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1. Introduction

Amazonian Dark Earths (ADE) are anthropic soils (Anthrosols) characterized by darker colour and enrichment in carbon (C) and other nutrients, particularly calcium (Ca) and phosphorus (P). These soils also exhibit higher pH and cation exchange capacity (CEC) and frequently evidences of human activity (e.g. ceramic artefacts and charcoal fragments) when compared to the carbon- and nutrient-poor adjacent soils from the Amazon basin (Glaser et al., 2001; Kämpf et al., 2003; Kern et al., 2017; Sombroek, 1966). Radiocarbon dating indicated that these soils were formed between 2500 and 500 before present and are of pre-Columbian origin (Neves et al., 2003). Possible carbon and nutrients sources are: terrestrial/aquatic plant biomass, human/animal excrements/bones and charcoal/ash residues of incomplete combustion (Glaser, 2007).

Due to the prevalence of weathered clay minerals (e.g. kaolinite) and iron and aluminium oxides in these soils, their ability to retain nutrients depends mainly on soil organic matter (SOM). However, high temperature and precipitation in the tropics accelerate the decomposition of SOM. Studies have suggested that high carbon content in ADE are related to the black carbon (BC) content in these soils (Glaser, 2007; Glaser et al., 2001). BC has been claimed to be one of the most stable forms of carbon found in soils due to its poly-condensed aromatic structure that makes it more difficult to be decomposed by soil microorganisms (Glaser, 2007; Haumaier and Zech, 1995; Novotny et al., 2007). In addition, high CEC in ADE is also likely to increase primary production due to higher soil fertility. Therefore, SOM input of non-BC origin is also expected to be increased in ADE. Despite tropical conditions, ADE have intriguingly remained highly fertile after abandonment of sites following European colonization. Current research indicates that the extent and intensity with which Amerindians occupied and

transformed the Amazon is far more complex than previously assumed due to possible environmental limitations (Kern et al., 2017). Studies on ADE empower the understanding of complex pre-Columbian cultural development in the Amazon and may also provide insights for future sustainable agricultural practices in the tropics.

ADE are highly variable in size, depth and soil physico-chemical characteristics not only among different sites, but also within a single site. This variation is mainly caused by the diversity and complexity of pre-Columbian settlements (Costa et al., 2013; Costa and Kern, 1999; Kern et al., 2015). The debate whether ADE were formed intentionally (for agricultural purposes) or unintentionally (as the unintended consequence of waste deposition), seems to be diminishing in relevance. Currently, scientists tend to perceive the formation of ADE (and other Anthrosols) as the inevitable outcome of daily activities throughout years of past human occupations (Fraser et al., 2014). The persistence of anthropic markers likely depends on frequency and intensity of occupation of the site that may lead to a regime shift which manifests as ADE formation (Browne-Ribeiro, 2016).

The differentiation between ADE and the adjacent soil is commonly done by non-quantitative field observations based on soil colour and the presence of archaeological remains (i.e. ceramic and charcoal fragments). Costa et al. (2013) reported that ADE characteristics could fit several qualifiers for Anthrosols. Recently, the pretic horizon has been proposed as an attempt to classify ADE systematically and better accommodate ADE within Anthrosols (IUSS Working Group WRB, 2015). This is important because it takes into consideration quantitative data rather than vague descriptive data. The pretic horizon is a dark surface horizon that among other criteria is characterized by high contents of carbon and nutrients.

Geochemical signatures reflect the variation of major elements within and beyond the limits of anthropic areas that were previously

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defined by visual evidence. Schmidt et al. (2014) reported a widespread pattern of past human occupation where terraces of domestic areas (e.g. houses or yards) are surrounded by waste disposal areas as middens that build up into mounds over time. Different geochemical signatures can be linked to past land use and occupation (Costa et al., 2013). However, interpretation of these patterns may be hindered by site-inherent complexity of past settlements and current land use and occupation (Kern et al., 2015). Moreover, anthropic soils likely exhibit these complexities both in small- and large-scale analyses. Hence, we still lack a solid understanding of the specific mechanisms that led to the formation and diversity of ADE (Schmidt et al., 2014). Limited and localized soil sampling is unlikely to elucidate those mechanisms in highly variable areas.

Spatial modelling techniques can be of great use to study the structure of the spatial variation of soil properties as it considers the continuity of spatial phenomena and the deterministic effect of environmental conditions. For soil scientists, it is a great tool to visualize how soil properties can vary greatly both horizontally and vertically. For archaeologists, it is a great tool to infer on the location of specific activities in the past. Significant progress in spatial modelling techniques was possible due to recent advances in data processing. However, several methodological hurdles are still evident, especially in large areas with high spatial variation between soil properties and environmental covariates (Song et al., 2016). These hurdles can be of great importance in anthropic areas where abrupt and gradual transitions can be expected horizontally and vertically due to the complexity of settlements. Therefore, it is important to include uncertainties of predictions when using these techniques.

Correlation between soil variables is not only dependent on the distance between sampling points, but also on their location. Therefore, environmental conditions may show a trend across a study area. Stochastic simulation of spatially distributed soil properties can be used for better predictions as it preserves the structure of the spatial variation, whereas kriging usually smoothens (Heuvelink and Webster, 2001). Predictions may be improved by using exhaustive environmental covariates (Lark and Webster, 2006). However, including several covariates is not always related to an increase in prediction accuracy (Samuel-Rosa et al., 2015).

