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Remediation/Restoration

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Remediation and rehabilitation of Fundão dam taillings

Rehabilitation of a Riparian Site Reached by Contaminated Tailings from the Fundão Dam Using Different Remediation Strategies in Brazil

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Abstract: The tailings spilled by the Fundão dam rupture in the Doce River basin (Brazil) presented high pH, elevated sodium and ether-amine contents as well as low soil organic matter. Aiming at alleviating these toxic compounds, we established two remediations strategies: T1-phytoremediation with tolerant native species of the Atlantic Forest cultivated on scraped sediment plus the incorporation of organic matter (OM) and T2-phytoremediation with native species plus superficial deposition of OM. The experimental site (ES) was compared with a degraded site (DS) reached by dam tailings and a preserved site (PS), a fragment of preserved Atlantic Forest. After twelve months, plants showed an outstanding growth, especially in T1(~ 4 m) and the remediation procedures resulted in a significant decrease of pH (from 8.0 to ~ 6.0), sodium (from 154 mg/kg to 22–35 mg/kg) and electrical conductivity as well as ether-amine (from 6.0 mg/kg to 0.5 mg/kg) in both treatments. By contrast, ammonium, a product of ether-amine degradation, showed a significant increase in ES, followed by a significant increase of nitrate and improvement of soil microbial populations assessed by phospholipid fatty acid analysis. The treatments also improved the soil fertility in ES, as estimated by soil nutrients, cation exchange capacity (CEC) and soil aggregation. Based on the analysed parameters, a principal component analysis (PCA) showed that samples from DS and PS clustered in an opposed position and those from ES grouped in an intermediate position but closer to PS. Overall, our results demonstrated that the remediation procedures adopted were effective and allowed the rehabilitation of a riparian forest reached by dam tailings contaminated with sodium and ether-amine.

Graphical Abstract

Keywords: ether-amine, Mariana, Samarco, sodium, pH, phytoremediation, reclamation

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INTRODUCTION

The rupture of the Fundão Dam, that belonged to the mining company Samarco S/A, located in the town of Mariana (Brazil), has impacted the Doce River basin, spreading around 43 million m^3 of tailings derived from the iron ore mining beneficiation (Santos et al. 2019). The Fundão dam was used to store the tailings after the iron ore beneficiation performed by the Samarco company via the flotation technique (Batisteli 2007; Filipov et al. 2014), resulting in the production of “superconcentrates” and highly purified hematite products (Peres and Mapa 2008). The reverse cationic flotation technique was employed by Samarco (Araujo et al. 2005; Batisteli 2007; Peres and Mapa 2008) to separate iron minerals from quartz and clays (Batisteli 2007). The main reagents used in this separation process were starches as oxide depressant and amines (Araujo et al. 2005; Filipov et al. 2014) as a cationic collector of quartz minerals as well as a froth stabiliser (Batisteli 2007). It is estimated that Samarco used around 1500 tons per year of amines in this process (Batisteli 2007).

Ether-amine is a highly efficient primary fatty amine that can perform flotation in alkaline pH by the addition of NaOH. Under such alkaline conditions (pH higher than 8.5), the cationic and the molecular species of the amine become stabilised (Araujo et al. 2005; Filipov et al. 2014). In light of this process, Santos et al. (2019) found high contents of sodium (mean of 70 mg/kg) and ether amine (10–20 mg/kg) as well as high pH (8.0) in sediment samples collected along the Doce River basin reached by the dam tailings in 2017. In addition, these authors confirmed the presence of monoamine and diamines in the tailings by chromatographic analyses and, Almeida et al. (2018) also detected the presence of primary and secondary amines and amides in the Fundão tailing. The sediments from the dam presented low fertility patterns with very low organic matter content and the affected site showed an overall plant mortality and low resilience capacity (Santos et al. 2019).

Sodium toxicity may inhibit plant growth, induce senescence and cause plant mortality either by osmotic stress or altering the abscisic acid synthesis, which closes stomata and compromises the photosynthesis (Zhu 2001). Similar to sodium, high pH has been described to cause ecotoxicity to fluvial sediments as observed in bauxite processing residue (red mud) in Hungary (Klebercz et al. 2012). It is well known that sodium may also affect soil properties (Quirk 2001) that assure soil structure such as pH, Cationic Exchange Capacity (CEC), electrical conductivity, soil permeability and aggregation, leading ultimately to plant growth inhibition (Abbas et al. 2013). Furthermore, the sodium and alkaline stresses may also alter the soil microbial community (Frostegåard and Bååth 1996) and cause biodiversity loss (Canfora et al. 2014). Indeed, Santos et al. (2019) confirmed the strong impact of Fundão tailings on soil microbial community.

Amines, in turn, may be mineralised to ammonia/ammonium (NH_3/NH_4^+), which, at high levels, may be toxic to native and agronomic species, causing leaf chlorosis, leaf curling and, ultimately, plant growth suppression, crop decline (Britto and Kronzucker 2002; Roosta and Schjoerring 2007; Coskum et al. 2013) and mortality (Britto et al. 2001; Britto and Kronzucker 2002; Britto and Kronzucker 2013). On the other hand, plant species tolerant to nitrogen overload may exhibit symptoms of toxicity, particularly excessive vegetative growth with stem elongation (Brady and Weil 2007). By contrast, metals levels found in the Fundão tailings did not differ from preserved sites that were not reached by tailings (Santos et al. 2019) nor from the region background soil both before (Santolin et al. 2015) and after (Santos et al. 2019) the dam rupture. Therefore, ether-amine and sodium are the only toxic contaminants identified in dam tailings.

Remediation of toxic compounds may be achieved by removing the pollutants or by reducing their mobility and availability. For that aim, several physico-chemical techniques and products such as extraction (Fedje et al. 2017), surfactants (Mao et al. 2015), chelators (Beiyuan et al. 2018), adsorption, catalysis, ionic exchanges, oxidation and reduction processes (Song et al. 2017) have been widely employed. One of the most common chemical remediation strategies for contaminated soils is the use of surfactants or charged organic materials as humic acids (Conte et al. 2005), biochar or activated carbon (Choi et al. 2014). Indeed, the amendment with organic matter has been proven one of the most successful remediation strategies (Wang et al. 2017) to buffer soil pH and to improve the adsorption of ionic compounds (Santibanez et al. 2008, Gomes et al. 2014). Among the physical remediation procedures, shallow or deep soil scraping or excavation (EERC 2016; Dornbusch et al. 2020) have been recommended to remove or minimise the effect of superficial toxic compounds such as sodium (Dornbusch et al. 2020).

Bioremediation refers to the remediation of pollutants promoted by biological elements such as plants and microorganism via absorption, transformation and degradation reactions. It includes the bio-stimulation, phytoremediation and biodegradation. Bio-stimulation is based on the inoculation of plant-growth promoting microorganisms (Zhou et al. 2016); phytoremediation refers to the pollutant sequestration by plants via different mechanism such as phyto-accumulation, phyto-filtration, phyto-volatilization and phyto-stabilization (Ali et al. 2013; Gomes et al.

2014; Zhou et al. 2016; Biswas et al. 2015; Song et al. 2017), while the biodegradation relies on the microbial degradation of pollutant mediated by plant rizosphere effect (Ali et al. 2013).

