



## Article

# Restoration Techniques Applied in Open Mining Area to Improve Agricultural Soil Fertility

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**Abstract:** Open pit mining causes damage in natural and rural regions; that is why soil restoration is necessary in order to recovery soil–plant systems. The application of waste can be a good solution for rehabilitation, and it clearly complies with the circular economy and the zero-waste strategy. This study was carried out in a quarry restoration area in the southeast of Spain, where experimental plots were designed and fertilized with different amendments (commonly used inorganic fertilizer N-K-P, pig slurry, pruning waste and urban solid wastes) with the objective of studying ways to improve the restoration of the soil by using these residues and increase the soil fertility before planting. The treatments applied were evaluated in the short term (two and four months from their addition to topsoil) and medium term (nine months) in order to determine if the restored soils will be adequate for agriculture based on nutrients' availability. The results showed that in all the treatments, the pH exceeded 8.5 due to the nature of the soil matrix, but after 9 months of the application, in the plots treated with NPK and pig slurry, the pH decreased. In general, with the application of the treatments, soil macro- (N, P, K, Na, Ca and Mg) and micro-nutrients (Fe and Cu) were increased. However, pig slurry and urban solid waste favored N and P, respectively.

**Keywords:** circular economy; organic amendments; pig slurry; recycled waste; soil restoration; semi-arid ecosystems; zero waste



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## 1. Introduction

Open-pit mining is an important activity basic for the development of human society [1,2]; moreover, it occupies an important position in global economic development [3]. At the same time, mining is considered the most destructive anthropogenic activity and affects ecosystems [4], causing damage to both geomorphology and geological structures and leading to habitat loss [5].

Mining activity significantly affects the environment due to the pollution of air, soil, surface and groundwater, landscape degradation and damage to biodiversity [6–10]. The problem of environmental degradation is aggravated in arid and semi-arid climates and, in particular, in the Mediterranean ecosystems, due to the scarcity of water and torrential rainfall, high solar radiation and low plant cover that favors the erosion processes [11].

Surface mining establishes new infrastructure, fragments natural habitats and is thus acting as a threat to biodiversity [12,13]. In addition to the habitat loss caused by open-pit mining, it alters the soil and topographic structure [14] and disrupts surface and subsurface water regimes [15]. Regarding the ES (ecosystem services) loss, open-pit mining development has also led to a series of ecological and environmental issues, such as air pollution, surface seepage, soil quality decline and soil erosion [15–17]. As a result, agricultural and environmental rehabilitation are challenges in these areas in order to improve landscape recovery and combat global warming.

In the Region of Murcia (Spain), the exploitation of geological resources was always linked to the extraction of minerals and rocks. Specifically, the last one is represented by the extraction of marble, marble limestone and limestone aggregates [18]. Currently, the mining regulations require the rehabilitation of the land after exploitation [19]. In areas degraded by open-pit mining in arid and semi-arid zones, ecological restoration is aimed to support the development and establishment of long-term, healthy and self-sustaining ecosystems with good vegetation cover [20].

Therefore, the restoration of the quarries is necessary to accelerate the recovery of the soil–plant system, and a way of solving these problems could be the use of organic amendments [21]. The model of sustainable development of the mining requires responsible land use and the restoration of affected soils [22–25]. To achieve greener mining with fewer impacts, it is necessary to reduce the pollution and affections produced by activities in the air and water and those produced by noise as well as the need to recover the land and its uses (forest, agriculture, landscape...) after the finishing of the activities on the exploitations [26]. Land cover change is the main force driving changes in ES [27,28]. Understanding the response of ES to changes in open-pit mining is vital for achieving ecosystem sustainability and thus requires more attention [3]. One of the most important services is food production, and mining restoration would be an opportunity to recover agricultural soil uses after the exploitation activities and maintain the economy of a region after the end of the mining exploitation. This is the case of some quarries exhausted to obtain cement that have undergone transformations to maintain cultivated areas in the southeast of Spain.

Soil properties constitute the material basis for soil services, so a sustainable perspective needs to be borne in mind when analyzing multiple soil services and to bring about an overall soil functioning improvement and sustainable development [29]. Arid and semi-arid ecosystems cover a significant area of the planet, around 45% [30], and they are expected to increase in the future due to negative climate change effects. So, there is a need to restore these ecosystems through methodologies that are appropriate and successful. Thus, in mining areas under arid and semi-arid climates, it is appropriate to address this recovery with organic amendments as it increases organic matter content, improves water retention and stimulates soil microorganisms compared to unamended soils [31]. Knowing these positive effects, soil nutrient status is the next step to ensure a good development of plant cover both for agriculture or landscape.

