Small-scale spatial variability of Technosol properties in a chronosequence of reclamation of dredged sediment landfills *(Running title: Spatial heterogeneity of reclaimed mining waste)*

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Data availability

The datasets generated during the current study are available in the Data Portal of the Center for Development Research - University of Bonn [https://www.zef.de/header/data-portal.html]

Author contributions

All authors contributed to the study conception and design. Material preparation and data collection were performed by Bibiana Betancur-Corredor, Juan Sebastian Perez-Jimenez, Santiago Cardona-Gallo and Juan Carlos Loaiza-Usuga. Data analysis and draft of the

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Abstract

Active reclamation is often necessary to ensure a transformation of mining waste into Technosols -“soils dominated or strongly influenced by human made material” - and restore its utility and environmental value. The objective of this study was to assess the spatial variation of the physicochemical properties of a Technosol forming on dredged sediment landfills left by alluvial gold mining in a chronosequence of reclamation (0, 4, 8 and 12 years). We hypothesized a higher spatial dependency of most soil properties with increasing time of Technosol formation and an overall homogenization of the soil resulting from pedogenetic processes. Our results showed that most of the investigated physical and chemical properties changed significantly among Technosols of different ages. The content of organic matter, phosphorus, and exchangeable cations showed the highest spatial variability in Technosols of all ages. In older Technosols, most soil properties showed less spatial variability than in younger Technosols. A multivariate geostatistical assessment allowed the delineation of spatial clusters i.e. homogeneous zones with distinctive physicochemical properties within areas of the chronosequence. This spatial clustering showed that reclamation and Technosol formation led to spatially-dependent fragmentation processes reflected in more and smaller homogeneous zones in the oldest Technosol assessed in the chronosequence. From the perspective of reclamation management, understanding the spatial variability of highly heterogeneous Technosols where substantial changes can be observed within small distances can support the development of reclamation strategies suitable to the characteristics of each field as well as the determination of its potential uses.

Keywords: Technosol reclamation, mine spoil restoration, spatial heterogeneity, early pedogenesis, space for time substitution.

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1 Introduction

Natural succession frequently fails to successfully reestablish the vegetation cover in sites heavily disturbed by mining (Ruggles *et al.*, 2021), or occurs at slower rates than expected due to poor substrate conditions (Gilland & McCarthy, 2014). In mining waste deposits, active reclamation is often necessary to ensure a transformation of the mining waste into Technosols (“soils dominated or strongly influenced by human made material” (IUSS, 2014; Schad, 2018)) and restore its utility or environmental value (Otremba *et al.*, 2021). In Colombia, several approaches have been developed to reclaim dredged sediment landfills resulting from alluvial gold mining activities through reforestation and agroforestry (Castellanos Barliza & Leon Pelaez, 2010; Mosquera *et al.*, 2008; Thomas, 2014). The main objective of reclamation is to restore native flora and fauna and support settlers in the mining-affected areas through wood, food and grazing opportunities (Betancur-Corredor *et al.*, 2018). The reclamation projects are of crucial importance in some regions of the country, as in 2016, approximately 79,000 ha were covered by dredged sediments (UNODC, 2016), with an increase of 44,746 ha between 2001-2014. These trends are expected to continue as the Colombian gold production continues to increase (UPME, 2021). Furthermore, 24,450 of natural forest and secondary vegetation are lost per year to the deposition of dredged sediments (UNODC, 2016).

The nature of the technogenic material that forms the dredged sediment landfills poses challenges for the spontaneous colonization by plants (Betancur-Corredor *et al.*, 2020). As these dredged sediments have been heavily washed for ore extraction, they are characterized by low macronutrient levels and low pH. Also, there is a discontinuity between surface and deeper layers of the soil profile as the deposition process takes place in layers (Bini & Bech, 2016). Furthermore, the physicochemical conditions of the dredged sediment landfills show great variability which can be related to the characteristics of the spoils, mining operations and stacking (Monterroso *et al.*, 1998). The spatial and temporal variability of the natural undisturbed soils results from the interaction of natural processes and management practices (Panakoulia *et al.*, 2017). In Technosols forming on dredged sediment landfills, variability also depends on the type of dredging machinery and on the properties of the parent material (Shlyakhov & Osipov, 2004).