Aiming at reclaiming ether-amine and sodium toxicities as well as alkaline and infertility conditions, we established a remediation model to rehabilitate a riparian area reached by the contaminated tailing using the following three co-strategies: phyto, physical and chemical remediation procedures. The early results showed that this model was effective in remediating the tailings contaminated with sodium, ether-mine, high pH and low organic matter and fertility, aside from creating a forest fragment with native species of the Brazilian Atlantic Forest (Scotti et al. 2020). Therefore, the aims of the present study were to evaluate the effectiveness of this functional remediation model over 1 year to remove the contaminants and to assess the land rehabilitation success through biotic and abiotic indicators.

MATERIAL AND METHODS

Study sites

The study site is located in the region of Minas Gerais state known as the “Quadrilátero ferrífero,” comprising banded iron formation (BIF) outcrops of the Cauê formation (Spier et al. 2007), which has the largest Fe ore deposit in Brazil. The climate of the region is characterized by mild summers and low annual average temperatures (17.4–19.8 °C).

This site is situated along the Doce River basin, which was reached by Fundão tailing (Figure 1 A). The studied areas are located in the Paracatu de Baixo district that belongs to Mariana town and consisted of the following riparian sites: a preserved site (PS) along the Lavras Velhas River (20°20'31.34"S 43°17'13.61"W) that is a fragment of preserved riparian Atlantic Forest, a degraded site (DS) located in the Gualaxo River, reached by the dam tailings (20°17'55.79"S 43°14'07.31"W) in the Paracatu de Baixo District (without management, as shown in Figure 1 B) and an experimental site (ES) located at the same river where the remediation procedures were installed over a 2 m sediment layer.

Plant species

Twelve woody species of the Brazilian Atlantic Forest previously screened for tolerance to the contaminated sediment (Scotti et al. 2020), belonging to the Anacardiaceae, Bignoniaceae, Fabaceae and Malvaceae families were transplanted to the field (Table 1). Plants were assessed in relation to survival and height growth after 6 and 12 months after transplantation.

Remediation Procedures

For the remediation procedure, three main strategies were used: 1- Phytoremediation using tolerant plant species to sodium and ether amine toxicity under field conditions; 2- Physical remediation where the sediment was scraped (EERC, 2016) to a depth of 20 cm (top layer) removing a superficial sediment layer (Supplementary Information, Figure S1-A) aiming at reducing ether-amine and sodium toxicity and 3- Chemical remediation using plant-derived organic matter with a pH around 5. The OM was composed primarily of pine bark mulch (Blanco-Garcıa and Lindig-Cisneros 2005) mixed with decomposing litter of different plant species (e.g., *Handroanthus heptaphyllus* and *Jacaranda mimosifolia*). The chemical composition of the dried OM was as follows: 1.28% Ca, 0.22% P, 2.1% K, 0.3% Mg and 2.1% N. Additionally, the dried OM had a C/N ratio of 24 and a pH of 5. The OM was added at a rate of 50 kg per plot in a 1:4 (v:v) proportion. This procedure of OM amendment was performed to chelate sodium and ether amine and to overcome the soil infertility as well as to reduce soil pH.

Experimental design

The experimental design used in ES was composed of two blocks of 1500 m^2 (60 m × 25 m) with two treatments or plots/block (15 × 25 m) as shown in Figure 1C. Four plots of 75 m^2 (15 × 25 m) were established in the reference sites (DS and PS). The remediation treatments established in ES were as follows: T1-Scraping to a sediment depth of ~20-30 cm (top layer) with incorporation into the sediment plant-derived organic matter (OM) before transplantation of woody species; T2-intact sediment was covered by a superficial layer of OM before transplantation of selected woody species (Figure 1C). Each plot was cultivated with 12 woody species of the Brazilian Atlantic Forest using a spacing of 3 × 3 m (Supplementary Information, Figure S1-B).

Physical and chemical analyses of sediment and soil

Samples were collected at a depth of 20 cm from DS and PS (2017) and in each study site (ES, DS) in 2018 at six and twelve months after transplantation. In the ES, eight mixed samples per plot were collected for all soil analyses. The experimental design was composed of eight mixed samples per plot or treatment × 2 treatments per block × 2 blocks, totalling 32 samples. In the DS and PS, 2 samples per plot × 4 plots totalling 8 samples per site were collected. (Supplementary Information, Figure S1-C). The sediment and soil samples were sieved with a 2-mm mesh and analysed for physical and chemical properties (Embrapa 1997) following the standardized protocols (USDA 1967), including for the toxic elements considered in this research: sodium and ammonium. Besides textural analysis, the experimental site was compared with the control sites in terms of the cations content, cation exchange capacity, organic matter content and soil pH, among other parameters. The samples were kept cool (4 °C) during sampling and transport and were maintained at 2 °C until processed. The N- ammonium and nitrate contents were determined by using the steam distillation method of Bremner and Keeney (1965) where 10 g of soil was used for extraction with 2-mol L^{-1} KCl. Soil aggregation measurements were carried out to estimate the proportion of water-stable aggregates (WSAs), using standard methods of Kemper and Rosenau (1986).

Ether amine

The ether amine quantification in soil and sediment samples was performed using the colorimetric bromocresol green methodology described by Araújo et al. (2009) and Santos et al. (2019) using as ether amine standards Flotigam EDA 3 and Fotigam 2835-2 The detection was performed using a spectrophotometer (Shimadzu UV-160A).

Measurement of residual sodium and ether amine

The residual fraction of pollutants sodium and ether amine was measured by calculating the removal rate (Song et al. 2017) using the following equation:

Removal rate (%) = $(1 - A\ B) \times 100$, where A is the residual fraction of pollutants after remediation and B is the total amount of pollutants before remediation.

Microbial Biomass Estimation via Phospholipid fatty acids (PLFA)

Phospholipid fatty acids (PLFAs) were extracted from lyophilised soil according to the Bligh and Dyer method (Bligh and Dyer 1959), and the lipid fractionation was performed according to Gehron and White (1983) and White et al. (1979). The extracts were analysed using a gas chromatograph with flame ionisation detection (HP Model 5890 Series 2 chromatograph). Fatty acid chemical structures were verified by GC–MS (HP Model 5890 Series 2 gas chromatograph and HP Model 5971 mass selective detector), and PLFAs were expressed as equivalent peak responses to the internal standard. Peak areas were converted to PLFA per microgram of dry soil (absolute abundance). As reported by literature (Bossio et al. 2006), normal saturated fatty acids were used to measure the total bacterial biomass. Branched-chain PLFAs represented the Gram-positive bacteria and monoenoic acids indicated the gram-negative bacteria. Thus PLFA structures of the microbial communities from the studied sites were characterized as following: 1- Gram- positive bacteria: 14:0, iso i15:0, ai15:0, i 15:1 ω 9c, i 15:1 ω 6c,,i16:0, i17:1 ω9c, i 17:0, ai 17:0 and i17:0,i18:0; 2- Gram-negative bacteria: 16:1 ω9c, 16:1 ω7c, 16:1 ω6c, 16:1 ω5c, C 15:4 ω3c, C 17:1 ω8c, C17:0 cyclo ω7c, C 17:0, C 18: 1 ω5c, C 19:0 Cy ω; 3-Actinomycetes**:** 17:1 ω7c 10meth, 17:0 10 meth, 16:0 10-methyl, C 18:1 ω7c 10 meth, 18: 0 10 meth; 4-Fungi: C 18:2 ω6c, C 18:1 ω9c, C 18:1 ω7c, and AMF fungi: 16:1 ω5c, C 20: 3 ω 6c, C 20:4 ω 6c; 5- Eucaryotes**:** 20:1 ω9c, C 20:0, C 21:1ω3c, C 22:0, C 24:0 (Bardgett et al. 1996;. Frostegår and Bååth 1996; Zelles 1997; Zogg et al.1997; Bååth and Anderson 2003; Bossio et al. 2006).

