DECIPHERING THE EVOLUTION OF AGRARIAN TECHNOLOGIES DURING THE LAST ~1600 YEARS USING THE ISOTOPIC FINGERPRINT (δ^{13}C, δ^{15}N) OF A POLYCYCLIC TERRACED SOIL

CRUZ FERRO VÁZQUEZ [1], SERAFÍN GONZÁLEZ PRIETO [2], ANTONIO MARTÍNEZ CORTIZAS [3] &
FELIPE CRIADO BOADO [4]

Abstract:
We analyzed the isotopic (^{13}C and ^{15}N) composition of a polycyclic terraced soil located in Santiago de Compostela (NW Spain) and compared it with previous results on total aluminum, iron and silicon and their fractionation by selective dissolution techniques. The aim was to recognize the imprints of land management changes, with particular attention to fertilization techniques applied during the use history of the terrace (~1600 y). The buried paleosol, found below the terraced layers, is considered to preserve the soil properties prior to the terrace construction. The isotopic composition (^{13}C, ^{15}N) provided evidence of extensive land use previous to the construction of the terrace, with the utilization of fire as liming and clearance tool. In the Late Antiquity and Early Medieval Ages the soil use was more intense and amendments with vegetal remains from nitrogen fixing shrubs were likely applied. Since the Early Middle Ages, animal wastes were used as a way to maintain or increase soil fertility because of an intensification of the agrarian use.

Keywords: soil management, fertilization, isotopic composition, terraces, Middle Ages

Resumen:
Descifrando la evolución de las tecnologías agrarias durante los últimos ~1.600 años utilizando la huella isotópica (δ13C, δ15N) en un suelo aterrazado policíclico
Hemos analizado la composición isotópica (^{13}C y ^{15}N) de un suelo policíclico aterrazado situado en Santiago de Compostela (NO de España) y la hemos comparado con resultados geoquímicos previamente obtenidos de aluminio, hierro y silicio totales, así como las fracciones en las que se distribuyen estos elementos, mediante técnicas de disolución selectiva. El objetivo era reconocer las señales de los cambios de manejo del suelo agrícola, con especial atención a la aplicación de técnicas de fertilización, durante la historia de uso de la terraza (~1600 años). Se asume que el paleosuelo enterrado, que se conserva bajo los niveles de aterrazamiento, preserva las propiedades del suelo previas a la construcción de la terraza. La composición isotópica (^{13}C y ^{15}N) proporcionó evidencias de un uso extensivo del suelo con anterioridad a la construcción de la terraza, con la utilización del fuego como principal herramienta para el encalado y el clareo del terreno. Durante la Antigüedad tardía y la Alta Edad Media el uso del suelo se intensificó y se introdujeron técnicas de fertilización basadas en la adición de restos de vegetales de arbustos fijadores de nitrógeno. A partir de la Alta Edad Media se detecta el uso de abonos de origen animal, como medio para mantener o aumentar la fertilidad del suelo, ante una creciente intensificación del uso.

Palabras clave: manejo del suelo, fertilización, composición isotópica, terrazas, Edad Media

1. INTRODUCTION

The agricultural landscape of NW Spain is mainly inherited from ancient agricultural practices. Its origin can be traced back, at least, to the VI century AD (BOUHIER 1979; CRIADO BOADO 1989; GUITIÁN RIVERA 2001; BALLESTEROS-ARIAS *et al.* 2006a), although some of its features might have originated in the Iron Age (PARCERO-OUBIÑA 2006) or even the Late Bronze Age (MARTÍNEZ CORTIZAS *et al.* 2009). In this traditional agricultural system, the land is divided between that used for intensive agriculture and plow (*agro*) and land used for growing slash and complementary activities (*monte*), configuring what some authors called "concave landscape" (BALLESTEROS-ARIAS *et al.* 2011).

Bouhier (1979) was one of the few authors dealing with the agrarian geography of NW Spain, in its broadest sense, describing the traditional Galician system and deepening in the main causes of its current configuration. Other noteworthy research on this issue was developed by Ballesteros-Arias *et al.* (2006a, 2006b, 2010) from an archaeological point of view, drawing conclusions about the land division systems, the constructed landscape and the spatial distribution of the agrarian activity that greatly improved the comprehension of the current landscape and the history of its evolution. But the lack of direct archaeological evidence frequently limited the achievement of conclusions on the management techniques and the technologies that

[1] Instituto de Ciencias del Patrimonio. Consejo Superior de Investigaciones Científicas . Espanha. Email: cruz.ferro@incipit.csic.es
[2] Instituto de Investigaciones Agrobiológicas de Galicia. Consejo Superior de Investigaciones Científicas. Espanha. Email: serafin@iiag.csic.es
[3] Departamento de Edafología y Química Agrícola. Universidad de Santiago de Compostela. Esapnha. Email: antonio.martinez.cortizas@usc.es
[4] Instituto de Ciencias del Patrimonio. Consejo Superior de Investigaciones Científicas . Espanha. Email: felipe.criado-boado@incipit.csic.es

allowed this evolution. Historical sources on this topic are also almost absent. In this context, the study of environmental archives is useful to get indirect knowledge about the evolution of the agricultural management. However, the studies addressing the agricultural evolution from a palaeoenvironmental point of view are still insufficient for this area and, to the extent of our knowledge, only a few are based on the study of the soil properties (LÓPEZ-SÁEZ *et al.* 2003; MARTÍNEZ CORTIZAS *et al.* 2009; KAAL *et al.* 2011, 2013).