In spite of the many challenges for reclamation, the technogenic surfaces have the potential of recovering some of the ecosystem services expected for healthy soils, such as carbon sequestration, nutrient provision and sustainment of biological activity even after short periods of reclamation (Ruiz *et al.*, 2020a). The Technosols often have a natural reserve of nutrients coming from primary materials, likely originated from mineral weathering (Ruiz *et al.*, 2020b). Furthermore, the afforestation of mine waste can generate economic, social and environmental benefits to the population in the region (Ahirwal & Pandey, 2021). The content of fine soil fractions, moisture, organic carbon (Bandyopadhyay *et al.*, 2020), as well as aggregate stability (Fourvel *et al.*, 2019) are important factors that control the capacity of the Technosol to recover functional nutrient dynamics. In some cases, ameliorative measures i.e. amendments are needed to improve the substrate for Technosol formation (Carabassa *et al.*, 2020), and toxic effects on faunal groups may be observed in some cases, due to high salinity, low pH or toxic substances (Vezzone *et al.*, 2020).

Chronosequences i.e. space-for-time substitutions are fundamental tools for studying environmental change in a spatial sequence of soils in which the only factor of formation that varies is the age of the surface on which the soil was formed (Phillips, 2015). The characterization of chronosequences is often used to examine to pedogenetic evolution occuring in Technosols of different ages: Ortega *et al.* (2022) detected advanced structural development and nutrient contents in 15 year-old Technosols forming on iron mine tailings, through a chronosequence analysis of micromorphological, physical and chemical properties; Pihlap *et al.* (2021) used a chronosequence approach to study soil aggregate formation in young agriculturally reclaimed Technosols, and determined that fresh soil organic matter contributes predominantly to macroaggregate formation; Wang *et al.* (2018) used a chronosequence approach to assess reclamation of gold-mining tailings and determined that the organic carbon accumulation is restricted in the long term by the total nitrogen content. Chronosequence approaches have also been used to assess the recovery after disturbance by estimating the spatial variability of the thickness of organic layers in forest stands (Boerner *et al.*, 1998) as well as spatial patterns of nutrient availability and organic carbon after mining disturbances (Bens *et al.*, 2006).

Technosols have been studied at different spatial scales, from micromorphological analyses (Watteau *et al.*, 2019) and lysimeter studies (Tifafi *et al.*, 2017) to field-based physicochemical characterizations (Leguédois *et al.*, 2016), and at different temporal scales, from a few months of formation (You *et al.*, 2018) to several decades (Coussy *et al.*, 2017). However, to the best of our knowledge, the assessment of the changes in spatial heterogeneity of physicochemical properties in a chronosequence of Technosol reclamation has not yet been conducted. To better manage, classify and map highly disturbed areas, it is necessary to perform a temporal and spatial characterization of the substrate properties that shows the magnitude of spatial patterns and relations, which would not be determined using nonspatial statistical methods (Demyan & Smeck, 2021). Mining waste deposits are often characterized by high spatial variability with no spatial dependence (Keck *et al.*, 1993). The estimation of the spatial dependence of soil properties and their multivariate covariance simultaneously can elucidate complex interactions among several soil properties in a spatial context (Dray *et al.*, 2008). Understanding the patterns of spatial variability is particularly important for mining-derived residues, as they exhibit high variability even in short distances of less than 100 m (Haering *et al.*, 2005).

The objective of this study was to assess the spatio-temporal changes of the physical and chemical properties of a Technosol forming on a deposit of dredged sediments deposit left by alluvial gold mining in a 12 year chronosequence. The specific objectives of this study were to implement a chronosequence approach to (1) assess the spatial heterogeneity of the physicochemical characteristics of Technosols of 0, 4, 8 and 12 years of formation on dredged landfills, (2) to examine the spatial dependence of the Technosol properties, (3) to assess overall spatial variation of the Technosol using multivariate geostatistical techniques. We hypothesize (1) a higher spatial dependency of most soil properties with increasing time of vegetation establishment and (2) an overall process of homogenization of the soil resulting from pedogenetic processes.