Statistical analysis

The studied variables were compared among the preserved, disturbed and experimental sites. ANOVA was used for the variables with normal distributions, and a Kruskal-Wallis test was used for the variables with non-normal distributions and a correlation matrix was performed for some variables. Multiple comparisons were performed with a Tukey test and a Nemenyi test for the variables with normal and non-normal distributions, respectively, using R statistical software R Studio Team (2017). A principal component analysis (PCA) based on a variance analysis was used to select the most significant variables associated with the environment impact and remediation process such as: sodium, ether-amine, soil physical, chemical and fertility parameters and metals. The total variability was expressed in terms of the two main components that better explained the influence of each study variable over rehabilitation process in each site, demonstrated by a scatter plot using the BioVinci software (BioTuring, San Diego, CA, USA). The significance level was set at 5% ($P \leq 0.05$) for all analyses.

RESULTS AND DISCUSSION

Plant growth

Under field conditions, the woody species selected for tolerance to the sediment toxicity showed the highest plant survival and better growth performance in T1 than T2, at both six (Scotti et al. 2020) and twelve months (M) after planting (Figures 1 D, E and Supplementary Information, Figure S1-D). Although the plant growth in T2 was worse in the first six months of planting in relation to T1, at 12 months there was some growth recovery in this treatment, reaching a similar height to T1 at 6 months (Supplementary Information, Figure S2). The plant growth differed among botanical families along the time and treatments (Table 1). Except for the Bignoniaceae species, which did not grow with time independent of the treatment, plants of the sub-families Faboideae (e.g., *Platypodium elegans*) and Mimosoideae clade within the Caesalpinoideae (e.g., *Mimosa bimucronata*) grew equally well at 6 and 12 months in T1. However, in T2, these plants presented a better growth performance only at 12 months (Table 1). It is noteworthy that plants from the Fabaceae family, especially the sub-family Caesalpinoideae (e.g., *Senna alata and Libidibia ferrea*) had outstanding growth until 12 months after transplantation (Table 1, Figures 1 D and E). Such an outstanding growth performance may be understood as a symptom of phyto-toxicity caused by the N overload (Brady and Weil 2007). Indeed, the phytoremediation with tolerant species has been pointed as an effective strategy to remediate not only N-ammonium (Glenn et al. 2016; Kinidi and Salleh 2017) but also sodium (Abbas et al. 2014) and alkalinity (Qadir et al.2002).

Remediation of the sediment toxic compounds

Elevated soil pH causes the loss of soil structure resulting in the increase of erodibility, soil impoverishment and impairment of plant growth (Bronick and Lal 2005). Therefore, the high pH found in the sediments (Table 2) might be one of the toxic factors that compromised the growth of the non-tolerant native plants since they are adapted to the pH of the Atlantic Forest soils (pH~ 5.5). Sediment pH remained elevated (~8.0) in samples collected in DS along 2017 and 2018 in comparison to PS (Table 2, Figure 2A). The remediation procedure adopted in T1 resulted in a pH reduction (~ 6) after 6 and 12 months, but in T2, there was a slower but insignificant pH drop in relation to T1 after 12 months (Figure 2A). Such pH reduction may be attributed to the effect of both phytoremediation and organic matter amendment.

The main phytoremediation strategy displayed by tolerant plant species to reduce the pH of alkaline soils is the release of H^+ ions from roots (Schubert et al. 1990; Austruy et al. 2014) and the exudation of organic acids (Haigang et al. 2013;

Luo et al. 2014). Such rhizospheric acidification effect (Marschner et al. 1987) may be enhanced in the presence of nitrogen compounds (Luo et al. 2014), making the leguminous species particularly attractive candidates for remediation of alkaline soils. In fact, successful phytoremediation procedures for pH reduction have been previously reported (Huang et al. 2013), including in the alkaline red-mud tailings derived from the rupture of a dam in Hungary (Alshaal et al. 2013) as well as in salt-affected soils (Abbas et al. 2014). The amendment with humified OM may have largely contributed for pH decline in ES since OM with pH ~ 5 might have buffered the tailing pH (Mackowiak et al. 2001), likely due to its large number of weakly acidic functional groups (carboxylic acid and phenolic). Thus, the chemical remediation success using OM such as pine bark (Conde-Cid et al. 2020) which shows a high lignin content, is related to the formation of humic substances enriched of negative charges (Simansky and Bajcan 2014; Kimura et al. 2017).

The high pH in DS derives from sodium used in the reverse cationic flotation process for iron ore beneficiation (Neder 2005) and, similarly to pH very high sodium levels were found in DS and these remained at the same levels from 2017 to 2018 (Table 2, Figure 2 B). The remediation procedures in ES were efficient at reducing Na levels, initially more in T1 (6 months) and after 12 months in both treatments (Table 2 and Figure 2 B), albeit these levels are still higher than PS (Figure 2B). These results suggest that the scraping (T1) method favoured the rapid establishment of plants at the first six months followed by reducing the sodium content (Scotti et al. 2020) but this procedure is not essential for sodium remediation success in the long-term, since sodium content was equally reduced in both treatments after 12 months. On the other hand, the sodium movement in soil promoted by water evaporation during the dry winter season (Scotti et al. 2020) can explain the oscillation of sodium levels along the time as verified in T1 (Figure 2B). The superficial accumulation of Na due to the water evaporation may be easily and inexpensively detected by the Na flame test (Scotti et al. 2020), what may help land managers and farmers to assess whether the soil scrapping is needed or not. If the test is positive, the scrapping is recommended to assure plant establishment in the first year. Considering the higher cost of scrapping with tractor, manual plowing may be an attractive alternative procedure for small farmers to reduce the superficial Na (~5 cm depth) before planting.

Excessive levels of sodium can compromise plant water and nutrient uptake (Nouri 2017), promoting soil swelling and decreasing soil permeability (Mohanty et al. 2015) and ultimately the plant growth. Since it is well known that the organic matter may also remediate sodium through its ability to chelate this ion (Robbins 1986; Lax et al. 1994; Ashworth 2007), we may also consider the organic matter amendment as an efficient remediation tool for Na overload since the sodium content was negatively correlated with soil organic matter and plant growth (Supplementary Information, Table S1). Indeed, numerous studies show the effect of organic amendment to alleviate Na toxicity in soil and sediments (Conte et al. 2005; Tejada et al. 2006; Wang et al. 2017) such as salt-affected sites (Ashworth 2007) and alkaline bauxite red mud wastes (Fuller et al. 1982; Wong and Ho 1994; Courtney and Timpson 2005; Courtney et al. 2009; Xue et al. 2016, Bray et al. 2018). Cha-um and Kirdmanee (2011) also reported that OM supply not only decreased pH and Na but also reduced the electrical conductivity (EC). Similarly, we found that EC remained high in DS from 2017 to 2018 due to the presence of Na overload in comparison to PS (Table 2) but declined in ES in both treatments (Table 2), whose drop was proportional to Na decline as shown in Figure 2C, with a correlation index of 0.74. This result highlights again the role of woody species and OM amendment for Na remediation rather than the scrapping treatment. However, it is noteworthy that EC found in the affected site is not typical of saline or sodic soils (Brady and Weill 2007, Nori et al. 2017) since this is a sodium-contaminated sediment (Santos et al. 2019).