Soils are an essential resource for the development of human communities and are affected by land use. The imprints of environmental and land use changes, when preserved, can be extracted and interpreted to get information about the kind and intensity of the activity the soil supported. In particular, colluvial soils are capable to store information of the environmental changes occurred during the deposition of the parent material and the soil formation after their stabilization (MARTÍNEZ CORTIZAS, 2002; KALIS *et al.* 2003; LEOPOLD & VÖLKEL 2007). Previous research has successfully used colluvial soils for reconstructing deforestation, pollution and fire use history and past climatic or vegetation changes (KAAL *et al.* 2008, 2011, 2013; MARTÍNEZ CORTIZAS *et al.* 2009; LÓPEZ-MERINO *et al.* 2012).

We applied this capability of colluvial soils for the reconstruction of land-use evolution by the analysis of a polycyclic soil sequence in an agricultural terraced system in Monte Gaiás (Santiago de Compostela, NW Spain). Because they are human-made, terraced soils are not standard colluvial soils. Nonetheless, their formation process is equivalent, involving successive depositions of soil material that result in a sequence of A horizons over the original *in situ* edaphic cycle (i.e paleosol). This allows us to extrapolate the demonstrated suitability of colluvial soils as paleoenvironmental archives to Monte Gaiás terrace soils.

In a previous investigation (FERRO-VÁZQUEZ *et al.* 2014) we studied the pedogenetical modifications in Monte Gaiás in relation to the land-use changes along time, using data on soil elemental composition and fractionation of aluminum, iron and silicon. We concluded that the changes in soil properties resulted first from the construction of the terrace, which changed the edaphic environment, and secondly from the intensification of the agricultural management. Also, signals of fertility amendments were found, although it was not possible to identify what forms of fertilization were used. Fertilization is a crucial issue for landscape evolution because it allows the intensification of farming and enables the continuous exploitation of the soil maintaining its productivity, thus providing the means to supply a largest population. Therefore, it involves a change in the cultural landscape in the broadest sense, with the adoption of new forms of land use and new settlement patterns and the emergence of new sociological, economic and political conditions. From this point of view, the knowledge on the use of fertilization techniques and their evolution provides tools for understanding the processes of cultural change.

Studying isotopic (^{13}C and ^{15}N) patterns in the soil might help to detect the existence and kind of fertilization techniques since they may reveal the processes that control the carbon and nitrogen cycles, which are related with land use. Stable isotopes measurements at natural abundance levels have demonstrated to be a powerful research tool in environmental sciences (HANDLEY & SCRIMGEOUR 1997; YAKIR & STERNBERG 2000; ROBINSON 2001). In the case of soils, δ^{13}C has been usefully employed to monitor long-term intensive land use effects on soil organic matter (SOM) (KALBITZ *et al.* 2000). Soil δ^{13}C is known to be affected by the isotopic signature of inputs, fractionation during organic matter microbial decomposition and soil characteristics (mineralogy, clay content and pH) (EHLERINGER *et al.* 2000; KRULL & SKJEMSTAD 2003; DIJKSTRA *et al.* 2006). In this sense, Balesdent *et al.* (1987) found that land-use changes that alter the isotopic signature of the litter inputs resulted in an effective labeling of soil organic matter. Plants δ^{13}C vary from -12 ‰ for C4 plants to -25 ‰ in plants having a C3 metabolism (BALESDENT *et al.* 1987; LEFROY *et al.* 1993), while animal manures δ^{13}C range from -30 ‰ in those derived from cattle fed with C3 plants to -20 ‰ in manures derived from cattle fed with C4 plants (GLASER *et al.* 2001; BOL *et al.* 2003; ANGERS *et al.* 2007). On the other hand, fertilization also affects the decomposition rate and the predominant pathways, indirectly modifying the size and composition of microbial communities and altering the δ^{13}C of the soil (HEIL *et al.* 2000; KRULL & SKJEMSTAD 2003; DIJKSTRA *et al.* 2006).

Soil δ^{15}N values reflect the net effect of biotic and abiotic environment on nitrogen-cycling processes (DAWSON *et al.* 2002), being influenced by the quantity and quality of SOM inputs, nitrogen sources and isotopic fractionation during nitrogen transformations (NADELHOFFER & FRY 1988). Different nitrogen sources would have different δ^{15}N fingerprints, with animal manures and slurries being generally more enriched in ^{15}N than soils (GLASER *et al.* 2001; SENBAYRAM *et al.* 2008) and these more enriched than plant remains (FRY 1991; NADELHOFFER & FRY 1994; HÖGBERG *et al.* 1995, HÖGBERG 1997, GÓMEZ-REY *et al.* 2013a) or ashes. Thus, the type of fertilizer applied is expected to differently alter the δ^{15}N values of soils (GLASER *et al.* 2001, CHOI *et al.* 2003a, WAZKA *et al.* 2006), especially as a result of long-term land use patterns (GERZABEK *et al.* 1999; ANTIL *et al.* 2005). Also, the addition of nitrogen-containing fertilizers influences the main nitrogen dynamics, which is reflected in soil δ^{15}N (HÖGBERG & JOHANNISSON 1993; HÖGBERG 1997; ROBINSON, 2001). Koerner *et al.* (1999) reported

that the mean $\delta^{15}N$ of soil increased with the intensity of former land use and that the $\delta^{15}N$ of understory plant and soil appear to be excellent tracers of previous land use in forests, and could be used in historical studies. González-Prieto *et al.* (2013) also found that $\delta^{15}N$ is a potential tracer of changing land use because it allows discriminating among soils with similar carbon and nitrogen stocks but under differing land use. A dual isotope approach with carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) is particularly useful in studying SOM sources (PETERSON & FRY 1987).

