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Medium-Term Effect of Organic Amendments on the Chemical Properties of a Soil Used for Vegetable Cultivation with Cereal and Legume Rotation in a Semiarid Climate

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Abstract: The response of a Petrocalcic Palexeroll dedicated to the cultivation of vegetables (*Apium graveolens* L.) with a rotation of a mixture of cereals and legumes when organic amendments are added has been studied; specifically, three fertilization treatments were experimented with: two organic (LSM and COA) and one with chemical fertilizers (I). The LSM and COA plots were managed according to the regulations governing organic production, while in I, conventional agricultural production practices were used. Over a three-year period, monthly samples were taken from the topsoil and analyzed for organic carbon (OC), total nitrogen (TN), C/N ratio, total P (P), electrical conductivity of the saturation extract (ECext) and extract ions, pH in water (pHw) and 1 M KCl (pHKCl), cation exchange capacity (CEC), exchangeable bases (Mg^{2+}, K^+, Na^+), and assimilable elements (Fe, Cu, Mn, Zn). The results obtained indicated significant differences between the treatments for the OC, TN, C/N, and P; specifically, in the case of OC the mean concentrations were 22.2 > 20.1 > 17.5 g kg^{-1} for the LSM, COA, and I, respectively. The addition of organic amendments also improved the soil function for food production, evidenced by the higher concentrations of K, Mg, and micronutrients. The ECext and extracted ions were also sensitive to the treatments such that the sequence of ECext in the third year was COA = LSM > I, with values of 5.0, 4.8, and 3.3 dSm^{-1}, which forced a rethink of the doses of the amendments applied. These results suggest that the LSM-based agronomic model had a beneficial effect on the soil properties and contributed to its function as a C sink.

Keywords: organic fertilization; sustainable management; carbon capture; organic farming

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1. Introduction

The agricultural practices and soil management model implemented in developed countries since the second half of the 20th century are causing the accelerated degradation of this natural resource and the ecosystems affected. For this reason, international organizations such as the FAO, the European Union, and the International Union of Soil Sciences (IUSS), among others, are promoting more environmentally friendly agricultural models and developing regulations for their effective application. In this sense, many studies have shown that intensive agricultural activities, oriented exclusively towards obtaining higher economic yields, have led to the progressive degradation of the environment, with the consequent loss of productive capacity [1–3]. This degradation is reflected in the soil in many ways, which includes episodes of physical degradation such as erosion [4,5], decreasing the soil porosity and hindering the exchange of greenhouse gases [6,7], as well as reducing the water retention capacity [8].

These intensive agriculture models also originate chemical degradation processes, as is the case for soil salinization and contamination, especially in semiarid regions similar to the area where this study was conducted [9], as well as to the appearance of nutritional deficiencies [10]. Therefore, the development and implementation of agricultural models that incorporate the objectives of the economic profitability of farms and others related to the preservation of natural resources, especially soil, would be highly recommended; incorporating sustainable agriculture techniques oriented towards a circular economy model [11], where most of the waste generated on farms is used [12–14], is one such example. In this sense, the promotion of the circular economy is one of the priority objectives to protect our planet and alleviate the greenhouse effect that has been gradually increasing in recent years [15]. One of the most widely used practices in sustainable agriculture is the use of organic fertilizers as a substitute for synthetic mineral fertilizers. Organic matter (OM) is a scarce component in the soils of semiarid climates, such as those of Southern Spain, and yet it is probably the most influential component of the properties of these soils. Thus, the use of organic amendments increases their quality and fertility [16,17] and has positive effects on production by providing essential macro- and micronutrients for crop development [18], thereby increasing the yield [19]. It also improves physical properties, such as the structure [20,21], as well as the chemical [22] and biological [23] properties of the soil.

In addition, it has been shown that the use of organic amendments favors the soil's role as a carbon sink [24,25]; although, when animal manure is used the impact on soil SOC is highly variable and depends on factors that are not yet well understood. In this regard, international agreements, particularly the 2006 Intergovernmental Panel on Climate Change (IPCC) guidelines, recommend that countries report changes in SOC stocks as a result of manure application for national greenhouse gas inventories.

On the other hand, when a reduction in the OM content occurs it causes a deterioration of the soil's physicochemical properties and, consequently, loss of productivity in the medium and long term [26,27]. The implementation of sustainable agricultural practices is accompanied by changes in most properties, which can affect soil fertility and alter the availability of nutrients for crops, all as a consequence of physical and chemical changes in rhizospheric environments [28]. As a result, it is necessary to monitor the soil properties that are most sensitive to soil management. These properties can be chemical quality indicators, such as organic carbon (OC), total nitrogen (TN), Olsen phosphorus (P), electrical conductivity (ECext), pH, cation exchange capacity (CEC) and exchangeable bases, and assimilable elements, to help ensure that the transition of management change is conducted properly, ensuring the soil quality and productivity. Some of these quality indicators are rapid response indicators [29], since the interpretation of their results allows for the prediction of soil quality evolution, because they are sensitive to soil management and agronomic management.

To carry out this study, a 2.5 km^2 farm was selected, located in the NE of the province of Granada (Southern Spain), where several projects on the sustainable management of its water, soil, and livestock resources were developed [29]. The farm had a plot of almond trees, which occupied most of the farm, and a plot of cereals, both rainfed. There was also a plot with water available for irrigation, dedicated to the cultivation of vegetables. The farm was completed with a flock of sheep of the Segureña breed.

In this part of the farm, where irrigation water was available, an experimental design was implemented to validate the agronomic and environmental impact of the application of sustainable agricultural techniques, including the use of organic amendments, whose production and crop quality results were published by Sanchez-Navarro et al. [30]; the environmental results are the subject of this article. Due to the marked continental character of the climate of the area, with winters in which minimum temperatures can drop below 0 °C for several months and very hot and dry summers, a farm management system was designed that combined a rotation of rainfed arable crops between October and May, based on cereals and legumes, with a cycle of outdoor vegetables (celery), taking advantage of the summer period.

In the starting hypothesis we considered that the use of organic amendments, as the basis of nutritional inputs, should have a positive impact on soil quality indicators compared to conventional inorganic fertilization and, hence, on some functions of the soil, such as food production and its role as a carbon sink. In addition to the use of other agricultural practices, such as crop rotation and extensive grazing, a model for the integrated and sustainable management of natural resources was implemented which, in economically depressed and depopulated areas such as the one where this study has been carried out, can represent a tool that optimizes the economic viability of farms.

The objective of this study was to determine the influence of the application of two types of organic amendments on sustainably managed crop soils versus conventional fertilization based on chemical fertilizers and intensive soil management on some soil quality indicators, their evolution in the study period, and the effect on soil functions.