Ether-amine naturally declined in DS from 2017 to 2018 in the superficial layer (Table 2, Figure 3A), which could be attributed to the natural biodegradation by microbial action, wash-out or destabilization promoted by pH reduction (Araujo et al. 2010). However, the remediation treatments were significantly more efficient at ether-amine reduction than the natural biodegradation (Figure 3A). The remediation procedure in T1 was initially significantly more effective in ether-amine reduction as compared to T2 at 6 months, but at 12 months there was a stabilisation of ether-amine concentrations in both T1 and T2 (Figure 3A). These levels, however, were still higher than those of PS, where it was not found (Figure 3A).

Sediment pH reduction seems to be the key factor responsible for ether-amine destabilisation. The pH of 8.5–9.0 is optimum for reverse flotation with ether-amine because at this pH range quartz particles become negatively charged reacting with ether-amine, showing low dissociation capacity and solubility (Neder 2005). In contrast, ether-amine becomes dissociated when the pH is lowered to ~ 5, becoming soluble in aqueous solution (Araujo et al. 2009). Therefore, the pH drop in ES may have favoured the ionisation of secondary and tertiary amines, increasing its availability to biodegradation (Araujo et al. 2010). In addition, root organic acids exudation by the rhizospheric effect may be enhanced in the presence of N compounds (Luo et al. 2014) such as ether-amines. Thus, we hypothesise that ether-amine itself may have favoured rhizosphere acidification which, associated with the OM acidification, promotes ether-amine chemical degradation, constituting a possible positive feedback mechanism for ether-amine remediation. Indeed, the treatments of phyto- and chemical remediation adopted were more effective at removing ether-amine than sodium, whose residual fractions were around 10% and 50% respectively (Figure 3B).

The natural products of ether-amine biodegradation (Araujo et al. 2010) are N-compounds such as ammonia (NH_3) or N-ammonium (NH_4^+). However, most plant species are sensible to ammonia/ammonium (NH_3/NH_4^+) overload (Britto and Kronzucker 2002) largely due to the inability of plants to restrict the primary entry of NH_4^+ by the aquaporin (AQP) transporter family (Kozono et al. 2002), leading to several downstream toxicological events (Britto et al. 2001). Under the particular conditions of high pH and high N supply, uncharged ammonia (NH_3) is likely to rapidly enter into plant root cells (Coskun et al. 2013) mediated by AQP (Kozono et al. 2002), which may result in further N toxicity. Thus, ammonia/ammonium toxicity may partially explain the mortality of adult trees in the field reached by the contaminated tailings (Santos et al. 2019).

Relatively elevated ammonium content was found in the sediment in DS-2017 (nearly 50% of that found in PS), which likely derived from the natural degradation of ether-amine (Table 2, Figure 3C). Considering that there is no vegetation in DS and the microbial communities (total biomass C) were highly depleted in DS during 2017 and 2018 (Table 3), the natural decrease of ammonium in DS during 2018 (Figure 3C) may be explained by soil leaching or probably to the abundant colonisation of Basidiomycota fungi (Supplementary Information, Figure S1-E) observed in this site. These fungi belong to a chemo ecological group stimulated by ammonia-rich substrate, known as ammonia fungi (Sagara 1975, Raut et al. 2015). However, such ammonium decline in DS was not followed by nitrate formation (Figure 3D) nor it was followed by an increase in the total bacterial population (Tables 3 and 4). In contrast, in ES there was also a reduction in ammonium content at 6 months in both treatments (Figure 3C), which may be explained by its oxidation as revealed by the strong increase of nitrate in both T1 and T2 (Figure 3D). Indeed, there was a significant increase in the total microbial biomass and Gram-negative bacteria, which includes the nitrifier population, especially in T1, improving soil fertility (Table 3 and 4).

Nevertheless, at 12 months, there was a decrease in nitrate content in ES concomitant with an increase of ammonium (Figures 3C and D). Such reduction of nitrate may be explained not only by the outstanding plant growth (Table 1; Figures 1D and E), which may have used nitrate as N source, but also leaching as nitrate is the most soluble chemical N form. In addition, the decomposition of the abundant litter-fall observed at 12 months (Supplementary Information, Figure S1-F) may be a novel source of ammonium input to the sediments as observed in Table 2 and Figure 3C, since at this time ether-amine concentration remained at the same levels as at 6 months (Table 2, Figure 3A). This idea is corroborated by the significant correlation between plant height growth and soil ammonium (Supplementary Information, Table S1). Thus, our results suggest that the N overload derived from ether amine was phyto-remediated by the woody vegetation, explaining its rampant growth and the soil N cycle appears to have been under reconstruction. Aside from the excessive vegetative growth, the plants showed stem elongation (Supplementary Information, Figure 1E), which are known symptoms of N toxicity (Brady and Weil 2017).

Despite few works report N remediation efforts (Jordan et al. 2008; Miao et al. 2013; Glen et al. 2016; Kinidi and Salleh 2017) because N is not a common soil contaminant but rather soils are usually N-deficient, some authors have employed the phytoremediation with native species and/or the stimulation of nitrification process to remediate ammonium-contaminated uranium mining sites (Miao et al. 2013; Gleen et al. 2016).

Rehabilitation assessment

Soil fertility, aggregation and microbial populations

Table 2 shows the mean values of soil fertility parameters and Tables 5 and 6 show the pairwise comparisons among treatments. The sites affected by the tailings (DS) presented a very low fertility pattern in 2017 and 2018 relative to PS (Tables 2 and 5). There was only an increase in K, Ca and Mg in DS 2018 relative to 2017, which was possibly a result of the leaching process since CEC and OM did not show a proportional increase (Tables 2 and 5). By contrast, ES showed an overall significant improvement of nutrients (K, Ca, Mg and P) at 6 months relative to DS-2017 due to the initial fertilisation and OM amendment (Tables 2, 5 and 6). However, the levels of Mg in ES were similar to DS-2018 in T2-12M. Calcium was found at relatively high levels in ES but similar to both DS and PS (Tables 5 and 6), which could be explained by the fact that it is also used in ore mine beneficiation during reverse cationic flotation to activate quartz (Filippov et al. 2014). In contrast, potassium and phosphorus suffered a marked depletion at 12 months after transplantation (Tables 2, 5 and 6), independent of the treatment. Similar to the decrease of ammonium and nitrate at 12 months, the depletion of K and P observed in ES was likely a consequence of the phytoremediation procedure that resulted in excessive vegetative growth.

Organic matter increased equally in ES-T1 and ES-T2 relative to DS 2017 and 2018. Nevertheless, the improvement of OM in T1 was greater than T2 along the time (Tables 2, 5 and 6), highlighting the effectiveness of the incorporation of OM to the sediment, resulting in a better plant growth performance in T1 than T2 (Supplementary Information, Figure S2). This improvement of soil fertility pattern as expressed by the CEC was significantly increased in both treatments of ES along the time (Tables 2 and 6). The CEC in T1 was higher than T2 at 6 months but after 12 months, there were no differences between treatments. The CEC is mainly determined by the negative charges of colloidal clays or humified OM (Bronick and Lal 2005; Brady and Weil 2007). Once the sediment showed a lower content of clay in comparison to sand and silt and high density (Supplementary Information, Table S2), the CEC was certainly provided by the supply of OM, more efficiently in T1 than T2, as confirmed by the positive correlation between OM and plant growth (Supplementary Information, Table S1). Similarly, the success of several remediation efforts of contaminated sites has been achieved by the OM amendment with the increase of CEC (Tejada et al. 2006; Song et al. 2017).