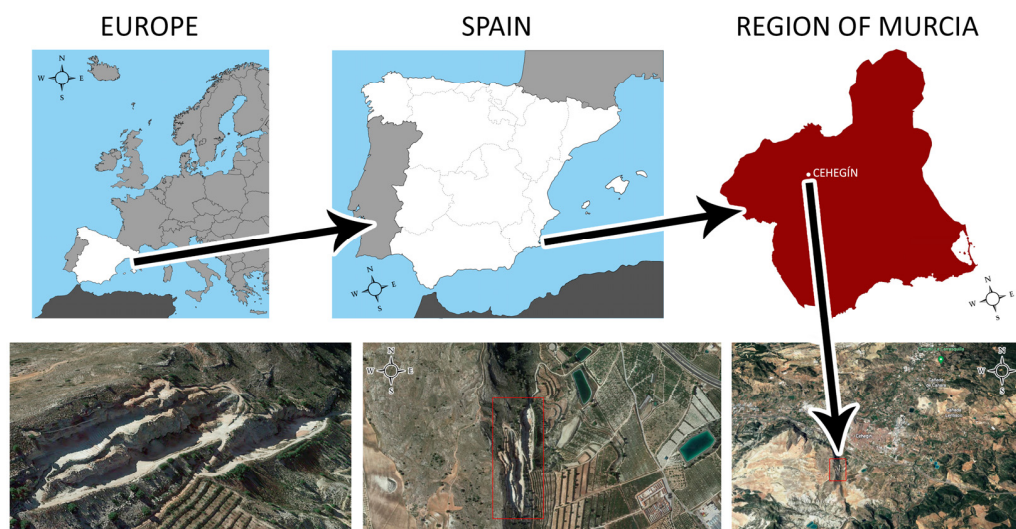
In addition to the degradation of the environment, the generation and agglomeration of waste is one of the greatest global concerns. For this reason, the EU (European Union) has created a zero-waste strategy through the circular economy [32]. For the recovery of soils and landscape after mining activities, a good option is to use organic wastes. In this way, its accumulation is avoided, and it contributes to the circular economy and the European Zero Waste strategy [19,33,34]. The recovery of the soil, the implementation of productive activities such as agriculture and the improvement of the landscape are objectives derived from the application of organic waste and the implementation of the circular economy in the restoration of quarries.

The aim of this study was to evaluate the improvement of soil restoration in open-pit mining areas by using wastes as a part of the zero-waste strategy. It is intended to evaluate the success of different restoration treatments by using four treatments (inorganic fertilizer NPK 15-15-15, pig slurry, pruning waste and urban solid wastes) on the physicochemical properties and nutrient availability of the soil in limestone quarries, trying to facilitate the implementation of rain-fed agriculture. These amendments have been applied because of their presence and high production in the Region of Murcia and the problem of their elimination. The treatments applied are intended to be evaluated in the short term (two and four months from the addition to the soil) and in the medium term (nine months) in order to determine if the restored soils will be suitable for agriculture landscape recovery based on the availability of nutrients. In addition, degraded soils without the application of any treatment were selected as reference and for comparison with the treated plots.

## 2. Materials and Methods

### 2.1. Study Area

The study was carried out on a limestone quarry, whose activity is the extraction of ornamental rock and carbonate aggregates, with an average slope of 8%, situated in the southwest of Cehegín, Murcia, Spain. The quarry is located at latitude  $38^{\circ}4'42.47''$  N and longitude  $1^{\circ}48'23.65''$  W (Figure 1).



**Figure 1.** Location of the limestone quarry in Europe.

According to the geological description, the quarry is part of the outcrop of the Zegrí Formation, Veleta Formation and Upper Ammonitic Rosso Formation (Betic mountain ranges in the southeast of Spain). The lithology of the study area is mainly gray and red limestone strata interspersed with some red marls, and limestones and limestones with flints belonging to the Veleta Formation, all of which are superimposed on gray limestones belonging to the Zegrí Formation (Toarcian–Domerian transition). On the surface, there could be found a large amount of ammonite, belemnite and foraminiferal fauna. Analyzing the hydrology of the quarry, about two kilometers to the south of the quarry, the river Quipar and a little closer Cañada Lengua ravine, which is a tributary of this river, are found. The ravines in the area are shallow, increasing in depth as the slope decreases, and run through loamy materials.

The climate is Mediterranean (Csa) with a hot summer according to the Köppen–Geiger climate classification [35], the coldest months reach, as a minimum temperature  $-3^{\circ}\text{C}$ , the maximum being  $18^{\circ}\text{C}$ . The summer is hot, with the average of the hottest month exceeding  $22^{\circ}\text{C}$ . Precipitation exceeds evaporation, and there is seasonal rainfall. The summer is dry, so the minimum rainfall is quite marked and coincides with the period of highest temperatures. The rainiest seasons are autumn and spring.

The land use of the area is basically rain-fed agriculture and scrubland, composing a traditional Mediterranean landscape. The following vegetation units have been found in the area: disperse pine forest, scrubland (rosemary, esparto grass. . .), rocky vegetation, bare soil and rain-fed crops. The main species of natural plants presented are the following: *Juniperus oxycedrus*, *Rosmarinus officinalis* L., *Stipa tenacissima* L., *Pinus halepensis* Miller and *Olea europaea* var. *Sylvestris*. The main rain-fed crops were esparto grass and hemp but under abandonment because of the low economic profitability, facilitating the soil erosion accentuated by the loss of vegetation cover.

### 2.2. Experimental Design

The study was carried out on one of the four quarry faces exploited in the area on a bench of 8.2 m high (wall) and 105 m long (front). After the selection of the study area

and the information collected, the plots were established. The first task was the removal of all the materials that cannot be used in the conditioning and subsequent leveling of the area. The area was filled with tailings as the terrain was irregular, and the surface was horizontally leveled (slope under 0.5%). The work was carried out following the relieve contour lines to avoid erosion. After that, 380 m<sup>3</sup> of topsoil was added from old alluvial terraces of long-time abandoned fruit cropping areas, with an approximately depth of 20 cm, covering an area of 2500 m<sup>2</sup>.