This investigation attempts to provide information on the evolution of the fertilization practices in Monte Gaiás terraced soils by studying the changes in the $\delta^{13}C$ and $\delta^{15}N$ isotopic fingerprint, together with the chemical properties and the previous knowledge about paleoclimatic conditions and archaeological information.

2. MATERIALS AND METHODOLOGY

The soil studied here (Fig. 1) was sampled in 2001 in the context of the archaeological excavation for the perceptive archaeological impact assessment and correction, previous to the construction of the Galician City of Culture, in Santiago de Compostela (Galicia, NW Spain).

Fig. 1. Location, topographical context and scheme of CC1 soil. The morphologic features of the soil monolith reflect the polycyclic pedogenesis.
Fig. 1. Localización, contexto topográfico y esquema del suelo CC1. La morfología del suelo refleja la edafogénesis policíclica.

The archaeological work reported an expansive process in this area in the Early Middle Ages (BALLESTEROS-ARIAS *et al.* 2006a), which marked the beginning of a deep transformation of the agricultural landscape, involving new features and techniques that potentially favoured the intensification of the agricultural activity. One of the stronger human interventions was the correction of the slope by building artificial horizontal surfaces, i.e. terraces, providing a new topography, which facilitates agricultural exploitation and improves productivity. For this purpose, the original middle slope was excavated and the extracted material was used to create, down in slope, the flat surface of the terrace by episodic contributions, in a construction system called "clearance-embankment with episodic filling" (BALLESTEROS-ARIAS *et al.* 2006a). Therefore, the infilling material of the terraced layers is thought to come from the slope material.

The City of Culture-1 soil sequence (hereafter referred to as CC1) is located in the E-NE slope of "Monte Gaiás" at 250 m a.s.l. (Fig. 1). CC1 develops on an amphibolitic colluvium. Present climate can be characterized as mild and humid with an average annual temperature of 12.3 ºC, total annual rainfall 1915 mm, and relatively low potential evapotranspiration (50-100 mm in winter and >300 mm in summer), which results in a positive water balance (600-800 mm) (MARTÍNEZ CORTIZAS & PÉREZ-ALBERTI 1999). Current vegetation is shrubland at the top of Monte Gaiás and its surrounding slopes, whereas crop and pasture dominate in terraced areas. Under these environmental conditions, basic and metabasic rock materials (gabbros, amphibolites, basic granulites), which are rich in weatherable minerals, naturally generate soils with relevant amounts of highly reactive components, such as poorly crystalline mineral constituents and organo-metal complexes (MACÍAS *et al.* 1978; GARCÍA PAZ *et al.* 1986; GARCÍA-RODEJA *et al.* 1987). This pedogenetic process is known as andosolization, and leads to soils with andic properties (IUSS-WRB 2007): low bulk density, high water retention potential, low permanent charge, an exchange complex dominated by variable charge surfaces, and high anion retention capacity. A highly stable soil organic matter (SOM), due to the predominance of stable organoaluminic

complexes and the adsorption of organic compounds on low-ordered minerals (BOUDOT *et al.* 1989; ZECH *et al.* 1997), also contributes to the andic character. This type of SOM is usually abundant in the surface horizons of andic soils (MARTIN & HAIDER 1986; ARAN *et al.* 2001).

CC1 is a thick polycyclic soil (300 cm deep), in which four different stratigraphic/soil formation cycles were distinguished: 1Aam (0-60 cm), 2Aam (60-150 cm), 3Aam (150-232 cm), 4Amo (232-252), 4Bw (252-285 cm), 4C (>285 cm). Continuous samples of 10 cm in thickness were taken from the surface to a depth of 223 cm, and of 5 cm in thickness from 223 cm to the bottom of the soil.

No relevant soil losses by erosion were detected, except in the transition from the 4A to 3A horizon, where there is a chronological evidence for truncation: the radiocarbon age of the upper sample of the paleosol differs in almost a millennium from the age of the bottom of the 3A layer. As a result of the truncation, 4A horizon is 20 cm thick, which impedes its classification as a melanic horizon, despite fulfilling the other IUSS-WRB (2007) criteria. Nonetheless, as andosolization takes place only in surficial horizons, and considering that 4A horizon has andic properties, it is deduced that the truncation has not removed all the buried epipedon. It is a dark, organic rich soil horizon with loamy texture and moderately well-developed aggregate structure. Horizons 1A, 2A and 3A are terraced layers and are classified as anthric horizons (IUSS-WRB 2007). They have a lower content of

organic matter than 4A. The 2A and 3A horizons have slightly higher sand content but loamy textures. The finer soil texture is found in the 1A horizon, which has the higher clay content. The terraced layers have moderate, subangular blocky structure.