Besides, OM input stimulates the soil microbial populations and their enzymatic decomposing activity (Singer et al. 2003; Tejada et al. 2006, Song et al. 2017). The microbial biomass was strongly inhibited in the sediments, reaching levels as low as 2.2 nmol C g^{-1} in DS (2017) compared to levels of 136 nmol C g^{-1} soil in PS (Tables 3 and 4) confirming the results of Santos et al. (2019). The remediation procedures stimulated the total microbial biomass as well specific groups, such as

Gram-positive, Gram-negative, total fungi, arbuscular mycorrhizal fungi (AMF) and actinomycetes, more abundantly in T1 than T2 (Table 3), reinforcing the efficacy of OM incorporation. The Gram-positive bacteria were particularly favoured not only in in ES but also in DS-2018, likely because several Gram-positive bacteria are halophytes (e.g., 15:0 iso and anti iso 17:0 PFLA signatures) and growth well at high pH (Aciego Pietri and Brookes 2009). On the other hand, Araujo et al. (2010) demonstrated that some anaerobic Gram-negative bacteria such as *Serratia marcescens* and *Enterobacter cloacae* could degrade ether-amine under laboratory conditions. The bacteria/fungi ratio (Table 3 and 4) shows that bacteria rather than fungi were significantly increased in T1 than T2, particularly the Gram-negative and actinomycetes groups (Tables 3 and 4), which include both the nitrifying and the ether-amine degrading bacterial communities. The presence of such bacterial communities may also explain the decrease of ether-amine and the increase of nitrate in ES. In contrast, the PLFA AMF population (16:1ω 5c PFLA signature) remained lower in ES (Tables 3 and 4) even after 12 months of remediation relative to the total fungal population and mainly to the bacterial group. Overall, our results show that the bacteria/fungi ratio (Table 3 and 4), which was particularly influenced by the bacterial group, appears as a sensitive indicator to assess the rehabilitation of the areas affected by the tailings. Thus, the soil bacteria population, rather than fungi, drives soil remediation and soil quality improvement during the first 12 months.

The dam sediments also showed a marked loss of physical structure (Tables 5 and 6; Supplementary Information Table S2). It is well known that soil physical structure is related to its properties of carbon stabilisation, soil porosity, water infiltration, aeration, hydraulic conductivity and resistance to erosion, which ultimately determine soil productivity (Bronick and Lal 2005; Brady and Weil 2007).

These functions depend on the substrate stabilisation, which is mediated by soil aggregation and plant roots (Wang et al. 2014). Since the humic organic matter plays a key role in soil aggregation (Simansky and Bajcan 2014; Kimura et al. 2017), the OM supply in ES associated with the effect of plant root system resulted in a significant improvement in soil aggregation in ES relative to DS, particularly in T1 (Tables 2, 5 and 6). Aggregate stability has been considered one of the most consistent soil quality indicators (Shukla et al. 2004; Song et al. 2017), especially for the rehabilitation of riparian sites (Kimura et al. 2017; Santos and Scotti 2018). The increase of soil aggregation by the incorporation of OM has also been reported by Abbas et al. (2014) in the phytoremediation efforts of the red mud from bauxite mine.

Regarding the content of the metals, it has been shown that they do not differ from the region background either before (Santolin et al. 2015) and after (Santos et al. 2019) the Fundão dam rupture. Indeed, the remediation procedure did not alter the levels of the metals as Cu, Fe, B and Zn, which showed similar levels to the preserved site (Tables 2, 5 and 6). Considering the forest rehabilitation success achieved and the fact that metals contents did not differ in ES in relation to DS after 12 months of rehabilitation, the putative toxicity of metals may be discarded for rehabilitation purposes. For Mn, relatively high levels (208.7 and 269 mg kg^{-1} in 2017 and 2018 respectively) were found in DS (Table 2), contrasting with the low levels (20 mg kg^{-1}) found in the preserved site of the Lavras Velhas River. However, high levels of Mn (400 mg kg^{-1}) were also registered in soils from the preserved soils of a riparian forest in the Carmo River, an affluent of the Doce river (Figure 1A), which was not reached by the dam tailings (Santos et al. 2019). Such differences between preserved sites were attributed to the fact that the Carmo River receives sewage discharge from Mariana town, while the Lavras Velhas is a very pristine site. Accordingly, Santolin et al. (2015) reported high Mn content in the Doce river basin before the dam rupture, which was attributed to domestic and industrial discharges to the river. Although the Mn content in T2 was higher than T1(Tables 2 and 6), such Mn levels are not considered toxic to plants (Gupta et al. 2008) and may not be related to the Fundão dam rupture but rather to industrial sewage discharge to the river as observed by Santolin et al. (2015).

Principal component analysis (PCA)

The first two axes of the principal component analysis (Figure 4) based on the studied variables (Table 2) in each site explained 58.8% of the total variation. Component 1 accounts for 46.46% of the variation and was mainly influenced by the variables OM, CEC, N-NH_4, N-NO_3, K and Mg, which particularly drove the clustering of PS samples. Component 2 explained 12.42 % of the variation, and it was influenced by the variables sodium, ether-amine, pH, and electrical conductivity, which were the main determining factors for the segregation of samples from DS in the PCA. Meanwhile, the samples from DS and PS clustered in an opposed position in the PCA; those from ES grouped in an intermediate position but closer to PS, especially the samples of ES-T1 at 6 and 12 months. These results suggest that the rehabilitation in ES is evolving towards the conditions of PS samples.

CONCLUSION

The remediation success of a riparian site contaminated with tailings containing high levels of sodium and ether-amine was achieved by the employment of three co-strategies: the phytoremediation with tolerant native woody species, sediment scraping and organic matter amendment. Such strategies resulted in the reduction of the toxic compounds concomitant with the establishment of a forest fragment with native woody species. These procedures may provide the basis for the rehabilitation not only of riparian sites but also crop sites reached by dam tailings across the Doce River. In addition, this technology may help reclaim other sites reached by dam tailings elsewhere, particularly alkaline bauxite red mud wastes, salt-affected sites and ammonium-contaminated uranium mining sites.

Supplemental Data—The Supplemental Data are available on the Wiley Online Library at DOI: 10.1002/etc.xxxx.

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Author contributions statement— A.R. Gomes, A. Antão, A.G.P. Santos’ T.J. Lacerda, A.S. Isla, C.H. Santos and M.B. Medeiros established the field experiments, performed the laboratory and field analyses and wrote the manuscript; S. Alvarenga, E.C. Rigobelo conceived and designed experiments and M.R. Scotti conceived and designed experiments, analyzed data and wrote the manuscript.

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FIGURES

Figure 1: A-Location of Fundão dam (Minas Gerais / Brazil), Mariana town and the experimental site at Paracatu de Baixo. **B:** Disturbed site in 2017 before transplantation at Paracatu de Baixo (Mariana/ Minas Gerais). **C**: Schematic drawing of the experimental design established in ES with treatments 1 and 2; **D and E:** Forest at 12 months after remediation treatment, highlighting the outstanding plant growth.