Therefore, the aims of this study were to: (i) predict the Total C, Total Ca and Total P stocks using an environmental covariate (including the uncertainties of predictions) and (ii) use the pretic horizon criteria to classify pretic and non-pretic areas and evaluate their relative contribution to the total stocks.

2. Material and methods

2.1. Study area

The study area (~9.4 ha) is situated on the north margin of the Solimões river (Amazon river), in the municipality of Iranduba, Amazonas state, Brazil (03°14'22" - 03°15'47" S and 60°13'02" -60°13′50" W) (Fig. 1). Regional climate is classified as Aw according to Köppen classification (tropical rainy). Local annual mean temperature is 26.7 °C, annual mean rainfall is 2100 mm and relative humidity is about 80%. The local slope is flat to undulated and the site is located above existing watercourses. The most common soil classes in the region are Xanthic Ferralsols, Plinthic Ferralsols, Pisoplinthic Plinthosols and Xanthic Acrisols (Macedo et al., 2017). The study area is locally known as the Experimental Research Station of Caldeirão - Embrapa Western Amazon. Part of the study area (~30%), located in the west (W) and southwest (SW) sectors, is composed of a forested area that has not been cultivated for over 40 years. The remaining part (\sim 70%), was converted into an experimental field where several crops have been cultivated and soil management practices have been applied over the past 40 years.

2.2. Soil sampling and analysis

Soil samples were collected from five 20-cm soil layers (from 0 to 100 cm) with a manual post hole digger (sample volume = 0.0063 m^3) at n = 53 georeferenced points (~5 m horizontal precision) placed in a grid of about 10 to 60 m spacing (265 soil samples). Ceramic fragments (> 2 mm) were weighed and quantified. Samples were air-dried, sieved through 2 mm mesh, homogenized and stored in plastic bags at room temperature prior to analyses. Samples were analysed for: Total C, Total Ca, Total P, Exchangeable Ca + Mg, Extractable P, soil pH, potential CEC (at pH = 7.0) and clav content. Total C was determined using an elemental analyser (PerkinElmer 2400 Series II) where acetanilide was used as reference material. Total Ca and Total P were determined at Geosol laboratories by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES), where samples were previously digested with a multi-acid solution (HCl, HNO3, HF and HClO4). Extractable P, K, Na (Mehlich 1), Exchangeable Ca + Mg (1 M KCl) and H + Al (0.5 M calcium acetate at pH 7.0) were also determined. Soil pH was determined in water (soil:water ratio of 1:2.5), potential CEC (at pH = 7.0) was defined as the sum of exchangeable cations (K, Na, Ca + Mg) plus acidity (H + Al) and the clay content was determined by the pipet method after organic matter removal with hydrogen peroxide. Detailed description on the methods are described in Embrapa (2017).

2.3. Spatial modelling

The spatial variation of soil properties was modelled as a function of fixed (deterministic) and random (stochastic) effects. The fixed effects describe the part of the spatial variation of a soil property that can be explained using spatially exhaustive covariates (Heuvelink and Webster, 2001). Spatial data covering the entire study area that can be related to the environmental conditions that likely influenced the observed large-scale patterns of spatial variation (> 50 m in our study area) can be used as covariates. Here, we assumed that past anthropic activities that caused enrichment of carbon and nutrients likely occurred closer to the river, despite specificity among activities. In addition, the current land use in the SW sector (forested area) is also expected to have caused some enrichment of organic matter due to SOM input, whereas the current land use in the NE (agronomic experimental field) is expected to have caused some impoverishment of organic matter due to cultivation. Therefore, the largest enrichment of carbon and nutrients likely occurred in the SW sector with decreasing enrichment gradient towards the NE sector. Because of the spatial association between these conditions, we chose to use one covariate to serve as their surrogate, which we defined as the expected anthropic enrichment gradient (Fig. 2a).

The understanding that this covariate could explain the large-scale spatial variation of soil properties was formalised by individually calibrating depth-wise linear regression models with soil property as dependent variable and the covariate as the independent variable. For an arbitrary soil depth (*d*), such linear regression model was defined as:

$$Y(\mathbf{s}i, d) = \beta_{0d} + \exp[\mathbf{x}(\mathbf{s}_i, d)]^{\mathrm{T}} \beta_{1d} + \varepsilon(\mathbf{s}_i, d), \text{ with } i = 1, 2, ..., n,$$
(1)

where the β 's are the estimated linear regression model coefficients conditional on the soil property (*Y*) and the covariate data (*x*) at the observation locations (s_i , d). The covariate is expressed in exponential form to emphasise the combined effect of past and current land use and occupation on enrichment of carbon and nutrients nearby the margin of the river.

In Eq. (1), $\varepsilon(s_b, d)$ is the spatially auto-correlated difference between the fitted and observed values of the soil property (regression residuals) (Heuvelink and Webster, 2001). For an arbitrary soil depth (*d*), the structure of this spatial autocorrelation was analysed using the autovariogram:

$$\gamma_d(\boldsymbol{h}) = 0.5 \operatorname{mean}\{[\varepsilon(\boldsymbol{s}_i, d) - \varepsilon(\boldsymbol{s}_i + \boldsymbol{h}, d)]^2\}$$
(2)

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