2. Material and Methods

2.1. Experimental Design and Soil Sampling

The study site, with the UTM coordinates 30S 535450-4192119 (Figure 1), is characterized by a continental Mediterranean climate with a mean annual temperature of 12.7 °C and a thermal oscillation of 15 °C. It is located in the municipality of Huéscar (Granada, Spain) in an agricultural field planted with celery (*Apium graveolens* var. *dulcis* cv. "*Golden Spartan*"). The annual precipitation is 400 mm, with the rainfall being concentrated in the spring and autumn, while winter and summer are predominantly dry. The annual ETP is 599 mm, and so the annual water deficit is approximately 200 mm.

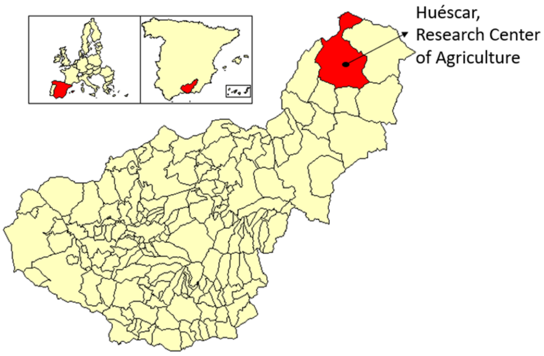

Figure 1. Geographical location of the study area.

The soil has an Ap1-Ap2-Bw-Ckm profile, which can be classified as Petrocalcic Palexeroll [31] and Petrocalcic Kastanozem (Cambic, Loamic) [32]. Some characteristics of the soil, before starting the experiment, are shown in Table 1.

Table 1. Characteristics of the soil at the beginning of the experiment.

Horizon.	Depth (cm)	OC	TN	C/N	pH_w	pH_{KCl}	EC	CEC	$CaCO_3$
Ap1	0–35	21.6	3.1	7.0	8.2	7.6	1.1	11.1	453
Ap2	35–52	12.4	1.7	7.3	8.3	7.6	1.5	12.3	402
Bw	52–65	13.6	1.7	8.0	8.4	7.5	1.4	18.5	349
Ckm	65	-	-	-	-	-	-		875

OC, organic carbon, g kg^{-1}; TN, total nitrogen, g kg^{-1}; pH_w, pH soil–water 1:1; pH_{KCl}, pH soil–KCl 1:1; EC, electrical conductivity, dS m^{-1}; CEC, cation exchange capacity, $cmol^+$ kg^{-1}; $CaCO_3$, calcium carbonate equivalents, g kg^{-1}.

Three fertilization treatments were tested: two organic treatments; local sheep manure (LSM) (Table 2) and a commercial organic amendment (COA) called ECOMAÑAN

(F0001491/2020) (Table 3); and one inorganic treatment (I), which was considered the control. In the LSM treatment, local sheep manure was applied at a rate of 1.7 kg m^{-2}, while in the COA treatment a commercial organic amendment based on sheep manure and peat was applied at a rate of 0.7 kg m^{-2}. The inorganic or mineral treatment (I) involved conventional chemical fertilizers (such as 15-15-15, NH_4NO_3, KNO_3, K_2SO_4, H_3PO_4, $Ca(NO_3)_2$, NH_4NO_3, and $Mg(NO_3)_2$ solution) applied, both as a basal dressing and in drip irrigation systems, at the levels recommended by various authors [33] for this crop, as listed in Table 4. The fertilizer doses were calculated considering two precepts. The first was that treatment I should be carried out taking as a reference the fertilizer recommendations existing in the bibliography [33], especially in similar soils. The second was the imposition of European regulations governing organic farming, which limit the amount of N ha^{-1} to a maximum of 170 kg.

Table 2. Chemical analysis of the sheep manure (LSM), with the nutrients given on a dry matter basis.

Year	M	pH	EC	C/N	OC	TN	P	K	Ca	Mg
1	64.8	8.1	5.5	20	481.1	24.0	6.1	19.6	18.0	3.7
2	59.7	8.0	5.3	20	445.7	22.3	6.1	31.0	26.6	2.8
3	61.1	8.3	5.2	19	439.0	23.1	6.1	25.4	21.1	4.2

M, % moisture; pH, pH 1:10; EC, electrical conductivity 1:5, dS m^{-1}; OC, organic carbon, g kg^{-1}; TN, total nitrogen, g kg^{-1}; P, total phosphorus, g kg^{-1}; K, total potassium, g kg^{-1}; Ca, total calcium, g kg^{-1}; Mg, total magnesium, g kg^{-1}.

Table 3. Chemical analysis of the commercial organic amendment (COA), with the nutrients given on a dry matter basis.

M	pH	EC	C/N	OC	TN	ON	P	K	Mg	Fe	S
14.0	6.0	9.6	11.8	232.6	23.3	19.8	10.2	9.6	1.4	11.6	41.9

M, % moisture; pH, pH 1:10; EC, electrical conductivity 1:10, dS m^{-1}; OC, organic carbon, g kg^{-1}; TN, total nitrogen, g kg^{-1}; ON, organic nitrogen, g kg^{-1}; P, total phosphorus, g kg^{-1}; K, total potassium, g kg^{-1}; Mg, total magnesium, g kg^{-1}; Fe, total iron, g kg^{-1}; S, total sulfur, g kg^{-1}.

Table 4. The N, P, K, Ca, and Mg applied to the inorganic and organic plots (kg ha^{-1}).

Year	LSM					I					COA				
	N	P	K	Ca	Mg	N	P	K	Ca	Mg	N	P	K	Ca	Mg
1	165	46	264	107	22	231	36	431	117	28	161	72	175	95	8
2	165	42	268	182	19	298	42	534	132	36	153	62	151	92	8
3	169	40	202	140	28	241	37	461	110	34	156	62	92	90	8
Mean	166	43	245	143	23	257	38	475	120	33	157	65	139	92	8

A completely randomized experimental design, as described by Little and Hills [34], with three treatments and two replicates was used, resulting in a total of 6 plots, each 64 m^2. Each plot was sampled monthly, between July and October, during the three years of the experiment, with a total of 72 samples of the arable soil layer (10 cm). Each sample was composed of subsamples from four different points of each plot. The samples were air dried and sieved to 2 mm for subsequent analysis in the laboratory.