Since burial seals soils from subsequent diagenesis (SSS-USDA 2010), we assume that horizons 4A, 3A and 2A preserved the properties that they had in the moment of burial, including features associated to cultivation and manuring, and that were not significantly affected by bioturbation (DAVIDSON, 2002). In this sense, in a previous work (FERRO-VÁZQUEZ 2004), the chemical features of CC1 were compared with those of a colluvial soil located a few kilometres distant from Monte Gaiás, developed on the same parent material and with similar geomorphic position, which was not farmed until the last decades. The chemical properties of the undisturbed part of this soil were coherent with the properties of 4A horizon of CC1. So it is thought to exemplify the pre-terracing slope soil characteristics. This enabled its use as reference material for the comparison with the terraced soil layers that have an anthropogenic origin and have been exposed to agricultural management.

Table 1 summarizes the chemical properties of the soil. The soil was acidic, with pH_{KCl} values (3.5-5.5) indicating high contents of inorganic reactive compounds in the 3A soil horizon and in the paleosol. This is supported also by the pH_{NaF} values (9-11) and by the percentage of phosphate

Table 1. Main physico-chemical properties of CC1 polycyclic soil (mean±standard deviation). These data are described in detail and discussed in Ferro-Vázquez *et al.* (2014).

Table 1. Propiedades físico-químicas generales del suelo CC1 (media ± desviación estándar). Los datos de esta tabla se describen y discuten en detalle en Ferro-Vázquez *et al.* (2014).

Hz	n[a]	pH_w[b]	pH_K[c]	pH_F[d]	C[e]	Al_T[f]	Fe_T[g]	Si_T[h]	P[i]	P_{ret}[j]	Sand[k]	Silt[l]	Clay[m]
1A	6	5.0±0.16	4.3±0.0	9.8±0.3	27±5.3	101±12.3	177±11.8	135±63	0.9±0.1	81.1±0.5	37.6±2.2	42.2±1.2	20.2±1.7
2A	7	4.6±0.08	4.4±0.1	10.5±0.2	28±2.8	106±08.0	155±12.4	142±42	1.2±0.3	85.4±3.6	45.6±7.6	37.2±7.9	17.3±4.7
3A	10	4.6±0.06	4.5±0.1	11.0±0.1	42±8.7	89±10.5	141±6.7	128±91	1.4±0.3	92.9±1.7	46.0±2.3	38.0±4.3	16.1±2.6
4A	4	4.8±0.05	4.6±0.1	11.0±0.4	48±29.6	91±17.7	169±18.3	112±88	1.0±0.2	96.7±1.6	42.3±4.2	40.3±5.6	17.4±7.6
4B	5	5.0±0.07	4.2±0.0	10.1±0.1	9±2.4	93±2.8	191±9.5	123±92	n.d.	91.4±2.9	38.9±3.2	38.0±10.2	23.1±7.4
4C	4	5.0±0.03	4.0±0.0	9.5±0.2	4±0.5	84±9.2	212±9.6	120±88	n.d.	81.9±1.0	25.4±3.8	53.3±3.2	21.3±5.0

[a] Number of samples in each soil horizon
[b] pH in water (GUITIÁN & CARBALLAS, 1976)
[c] pH_K in saturated KCl solution (GUITIÁN & CARBALLAS, 1976)
[d] pH_w in NaF 1M solution (FIELDES & PERROT, 1996)
[e] Total C content expressed in g kg^{-1}
[f] Total Al content expressed in g kg^{-1}
[g] Total Fe content expressed in g kg^{-1}
[h] Total Si content expressed in g kg^{-1}
[i] Total P content expressed in g kg^{-1}
[j] Amount of P retained (Blakemore et al, 1981) expresed in %
[k] Proportion of the fine earth size >0.050 mm
[l] Proportion of the fine earth 0.050 mm>size>0.002 mm.
[m] Proportion of the fine earth 0.002 mm <size
n.d..Below the detection limit of the analytical technique

retention, which increases with depth from 80% in the samples of the 1A horizon to 99% in 4A horizon. Soil carbon content was considered entirely as organic C given the lack of carbonates. It was higher in the paleosol than in the terraced horizons, except for the present soil surface, probably as a result of biogenic enrichment and recent organic amendments. The highest P contents are found in 2A and 3A. Phosphorous depth variation is independent of the phosphate retention capacity, pointing to the addition of some P containing amendment. Total Si content ranged from 100 to 150 g kg^{-1}, total Al from 72 to 150 g kg^{-1} and total Fe from 130 to 200 g kg^{-1}. The Al and Fe fractionation showed a different pattern after the terrace construction, evidencing more weathered edaphic materials in the terraced layers despite their younger age. This process was attributed to the accelerated mineralization due to the intensification of agriculture (FERRO-VÁZQUEZ *et al.* 2014), leading to the loss of the andic character of the original soil, and to a shift towards a progressively more degraded soil stage in the more recent terraced soil horizons.

2.1. Isotopic measurements

The ^{13}C and ^{15}N isotopic signatures were determined on ground samples (<100 μm) by isotopic ratio mass spectrometry, using an automated CN analyzer coupled on-line to a Finnigan MAT Delta-C isotope-ratio mass spectrometer. All determinations were performed in duplicate and an isotopic and an elemental reference material were included in each

set of 10 samples to check the accuracy of the results. Drift correction was made against internal standards during the run. Results are given in $\delta^{13}C$ and $\delta^{15}N$, relative to the PDB fossil and air N_2 isotopic composition, respectively.