2 Materials and methods

2.1 Study area

Our study site (Figure 1, 7°48'11"N - 7°51'30"N, 74°48'10"W - 74°46'42"W, Altitude: 38.4 - 66 m) is located in the alluvial gold mining area of El Bagre, Antioquia (mean annual temperature: 28°C, mean precipitation: 2000-4000 mm). The dredging operations take place in areas with low-lying forest that remain flooded during the rainy season (April - October). The dredged sediment landfills consist of rock fragments coming from material extracted by bucket dredges, as well as sand from material extracted by suction dredges. The washed dredged sediments form dumps, which modify the channels, beds and slopes of river valleys (Egidarev & Simonov, 2015). The dredged sediments landfills deemed suitable for reclamation are flattened with heavy machinery to a slope below 10%. The main types of reclamation sites are gravel and sand landfills. These landfills are characterized by a heterogeneous texture, with gravel of varying diameter, and very poor water retention capacity. Depending on the site location and stability of the landfills, some specific areas are selected to be used as farmlands, in which woody, herbaceous species and perennials are planted with crops and livestock. The soils forming on the dredged sediment landfills are classified as *Spolic-Relocatic Dystric Ochric Technosols* (Wartenberg, 2015). The micromorphological details and profile descriptions of these deposits can be found in (Betancur-Corredor *et al.*, 2020).

A chronosequence was assessed to investigate the spatial and temporal soil dynamics of areas of different ages undergoing reclamation. A series of reclaimed areas established after 4, 8 and 12 years of the establishment of vegetation cover named as T4, T8 and T12. An area without vegetation was selected as a reference (T0). T0 consists of 46.6 ha, largely covered by spontaneous herbaceous vegetation, mostly *Rubus ulmifolius*. A large proportion of this reclamation area was bare soil. T4 consists of 49.6 ha with an important fraction of spontaneous vegetation and grasses. T8 consists of 46.2 ha covered mostly by grass and tree plantations of native species. T12 consists of 54.9 ha mostly covered by single-species stands. The most abundant species in the four areas of the chronosequence are *Acacia mangium* (Fabaceae), *Cecropia peltata* (Urticaceae), *Casearia* sp. (Salicaceae) and *Vismia* sp. (Hypericaceae).

2.2 Experimental design and sampling

A sampling grid consisting of 312 sampling points spaced 100 m apart (78 sampling points for each of the four reclamation sites) was created (Figure 1). In each area, once cell of the grid was randomly sampled in a smaller grid pattern of 25 m spacing. An auger (10 cm diameter) was used to sample sand deposits. In gravel deposits, a shovel was used. Samples were taken at a 0-10 cm depth, passed through a 2-mm sieve and air dried before laboratory analysis. The following soil analyses were conducted: pH (potentiometric method 1:1 water), organic matter (OM; Walkey-Black method), total nitrogen (N; Kjeldahl method), available phosphorus (P; modified Bray-II method), calcium (Ca), magnesium (Mg), soil texture (Bouyoucos hydrometer), bulk density (BD; soil cores dried at 105°c for 48 hours), penetration resistance (PR; penetrograph Eijkelkamp, Netherlands), sampling coordinates (portable GPS (Garmin eTrex 10, Germany)).

2.3 Data analysis

The data collected on soil physicochemical properties were strongly non-normal and resistant to transformation. For this reason, non-parametric tests were chosen for this study. Permutational analyses of variance were conducted to compare each soil parameter across the different Technosol ages. The $p-values$ were calculated on 10000 permutations of the data using the *adonis* function of the R *vegan* package (Oksanen *et al.*, 2020). Pairwise multilevel comparisons (Martinez, 2020) were conducted among the different Technosol ages to assess significant differences among the different sites of the chronosequence.

2.3.1 Geostatistical analysis of soil properties

A general nonparametric procedure was conducted to model the spatial correlation (Fernández-Casal *et al.*, 2018). For each soil property, semivariograms were calculated using local polynomial kernel smoothing of linearly binned semivariances, using the *np.svar* function of the *npsp* R package (Castillo-Páez *et al.*, 2019). Maximum likelihood estimation of the parameters of the theoretical variogram from the best-fit model (spherical, exponential and Gaussian) was conducted: nugget variance (τ^2), partial sill (σ^2), practical range (ϕ), anisotropy angle (ψ_A), and anisotropy ratio (ψ_R). These estimated parameters were then used for spatial interpolation of the soil properties to analyze their spatial distribution (Bivand *et al.*, 2008). To define classes of spatial dependence of each soil property, we calculated the nugget to sill ratio (NSR (%)): (1) Strong spatial dependence $NSR < 0.25$, (2) moderate spatial dependence ($NSR : 0.25 - 0.75$), (3) weak spatial dependence ($NSR > 0.75$) (Zimmermann *et al.*, 2008).