Over the three years of the experiment, the celery crop was rotated with cereal (*Avena sativa*, L.) and legume (*Vicia sativa*, L.) mixtures, in a 1:3 ratio, in the winter and spring. The celery was sown in the first week of May. Transplanting took place in the third week of June at a density of 12 plants m^2 and harvesting at the end of October for all crop cycles. Before sowing the celery, the fields were prepared and ploughed with a chisel to ensure homogeneous conditions for both soil and irrigation. The fields were also plowed with a disk harrow to a depth between 10 and 15 cm before sowing the cereal and legume crops at the end of January. Both the plant residues from the cereal and legume crop and the celery crop were used as pasture for livestock and, therefore, only the roots of the plants and the lower part of the stem were incorporated into the soil.

The organic plots received organic amendments before planting. The mineral plot received synthetic fertilizer applied, according to values recommended for celery crops, through drip irrigation systems [33]. The doses (kg ha^{-1}) of the nutrients (N, P, K, Ca, and Mg) applied to the inorganic and organic plots are shown in Table 4. Commercial pesticides were not used during this study on any of the experimental plots, and only products authorized for organic agriculture (R (EC) 834/2007) were used for pest and disease control.

2.2. Analytical Methods

The analytical methods used were as follows: OC and TN contents were determined in an elemental analyzer (Leco, model CHNS-932) and the pH in a 1:1 suspension of soil in water (pHw) and in a 1:1 suspension of soil in 1 M KCl (pH_{KCl}) [35]. A soil-saturated paste was prepared for ECext determination [36]. The total carbonates ($CaCO_3$) were determined by volumetric analysis using a Bernard calcimeter [36]; the cation exchange capacity (CEC) by means of the method described in [37]; the Na, K, and Mg cations extractable with ammonium acetate (AcNH4-Na, K, and Mg) using atomic absorption [36]; and P using Watanabe and Olsen's method (Olsen-P) [35]. The Fe, Cu, Mn, and Zn were determined using atomic absorption after extraction with a solution of 0.05 M DTPA, 0.01 M $CaCl_2$, and 0.1 M triethanolamine at pH 7.3 (DTPA Fe, Cu, Mn, and Zn) [35].

The estimation of the total carbon capture was estimated from the OC in each treatment, according to the equation:

$$OCc = OC.BD.depth.10$$

where OCc is the organic carbon capture in Mg ha^{-1}; OC (g kg^{-1}); BD is the bulk density (1.25 g cm^{-3}); and the depth is 0.10 m (depth of the soil sampled (m)).

The efficiency (E) for each treatment was calculated using the following equations:

$$E(LSM,\ I) = \frac{OCcLSM - OCc\ I}{OCcLSM}100$$

$$E(LSM,\ COA) = \frac{OCcLSM - OCc\ COA}{OCcLSM}100$$

$$E(COA,\ I) = \frac{OCcCOA - OCc\ I}{OCcCOA}100$$

The efficiency (E) estimates were made using carbon capture data from the third year of the study.

2.3. Statistical Methods

Statistical analyses were made using the R software Development Core Team 2009 [37] to obtain the general statistics and analysis of variance (ANOVA). Thus, when the data met the requirements of normality and equality of variances, Tukey's test was performed to locate the differences. However, when the data did not fit a normal distribution and the logarithmic transformation did not, a situation more frequent in the data of this work, the Kruskal–Wallis rank sum test was applied between the data series of the three plots to determine the significance of the possible differences between trials reflected in the text, when appropriate, as the X^2 statistic, the degrees of freedom (df), and the p-value. Next, the Wilcoxon test was performed contrasting pairs of trials to locate when these were significant, a situation that is reflected by the corresponding W statistic and its p-value. A significance level of 0.05 was established for all tests.

3. Results

3.1. pH, $CaCO_3$, Organic Carbon, Total Nitrogen, C/N Ratio, and Olsen P

Regarding the general soil parameters, it should be noted that for the pH values, both in water and KCl, the differences recorded were not statistically significant (*p*-value = 0.342 and 0.949, respectively), obtaining mean values of 8 and 7.5 for pHw and pH_{KCl}, respectively. The $CaCO_3$ also showed no differences between treatments, neither was any change observed over the 3 years of the experiment, obtaining mean values of 550 g kg^{-1}.

As can be seen in Table 5, OC presented significantly higher values in the LSM than in the other two treatments (W = 538, $p < 0.001$), with a significant increase in this constituent during the trial period, while in the I and COA it remained unchanged. In the COA, the OC content was intermediate between I and LSM and remained approximately constant throughout the experiment. Finally, it should be noted that, although the OC values were not as high as in the LSM, significant differences were observed with I (W = 57, $p < 0.001$).

Table 5. Content and evolution of the OC (g kg^{-1}), TN (g kg^{-1}), C/N, and Olsen P (mg kg^{-1}).

	OC			TN			C/N			P		
	LSM	**I**	**COA**	**LSM**	**I**	**COA**	**LSM**	**I**	**COA**	**LSM**	**I**	**COA**
Year 1	20.7 c	17.8 a	20.4 a	3.0 c	2.9 a	3.4 a	6.9 c	6.2 c	5.9 c	44.3 b	27.1 b	23.5 b
Year 2	22.3 b	17.8a	19.8 a	2.6 b	2.1 c	2.4 c	8.6 a	8.1 a	8.2 a	50.8 a	31.2 a	32.9 a
Year 3	23.7 a	16.8 a	20.2 a	3.2 a	2.3 b	3.1 b	7.4 b	7.7 b	6.8 b	52.0 a	33.1 a	34.6 a
Mean	22.2	17.5	20.1	2.9	2.4	3.0	7.7	7.3	6.9	49.0	30.5	30.3
SD	4.4	1.6	0.3	0.4	0.5	0.5	0.7	0.8	1.1	4.1	3.0	5.9
p-Value *	5.85×10^{-9} ***			0.4×10^{-2} ***			0.04 **			2.56×10^{-7} ***		

SD, standard deviation. * $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$. "a", "b", and "c" indicate significant differences between years at the 95% confidence level.

As for total nitrogen (TN), higher values were recorded during the three years in the LSM and COA (Table 5), such differences being significant between LSM and I (W = 459, *p*-value = 0.0004) and between I and COA (W = 132, *p*-value = 0.0013). There was no clear trend in this variable throughout the trial period, as it decreased between the first and second years and increased from the second to the third years in all treatments.

As for the C/N ratio, significant differences were also detected between the LSM and COA (W = 408, *p*-value = 0.013), with the highest values corresponding to the LSM, with an average of 8, while the lowest values were found in the COA plot, where the average value throughout the experiment was 6.9. The evolution of the C/N ratio over the three years did not show a fixed trend either, increasing between the first and second years and decreasing between the second and third, although the value at the end was statistically higher than at the beginning.