2.2. Dating

Five samples, from the 1A, 2A, 3A and 4A horizons (45, 105, 165, 223 and 236 cm depth), were dated by their ^{14}C content. Before radiocarbon analysis, sand, roots and other undecomposed organic remains were removed from the samples by sieving through a 50 μm mesh. Radiocarbon age determinations were obtained in the alkali-soluble fraction that is thought to provide the most accurate date for this kind of soils (TONNEIJCK *et al.* 2006). Accelerator mass spectrometry (AMS) was performed at the radiocarbon facility of the Ångstrom Laboratory (Uppsala-Sweden) and the CNA (Sevilla-Spain) and the radiometric datings were carried out in the Instituto Rocasolano-CSIC (Madrid-Spain). Conventional ages were calibrated using Calib 7.0 and the IntCal13 curve (STUIVER & REIMER 1993; REIMER *et al.* 2013). The radiocarbon dating results were previously published by Ballesteros-Arias *et al.* (2006) and Ferro-Vázquez *et al.* (2014), and are given in Table 2 as 2σ cal years BP. The ages obtained showed consistency with radiocarbon measurements obtained for other terraces of the same system (BALLESTEROS-ARIAS *et al.* 2011), and also with stratigraphy and field observations.

Table 2. Results of radiocarbon measurements (AMS of SOM after an acid-alkaline-acid treatment).
Tabla 2. Resultados de las dataciones por radiocarbono (AMS de la materia orgánica tras la aplicación de un tratamiento ácido-alcalino-ácido).

Sample	Depth (cm)	Horizon	^{14}C age y BP	Calibrated age BP (2σ range)[a]	Calibrated age BC/AD (2σ range)[a]	Laboratory Code
CC1-04	45	1A	815±45	670-797 cal BP	1153-1280 cal AD	CNA-1575
CC1-10	105	2A	1130±45	958-1155 cal BP	775-992 cal AD	Ua-21690[b]
CC1-16	165	3A	1455±45	1288-1415 cal BP	535-662 cal AD	Ua-20001[b]
CC1-22	223	3A	1485±45	1299-1421 cal BP	529-651 cal AD	Ua-20002[b]
CC1-24	236	4A	2334±31	2310-2440 cal BP	491-361 cal BC	CSIC-1947[b]

[a] Calib 7.0 (STUIVER & REIMER, 1993) with the IntCal13 curve (REIMER *et al.* 2013)
[b] From Ferro-Vázquez *et al.* (2014)
[c] From Ballesteros-Arias *et al.* (2006a).

2.3. Data Handling

Statistical analyses were carried out using SPSS19 software, after data transformation to Z scores ([$Z_i=(X_i-X)/SD$], where X_i is the value of the variable for the i sample of CC1 soil, and X and SD are the mean value and the standard deviation) to avoid scaling effect and provide average centering (ERIKSSON *et al.* 1999).

Co-variation among the different variables was tested using Pearson correlation analyses. In the text determination coefficients (r^2) are provided

and are statistically significant (p< 0.05) if not otherwise stated. Scatter plots were examined for evidence of non-linear association, heteroscedasticity and presence of outliers or non-homogeneous groups.

Multiple stepwise regression analysis was used to determine the soil properties better explaining the variability of the $\delta^{15}N$. Candidate predictor variables comprised total C and N, C/N ratio, pH_{H2O}, pH_{KCl}, pH_{NaF}, effective cation exchange capacity (eCEC), sum of base cations (SB) and Al saturation (S_{Al}) of the exchange complex, total Al,

Fe, Si and P concentration (Al$_T$, Fe$_T$, Si$_T$, P), Al and Fe extracted with Na-pyrophosphate (Al$_p$ and Fe$_p$) and NH$_4^+$ oxalate/oxalic acid, Al and Si extracted with NaOH (Al$_n$, Si$_n$), Fe extracted with Na-dithionite-citrate (Fe$_d$), and δ^{13}C. An explanation and a detailed description on the depth variation of the variables related to the Al, Fe and Si fractionation are given in Ferro-Vázquez *et al.* (2014). The calculations were made considering the samples from 1A, 2A, 3A and 4A horizons (the present and buried epipedons) and excluding the samples from 4B and 4C because they are subsurface horizons, which had not been affected by andosolization. Multiple regression models were built with a significance level of p=0.05 for variable entry. Within all-possible regressions the model maximizing the adjusted R^2, minimizing the mean square error and including only variables with a tolerance higher than 0.6 was selected. Cook's distance tests and leverage analyses were run for the detection of outliers.

Calibrated radiocarbon dates were used to produce an age-depth model, using Clam (BLAAUW 2010). The best fit was obtained with linear interpolation assigning to the top sample the year of sampling (Fig. 2). According to this model, the 232 cm corresponding to the terraced soil layers represents the last ca. 1600 years. The sequence has a particularly good resolution for the Late Antiquity and Medieval periods.
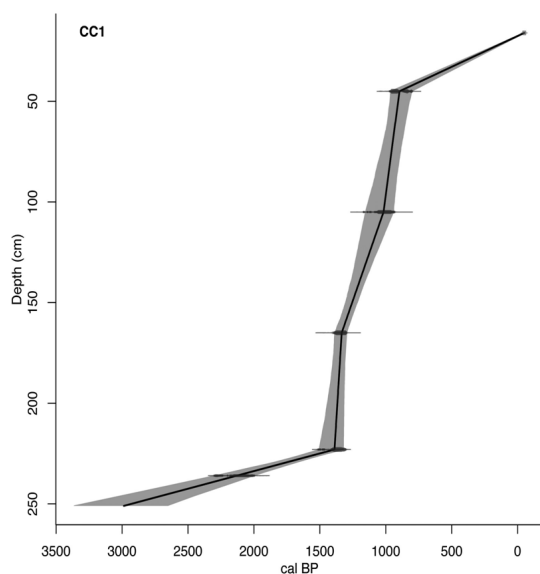
Fig. 2. Linearly interpolated age–depth model of CC1 soil, using Clam.R (BLAAUW, 2010). Blocks in the radiocarbon ages represent the 95% confidence level in radiocarbon dates calibration, and the grey-shaded area the highest density ranges.
**Fig. 2.** Modelo edad-profundidad del suelo CC1, calculado mediante interpolación lineal usando Clam.R (Blaauw, 2010). Los bloques en las edades radiocarbónicas representan el 95% de nivel de confianza en las calibraciones y el área sombreada en gris los rangos de mayor densidad de probabilidad.