Moran's I statistic was used to analyze the spatial autocorrelation of the variables. If the global Moran's index (which varies from -1 to 1) is negative, it shows that a variable has a negative spatial autocorrelation, and if the value is positive, it indicates positive spatial autocorrelation (Bivand *et al.*, 2008). The contribution of each observation to the global indicator was determined by the estimation of the local Moran's index (Anselin, 2019). The *moran.test* and *localmoran* functions of the *spdep* R package were used for calculations of global and local Moran's I test (Bivand & Wong, 2018). For further geostatistical analyses, the influential points detected by the estimation of the local Moran's I test were removed from the database. The interpolated maps with a spatial distribution of each soil physicochemical property across the four areas of the chronosequence can be found in the supplementary material (Supplement 4).

2.4 Multivariate site classification

Wartenberg (1985) developed a strategy for multivariate exploratory analyses of spatial patterns, elaborating on methods from principal component and factor analyses. A database of predicted soil properties was created to store the results of spatial interpolation (kriging). A principal component analyses (PCA) was conducted on the predicted soil properties with the *dudi.pca* function of the *ade4* R package (Chessel *et al.*, 2004). A multivariate spatial correlation analysis of the principal components was conducted, by using a multivariate extension of the univariate method of the spatial autocorrelation analysis with the *multispati* function of the *ade4* R package. A fuzzy c-means algorithm was applied to the two first principal components to form the different spatial clusters; each cluster corresponds to a zone with homogeneous soil properties. The analysis was performed using the *cmeans* function of the *e1071* R package (Meyer *et al.*, 2014). The optimal number of clusters was calculated based on a summarizing index ($SI = \sqrt{XB^2 + PE^2 + PC^{-2}}$) proposed by Galarza *et al.* (2013), which summarizes the results of known cluster validity indices such as Xie-Beni (XB) (Xie & Beni, 1991), Fukuyama-Sugeno (FS) (Kwon, 1998), partition coefficient (PC), and partition entropy (PE) (Tang *et al.*, 2005). This methodology for spatial clustering was adapted from Córdoba *et al.* (2016) for the use of non-parametric geostatistical techniques for spatial interpolation, which was necessary due to the highly skewed sets of values of soil properties (See Supplementary material - Supplement 1).

3 Results

3.1 Physical and chemical changes

The inherent heterogeneity of these Technosols was reflected in the high standard deviation of values of each soil property. The soil was strongly acidic, with a predominant sandy fraction. The values for pH, N, BD and PR were symmetrical in contrast to textural parameters such as sand (skewness = -1.18) and clay (skewness = 2.10), which were highly skewed. Density graphs for each parameter in each reclamation area were provided as supplementary material (Supplement 1).

Most of the investigated physical and chemical properties had significant changes among Technosols of different age (Table 1). The median pH changed significantly among Technosols of 0 and 4 ($p = 0.002$), 0 and 8 ($p = 0.001$), 0 and 12 ($p = 0.001$), 4 and 12 ($p = 0.01$) years of age, with lower values in older Technosols. The median content of N and OM changed significantly among Technosols of 0 and 4 ($p = 0.001$), 0 and 8 ($p = 0.001$), 0 and 12 ($p = 0.001$), 8 and 12 ($p = 0.01$) years of age, with lower values in older Technosols. The median content of P was significantly higher in Technosols of 4 and 8 years of age, and significantly lower in Technosols of 12 years of age. The median content of K changed significantly among Technosols of all ages ($p < 0.05$), with the exception of Technosols of 0 and 12 years of age, and was significantly higher in Technosols of 4 and 8 years of age. The median Ca content changed significantly between Technosols of 0 and 4 years of age ($p = 0.016$). The median contents of Mg changed significantly among Technosols of all ages ($p < 0.04$), being significantly higher in older Technosols. The median Na contents changed significantly among Technosols of 0 and 4 ($p = 0.029$), 4 and 8 ($p = 0.005$), 4 and 12 ($p = 0.02$) years of age, and were significantly higher in Technosols of 4 years of age. The soil texture was significantly different in Technosols of 4 years ($p < 0.03$) compared to all other Technosol ages, with significantly higher silt and clay content, and lower sand content. The BD and PR were significantly ($p < 0.004$) different among Technosols of all ages. The BD was significantly lower in older Technosols and the PR did not show a clear pattern of variation with Technosol age.