The Olsen phosphorus (P) showed that the LSM treatment presented significant differences with respect to I (W = 529, *p*-value = 7.05×10^{-7}) and COA (W = 408, *p*-value = 6.59×10^{-6}) such that the average of this macronutrient was higher in the LSM. As for its evolution during the experiment, a statistically significant increase was observed.

3.2. Cation Exchange Capacity and Exchangeable Bases

Regarding the CEC, Figure 2A shows the evolution of this property in each of the trials over the three years of the experiment. As can be seen, the highest values were obtained in the COA and the lowest in the I, while the LSM had intermediate values. Likewise, the statistical analysis showed significant differences between all treatments (LSM-I:W = 497.5, *p*-value = 1.449×10^{-5}; I-COA:W = 67.5, *p*-value = 5.511×10^{-6}). As for its evolution throughout the trial, no significant differences were observed between the different years.

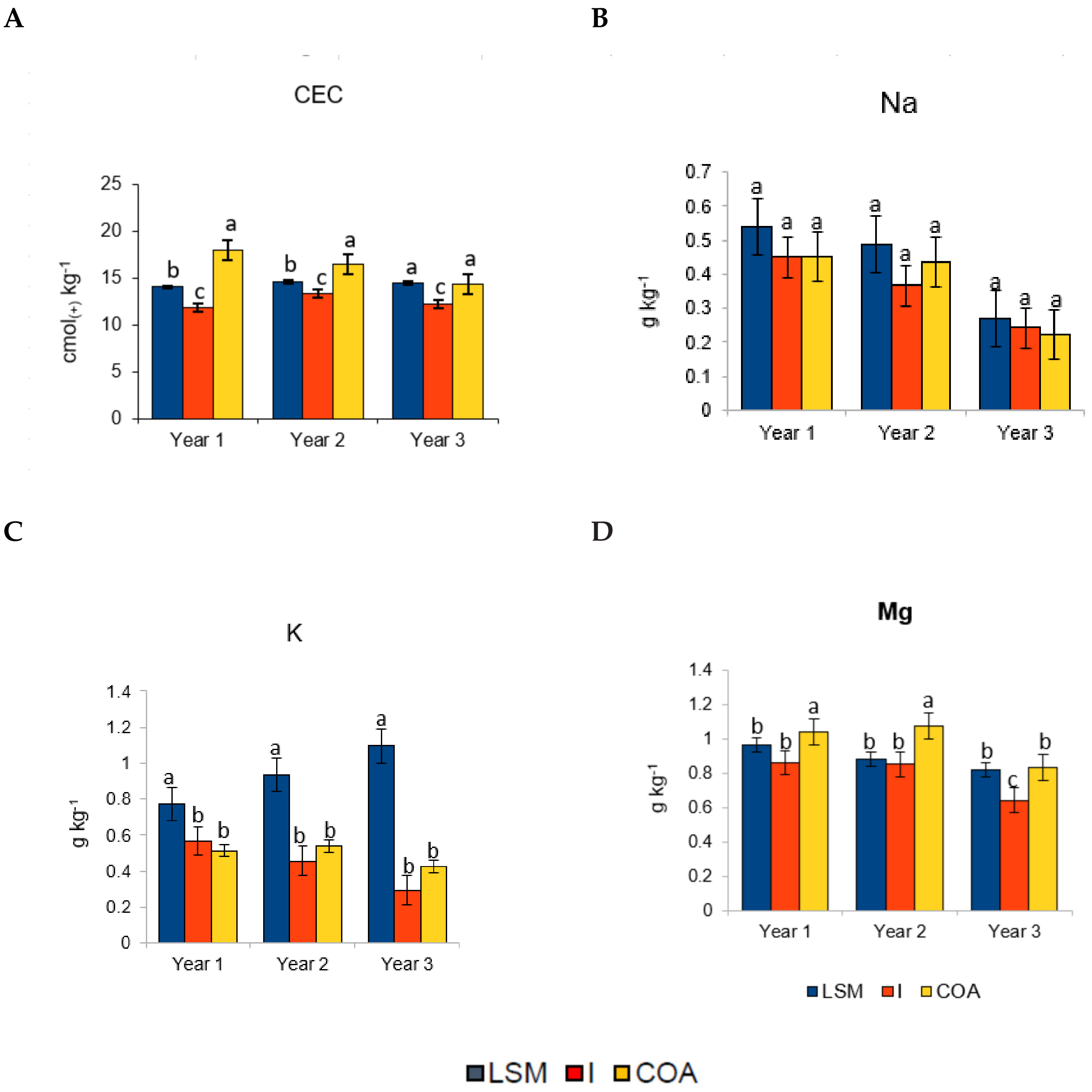

Figure 2. Evolution of the CEC (**A**) and Na, K, and Mg exchangeable cations during the experiment, (**B**–**D**) figures, respectively. $^{a–c}$ Significant differences at 95%.

As for the bases of change, Na (Figure 2B) did not show significant differences throughout the study between treatments, although a tendency to decrease throughout the trial was observed. The potassium content, however, was significantly different, at 99%, between the treatments (LSM > I > COA), with the statistical significance of these differences being as follows: between the LSM and I, W = 501.5, *p*-value = 1.115×10^{-5}; LSM and COA, W = 518, *p*-value = 2.203×10^{-5}; and no statistical significance between the I and COA. As for the evolution of the experiment over the years, an inverse trend was observed between the LSM and the rest, since in the former the potassium increased from 0.77 in year 1 to 1.09 g kg^{-1} in year 3, while in the I the contents decreased from 0.56 to 0.29 g kg^{-1}, respectively, and in the COA they remained more or less stable with a mean value of 0.49 g kg^{-1}, as shown in Figure 2C.

The assimilable Mg values also showed differences between treatments (Figure 2D); thus, between the LSM and COA a statistical significance of W = 394 with a *p*-value = 0.02948 was obtained, while between the I and LSM there was W = 186.5 with a *p*-value = 0.0372, and in the I-COA pair the values were W = 99 with a *p*-value = 0.0001006. Finally, over the three years the tendency was to remain constant, except in the treatments I and COA, where a decrease was observed between the second and third years.

3.3. Electrical Conductivity of the Soil Extract Saturation and Ion Content in the Solution

The ECext showed different trends according to the treatment applied, increasing slightly in the LSM throughout the experiment (Figure 3A), from 4.5 in year 1 to 4.8 dS m^{-1} in year 2, with the mean value in this treatment being the highest of the three (4.5 dS m^{-1}). In trial I, the ECext decreased, especially in the second year, with a mean value of 3.5 dS m^{-1}. The mean value in the COA was intermediate between the other two trials (4.2 dS m^{-1}), although the tendency in this trial was to increase throughout the experiment. The differences between the three treatments, with two degrees of freedom (df = 2), were significant with a *p*-value of 0.003894 ($X^2 = 11.0966$, df = 2, *p*-value = 0.003894), specifically between the LSM and I (W = 118.5, *p*-value = 0.001556) with a 99% degree of significance, as well as between I and COA (W = 1075, *p*-value = 0.02958), while between the LSM and COA trials there were no differences.