Once the area was ready, it was divided into 55 experimental plots of 5 × 4 m, leaving a separation of 1 m between them. The inorganic and the three organic amendment treatments were placed on the superficial layer of the soil and mixed with the topsoil with a mechanical backhoe; the soils that were not treated also carried out this mechanical process but without applying any amendment, keeping the characteristics of the original soil from the abandoned alluvial terraces.

The treatments were established and randomized on the bench. Each of the treatments was applied in 11 plots, and 11 control plots were left without any treatment. The treatments applied were the following (see also Table 1):

- (i) The addition of granulate inorganic fertilizer (NPK 15-15-15) in the amount of 1 kg per plot; this mineral fertilizer is composed as follows: 15% TN (total nitrogen), 15% total phosphorus (P<sub>2</sub>O<sub>5</sub>) and 15% water-soluble potassium (K<sub>2</sub>O), and it also contains 25% of sulfur (SO<sub>3</sub>). This treatment was applied since it is a common agricultural fertilizer that is easy to apply (since it is a solid granule); in addition, it contains the 3 elements most demanded by plants from the soil, and its slow dissolution allows the relatively slow release of nutrients into the soil.
- (ii) The addition of pig slurry in the amount of 5.4 kg per plot based on the contribution of nitrogen equivalent to the inorganic fertilizer. The application of pig slurry is of special interest in regions where the management of the pig industry represents a serious environmental problem due to its large volume produced, like the Region of Murcia. One of the main concerns is how to treat them in an environmentally sound way [36]. The pig slurry was obtained from a pig farm in the Murcia region, located in Fuente Álamo, where conventional purification systems are used, in which the solid is separated from the liquid. The slurry obtained on the farm was transported to the treatment plots where it was applied.
- (iii) The incorporation of composted urban solid wastes (USWs) from the Cañada Hermosa waste treatment center in the northwest of the Region of Murcia in the amount of 60 kg per plot. The Cañada Hermosa treatment center is a benchmark for innovation, and it changes the vision of waste as garbage to turn it into resources [37], which is in continuous progress to the circular economy.
- (iv) The application of woody pruning waste (variable size over 5 to 20 cm long), from olive trees, vines and almond trees of the region, in the amount of 60 kg per plot. The almond tree represents more than 12% of the farmland in the Region of Murcia, and it is intended to take advantage of these crops for restoration and mitigate the problem of abandoning agriculture or carrying out uncontrolled incineration [38]. The vineyard and the olive trees are in regression due to abandonment, especially in marginal areas and terraces. The abandonment of crops and soil conservation measures cause a collapse of the terraces and the appearance of different forms of associated erosion [38,39]. The composition of the pruning remains was equally divided between remains of olive trees, vines and almond trees from the farmland in the Region of Murcia. These remains were crushed with a tractor shredder and transported to the treatment plots on which these residues were spread.

The rest of the plots, without any fertilization or amendments added, were the control treatment. These consisted of soil situated on the carbonate tailings, which were used for leveling the terrain, as the rest of the treatments, but in this case, this soil was used to understand the evolution of the properties without any intervention, considering that the

substrate is close to an agricultural non-treated soil [40]. All the organic treatments were applied directly to the plots and mixed with the topsoil.

**Table 1.** Name assigned to each treatment.

T0	T1	T2	T3	T4
Control	Inorganic fertilizer NPK	Pig slurry	Urban solid wastes	Woody pruning wastes

Soil samples were taken at a 10 cm depth for each plot and treatment (11 samples per treatment), at two (S1), four (S2) and nine (S3) months from the beginning of the fertilization treatments (at short and medium term). Samples were taken to know the state of the soil in the first stages and at the end of a reasonable period to achieve some stability previously to planting crops.

### 2.3. Methods of Soil Analysis

Soil samples dried at room temperature were sieved at 2 mm. After that, pH was determined in a soil/water extraction (ratio of 1:2.5 *w/v*) [41,42]. The CaCO<sub>3</sub> equivalent was determined by using the FAO volumetric calcimeter method [43]. Clay, silt and sand were measured with the Bouyoucos method based on Stokes' law [44]. Soil nitrogen was determined by the Kjeldahl method as well as phosphorus by using the Olsen method [45,46].

K, Ca, Mg and Na were determined in the soil by using the ammonium acetate 1 N extraction, and Fe, Cu and Zn were determined by the DTPA-TEA method [47]. After the extraction, Ca, Mg and micronutrients were measured by atomic absorption spectrometry and Na and K were measured by atomic emission spectrometry.

### 2.4. Organic Wastes

The same parameters were analyzed in the organic amendments, pH and nutrients, following the Official Methods of the Ministry of Agriculture of Spain [48], according to Royal Decree 824/2005 and Royal Decree 506/2013 for organic by-products. The pH level was measured in an aqueous extract 1:5 *w/v* [49]; nitrogen was determined by the Kjeldahl method. The elemental composition of K, Ca, Mg and Na and micronutrients Fe, Cu and Zn were determined by atomic absorption or emission spectrometry in the aqueous solutions resulting from dissolving with HCl (1:1) the ashes from the calcination of the organic samples. Phosphorus was determined by the spectrophotometry at 440 nm indicative for the complex formed in the presence of ammonium molybdate and vanadate (yellow color in acid medium). The composition of organic amendments is given in Table 2.