3.2 Global spatial autocorrelation of Technosol properties

Table 1 shows the global Moran's I of the physicochemical properties of Technosols of different years of formation. The soil pH exhibited a strong spatial autocorrelation (>0.25) in T0. However, in Technosols of more than 4 years of formation, the spatial autocorrelation decreased, and in T12 it was not significant. The content of N and OM exhibited strong spatial autocorrelation in T0, T4 and T8, but much lower values in T12. A similar trend was observed for K and Ca content, which showed strong spatial correlation in T0, T4 and T8, but weak spatial correlation in T12. The P and sand content showed a strong spatial autocorrelation in Technosols of all ages. The silt and clay content exhibited moderate to weak spatial autocorrelation in T8. The BD showed strong spatial autocorrelation in T0 and T8, and moderate to weak spatial autocorrelation in T4 and T12, in contrast to PR which exhibited strong spatial autocorrelation in T4 and T12 and weak to moderate spatial autocorrelation in T0 and T8. The local autocorrelation of each soil property across the chronosequence can be found in the supplementary material (Supplement 2).

3.3 Spatial variability of Technosol properties across the chronosequence

Non-parametric semivariograms were calculated for each soil parameter in the four areas (See Supplementary material - Supplement 3), and theoretical models were used to find the best fit for each case (Table 1). The content of OM ($\sigma^2 = 12, \phi = 262.1m$), Ca ($\sigma^2 = 4.83, \phi = 163.1m$) and Mg ($\sigma^2 = 0.58", \phi = 202.8m$) exhibited the highest spatial variability in T0. In T4 the highest spatial variability was exhibited by P ($\sigma^2 = 34.32, \phi = 165.9m$), followed by Ca ($\sigma^2 = 2.61, \phi = 364.4m$), OM ($\sigma^2 = 2.31, \phi = 100m$) and Mg ($\sigma^2 = 0.38, \phi = 339.7m$). In the T8, the highest spatial variability was exhibited by P ($\sigma^2 = 126.6, \phi = 575.4m$), followed by OM ($\sigma^2 = 9.79, \phi = 307.6m$), Ca ($\sigma^2 = 1.03, \phi = 202.1m$) and sand content ($\sigma^2 = 0.7, \phi = 353.1m$). In T12, the highest spatial variability was exhibited by P ($\sigma^2 = 44.02, \phi = 768.6m$), followed by OM ($\sigma^2 = 2.74, \phi = 201.2m$), Ca ($\sigma^2 = 1.22, \phi = 119.2m$) and Mg ($\sigma^2 = 0.88, \phi = 526.7m$). Most of the soil properties had less spatial variability in the Technosol of 12 years of age, compared to the non-reclaimed area. Furthermore, the nugget to sill ratio (NSR) showed that in T0 most of the variables exhibited moderate to weak spatial dependency. In T4, only BD exhibited strong spatial dependency. In T8 most variables exhibited strong spatial dependency, with exception of Ca, Mg and PR. In T12 most variables exhibited weak spatial dependency, with exception of the textural variables.

3.4 Multivariate geostatistical assessment: spatial clustering

To summarize the variability of the soil properties, a multivariate spatial principal component analysis was performed (Figure 2) for the spatial clustering of the Technosol. A high percentage of the variability ($\approx 70\%$ - see Supplement 7) could be explained by the first two spatial principal components (sPC1: horizontal axis, sPC2: vertical axis) in Technosols of different ages, hence only the first two axes were used for the cluster analysis. The graphical display shows the spatial correlation structure between the variables used for spatial clustering in each area. In T0, the content of N, OM and Mg were positively correlated, and alongside pH and sand content were the most important properties explaining spatial variability at SPC1 level. In T4, sand content, P, pH and BD were positively correlated, as well as N, OM, K and Ca. In this area, all soil variables were important to explain the spatial variability at sPC1. In T8, the textural features, BD and P were the most important soil properties explaining spatial variability at sPC1 and SPC2. In T12, three groups of correlated variables could be observed, silt, clay and Mg; P, pH and sand content; N, OM, Ca and K. In this case P was the most important property explaining spatial variability at sPC1, and pH at sPC2. The spatial distribution of the eigenvalues of sPC1 shown in the maps displayed the presence of patches and a clustered structure within the areas of the chronosequence.