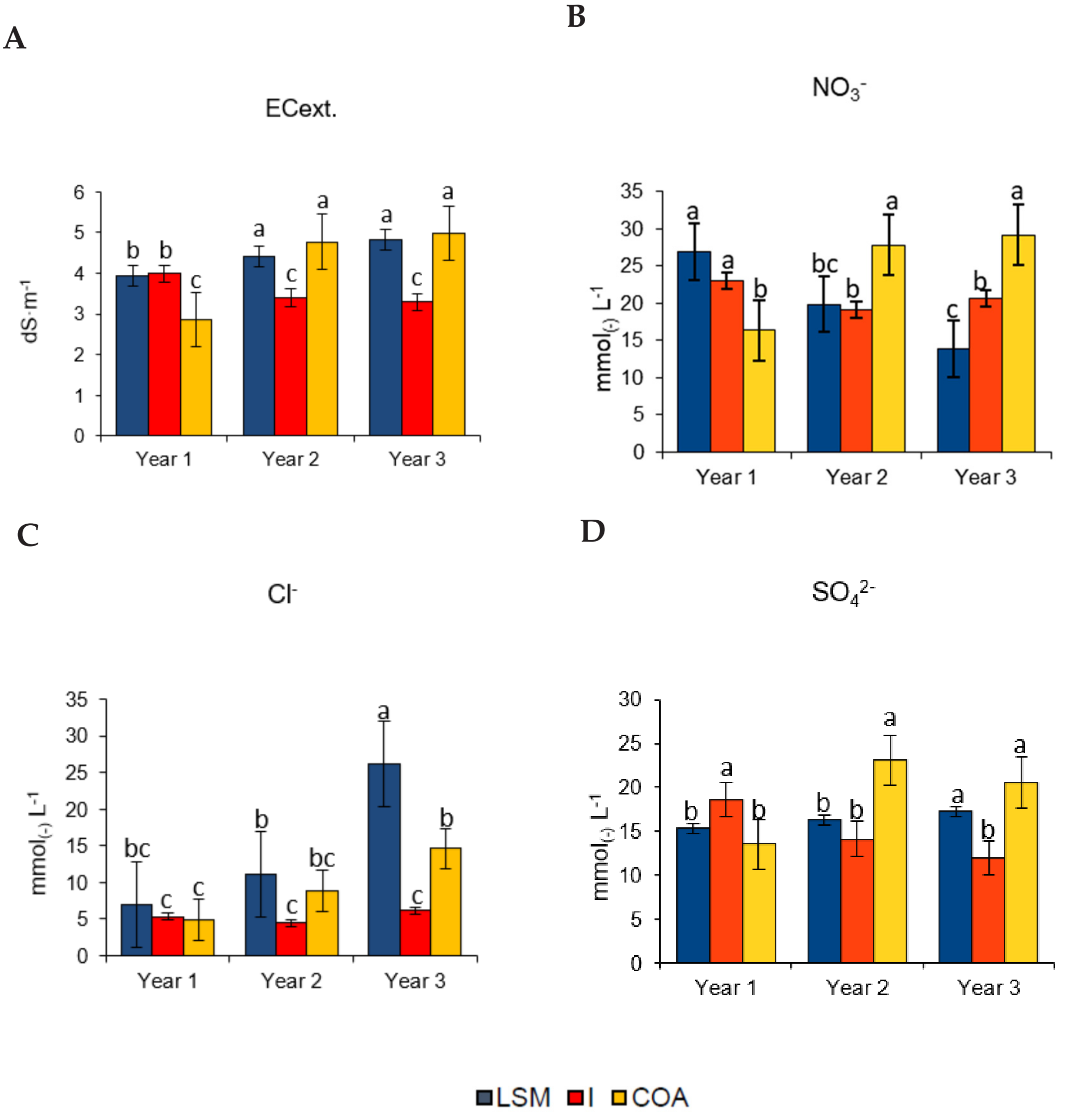

Figure 3. Evolution of the EC_{ext} (**A**), and NO_3^-, Cl^-, and SO_4^{2-} in the soil solution, (**B**–**D**) figures, respectively. $^{a–c}$ Significant differences at 95%.

As for the composition of the soil solution, NH_4^+, PO_4^{3-}, and CO_3^{2-} were not found, predominating instead as anions NO_3^-, Cl^-, and SO_4^{2-}, while in the group of cations Na^+, K^+, Ca^{2+}, and Mg^{2+} stand out. Thus, the mean NO_3^- content was higher than 20 mmol$_{(-)}$L^{-1} in the three trials, and there were no statistically significant differences among them ($X^2 = 0.517$, df = 2, p-value = 0.7722). Finally, a very different trend was observed between the time evolution of the LSM and COA (Figure 3B), since while the former tended to decrease throughout the experiment, the COA increased significantly between the first and second years, remaining stable from the second to the third year. The concentration of NO_3^- in the I remained more stable, and only a slight decrease was observed between years 1 and 2. As for Cl^-, in view of Figure 3C, it can be said that the large deviation in the results prevented establishing a defined behavior of this parameter; in spite of this, there were significant differences between the three treatments at the end of the experiment, with a degree of significance of 95% ($X^2 = 19.0197$, df = 2, p-value = 7.412×10^{-5}), highlighting those between the treatments LSM and I, with a significance of 99%. Likewise, a significant increase in the concentration throughout the cycle was observed in the three treatments. Finally, $SO_4^=$ had a behavior over time very similar to that of Cl^- (Figure 3D) such that it increased in the LSM and COA throughout the experiment, while in the control (I) it decreased from the second year onwards. The differences between the treatments were not as contrasted as in the case of Cl^- ($X^2 = 7.7486$; df = 2; p-value = 0.02077), but they were significant between the treatments I and COA (W = 599, p-value = 0.01149), where the minimum and maximum values were found, respectively.