3. RESULTS AND DISCUSSION

The common trend reported in the literature for δ^{13}C and δ^{15}N is to increase 1-3 ‰ with depth, (HÖGBERG 1997; EHLERINGER *et al.* 2000; HEIL *et al.* 2000; KRULL & SKJEMSTAD 2003). However, these data are generally referred to soils that have only one recognizable pedogenetic cycle. The isotopic depth variation in CC1 soil is not in agreement with this general trend, because it is a polycyclic sequence with at least 3 additions of edaphic material that evolved in different time periods under different management.

3.1. δ^{13}C

The δ^{13}C values ranged -26.6 ‰ to -24.5 ‰ (Fig. 3) and are coherent with the range reported for soils developed under C3 vegetation (ARROUAYS *et al.* 1995; BOUTTON *et al.* 1998; SPACCINI *et al.* 2000). The δ^{13}C depth variation is independent of the C content. The upper half of 4A has identical δ^{13}C than the whole 3A horizon (-25.6 ± 0.08 ‰), while the lower half of 4A shares with the 4B layer the highest δ^{13}C values of the soil (-24.7 ± 0.08 ‰). This abrupt change in ^{13}C abundance may indicate a shift in the ^{13}C isotopic signature of the C inputs into the soil or/and a different management. From previous research it is known that SOM decomposition was indeed higher in 3A with regard to 4A as a result of the introduction of the intensive agriculture (FERRO-VÁZQUEZ *et al.* 2014). Therefore an increase in ^{13}C abundance in 3A was already expected. Instead of this, δ^{13}C values remain low (similar to the upper
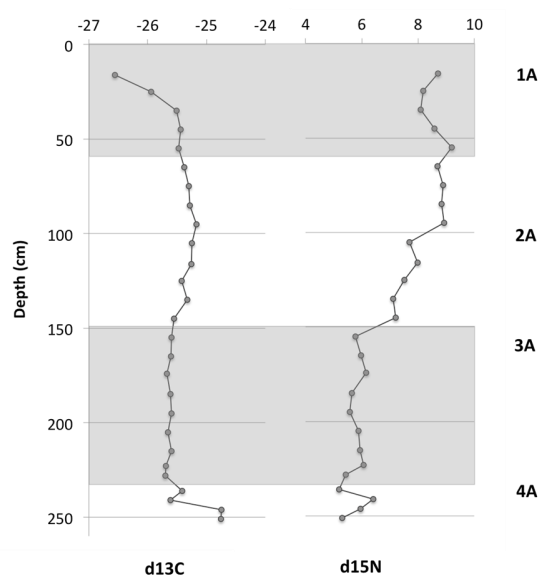
Fig. 3. Isotopic composition of the SOM in CC1 soil: depth variation of δ^{13}C and δ^{15}N
Fig. 3. Composición isotópica de la MOS en CC1: variación con la profundidad de δ^{13}C y δ^{15}N

samples of 4A), suggesting an input of non or poorly humified and ^{13}C depleted SOM that counteracted the effect of the increased decomposition. A change in vegetation yielding ^{13}C-depleted litter would be a possible explanation for this result. Also an amendment with some ^{13}C poor material could have occurred, e.g. the addition of *Ulex* spp. ashes, which have a lower $\delta^{13}C$ (-28 ‰, GÓMEZ-REY *et al.* 2013a) than the original slope soil (-25.1 ± 0.45 ‰).

The $\delta^{13}C$ values remained low and fairly unchanged (-25.6 ± 0.05 ‰) in 3A and increased slightly (0.5 ‰ average) but significantly (p<0.01) in 2A (-25.3 ± 0.08 ‰). In 1A the $\delta^{13}C$ decreases strongly (~1 ‰ in the upper 35 cm).

The increase in $\delta^{13}C$ in 2A and the lower part of 1A is coherent with an intensification of soil management, as pointed out by Ferro-Vázquez *et al.* (2014) from the data on Al and Fe fractionation. Nonetheless, an accelerated SOM decomposition leading to a $\delta^{13}C$ increase could have been promoted by other factors, such as: i) increased temperature; ii) addition of a ^{13}C enriched amendment (e.g. green or cattle manures derived from C4 plants, GLASER *et al.* 2001; BOL *et al.* 2003; KRISTIANSEN *et al.* 2005); and/or iii) an increase in microbial biomass (HEIL *et al.* 2000; KRULL & SKJEMSTAD 2003; DIJKSTRA *et al.* 2006), since it is generally ^{13}C enriched (1.6 ‰)

relative to the total C for soils that exhibited a C3-plant signature (DIJKSTRA *et al.* 2006).