The comparison of means for the soil parameters indicated statistically significant differences among all the clustered zones within each area of the chronosequence. In T0, three homogeneous zones were identified (Figure 3, Table 2): Zone T0-A had the highest mean content of N, OM, Ca, and Mg, the lowest mean sand and P content, and the lowest mean BD and pH. Zone T0-C had contrasting properties, with the lowest mean N, OM K, Ca, Mg contents, PR and pH, and the highest mean sand content. Zone T0-B exhibited similar chemical properties to Zone T0-C, and similar sand content but higher mean BD and PR. In T4, three zones were identified: Zone T0-A had the highest mean content of N, OM, K, Ca, Mg, and the lowest mean pH and sand content. Zone T4-C had the highest mean pH, lowest mean N, OM, K, Ca, Mg, the highest mean BD and sand content. Zone T4-B had similar textural features to Zone T4-A, and its chemical properties and BD ranged between those of Zones T4-A and T4-C. In T8, three zones were identified: Zone T8-A had the highest mean content of Ca and Mg, the lowest mean BD but interestingly, the highest mean PR. Zone T8-C had the highest mean sand content, the highest mean BD and lowest mean PR, and the highest mean content of N and OM. Zone T8-B, had the lowest mean OM, K, and Ca content, the highest mean P content and PR, and textural features that range between those of Zones T8-A and T8-C. In T12, four zones were identified: Zone T12-A had the highest mean N, OM, Ca, and the lowest mean sand and P content, and the lowest mean pH. Zone T12-D had the lowest N, OM, K, Ca and Mg mean content, the highest mean sand and P content, the lowest mean BD and highest mean PR. Zones T12-B and T12-C had chemical properties that range between those of zones T12-A and T12-D, but rather different textural and structural features i.e. higher mean sand content and BD, and lower mean PR in zone T12-C compared to T12-B.

4 Discussion

Our results show significant changes in P, K, and Na among the different years of establishment assessed in the chronosequence. The Technosols with more than 4 years of formation had significantly lower pH, lower nutrient and exchangeable cations than T0. This was the result of early pedogenetic processes such as mineral transformations, organic matter accumulation, changes in redox conditions (Huot *et al.*, 2015). The rapid and intense weathering reported during Technosol early pedogenesis is the result of a lack of equilibrium between the technogenic materials and the environmental conditions (Huot *et al.*, 2015). Particularly, in the case of dredged sediment landfills, the transition from a reducing to an oxidizing environment at the surface (Vandecasteele *et al.*, 2009). Séré *et al.* (2010) proposed two phases of Technosol pedogenesis: (1) a first phase (distinctive of Technosols) in which the technogenic materials suffer dramatic changes such as compaction, water release and intense weathering of soluble minerals and (2) a second phase characterized by processes similar to those of natural soils under the same pedoclimatic conditions i.e. decarbonatization and formation of aggregates due to biological activity. We found lower values of BD and PR in T12, compared to T0. One reason for this may be the development of a vegetation layer which increases soil porosity and maintains permeability of the soil (Burylo *et al.*, 2011). Additionally, as the vegetation grows, it consumes the available nutrients in the soil and the biogeochemical cycling of nutrients or the soil amendments may not be sufficient to replenish them (Rodriguez-Vila *et al.*, 2017).

The assessment of the spatial heterogeneity of each individual Technosol property shows that their spatial variability tends to be higher in younger Technosols. The high spatial variability of P content in the Technosols is explained by the primary origin of the alluvial deposits and the predominantly high content of sand. In young floodplain soils, the P retention and release is influenced more strongly by the parent material and sedimentation conditions