**Table 2.** Composition of organic amendments.

Parameter	Pig Slurry	USW Compost	Woody Pruning Wastes
pH	7.63	7.01	4.71
N (g/kg)	27.7	12.95	5.1
P (g/kg)	2.36	7.00	0.63
K (g/kg)	3.84	3.81	0.63
Ca (g/kg)	4.28	33.5	1.06
Mg (g/kg)	0.76	2.25	0.11
Na (g/kg)	0.88	1.3	0.016
Fe (mg/kg)	252	10050	33
Cu (mg/kg)	46.8	203	3.7
Zn (mg/kg)	85.2	611	9.2



There is a remarkable and important amount of N presented in pig slurry, which is followed by USW and woody pruning wastes. The last one has an acid pH, which will be associated with the presence of organic acids derived from the plant material. However, opposite to N, P was greater in the composted USW. Moreover, USW also had the highest concentration of Ca as well as the major content of Fe, Cu and Zn.

2.5. Statistical Analysis

The descriptive statistics, mean value and standard deviation were calculated for each treatment. Data were tested for homogeneity of variance, and differences between individual means were evaluated by Tukey’s post hoc test at  $p < 0.05$ . Mean values with the same letter in the tables are not significantly different. After that, a one-way ANOVA test was applied to compare the treatments [50].

3. Results

The results obtained for soil pH, equivalent  $\text{CaCO}_3$ , and texture fractions (clay, silt and sand) are presented in Table 3.

Table 3. pH, equivalent  $\text{CaCO}_3$ , clay, silt and sand for the first sampling period (S1), second sampling period (S2) and the third one (S3).

Treat. S1	pH		CaCO3 (%)		Clay (%)		Silt (%)		Sand (%)	
T0	8.9 ± 0.1	a	89 ± 1	a	32 ± 1	a	20 ± 1	a	48 ± 1	ab
T1	8.6 ± 0.1	b	72 ± 1	b	32 ± 1	a	21 ± 1	a	47 ± 1	b
T2	8.9 ± 0.1	a	85 ± 2	cd	32 ± 1	a	20 ± 1	a	48 ± 1	ab
T3	8.8 ± 0.1	a	85 ± 1	c	30 ± 1	a	20 ± 1	a	50 ± 1	a
T4	9.0 ± 0.1	a	87 ± 1	d	31 ± 1	a	22 ± 1	a	47 ± 1	b
ANOVA	***		***		ns		ns		ns	
Treat. S2	pH		CaCO3 (%)		Clay (%)		Silt (%)		Sand (%)	
T0	8.8 ± 0.1	a	89 ± 1	a	31 ± 1	a	21 ± 1	a	48 ± 1	a
T1	8.7 ± 0.1	a	70 ± 1	b	32 ± 1	a	21 ± 1	a	47 ± 1	a
T2	8.8 ± 0.1	a	81 ± 1	c	31 ± 1	a	20 ± 1	a	49 ± 1	a
T3	8.9 ± 0.1	a	84 ± 1	c	30 ± 1	a	20 ± 1	a	50 ± 1	a
T4	9.1 ± 0.1	b	88 ± 1	a	31 ± 1	a	19 ± 1	a	50 ± 1	a
ANOVA	***		***		ns		ns		ns	
Treat. S3	pH		CaCO3 (%)		Clay (%)		Silt (%)		Sand (%)	
T0	8.8 ± 0.1	a	89 ± 1	a	30 ± 1	a	20 ± 1	a	50 ± 1	a
T1	8.6 ± 0.1	b	76 ± 1	b	31 ± 1	a	21 ± 1	a	48 ± 1	a
T2	8.7 ± 0.1	a	81 ± 1	cd	32 ± 1	a	20 ± 1	a	48 ± 1	a
T3	8.9 ± 0.1	a	84 ± 1	c	30 ± 1	a	19 ± 1	a	51 ± 1	a
T4	9.2 ± 0.4	c	86 ± 1	a	30 ± 1	a	20 ± 1	a	50 ± 1	a
ANOVA	***		***		ns		ns		ns	

Mean values with the same letter in a column for each sampling period (S1, S2, S3) do not differ significantly according to Tukey’s test at  $p < 0.05$ ; ANOVA test: ns: not significant; \*\*\* significant at 0.001.

Soil pH showed significant differences ( $p < 0.001$ ) between all the treatments applied and the control plots (T0) both in sampling 1, 2 and 3 (Table 3). Throughout the nine months of sampling (S3), it was observed that the pH slightly decreased over time except in those plots amended with woody pruning waste (T4). Although this amendment had the lowest pH value (Table 2), the effect was not noticed in the soil.