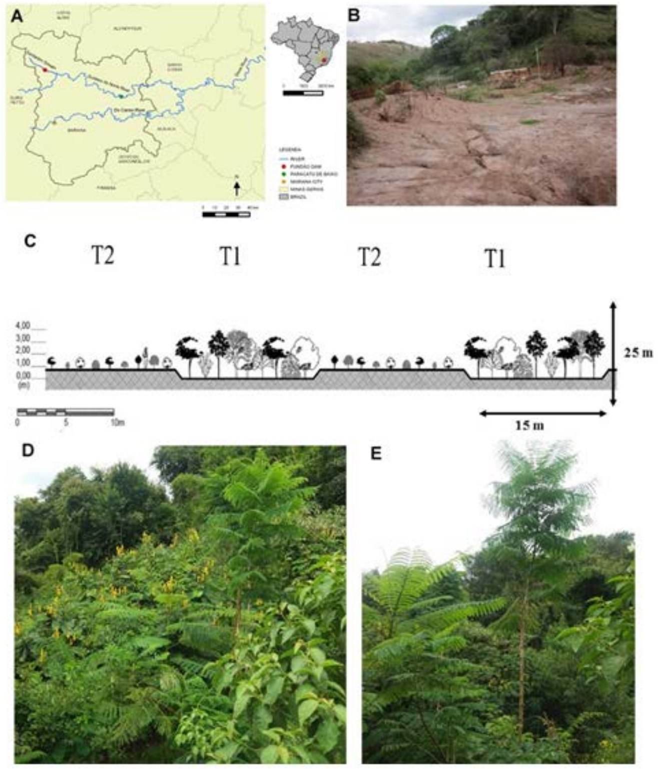

Figure 2-Comparisons of soil pH (**A)** and sodium content **(B**) among preserved site (PS- green), disturbed sites (DS- white) and experimental Site (ES T1- blue), and EST2- orange) at 6 and 12 months (M) after transplantation, as follows: DS 2017; DS 2018; ES-Treatment 1- 6M; ES Treatment 2- 6M; ES- Treatment 1- 12M and ES-Treatment 2 -12M. **(C)** Linear regression based on electrical conductivity and sodium content of samples from all the sites. The analysis of variance was performed using ANOVA or Kruskal-Wallis test and for pairwise comparisons it was used Tukey's or Nemenyi's test. Means followed by the same letters per line did not show significant differences (significance at p ≤ 5%).

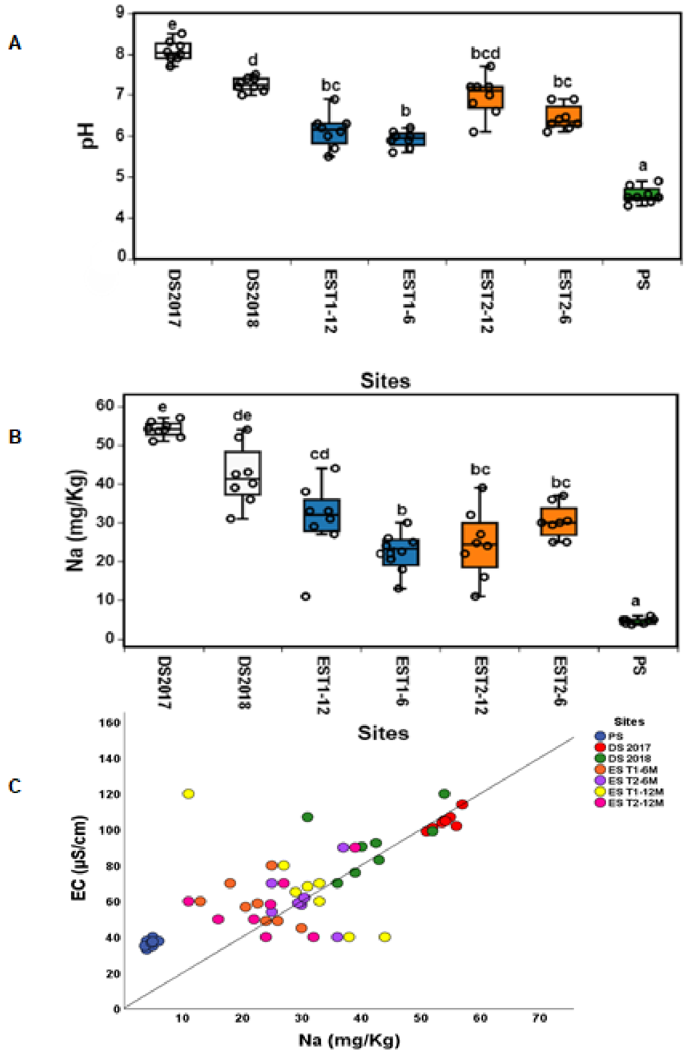

Figure 3: Comparisons of soil **(A)-** ether-amine content, **(B)-**Residual soil fraction (%) of the pollutants ether-amine and sodium, **(C)-** N-NH4, **(D)-**N-NO3 contents among soils of preserved site (PS-green); sediments from Disturbed site (DS-white); and Experimental Site (ES T1-blue) and ES T2 (orange) at 6 and 12 months (M) after transplantation as follows: DS 2017; DS 2018; ES-Treatment 1- 6M; ES Treatment 2- 6M; ES- Treatment 1- 12M; ES-Treatment 2 -12M. The analysis of variance was performed using ANOVA or Kruskal-Wallis test and for pairwise comparisons it was used Tukey's or Nemenyi's test. Means followed by the same letters did not show significant differences (significance at p ≤ 5%).

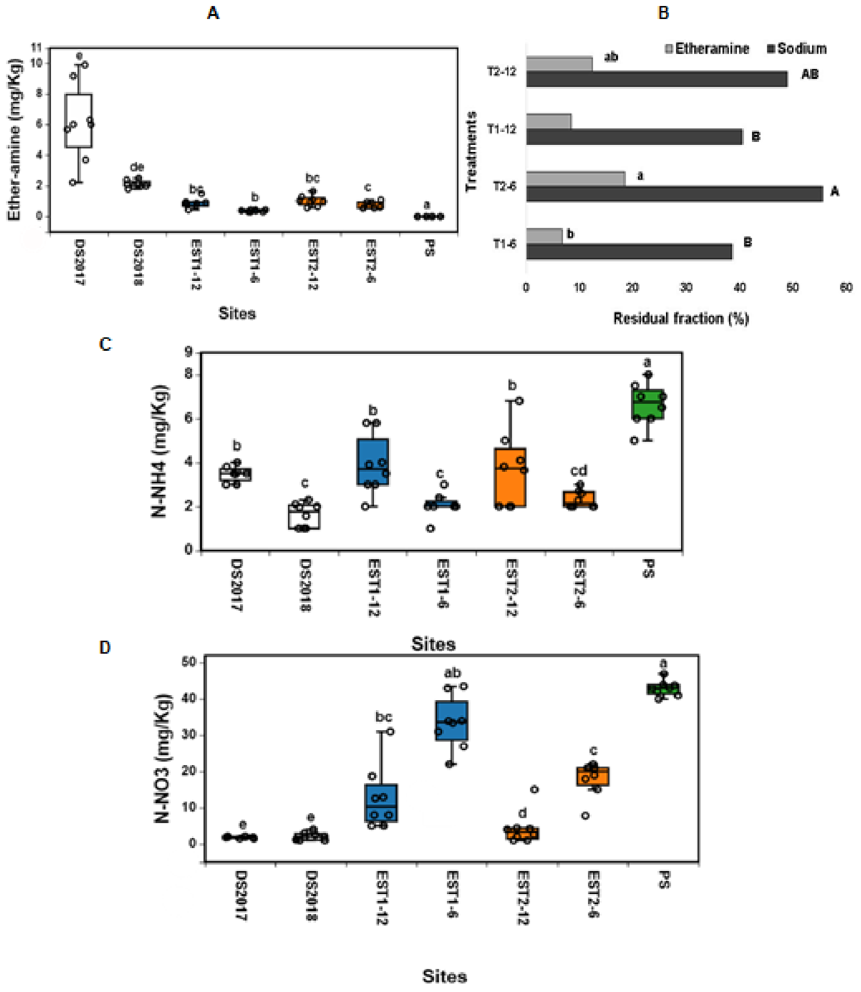

Figure 4: Principal component analysis based on soil and/or sediment properties measured at the preserved site (PS), the disturbed site in 2017 and 2018 (DS) and the experimental Site (ES), under treatment 1 (ES T1) and treatment 2 (ES T2), at 6 and 12 months after transplantation.