(Lair *et al.*, 2009). In alluvial soils, a fairly high proportion of the P in sand fractions is of primary origin, whereas in clay fractions is mostly the result of the organic and inorganic accumulation (Syers *et al.*, 1969). The increase of P spatial variability in T8 and T12 may be due to the establishment of vegetation cover (Chen *et al.*, 1998). Management practices can also influence P spatial variability. For example, the irregular supply of nutrients to the soil, can also result in heterogeneous plant growth and rates of P absorption (Corazza *et al.*, 2003). The high spatial variability of OM can be explained by the fact that these materials are a complex mixture of the materials deposited during alluvial-gold exploitation and natural recent OM, particularly in T12. The OM content in sediments often exhibits high spatial variability, especially in areas where gold mining has taken place (Nascimento *et al.*, 2012). Furthermore, OM is easily affected by climate, and in Technosols can easily be washed out (Komnitsas *et al.*, 2010). In T12, the higher OM spatial variability is explained by the incorporation of fresh OM from vegetation in the surface layer (Huot *et al.*, 2014a). The high spatial variability of OM, particularly in T0, can also explain the high variability of exchangeable cations observed in the study area. Alluvial soils were usually characterized by low K and Mg, therefore most of the ions occur in the horizons enriched with OM as consequence of the biological accumulation of these components (Bartkowiak & Dlugosz, 2010). The high NSR of N, OM and P in T12 compared to T0 may indicate that the spatial variability of these properties in T12 was primarily caused by stochastic factors e.g. vegetation establishment and reclamation activities.

The Technosols forming on dredged sediment landfills have distinctive features and their pedogenesis encompasses significant changes of physical, chemical and biological condition of the soils (Leguédois *et al.*, 2016). Most of the variograms show a considerable nugget effect, which implies that the variability of Technosol properties occurs at a smaller scale than the minimum lag distance assessed (Moral *et al.*, 2010). This nugget effect was more pronounced in T0, where extreme values were likely to be found, and significant variability could be observed in distances as small as 25 m. The heterogeneity of Technosols controls the occurrence of pedogenic processes, which often occurs at different scales, in interfaces between horizons, in cracks and in the rhizosphere (Huot *et al.*, 2014b). Furthermore, the establishment of a vegetation layer has extensive effects on soil nutrients and spatial heterogeneity of the Technosol properties (Wu *et al.*, 2021). The Technosols often exhibit a high rate of development in the first stages of weathering that can vary over short distances due to the their high diversity and spatial heterogeneity (Huot *et al.*, 2015). For this reason, a detailed spatial representation is essential for the understanding of the global effect of the multiscale interactions influencing the pedogenesis of a Technosol (Leguédois *et al.*, 2016).

The estimation of the Global Moran's index showed a strong positive spatial autocorrelation of the physicochemical properties of Technosol of all ages of formation. Our results suggest that in older Technosols the spatial autocorrelation of chemical properties reduces, which was probably a result of Technosol pedogenesis and of the establishment of a vegetation layer which induces spatially random processes on the soil. The positive spatial autocorrelation suggests systematic spatial variation of the Technosol properties (Smelser *et al.*, 2001) and that in each area of the chronosequence the Technosol properties form spatial clusters (i.e. high values cluster near high values; low values cluster near low values). This clustered structure may be the result of waste dumping for landfill formation, disturbance with heavy machinery as well as reclamation management (Wang *et al.*, 2020).

To elucidate how this clustered structure of the Technosols evolves throughout the chronosequence of formation, multivariate geostatistics and spatial clustering were conducted in Technosols of all ages. The spatial distribution of the sPC1 (Figure 2) shown in the maps indicates the presence of patches within the areas of the chronosequence which correspond to positive autocorrelation (Montano & Jombart, 2017) (consistent with Global Moran's index estimations). Several indices (see Supplement 8) were used to assess the appropriateness of the clustering algorithm, particularly the optimum number of clusters for each area of the chronosequence. However, the division of the experimental field into a specific number of homogeneous zones should not only depend on the statistical results of these tests, but also on a logical assessment of the delineated areas (Córdoba *et al.*, 2016) with respect to the feasibility of developing different management strategies for each zone by farmers or reclamation managers. This logical assessment depends on a better separation, less overlap and fragmentation among the delineated homogeneous zones. The number of delineated homogeneous zones within an area of Technosol formation depends on the inherent variability of the Technosol and the physicochemical properties selected for the clustering. Indirectly, it may also reflect the weather and vegetation type established in the Technosols, as well as the sensitivity of measurements of within-field variability (Vallentin *et al.*, 2020). The homogeneous zones produced for this study will probably change if data from different seasons is collected, as it would be possible to account for the effect of climatic factors on soil properties.