The equivalent  $\text{CaCO}_3$  in the soil, after two months of sampling did not showed significant differences between pig slurry and USW compost (T2 and T3) nor did it have significant differences between pig slurry (T2) and woody pruning waste (T4) (Table 3). In the sampling period 2 (S2) (Table 3), there were no significant differences between the control plots and the pruning plots, but there were significant differences between these plots and the rest of the treatments ( $p < 0.001$ ). After nine months of application of the amendments (S3), considering this time enough for an initial stabilization of the soil before planting, there were no significant differences between the plots treated with USW compost (T3) and the plots treated with woody pruning waste (T4). In fact, the presence of carbonates due to the nature of the substrate is too high, and it would be expected that within time, the carbonates presented in the subsoil and the topsoil will determine poor differences between treatments. Probably, the application of NPK 15:15:15 (T1) affected the carbonates, diminishing the presence in the topsoil by an initial acidification favored by the fertilizer.

Regarding the percentage of clay, silt and sand, no significant variation in the mineral fraction granulometry was observed in all the treatments (Table 3), although minor changes were noticed regarding the sand fraction in the plots treated with NPK (T1) that can be, in part, associated to an initial acidification due to the reaction of the NPK fertilizer with the soil matrix.

Considering the presence of macronutrients and their availability, significant differences were observed between treatments (Table 4). In the case of Kjeldahl nitrogen, after two (S1) and four months (S2), there were significant differences ( $p < 0.001$ ) between all treatments and control plots. After 9 months (S3), T1 and T3 had similar values, but there were significant differences ( $p < 0.001$ ) between NPK (T1) and USW compost (T3) and the rest of the treatments. After nine months of application (S3), in T0, T1 and T2, the soil N decreased, and in T3 and T4, it increased. In general, the N in the topsoil decreased except for in the plots of T3 and T4, where organic waste treatments were applied (Table 4).

P significantly varied between all treatments in both sampling 1 and 3 ( $p < 0.001$ ). In sampling 2, no significant differences between the NPK treatment (T1) and the woody pruning waste (T4) were observed, but there were significant differences ( $p < 0.001$ ) between these two treatments with the rest of the treatments. After nine months (S3), this nutrient decreased in the control plots, while in the plots treated with NPK (T1), pig slurry (T2) and USW compost (T3) increased at four months (S2) and decreased at nine months (S3), which was opposite to the behavior shown in the plots treated with woody pruning waste (T4) (Table 4). However, special attention should be given for this nutrient regarding the calcium carbonate nature of the soil and the possible lack of this nutrient to supply crops necessities because of a possible precipitation of calcium phosphates favored in high pH soil solution.

The available Ca varied between treatments, but the amount in all the treatments was expected to be high enough for plant nutrition, and this macronutrient showed the highest values comparing with the rest analyzed. The available Ca showed some differences in the treatments and along the time. However, treatment T2 (pig slurry) was that which favored the highest values. After nine months (S3), calcium availability decreased in the control plots and those treated with pig slurry (Table 4).

Although Mg varied with the treatments and the sampling period, these variations did not follow the same pattern like Ca. After two months (S1), Mg did not show significant differences between USW compost and woody pruning waste (T3 and T4) but showed significant differences ( $p < 0.01$ ) between both and the rest of the treatments. However, in sampling 2, Mg did not show significant differences between T2 and T3, but these differences appeared between the plots treated with pig slurry (T2) and those treated with woody pruning waste (T4) with the rest of the treatments. In sampling 3, the differences among treatments were reduced.

**Table 4.** Macronutrients (N, P, Ca, Mg, K and Na) for the first sampling period (S1), second sampling period (S2) and the third one (S3).

Treat. S1	N (g/kg)		P (mg/kg)		Ca (g/kg)		Mg (g/kg)		K (g/kg)		Na (g/kg)	
T0	0.11 ± 0.01	a	5.9 ± 0.2	a	7.84 ± 0.08	a	0.63 ± 0.01	a	0.13 ± 0.02	a	0.80 ± 0.04	a
T1	0.83 ± 0.02	b	10.4 ± 0.1	b	7.05 ± 0.05	b	0.75 ± 0.02	b	0.23 ± 0.01	b	1.95 ± 0.01	b
T2	0.94 ± 0.01	c	11.4 ± 0.1	c	8.36 ± 0.08	c	0.70 ± 0.01	c	0.24 ± 0.03	b	1.70 ± 0.02	c
T3	0.66 ± 0.03	d	12.1 ± 0.1	d	6.70 ± 0.10	d	0.62 ± 0.04	d	0.23 ± 0.01	b	2.17 ± 0.02	d
T4	0.48 ± 0.02	e	9.6 ± 0.4	e	6.70 ± 0.23	d	0.61 ± 0.01	d	0.2 ± 0.01	c	2.09 ± 0.01	e
ANOVA	***		***		***		**		***		***	
Treat. S2	N (g/kg)		P (mg/kg)		Ca (g/kg)		Mg (g/kg)		K (g/kg)		Na (g/kg)	
T0	0.11 ± 0.01	a	5.9 ± 0.1	a	7.14 ± 0.05	a	0.62 ± 0.01	a	0.13 ± 0.01	a	0.84 ± 0.05	a
T1	0.84 ± 0.01	b	8.0 ± 0.04	b	7.62 ± 0.14	b	0.73 ± 0.02	b	0.24 ± 0.01	b	1.94 ± 0.02	b
T2	0.75 ± 0.01	c	12.0 ± 0.1	c	7.63 ± 0.08	b	0.66 ± 0.01	c	0.24 ± 0.01	b	1.39 ± 0.01	c
T3	0.56 ± 0.02	d	15.3 ± 0.5	d	7.69 ± 0.10	b	0.65 ± 0.01	c	0.23 ± 0.01	b	1.96 ± 0.01	b
T4	0.41 ± 0.01	e	8.8 ± 0.1	b	7.00 ± 0.37	a	0.58 ± 0.01	d	0.17 ± 0.01	c	2.02 ± 0.02	d
ANOVA	***		***		***		***		***		***	
Treat. S3	N (g/kg)		P (mg/kg)		Ca (g/kg)		Mg (g/kg)		K (g/kg)		Na (g/kg)	
T0	0.10 ± 0.02	a	3.4 ± 0.1	a	6.80 ± 0.08	a	0.58 ± 0.06	ab	0.13 ± 0.01	a	0.81 ± 0.02	a
T1	0.77 ± 0.02	b	6.9 ± 0.1	b	7.28 ± 0.10	b	0.72 ± 0.08	b	0.23 ± 0.02	b	1.62 ± 0.02	b
T2	0.92 ± 0.01	c	13.9 ± 0.1	c	7.60 ± 0.03	c	0.66 ± 0.07	ab	0.20 ± 0.01	b	1.39 ± 0.01	c
T3	0.74 ± 0.02	b	11.8 ± 0.1	d	7.40 ± 0.09	d	0.64 ± 0.60	ab	0.23 ± 0.02	b	1.96 ± 0.01	d
T4	0.57 ± 0.01	d	9.6 ± 0.1	e	6.79 ± 0.16	a	0.55 ± 0.09	a	0.20 ± 0.01	b	1.75 ± 0.01	e
ANOVA	***		***		***		*		***		***	