The strong decrease in $\delta^{13}C$ values in 1A (Fig. 3) is probably related to the decrease of $\delta^{13}C$ in atmospheric CO_2 due to fossil fuel combustion and biomass burning in the last centuries (particularly since the Industrial Revolution), commonly known as Suess effect (SUESS 1955; REVELLE & SUESS 1957; LEVIN *et al.* 1989). Also, the addition of fresh OM from recent inputs could be contributing given that the terrace is still being cultivated nowadays.

3.2. $\delta^{15}N$

The $\delta^{15}N$ values ranged between 4.7 ‰ to 9.2 ‰ (Fig. 3), being the upper limit of the interval slightly higher than the $\delta^{15}N$ range of –1 ‰ to + 8 ‰ usually found in forest or cultivated soils (BINKLEY *et al.* 1985; NADLEHOFFER & FRY 1988; HADLEY & SCRIMGEOUR 1997; KOBA *et al.* 1998; GONZÁLEZ-PRIETO & VILLAR 2003). A multiple stepwise regression analysis was performed in order to determine which processes led to this $\delta^{15}N$ signature. The best model (Table 3) explains 79% of the variance and includes pH_{KCl} (63%), C/N ratio (9%) and $\delta^{13}C$ (7%).

Table 3. Summary of the selected regression model for the $\delta^{15}N$[a].
Tabla 3. Resumen del modelo de regresión seleccionado para $\delta^{15}N$[a].

R2	R2 corrected	error
0.887	0.786	0.757

Coefficients[a]

Modelo	Non standardized coefficients		Typified coefficients	Sig.	Tolerance
	B		Beta		
pHKCl	-0.953	0.116	-8.250	0.931	1.074
C/Nmasas	-0.614	0.197	-3.117	0.996	1.004
$\delta^{13}CTF$	0.358	0.138	2.597	0.934	1.071

a. The calculations are based only in samples from 1A, 2A, 3A and 4A horizons.

The pH_{KCl} indicates exchangeable acidity and its increase relative to the pH_w is used to infer a more positive charge in the soil mineral fraction, i.e, a higher capacity to adsorb anions. The main inorganic anionic N form in this soil is thought to be nitrate (under aerobic conditions, denitrification is not favoured and the nitrite amount is minor) and is originated by nitrification (the addition of nitrate amendments is a modern technique whose use became generalized only since the 1950s onwards). Nitrification is a highly fractionating process that produces ^{15}N-depleted nitrate at the expense of an ammonia source (SHEARER & KOHL 1986; HÖGBERG 1997), also contributing to the acidification of the soil (CURTIN *et al.* 1998; MORA & BARROW

1996; EGIARTE *et al.* 2006; VERDE *et al.* 2010). Therefore, ammonia addition (e.g. from fertilization) would lead to relevant amounts of ^{15}N-depleted nitrates. From the pH_{KCl} it is suggested that the soil anion retention capacity is low in the in the upper terraced layers (1A and 2A) as a result of the diminished amounts of reactive mineral surfaces, due to the terrace building method and to the more intensive management (FERRO-VÁZQUEZ *et al.* 2014), thus promoting the leaching of ^{15}N depleted NO_3^-. The soil $\delta^{15}N$ therefore would increase in the upper soil layers because the remaining enriched NH_4^+ is kept in soil, adsorbed in the exchange positions or even as non-exchangeable NH_4^+ in clay interlayers. This is in agreement to the

inverse correlation between $\delta^{15}N$ and other properties related to the reactivity of the soil amorphous phase [pH in NaF (pH_{NaF}, r^2=0.67) and phosphate retention (Pret, r^2=0.77)] in 1A, 2A and 3A horizons. In the same way, the soil N content showed a strong negative correlation (r^2=-0.759; p< 0.0005) to the $\delta^{15}N$ signature in the terraced soil layers (1A, 2A and 3A) pointing to an open N cycle with sustained N losses in the cultivated soil. Accordingly, the results of other investigations reported a high positive correlation between the ^{15}N abundance and N losses (HÖGBERG & JOHANNISSON 1993; GONZÁLEZ-PRIETO et al. 2013).

The variance that is not explained by the NO_3^- lixiviation, i.e. the $\delta^{15}N$ values detrended for pH_{KCl}, is partly explained by the C/N ratio and the relative abundance of ^{13}C isotope. The C/N ratio has a negative coefficient, meaning that an increase in OM with high C/N values would lower the $\delta^{15}N$. This makes sense since, as decomposition progresses, the concentration of N in SOM increases, while C concentration decreases in relation to their original amounts, at the same time producing the ^{15}N enrichment of the soil because the chemical and microbiological processes discriminate against the heavier isotope (KOERNER et al. 1999). The contribution of $\delta^{13}C$ to the regression model is probably reflecting differences in decomposition rates and/or changes in SOM composition, being temperature and humidity the two environmental factors with the greatest impact on the rate of decomposition of SOM (LEIRÓS et al. 1999; ZAMAN & CHANG 2004). Climatic conditions being equal, the kind and amount of organic matter influence the isotopic signature of the soil N (BARRACLOUGH et al. 1998). An increase of microbial biomass may also contribute, as mentioned above (HEIL et al. 2000; KRULL & SKJEMSTAD 2003; DIJKSTRA et al. 2006).