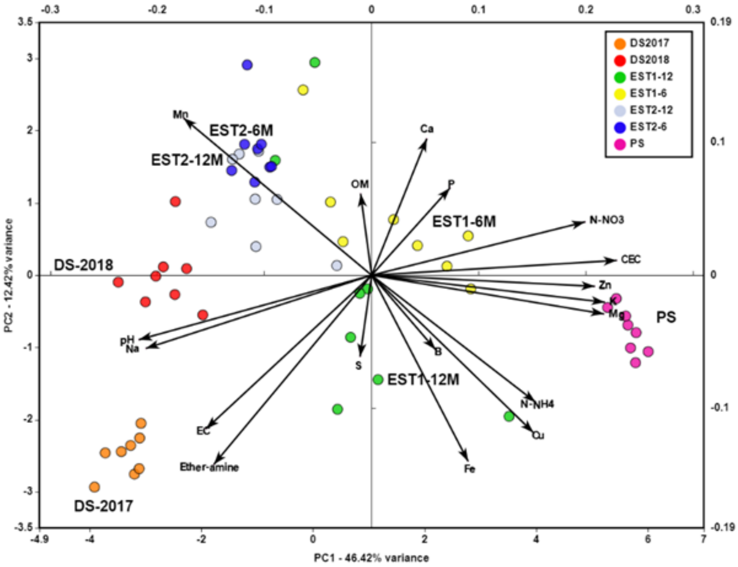

Table 1- Height growth (m) of plants per families at 6 and 12 months (M) after transplantation in the experimental site (ES) under treatments T1: (sediment scraping plus incorporation of OM) and T2: (OM amendment to intact sediment). The analysis of variance (ANOVA or Kruskal-Wallis test) was performed to compare differences of plant growth/family (in each line) between sites along the time using Tukey or Nemenyi's tests. Means followed by the same letters per line did not show significant differences (significance at p ≤ 5%).

Plant Family	T1 (m)-		T2 (m)	
	6 M	**12 M**	**6 M**	**12 M**
Anacardiaceae	2.62^{b}	3.41^{a}	0.82^{d}	2.2^{c}
Bignoniaceae	1.5^{a}	1.5^{a}	0.83^{b}	0.89^{b}

Fabaceae (Caesalpinoidaea)	2.6^{b}	3.4^{a}	1.07^{c}	1.5^{c}
Fabaceae (Faboideae)	3.1^{a}	3.2^{a}	1^{c}	2.4^{b}
Fabaceae (Mimosoidae)*	3^{ab}	3.5^{a}	1.0^{c}	2.2^{b}
Malvaceae	1.4^{c}	2.6^{a}	0.9^{d}	2.1^{b}

***Mimosoidae has been considered a clade within Caesalpinoidaea**

Table 2- Soil chemical analysis in the experimental site (ES) under two treatments (ES-T1 and ES-T2) compared to preserved (PS) and disturbed (DS 2017 and 2018) sites at 6 and 12 months (M) after transplantation. The analysis of variance was performed using ANOVA or Kruskal-Wallis tests (significance at p ≤ 5%). Values in bold were significantly different at p ≤ 5%.

Variables	**PS**	**DS (2017)**	**DS (2018)**	**ES T1 (6M)**	**ES T2 (6M)**	**ES T1 (12 M)**	**ES T2 (12 M)**	**p-values**
pH	4.58	8.05	7.34	5.91	6.46	6.1	7	**< 0.001**
K (mg/kg)	73.83	8.00	12.66	38.83	18.66	26.33	9.8	**< 0.001**
Ca (mg/kg)	274.00	182.16	290.5	327.83	252.50	396.7	326.2	**0.004**
Mg (mg/kg)	72.33	10.16	21.16	48.83	24.00	43	25.2	**< 0.001**
P (mg/kg)	6.57	4.83	4.16	11.83	10.50	8.33	6	**< 0.001**
S (mg/kg)	11.66	18.16	25.0	27.16	11.00	20.8	6.67	0.068
B (mg/kg)	0.18	0.16	0.1	0.20	0.15	0.28	0.22	0.081
Cu (mg/kg)	0.78	0.50	0.5	0.67	0.31	0.63	0.4	**0.006**
Fe (mg/kg)	192.35	182.16	105.6	153.50	97.33	182.5	122.8	**< 0.001**
Mn (mg/kg)	44.16	208.66	269.16	219.33	281.66	201	314	**0.001**
Zn (mg/kg)	4.60	0.38	0.43	1.81	0.91	1.95	1.71	**< 0.001**
OM (%)	8.65	0.96	1.00	2.27	1.49	2.28	1.58	**< 0.001**
Na (mg/kg)	4.83	54.167	44.8	22.667	30.500	31	24.8	**< 0.001**

Ether-amine (mg/kg)	0.00	6.113	1.98	0.373	0.712	0.86	1.02	**< 0.001**
E. conductivity (µS/cm)	36.88	104.7	92.5	58.8	62	68	58	**<0.001**
CEC (meq/100g)	5.27	0.97	1.3	3.4	2.07	3.25	2.58	**<0.001**
N-N0$_3$ (mg/kg)	42.83	1.83	2.03	33.42	17.97	12.6	4.27	**<0.001**
N-NH$_4$(mg/kg)	6.5	3.47	1.51	2	2.28	3.93	3.65	**<0.001**
Total porosity (%)	54.0	43.6	---	---	---	57.188	57.638	**<0.001**
Wet aggregate stability (%)	59.2	3.9	---	----	---	10.72	6.6	**<0.001**

Table 3- Phospholipid fatty acid (PLFA) concentrations(C-nmols/g) of soil microbial communities from Preserved site **(PS)**. disturbed site in 2017 and 2018 (**DS**) and the Experimental Site (**ES).** under treatment **1 (ES T1)** and treatment **2 (ES T2**). The analysis of variance was performed by Kruskal-Wallis test and pairwise comparisons were estimated using the Nemenyi's test. Means in bold were significantly different at p ≤ 5%. Values in bold were significantly different at p ≤ 5%.