Mean values with the same letter in a column for each sampling period (S1, S2, S3) do not differ significantly according to Tukey's test at  $p < 0.05$ ; ANOVA test: \* significant at 0.05; \*\* significant at 0.01; \*\*\* significant at 0.001.

Regarding soil K, in sampling 1 and 2, there were no significant differences between the plots treated with NPK, pig slurry and USW compost (T1, T2 and T3), and, in sampling 3, there are no significant differences between plots of the treatments T1, T2, T3 and T4. In the control plots, soil K is lower than in the rest of the plots where the amendments were applied, as it happened with the rest of nutrients except Ca and Mg, which would be due to the nature of soil matrix (Table 4). From two (S1) to nine months (S3) after the application of the treatments, the soil K in the plots treated with NPK, USW compost and woody pruning waste (T1, T3 and T4) maintained its availability, and in the plots treated with pig slurry (T2), it decreased (Table 4).

In the case of Na, significant differences were observed, as it would be expected because of the nature of the amendments and inorganic fertilizer used (T1). There were significant differences in soil Na between all the treatments and the control plots in both S1 and S3. In S2, there were no significant differences between the plots treated with NPK (T1) and those treated with USW compost (T3), but there were significant differences between T1 and T3 and the rest of treatments. In sampling 3, we observed a decrease since the first sampling in T1, T2, T3 and T4. In general, the highest values of extractable Na were obtained in treatments T3 and T4 (Table 4). All the treatments increased the availability of sodium.

There were significant differences in Fe between all the applied amendments and the control plots (T0) in both S1 and S3. In sampling 2, there were no differences in soil Fe between the plots treated with NPK (T1) and those treated with USW compost (T3), but there were significant differences between T1 and T3 and the rest of the treatments. From two months (S1) to nine months (S3), a decrease in Fe was determined in the control plots (T0) and T4, and in T1, T2 and T3, the availability of Fe increased. The treatments that provide the most available Fe were NPK (T1) (45 mg/kg) followed by pig slurry (T2) (42.6 mg/kg) (Table 5).



**Table 5.** Micronutrients (Fe, Cu and Zn) for the first sampling period (S1), second sampling period (S2) and the third one (S3).

<b>Treat. S1</b>	Fe (mg/kg)		Cu (mg/kg)		Zn (mg/kg)	
T0	26.8 ± 0.1	a	6.7 ± 0.1	a	2.6 ± 0.1	a
T1	40.2 ± 0.2	b	11.4 ± 0.1	b	2.3 ± 0.1	b
T2	41.3 ± 0.4	c	14.6 ± 0.1	c	3.6 ± 0.1	c
T3	37.6 ± 0.2	d	11.6 ± 0.1	b	4.4 ± 0.1	d
T4	34.0 ± 0.4	e	11.3 ± 0.2	b	2.8 ± 0.2	a
ANOVA	***		***		***	
<b>Treat. S2</b>	Fe (mg/kg)		Cu (mg/kg)		Zn (mg/kg)	
T0	23.3 ± 0.1	a	6.2 ± 0.4	a	2.4 ± 0.1	a
T1	38.6 ± 0.3	b	10.5 ± 0.1	b	2.4 ± 0.1	a
T2	40.0 ± 0.7	c	14.5 ± 0.7	c	4.2 ± 0.2	b
T3	38.1 ± 0.1	b	11.4 ± 0.3	d	3.5 ± 0.2	c
T4	33.7 ± 0.1	d	10.1 ± 0.1	e	2.6 ± 0.3	d
ANOVA	***		***		***	
<b>Treat. S3</b>	Fe (mg/kg)		Cu (mg/kg)		Zn (mg/kg)	
T0	24.1 ± 0.6	a	6.0 ± 0.2	a	2.7 ± 0.3	a
T1	45.0 ± 0.1	b	11.6 ± 0.3	b	2.6 ± 0.2	a
T2	42.6 ± 0.1	c	15.3 ± 0.2	c	4.7 ± 0.1	b
T3	38.8 ± 0.1	d	10.4 ± 0.1	d	3.3 ± 0.1	c
T4	31.9 ± 0.6	e	12.0 ± 0.1	b	2.1 ± 0.1	d
ANOVA	***		***		***	

Mean values with the same letter in a column for each sampling period (S1, S2, S3) do not differ significantly according to Tukey's test at  $p < 0.05$ ; ANOVA test: \*\*\* significant at 0.001.