As for the cations in the soil solution, Na^+ did not show a definite trend during the test period (Figure 4A), with an increase in the LSM and COA treatments between the second and third years and a decrease in I starting from the first year. Among the different treatments, at a significance level of 0.05, there were no statistically significant differences ($X^2 = 4.8915$, df = 2, p-value = 0.08666). K^+ showed significant differences among the three treatments ($X^2 = 17.6592$, df = 2, p-value = 0.0001463) (Figure 4B), with the sheep manure trial having the highest mean value of the three (5.6 mmol$_{(+)}$ L^{-1}). Regarding the evolution over the three years of the experiment, an increase in the LSM and COA was observed between the second and third years, while the opposite was observed in the I. The Ca^{2+} concentration in the soil solution was the maximum in the first year of the trial in the LSM and I, after which a decrease was observed, which stabilized in the third year; however, in the COA it remained stable over the 3 years (Figure 4C). On the other hand, no differences were obtained with a statistical significance of 95% between the treatments ($X^2 = 4.2368$, df = 2, p-value = 0.1202; df = 2, p-value = 0.102).

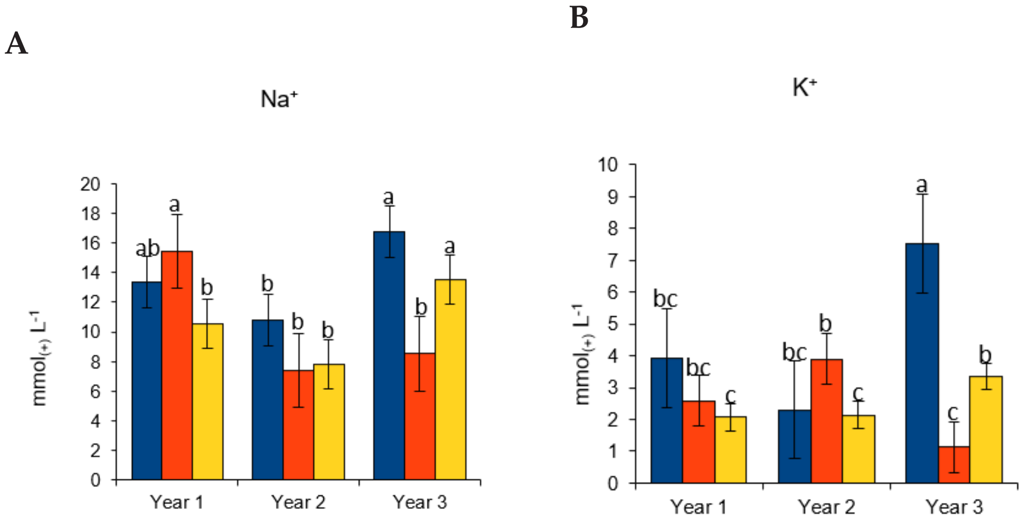

Figure 4. *Cont.*

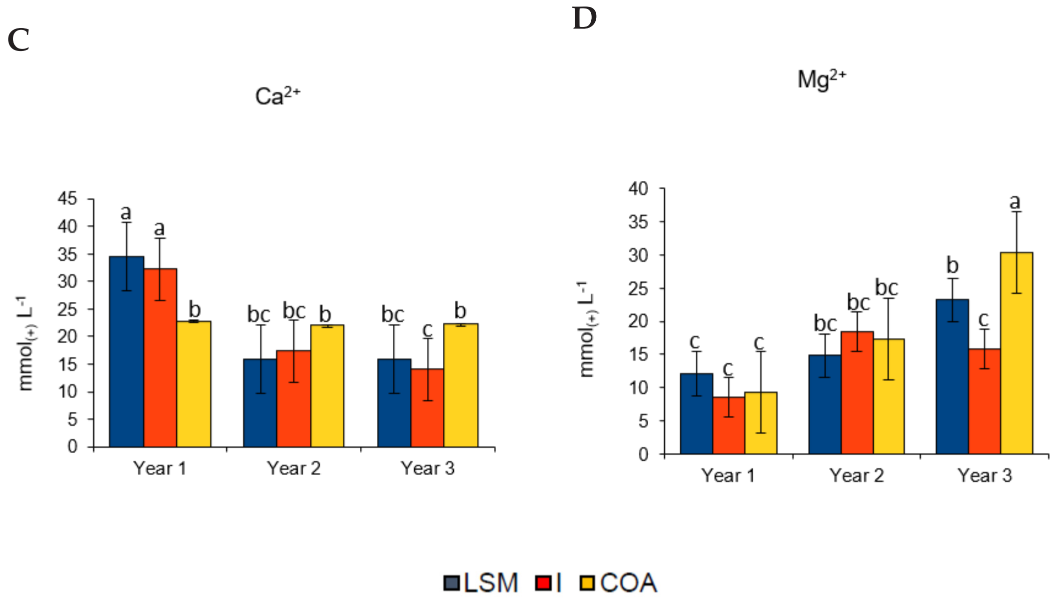

Figure 4. Evolution of the Na^+, K^+, Ca^{2+}, and Mg^{2+} in the soil solution, (**A**–**D**) figures, respectively. $^{a–c}$ Significant differences at 95%.

The Mg^{2+} concentration in the soil solution (Figure 4D) increased in the three treatments throughout the study period, especially in the LSM and COA where a statistically significant increase was observed over the three years, while in the I it only appeared between the first and second years and remained constant between this and the third year. Despite these trends, the values were very similar in the three treatments and, at a significance level of 0.05, lacked statistical significance (X^2 = 0.9594, df = 2, *p*-value = 0.619).

3.4. Assimilable Fe, Cu, Mn, and Zn

Of the assimilable elements, Mn and Fe were the most abundant, followed by Zn and Cu, as shown in Table 6. When compared among the three treatments carried out, the LSM showed the highest Fe content, while the I and COA had the lowest value. The differences were, therefore, significant between the LSM and I (W = 440.5, *p*-value = 0.001719) and the LSM and COA (W = 411, *p*-value = 0.01152). In the case of Cu, the treatment I had the lowest mean value, showing significant differences with respect to the treatments LSM (W = 392, *p*-value = 0.03278) and COA (W = 170.5, *p*-value = 0.01576). Except in the first year, Mn had a similar concentration in the three treatments such that no significant differences were observed in the accumulated values. Finally, Zn had the highest average value in the LSM, representing twice the concentration found in the trials I and COA. The statistical analysis confirms that the differences found were significant between the LSM and trial I at 99% (W = 428.5, *p*-value = 0.003885) and especially with COA (W = 459, *p*-value = 0.0004363).

Table 6. Assimilable element content and evolution (mg kg^{-1}).