The descriptive statistics of Technosol properties of each zone indicate that the delineation i.e. spatial clustering using physicochemical properties allowed the creation of homogeneous zones with significantly different values of each property. The use of soil physicochemical properties sampled in different locations of a field following a grid pattern was a suitable approach to delineate homogeneous zones (Farid *et al.*, 2016). The delineation process showed that in T12, there were more homogeneous zones than in younger Technosols, and the delineation process was much more fragmented in T12 in comparison with younger Technosols. This finding was in agreement with the lower values observed for the Global Moran's index of several properties in T12 which suggest less spatial clustering i.e. more randomness in spatial processes. This suggests that the estimation of spatial heterogeneity with geostatistical techniques from a multivariate perspective may be a more robust approach for the understanding of spatial processes as associations among variables may be precluded by independent analyses and improved inference can be obtained by analyzing the variable dependence (Zhang *et al.*, 2021).

5 Conclusions

The study of a chronosequence of Technosol formation on dredged sediment deposits allowed us to observe significant differences in physicochemical properties across sites of different ages, which can be interpreted as early signs of pedogenesis. Younger Technosols exhibited higher spatial variability of their physicochemical processes and higher spatial autocorrelation (clustering). In older Technosols, the effect of reclamation activities and establishment of vegetation cover plays a more important role in explaining spatial variability. The spatial clustering of the Technosols allowed the delineation of homogeneous zones with significantly different values of Technosol properties, and showed that reclamation and Technosol formation lead to spatially-dependent fragmentation processes reflected in more and smaller homogeneous zones in the oldest Technosol assessed in the chronosequence.

From the perspective of reclamation management, understanding the spatial variability in a chronosequence of Technosols formation can support the development of reclamation strategies suitable to the characteristics of each field and its potential uses. Areas with higher nutrient contents and better physical properties can be identified, as well as areas where protective vegetation is crucial to enhance soil fertility. This is especially required in these highly heterogeneous Technosols where the spatial variability analysis revealed that substantial changes in nutrient levels and organic matter can be observed even within small distances. Further studies need to be conducted to compare the seasonal variability of soil properties and its influence on the delineation process to better understand the spatio-temporal dynamics of the reclamation process.

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7 Statements and declaration

The authors have no relevant financial or non-financial interests to disclose.

8 Supplementary material

Electronic supplementary material available with the manuscript.

Table captions

- Table 1. Descriptive statistics and univariate geostatistical analysis of soil physico-chemical properties of Technosols of different age. Maximum likelihood estimation of parameters of theoretical variogram for nugget variance (τ^2), partial sill (σ^2), practical range (ϕ (m)), nugget to sill ratio (NSR), anisotropy angle (ψ_A) and anisotropy ratio (ψ_R). Units: N (%), OM (%), P ($mg\ kg^{-1}$), K ($cmol_{(+)}\ kg^{-1}$), Ca ($cmol_{(+)}\ kg^{-1}$), Mg ($cmol_{(+)}\ kg^{-1}$), Sand (%), Silt (%), Clay (%), BD ($g\ cm^{-3}$), PR (MPa).
- Table 2. Comparison of delineated homogeneous zones. Mean values of soil properties are shown for each zone. Units: N (%), OM (%), P ($mg\ kg^{-1}$), K ($cmol_{(+)}\ kg^{-1}$), Ca ($cmol_{(+)}\ kg^{-1}$), Mg ($cmol_{(+)}\ kg^{-1}$), Sand (%), Silt (%), Clay (%), BD ($g\ cm^{-3}$), PR (MPa).

Figure captions

- Figure 1. Study area showing the soil sampling points within the four areas undergoing reclamation for 4 (T4), 8 (T8) and 12 (T12) years, as well as the unreclaimed (T0) dredged sediment landfill selected for this study.
- Figure 2. Graphical display of the first two axes of multispatial principal component analysis with soil properties pH, N, OM, P, K, Ca, Mg, Na, sand, silt, clay, bulk density (BD), penetration resistance (PR) for Technosols of 0, 4, 8 and 12 years of formation.
- Figure 3. Homogeneous zones delineated in Technosols of 0, 4, 8 and 12 years of formation.

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