Regarding soil Cu, in S1, no significant differences were observed between the plots treated with NPK, USW compost and woody pruning waste (T1, T3 and T4), but there are significant differences ( $p < 0.001$ ) between T1, T3 and T4 and the rest of the treatments. In S2, significant differences ( $p < 0.001$ ) were observed between all the treatments and the control plots (T0). In S3, no significant differences were observed between the plots treated with NPK (T1) and those of woody pruning waste (T4), but there were significant differences ( $p < 0.001$ ) between T1 and T4 and the rest of treatments. From two (S1) to nine months (S3) after the application of the treatments, it was observed that soil Cu decreased in the control plots and T3 and increased in T1, T2, and T4 (Table 5).

The availability of Zn in the soil showed no differences between the control plots (T0) and those treated with woody pruning waste (T4) in S1, but there were significant differences ( $p < 0.001$ ) between T0 and T4 and the rest of the treatments. After four (S2) and nine months (S3) of the application of the treatments, there were no significant differences between T0 and T1, but there were significant differences ( $p < 0.001$ ) between T0 and T1 and the rest of the plots. From two (S1) to nine months (S3) after the application of the treatments, it is observed that the Zn of the soil decreases in T3 and T4 and increases in T0, T1 and T2.

The extractable Fe, Cu and Zn from S1 to S3, maintained, more or less in each treatment, the same amount of micronutrient availability. The T1 and T2 treatments showed the highest values for Fe and Cu, while Zn showed the highest extractable value in T2 (4.7 mg/kg), although the USW compost (T3) was where those micronutrients were presented in the highest elemental concentration in the waste composition (T3) (Table 5).

As shown in Table 5, the high content of Fe, Cu and Zn in the USW compost did not affect the quality of the treated soil; even lower availability was obtained in some cases comparing with the other treatments.

#### 4. Discussion

In all the treatments, even in the control plots (T0), the pH exceeds 8.5, and therefore, it is an alkaline soil due to calcareous matrix due to the nature of the area, with a high presence of calcium carbonate. In other studies [51,52], it was observed that the presence of calcium carbonate from the marble in the mine areas increased the pH level of the soil, remaining stable for at least one year. The values similar to those obtained in this study were found in a study carried out in 2016 [53], in which several combinations of organic amendments (sewage sludge and compost from domestic organic waste) and mulches (gravel and woodchip) were applied in a quarry in a semi-arid climate. In another study carried out in 2017 [54], the pH was increased to values close to neutrality due to the presence of carbonate in the wastes; marble waste and pig slurry were used to rehabilitate a mining waste dam. With the application of other waste, for instance, from the olive oil industry, an increase in the soil pH was also found [55,56] even in calcareous soils. In our study, after nine months (S3) from the addition of the amendments, it was observed that the plots treated with NPK and pig slurry (T1 and T2) slightly decreased the pH, but it was still lower than in the control plots (T0) (Table 3). Similar results were observed in a study carried out in 2020 [57], where the application of sewage sludge from urban waste water treatment plants and compost from domestic solid waste in degraded soils from calcareous quarries with a semi-arid climate reduces pH levels over time. In [58], the pH values were maintained between 8 and 8.74. On the contrary, in our experiment (Table 3), nine months (S3) after the application of the USW compost and woody pruning waste amendments (T3 and T4), the pH slightly increased.

The availability of nutrients such as potassium, calcium and magnesium would be associated with the calcium carbonate matrix because they are more mobile at pH above 6 [59]. In addition, it was shown that with the application of the organic amendments used and the inorganic fertilizer, soil macronutrients were considerably increased (N, P, K, Na to a greater extent and Ca and Mg to a lesser extent) in reference to the control plots (T0), as it was expected. However, it was observed that after nine months (S3), the content of these nutrients was still higher than in the control plots except for Ca and Mg (Table 4).

The increase in N content is very positive, since this macronutrient is an essential element for vegetation growth and for microbial communities, which are responsible for many of the necessary processes for plant nutrition. Therefore, its increase contributes to the improvement of soil nutritional conditions and to the growth and development of introduced crops [54] after the restoration process.

Similar results are observed in a study carried out in a limestone quarry in a semi-arid climate [53] in which organic amendments (sewage sludge and compost from domestic organic waste) were applied, where the soils showed higher N, P and K content than the soil without treatments. Another study [54] also showed that the application of amendments (marble waste and pig slurry) significantly increased soil N in an old mining waste dam recovery in a semi-arid climate. The application of pig manure increased soil N [59]. In an abandoned mine tailing under a semi-arid climate, the application of compost made from pig slurry also increased the N and nutrient (mainly K and P) contents in the soil solution [60]. The sewage sludge from urban waste water and compost from domestic solid waste also significantly modified soil chemical properties, increasing nutrient (N) availability in a limestone quarry with an arid/semi-arid climate [57].