	Fe			**Cu**			**Mn**			**Zn**		
	LSM	**I**	**COA**	**LSM**	**I**	**COA**	**LSM**	**I**	**COA**	**LSM**	**I**	**COA**
Year 1	9.9 a	4.2 a	3.9 b	1.5 b	1.1 c	1.4 c	13.7 a	11.4 a	11.3 a	3.2 a	1.6 a	1.4 a
Year 2	4.1 c	3.3 c	4.3 a	1.5 b	1.4 b	1.8 b	7.0 b	6.6 b	8.6 b	1.9 b	1.2 b	1.3 a
Year 3	4.8 b	3.8 b	4.2 a	2.6 a	2.0 a	2.4 a	3.1 c	4.2 v	3.3 c	0.6 c	0.6 c	0.4 b
Mean	6.3	3.7	4.1	1.9	1.4	1.8	7.9	7.8	7.7	1.8	1.1	1.0
SD	3.2	1.2	0.5	0.8	0.5	0.6	5.1	4.1	4.2	1.1	0.6	0.6
p-Value *		0.003 **			0.029 *			0.93			0.023 *	

SD, standard deviation. * $p < 0.05$; ** $p < 0.01$. "a", "b", and "c" indicate significant differences between years at the 95% confidence level.

Considering the evolution over the three years of the study, in all three treatments there was, in general, a considerable decrease in the concentrations of Mn and Zn, while Fe showed an uneven trend in the three treatments and the Cu concentration increased to a statistically significant extent.

3.5. Organic Carbon Capture

As can be seen in Table 7, the LSM treatment based on composted sheep manure, showed an increase in organic carbon capture (OCc) throughout the three years of the experiment, while in the COA an increase was observed only in the first year and a decrease in the rest, as in the case of I. Because of this, it can be said that the addition of organic amendments to the soil led to a significant increase in its capacity as a carbon sink, especially when comparing the LSM and I treatments where the greatest difference was obtained at the end of the trial, with 37.08% more OC per hectare than in I, as well as in the COA-I comparison where the efficiency ranged between 10 and 16%.

Table 7. OC captured by the soil in the different treatments (Mg C ha^{-1}) and comparative efficiency (E) between treatments (%).

	OCc			**E**		
	LSM	**I**	**COA**	**LSM, I**	**LSM, COA**	**COA, I**
Year 1	25.88 a	22.25 a	25.50 a	14.01 c	1.45 c	12.75 b
Year 2	27.88 b	22.25 a	24.75 b	20.18 b	11.21 b	10.10 b
Year 3	29.63 c	21.00 b	25.25 a	29.11 a	14.77 a	16.83 a
SD	1.88	0.72	0.38	7.59	6.90	3.39

SD, standard deviation. “a”, “b”, and “c” indicate significant differences between years at the 95% confidence level.

4. Discussion

4.1. pH, CaCO$_3$, Organic Carbon, Total Nitrogen, C:N Ratio, and Olsen P

The treatments tested caused changes in some soil properties, while others, such as pH and total CaCO$_3$ content, were not influenced by the treatments. As for pH, it was possible that it was not affected due to the buffering capacity of the soil because of the presence of amphoteric compounds, among which CaCO$_3$ stands out. This result coincides with that found in [38], although it differs from those of other researchers, who found a lower [39] or even higher pH [40,41] in plots that received organic amendments. The CaCO$_3$ content of this soil depends mainly on the limestone–dolomitic nature of the geological material, which is the main source that contributes this constituent to the soil and justifies the low relevance of irrigation water and organic amendments in the final CaCO$_3$ content.

The increases in the OC, TN, and Olsen P in the LSM and COA with respect to I were due to the contribution of organic amendments to the soil, either as manure (LSM) or as a commercial product (COA)—results that were also obtained by different authors [42–44]. Thus, in terms of the soil’s OC dynamics, the addition of organic amendments had beneficial effects in the short term on the soil quality [45] and, more importantly, in the long term [46]. Likewise, Herencia et al. [47,48] concluded that the OC content decreases less over time in ecological plots than in conventional plots and, in some cases, increases in OC have even been recorded in plots with sustainable management [49], thus contributing to improving their functions—among others, their fertility and infiltration capacity [50]—and supporting the starting hypothesis and objectives of this article.

Regarding N, several studies have concluded that soils managed with organic farming practices have a higher TN content than those managed with conventional agriculture from the first or second year of conversion [51,52], thus justifying the higher TN content found in LSM and COA, relative to I, and, therefore, its cataloging as a rapid response indicator. This behavior must be related to the effects of the tillage on the soil OM, which mixes and aerates its upper horizons [53], stimulating microbial decomposition and, therefore, favoring the increase in the mineral ions in the soil solution, including nitrogenous forms.

However, the evolution of TN over the three years of the study did not show a definite trend, since it decreased between the first and second years and increased between the second and third years, an aspect that we attribute to the crop rotations with cereal or a cereal–legume mixture that were carried out in the winter–spring periods and that served as pasture for the existing livestock on the farm. Thus, the N extractions carried out by the cereal intercropping between the first two years of the experiment considerably reduced the content of this constituent, while between the second and third years a cereal–legume rotation was carried out with the consequent incorporation of N into the soil from the fixation of atmospheric N_2 by legumes. In line with this trend, the C/N ratio adopted an inverse behavior to TN, although it remained at higher levels than at the beginning of the experiment due to the proportionally greater increase in the OC than in TN. Similar evolutions have been shown in similar soils and climatic conditions by Nardi et al. [54] and Sánchez-Navarro et al. [41], who indicated that the addition of manure to the soil conditions the evolution of OM, favoring the formation of high-quality humus.

The Olsen P reached the highest level in the LSM, a fact that can be attributed to the positive action of humic compounds on the availability of this macronutrient, as observed, among others, by Wang et al. [55] or Bermudez et al. [56], who reported an increase in the content of assimilable P after the application of humic substances to the soil. This is due to the contribution of organic phosphorus, which when degraded by soil microorganisms releases soluble phosphate compounds [57] and reduces the precipitation of P in the soil as Ca_3PO_4 [58] and also justifies the increase during the study period, in harmony with soil OM, as also observed by Sastre et al. [59] and Sánchez-Navarro et al. [49], among others.

4.2. Cation Exchange Capacity and Exchangeable Bases

As already found for other soil components, the increase in the CEC in the LSM and COA was due to the addition of organic amendments, since, as is widely accepted, this property depends on soil colloids (i.e., OM and clay) and, in the present case, the clay content did not change as a consequence of the treatments. These results are in agreement with those obtained in other studies [60–64], which describe an improvement in the CEC due to the addition of OM compared to soils treated with conventional fertilization. However, the fact that the highest CEC was obtained in the COA and not in the LSM, with the OC reaching the highest content in the latter treatment, must be related to the low degree of humification of the sheep manure, ascertained by its high C/N ratio.

