products contained >100 μg $kg^{-1}$ Cd and thus Cd levels that were higher than those of other food products (European Food Safety Authority, 2012). According to a report of the European Food Safety Authority (2012), chocolate confectionary products contributed on average 4.3% to the weekly Cd intake of the European population. Responding to public health concerns caused by these figures, the European Union (EU) defined maximum values for Cd in cocoa products imported to the EU to be enforced in 2019 (The European Commission, 2014). While protecting the health of consumers, these regulations are a potential threat to the livelihood of many cocoa-growing smallholder farmers, in particular for producers of high-value cocoa used in products with high cocoa contents, such as dark chocolate. Thus, there is a need to identify the factors that govern Cd accumulation in cocoa beans and to find measures to reduce its concentration.

While cocoa products from Latin America generally contain higher Cd concentrations than those from West Africa (Mounicou et al., 2003), considerable variation in bean Cd contents has also been found within Latin America, between as well as within countries (Chavez et al., 2015; Gramlich et al., 2017; Mite et al., 2010; Mounicou et al., 2003). The variability in Cd concentrations in cocoa beans from different sites has been attributed to different soil parent materials (Manton, 2013), but also other factors must be considered, such as Cd inputs with phosphorus (P) fertilizers (McLaughlin and Singh, 1999; Zarcinas et al., 2004) or soil properties influencing the availability of soil Cd for uptake by plants (Chavez et al., 2015).

Honduras is one of the countries where elevated Cd contents in cocoa beans were observed recently by the chocolate industry. Even though most cocoa farmers do not add organic or mineral fertilizers to their plantations and many farms are certified for organic production in Honduras (Fromm, 2013), applications of fertilizers and pesticides in the past are still a possible source of soil contamination Cd. Intensive banana production was widespread in the second half of the last century and many cocoa farms have been established on former banana plantations. Further inputs to be considered as potential sources, at least locally, are atmospheric deposition of emissions from industries in the vicinity of cocoa fields or diffuse pollution due to flooding or irrigation with polluted river water.

Apart from the 'total' soil Cd content, soil pH, texture and organic matter are important soil factors influencing Cd availability to plants (Adams et al., 2004; Fauziah et al., 2001; Kirkham, 2006). Increased Cd uptake has also been found in plants deficient in zinc (Zn) compared to plants with adequate Zn nutrition (Choudhary et al., 1995; Dar et al., 2012; Oliver et al., 1994). One explanation for this finding is competition of Zn and Cd for the same carriers or transporters in the root cell membranes (Hart et al., 2002). However, there are also studies that did not find an antagonistic effect of Zn on Cd uptake and the interaction between the two metals may even vary among different cultivars of the same plant species (Köleli et al., 2004; Nan et al., 2002; Sanaeiostovar et al., 2012). Cadmium accumulation was also found to vary with age and season in trees (Lettens et al., 2011).

Only a few studies have investigated the effects of soil and other environmental factors on heavy metal uptake by cocoa, especially under field conditions (Chavez et al., 2015; Fauziah et al., 2001; Gramlich et al., 2017; Ramtahal et al., 2016; Ramtahal et al., 2015). In two of the studies good correlations between plant Cd and 'total' soil Cd were found (Fauziah et al., 2001; Ramtahal et al., 2016). In a further study EDTA, DTPA and Ammonium-Bicarbonate-DTPA extractions were found to be suitable methods to predict the available Cd to cocoa plants (Ramtahal et al., 2015). In the two other studies from Ecuador (Chavez et al., 2015) and Bolivia (Gramlich et al., 2017), the 'available' soil Cd (Mehlich 3 or diffusive gradients in thin films (DGT) method) in combination with the soil organic matter were good predictors of the plant Cd content. An additional effect of the clay content was observed in one of them. While in the study from Bolivia only the leaf concentration could be modeled, in the one from Ecuador a good prediction of the bean Cd was achieved (Chavez et al., 2015; Gramlich et al., 2017). The reason for this difference may be that a much broader concentration range was covered in the study from Ecuador.

The objective of this study was to identify sources of Cd in the soils of cocoa plantations in Honduras and the factors governing its accumulation in cocoa. For this purpose, we carried out a survey on 55 cocoa farms in northern and eastern Honduras in which we determined Cd concentrations in cocoa leaves, pod husks and beans and analysed their relationships to a variety of soil and site factors at the scale of individual trees.

## 2. Materials and methods

### *2.1. Survey area*

Cocoa is grown in the northern and eastern parts of Honduras (Fromm, 2013), where the climate is mostly an equatorial monsoon climate (Am) according to the Köppen classification (Kottek et al., 2006). The annual temperatures vary between 20 and 35 °C in the low lands, with maxima in May and June, and the average monthly rainfall varies between about 100 mm in April and May and up to 500 mm in October, November and December.

Cocoa production is mostly in the hands of smallholder farmers in Honduras, with farm sizes ranging from less than one up to six hectares. Most farmers grow cocoa in agroforestry systems in addition to fruit and timber trees (Fromm, 2013). On the surveyed farms, we frequently found the legume tree Madreado (*Gliricidia sepium*), Caoba (*Swietenia macrophylla*), various species of the genus *Inga*, and fruit trees such as orange, mango or lemon. Most smallholder cocoa farms are managed organically. Mineral fertilizers were applied only on a few farms surveyed and not on a regular basis. The age of the sampled cocoa plantations varied greatly, from 3 to >30 years.

The survey was carried out in December 2014 and January 2015 on 55 cocoa farms in the political departments Santa Bárbara, Cortés, Atlántida, Yoro and Gracias a Dios (Figs. 1, S.1). The farms were selected to represent a wide range of soil types, site conditions (geology, topography, climate) and management schemes. The geological substrates included alluvial sediments, cretaceous intrusive and undifferentiated volcanic rocks, Cacaguapa schist, and rocks of the Matagalpa formation and Yojoa group according to USGS classification (US Geological Survey, 2016). We divided the alluvial sites into four geographic groups for the analysis of the results (wherever enough samples were available the division was made by catchment, Fig. 1). According to the FAO Classification Map of Honduras (1998), the soil types included Fluvisols, Nitosols, Cambisols and Regosols.

### *2.2. Soil and plant sampling*

On each farm, two cocoa trees with mature fruits were selected at random and their location recorded using GPS. A total of 10 medium aged leaves were collected from each tree by taking the 9th and 10th leaf of 5 different branches, counting from their tips. In addition, 1–4 mature pods, depending on availability, were collected from each tree, separated into beans and pod husks and combined to one composite