The micronutrients (Fe and Cu) also increased significantly with the application of the amendments used in this study (Table 5). Regarding Zn, it increased in the plots treated with pig slurry and USW compost (T2 and T3) after nine months (S3) from the application of the treatments. The soil treated with NPK (T1) has similar Zn values as the control plots (T0), and in the soils treated with woody pruning waste (T4), the Zn concentration in the

soil decreased from sampling 1 to sampling 2, and it was even lower than in the control treatment (T0). In a study carried out in an abandoned mining waste dam, it was observed that with the application of pig slurry and marble waste, the Zn concentration decreased after two years of the application of the amendments [54], and the application of compost made from pig slurry also reduced Zn concentration in the soil [60].

Long-term pig slurry applications have a cumulative effect on the availability of N and other nutrients [61,62]. The use of pig slurry as fertilizer increases the concentrations of N, K, P, Ca, Mg and Na [55]. The high nutrient content of pig slurry, mainly N and P, could be a positive for plant growth, as also shown in a study carried out in soil from a mining area in a semi-arid climate [63]. Furthermore, its use is of great interest, since it provides many nutrients to optimize the growth of the vegetation cover [37].

Woody pruning waste has been recognized as limiting the loss of water by evaporation, improving filtration and root growth in addition to establishing vegetation and reducing soil erosion [64–67]. The establishment of plant cover will provide great benefits particularly in arid and semi-arid areas [68]. Along with crop establishment, woody pruning waste increases the stability of soil aggregates [65]. Other strategies can be applied instead of woody pruning waste such as plastic or stones, which improve water filtration [69–71]. However, by applying pruning residues as mulch or incorporated to the topsoil, we avoid their uncontrolled incineration, since this incineration forms a source of disease and expels large amounts of CO<sub>2</sub> into the atmosphere without positive effect regarding the mitigation of the negative effects of climate change. Moreover, its abandonment in boulevards and vacant land helps the spread of diseases and pests and generates a significant risk when there is torrential rain [20].

USW compost induces an increase in TOC (total organic carbon) and glomalin content in the soil, as several works showed [72–74]. The increase in the TOC content of the compost is caused by the stable nature of the amendment [73]. The compost increases microbial biomass and modifies community composition to form a fungal-dominated community, and these fungi are capable of degrading more recalcitrant organic material [74]. In addition, the growth of a mulch on compost-treated soils can improve the uptake of cellulose and lignin into the soil [75]. By using USW compost, we reduce our carbon footprint, avoid incineration, reduce the risk of disease spread (because USW can attract rodents, animals, and insects and can act as disease vectors), and furthermore, we promote the circular economy. Moreover, as this work shows, the availability of plant nutrients is key to understanding the use of this composted matter.

Pig slurry has a significant amount of organic matter; furthermore, it supplies macronutrients such as N, P, K, Na, Mg and Ca that favor the development of vegetation. Even more, exchangeable cations (Ca<sup>2+</sup> and Mg<sup>2+</sup>) positively affect soil aggregation. Opposite to this positive effect, the presence of Na mainly causes the aggregates to be unstable and the clay particles to spread [76]. The spreading of the aggregates can form crusts on the soil; this generates a slow filtration of water into the soil and also causes the particles to move faster due to surface runoff. In addition, exchangeable cations (Ca<sup>2+</sup> as Mg<sup>2+</sup>), in limestone soils, decrease clay dispersion [77].

The addition of pig slurry increases soil fertility and therefore improves the colonization of vegetation [78]. It also increases N and TOC, improves the soil structure and supplies nutrients to plants microbial populations, which are very important for recovery of the ecosystems [79]. The increase in soluble labile organic matter in pig manure can generate greater microbial growth [80].

## 5. Conclusions

The addition of the amendments used in this experiment to the soil was very beneficial, increasing the availability of plant nutrients in the soil and therefore facilitating the achievement of soil conditions for the development of crops. Moreover, in this area, this strategy could favor the implementation and the recovery of rain-fed Mediterranean agriculture that also contributes to landscape recovery.

All of the treatments favored the availability of nutrients but with several differences. The inorganic fertilization, the application of NPK fertilizer, increased the N and P, as it was expected. However, pig slurry increased the soil N and USW of the P content; both nutrients presented in their composition in significant amounts, respectively. Moreover, the last one favored the presence of the micronutrients studied (Fe, Cu and Zn). However, the high presence of calcium carbonates in the soil matrix controlled the high availability of Ca and Mg. In general, the values obtained for the nutrients analyzed in the soil do not pose any risk to the environment, and their availability can favor the development of the plant cover. Several considerations should be undertaken if the recovery of the soil for agriculture has a main role of restoring the landscape or looking for higher yield, and in such a case, more studies should be conducted.

The use of these wastes in the restoration of mining areas is feasible. Recovering the soil–plant systems is a necessary strategy for landscape, agriculture and forestry in restored areas. In addition, if organic amendments are used, the development of the circular economy will be promoted as well as the zero-waste strategy.

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