As for the behavior of the exchange of Na, K, and Mg, the lack of a defined pattern was observed, attributable to the different adsorption/desorption balances to which the exchange bases were subjected in equilibrium with the soil solution. Thus, the higher K content in the LSM must be related to the high concentration of this element in the sheep manure, as can be seen in Table 2, the functional groups of which must be the maximum responsible for the CEC, as highlighted by McLean and Bledsoe [65]. However, the exchange of Na was not sensitive to the treatments, although differences in the concentration of the soluble Na^+ in the soil saturation extract were observed, as shown in Figure 3; this points to an increase in the concentration of this element in the LSM and COA at the end of the experiment and, therefore, is evidence of salinization and, subsequently, evidence of an incipient salinization of the soil. This was observed by other authors such as Hao and Chang [66], who attribute it to the more or less accelerated decomposition or mineralization of the organic remains, catalyzed by the cultivation work, with the consequent release of soluble ions.

4.3. Electrical Conductivity of the Saturated Soil Extract and Ion Content in the Solution

The soil salinity and the ionic composition of the solution were affected by the treatments tested such that the decomposition of the organic amendments caused an increase in the EC_{ext} [67], especially in the COA whose classification, according to Allison and Richards [68], went from nonsaline (<2 dS m^{-1}, Table 1) to moderately saline (4–8 dS m^{-1}). These changes in the soil EC_{ext} were related to the incorporation of soluble salts, either in the organic or inorganic form, which, according to the results, were more intense in the former and, therefore, ultimately responsible for this increase in the EC_{ext}, as well as soluble ions [69], as also described by Bulluck et al. [60], Martínez et al. [61], Larchevêque et al. [70], and Morari et al. [64]. This parameter can be considered a rapid response indicator for the studied soils [41]. Regarding ions, the NO_3^- dynamics suggest that the mineralization of the LSM was slower than that of the COA such that a decrease in the concentration was observed in the LSM over the three years of the study, while it increased in the COA. This behavior of the LSM versus COA should be taken into account to avoid an excess of NO_3^- in the soil solution and, therefore, not to favor the contamination that was observed in nearby areas in groundwater or surface aquatic ecosystems [71]. Cl^- and SO_4^{2-} were also sensitive to the different treatments such that their presence in the soil solution was regulated by the nature and composition of the organic amendments, as a source of inputs, and with the absorption or washing of these anions; their overall balance justified the increase in the concentration throughout the study period in the organic treatments, which was more pronounced in the LSM for Cl^- and in SO_4^{2-} for the COA. As for cations, the increase in the concentration of Na^+, K^+, and Mg^{2+} in the LSM and COA at the end of the experiment, together with the decrease in the I, confirms the strong relationship of these with the amendments and with their gradual release in the OM mineralization process, which are the cause of the incipient salinization; this is a process that must be taken into account in management programs to avoid soil degradation, a situation that was also described in a similar study over 20 years ago as carried out by Sánchez-Navarro et al. [49], where it was necessary to include a crop rotation in the management plan, as well as to modify the fertilization doses from the fifth annual period onwards, to stop soil salinization.

4.4. Assimilable Fe, Cu, Mn, and Zn

It is widely accepted that nutrient bioavailability is highly influenced by the content and type of soil OM due its capacity to form organometallic complexes [70,72–75], results that coincide with those presented in this work and which would justify the higher concentration of assimilable Fe, Cu, Mn, and Zn in the LSM and COA rather than in I. On the other hand, the levels of Fe, Mn, and Zn were abnormally high in the LSM treatment of the first year and must have their origin in the addition of residues from the iron and steel industry, rich in these elements, to the manure; this is a common practice to improve the manure used as an organic amendment for vegetables. Therefore, in line with the starting hypothesis, it can be affirmed that the chemical fertility of the soil improved significantly with the addition of organic amendments. This is very important for the good development of crops, since in these soils with basic pH values and high limestone contents most of the divalent cations are present in precipitated forms [38] and, therefore, very often crops are deficient in these elements.

4.5. Organic Carbon Capture

Special mention should be made of organic fertilization as a method for enhancing the soil's role as a carbon sink, since such OM inputs offset CO_2 emissions, especially in croplands in arid and semiarid regions. This enables compliance with the Kyoto Protocol [76,77] and the objectives set at the Paris Climate Conference (COP21), which establish the need to increase the OC content of agricultural soils by 4 per 1000, thus favoring the transition to resilient agriculture. On the other hand, they come in response to the guidelines provided by the 2006 Intergovernmental Panel on Climate Change (IPCC), which establishes the need for countries to carry out an inventory with changes in SOC stocks as a consequence of animal manure inputs for national greenhouse gas inventories. Similar results were reported in [78,79] and in studies that, in some cases, lasted more than 20 years [49] and validated the compatibility of the management system used throughout the study with the trends of sustainable agriculture [80,81], which the European Union is promoting in terms of soil protection. On the other hand, the decrease in OC in treatment I must be related to the accelerated mineralization of OM due to tillage [82] and that obviously must also have occurred in the LSM and COA treatments, although it was not detected, due to the OM provided in the amendments.

5. Conclusions

This study has shown that the use of organic amendments as fertilizers instead of inorganic fertilizers produced favorable changes in some soil chemical properties. Thus, during the study period, there was a clear increase in the OC content in the organically managed soils, while in the control plot (I) it remained fairly stable, with a slight tendency to decrease. Because of this, the addition of organic amendments to the soil, together with rotations with cereals and legumes in the cold seasons and the use of extensive grazing, is a cultivation technique that enhances the soil's function as a carbon sink. In addition, fertility indicators also improved, as is the case for the macronutrients N, P, K, and Mg and micronutrients; this is especially true for Fe and Cu, which, given their sensitivity to agronomic management, can be considered as rapid response indicators. This contributed to the improvement in the soil's ability to produce food, but with an additional added value—the preservation of the environment. Positive changes were also observed in the CEC and, therefore, in the soil's capacity to adsorb ions and act as a substrate capable of fixing and partially immobilizing pollutants, such as heavy metals and other positively charged metabolites.

On the other hand, the ECext was also affected by the agricultural activities carried out over the three years of the experiment, especially as a consequence of the increase in the concentration of Na^+ and Cl^-; they reached higher values in the organic treatments LSM and COA, than in I, an aspect that can trigger a salinization process and that makes it necessary to review the doses of fertilizers applied, as well as the crop rotations that can act as phytoremediators.

In summary, it can be stated that the agronomic management of farms based on organic amendments, crop rotation, and extensive grazing appears to be a model of sustainable agriculture that preserves the quality of the soil as a source of nutrients and promotes its ecological function as a C sink.

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