Variables	**Mean**					**p-value**
	PS	**DS (2017)**	**DS (2018)**	**ES T1**	**ES T2**	
Total C biomass	136.980	2.253	4.966	28.584	12.462	**< 0.001**
Gram + bacteria	39.690	0.953	1.398	10.769	4.491	**< 0.001**
Gram - bacteria	32.268	0.490	0.98	6.690	2.790	**< 0.001**
Actimomycetes	17.860	0.093	0.274	2.916	0.994	**< 0.001**
AMF Fungi	5.252	0.000	0.102	1.424	0.754	**< 0.001**
Fungi	22.000	0.357	1.280	6.470	3.043	**< 0.001**
Eukaryote	2.038	0.000	0.000	0.217	0.000	**0.001**
Gram +/Gram -	1.239	1.951	1.506	1.572	1.614	**0.005**
Bacteria/Fungi ratio	41.163	2.440	2.269	11.811	5.417	**< 0.001**

Table 4- Comparisons of PFLA (C-nmols/g) soil microbial biomarkers concentrations among the preserved site (PS). disturbed site in 2017 and 2018 (DS) and Experimental Site (ES). under treatment 1 (ES T1) and treatment 2 (ES T2). The analysis of variance was performed by Kruskal-Wallis test and pairwise comparisons were estimated using the Nemenyi's test. Means in bold were significantly different at p ≤ 5%. Values in bold were significantly different at p ≤ 5%.

Variables	**p-values* for pairwise comparison**									
	PS x DS(2017)	**PS x DS(2018)**	**PS x EST**	**PS x EST2**	**DS (2017) x DS (2018)**	**DS (2017) x EST1**	**DS (2017) x EST**	**DS (2018) x EST**	**DS (2018) x EST**	**EST x EST**
Total C biomass	**< 0.001**	**< 0.001**	0.23	**0.005**	0.641	**0.002**	**0.00**	**0.00**	**0.00**	**0.04**
Gram + bacteria	**< 0.001**	**< 0.001**	0.23	**0.005**	0.689	**0.002**	**0.00**	**0.00**	**0.00**	0.05
Gram - bacteria	**< 0.001**	**< 0.001**	0.23	**0.005**	0.641	**0.002**	**0.00**	**0.00**	**0.00**	**0.04**
Actimomycetes	**< 0.001**	**< 0.001**	0.23	**0.005**	0.698	**0.002**	**0.00**	**0.00**	**0.14**	**0.04**
AMF Fungi	**< 0.001**	**< 0.001**	**0.01**	**0.007**	0.549	**0.002**	0.05	**0.00**	**0.01**	0.08

Fungi	**< 0.001**	**< 0.001**	**0.02**	**0.006**	0.690	**0.002**	**0.00**	**0.00**	0.12	0.06
Eukaryote	**0.001**	**< 0.001**	**0.00**	**< 0.001**	1.000	0.298	1.00	0.21	1.00	0.17
Gram +/Gram	**< 0.001**	0.084	**0.02**	**0.009**	0.021	0.032	0.05	0.67	0.44	0.69
Bacteria/Fungi		**< 0.001**	**0.00**	**0.005**	0.714	**0.009**	**0.00**	**<**	**0.04**	0.04

Table 5-Comparisons of soil properties and nutrient contents from preserved site (PS), disturbed site in 2017 and 2018 (DS) and Experimental Site (ES), under treatment 1 (ES T1) and treatment 2 (ES T2), at 6 and 12 months after transplantation. The analysis of variance was performed by Anova or Kuskal-Wallis and for pairwise comparisons it was used Tukey's or Nemenyi's tests (significance at p ≤ 5%). Values in bold were significantly different at p ≤ 5%.

	p values										
Variables	**DS 2017 x**						**DS 2018 x**				
Nutrients	**DS (18)**	**ES T1 (6M)**	**T1 (12M)**	**ES T2 (6M)**	**ES T2 (12M)**	**PS**	**ES T1 (6M)**	**ES T1 (12M)**	**ES T2 (6M)**	**EST2 (12M)**	**PS**
K	**0.01**	**0.01**	**0.02**	**0.01**	1	**0.01**	0.04	0.01	1	1	0.01

Ca	0.58	**0.001**	**0.01**	**0.001**	0.3	**0.001**	1	1	1	1	0.15
Mg	0.01	**0.01**	**0.001**	**0.001**	1	**0.01**	0.6	0.3	1	1	**0,01**
P	1.0	**0.01**	**0.01**	**0.001**	1	**0.02**	0.01	0.01	0.04	1	**0.01**
S	1	1	0.7	**0.01**	0.18	**0.01**	1	0.6	0.01	0.03	1
B	0.32	0.52	0.14	1	0.53	1	0.01	0.7	0.5	0,1	1
Cu	1	1	0.19	0.06	0.33	0.49	0.7	0.21	0.06	0.29	**0.01**
Fe	0.51	1	0.98	0.12	0.1	0.05	0.02	0.03	0.7	0.49	**0.01**
Mn	0.09	1	0.79	**0.01**	**0.01**	**0.01**	1	**0.02**	0.1	0.12	**0.01**
Zn	1	**0.01**	**0.001**	**0.01**	**0.01**	**0.01**	**0.01**	**0.01**	**0.02**	**0.01**	**0.01**
OM(%)	0.6	**0.02**	**0.01**	**0.01**	**0.01**	**0.01**	**0.02**	**0.01**	**0.02**	**0.03**	**0.01**
CEC	0.19	**0.001**	**0.001**	**0.001**	**0.01**	**0.01**	**0.01**	**0.01**	**0.01**	**0.01**	**0.001**
Wet aggregate stability (%)	-	**-**	**0,001**	**-**	**0.001**	**0.001**	**-**	**-**	**-**	**-**	**-**

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Table 6-Comparisons of soil properties and nutrient content among preserved site (PS) and Experimental Site (ES), under treatment 1 (ES T1) and treatment 2 (ES T2), at 6 and 12 months after transplantation. The analysis of variance was performed by Anova or Kuskal- Wallis and for pairwise comparisons it was used the Tukey's or Nemenyi's test (significance at p ≤ 5%). Values in bold were significantly different at p ≤ 5%.

	P values									
Variables	**ES T1 (6 M) x**				**EST1 (12M) x**			**ES T2 (6M) x**		**ES T2 (12 M) x**
	ES T1(12)	**ES T2 (6)**	**ES T2 (12)**	**PS**	**ES T2 (6)**	**ES T2 (12)**	**PS**	**ES T2 (12)**	**PS**	**PS**
K	1	0.1	**0.02**	**0.01**	1	0.46	**0.01**	1	0.001	**0.001**
Ca	1	1	1	1	0.3	1	0.9	1	1	1
Mg	1	0.1	0.18	0.1	0.7	1	**0.02**	1	**0.001**	**0.001**
P	1	1	0.10	0.2	1	1	1	0.6	1	1
S	0.4	**0.06**	**0.02**	0.07	0.2	0.1	0.2	0.6	0.9	0.5
B	0.2	0.5	0.8	0.8	0.09	0.4	0.21	0.4	0.6	0.6
Cu	0.8	**0.01**	**0.03**	0.1	**0.01**	**0.02**	0.14	0.38	**0.001**	**0.001**
Fe	0.2	**0.02**	0.2	0.1	**0.001**	**0.02**	0.69	0.31	**0.001**	**0.001**
Mn	0.5	**0.03**	**0.02**	**0.01**	**0.001**	**0.01**	**0.01**	0.26	**0.001**	**0.001**
Zn	0.7	**0.01**	0.7	**0.001**	**0.001**	0.5	**0.01**	**0.03**	**0.001**	**0.001**
OM(%)	0.16	**0.04**	**0.04**	**0.001**	**0.002**	**0.001**	**0.01**	0.9	**0.001**	**0.001**
CEC	0.5	**0.01**	**0.001**	**0.001**	**0.001**	**0.01**	**0.01**	0.4	**0.001**	**0.001**
Wet aggregate stability (%)	-	-	-	-	-	0.6	**0.02**	-		**0.001**