In the 4A horizon, a strong oscillation in the $\delta^{15}N$ values was observed (+1.1 ‰ between 251-241 cm depth and -1.2 ‰ between 241-236 cm, Fig. 3). This could be related to deforestation events since Pardo et al. (2002) found that the increased $\delta^{15}N$ detected after clear-cutting of vegetation returned to near-initial values as the vegetation recovers. Fire also increases N losses that discriminate against the heavy isotope, leading to higher punctual soil $\delta^{15}N$ values (COUTO-VÁZQUEZ et al. 2006, 2011; GÓMEZ-REY et al. 2013b), which can progressively return to pre-fire levels as the N cycle becomes more 'closed'. The more open N cycle in these samples coincides with the aforementioned abrupt change in the $\delta^{13}C$, pointing to the possibility of a change in vegetation cover after disturbance. In this sense, the development of andic soils from basic igneous materials in temperate areas has been linked to rejuvenation processes (GARCÍA-RODEJA et al. 2004), which could have been related to colluvial mechanisms enhanced by deforestation.

Soil $\delta^{15}N$ increased only slightly from the upper part of 4A to 3A horizon (+0.1 ‰; +0.3 ‰ if the peak value at 241 cm is not considered, Fig. 3). The $\delta^{15}N$ values are lower than those expected from the anion retention, meaning that the other variables included in the regression model (C/N ratio and $\delta^{13}C$) have a lowering effect in the N isotopic signature. This could be explained by a slight disruption of the N cycle by a non-intensive use, which allowed high C/N ratios and a decrease in $\delta^{13}C$ values. However, this is unlikely considering the effort invested in terrace construction. Moreover, previous studies indeed reported an increase of the decomposition rate (FERRO-VÁZQUEZ et al. 2014). Therefore, if an intensive cultivation is assumed, these relatively low $\delta^{15}N$ values may be due to an input from a ^{15}N-depleted source respect to soil $\delta^{15}N$ signature which counterbalanced, at least partially, the N losses triggered by cultivation, such as, for example, legume green manure. In particular, *Ulex* spp. shrubs are leguminous plants that actively fix atmospheric N_2 and are ^{15}N depleted (SHEARER & KOHL 1993; COUTO-VÁZQUEZ et al. 2011) and have been extensively used in the traditional agriculture in NW Spain to maintain soil fertility (BOUHIER 1979).

In agreement with this, some Fabaceae shrubs that have been traditionally used in the agricultural management have high P contents (720 mg/kg in the foliar part of *Ulex micranthus*, 725 mg/kg in *Pterospartum tridentatum*, COUTO-VÁZQUEZ et al. 2011). Their addition to the soil could explain the data on P content, which are higher in the 3A and 2A layers than in the paleosol despite the latter has a larger phosphate retention capacity. In this sense, Haynes & Mokolobate (2001) found that organic residues could be used as a tool to reduce the rates of lime and P fertilizer required for optimum crop production on acidic, P-fixing soils. This interpretation is in agreement with the $\delta^{13}C$ variation described above, since an addition of plant remains would lower the soil ^{13}C abundance, and with the $\delta^{15}N$ decrease (-1‰) found by other authors due to the net influence of atmospheric dinitrogen inputs via legumes (MARRIOT et al. 1997).

The $\delta^{15}N$ values in 2A horizon are higher than expected from N losses by nitrate leaching. Again, a contribution of an external ^{15}N-enriched source is suggested. A possible explanation for this enrichment would be the use of animal manure which led to an increase in the $\delta^{15}N$ value of soil (CHOI et al. 2003a; SENBAYRAM et al. 2008), primarily due to the high ^{15}N abundance of these amendments (HÖGBERG et al. 1995; GLASER et al. 2001; SENBAYRAM et al. 2008) and secondarily because they are substrate for bacterially-mediated reactions where the lighter ^{14}N isotope is preferentially lost leading to soil ^{15}N enrichment (KOERNER et al. 1999).

Two strongest increases in soil $\delta^{15}N$ were recorded in this soil horizon: the first one (+1.68 ‰) in the uppermost sample of 3A and the four deepest samples of 2A (7.50 ± 0.36 ‰), and the second one just at the middle of 2A horizon (Fig. 3). This suggests that the use of this soil horizon may have had two different phases, which might be related to a crop change, a shift in the composition of the amendment or to a more frequent or intense fertilization. Alternatively, this two-step variation could indicate that this terrace layer had two phases of construction, albeit it hasn't been detected in other soil properties.

The high $\delta^{15}N$ values of the 1A horizon (Fig. 3) are likely due to an open N cycle with substantial N losses, since the anion retention in these samples is low. In spite of this, the $\delta^{15}N$ values are much lower than predicted for nitrate lixiviation. This indicates an addition of depleted N from the surface, probably from a recent inorganic input, since many investigations reported low $\delta^{15}N$ values for the chemical fertilizers more commonly used nowadays (e.g. CHOI *et al.* 2003b; VITORIA *et al.* 2004; KANSTRUP *et al.* 2011).

3.3. Inferred implications for the evolution of soil management

In Figure 4 the isotopic data are represented with respect to the interpolated age of the analyzed samples. The upper sample of the paleosol was dated to 490-360 cal BC (Table 1), corresponding to the end of the Iron Age. But the development of this *in situ* soil was probably longer, including the Roman Period, given the wide chronological gap between this sample and the bottom of the overlying soil layer (530-650 cal AD), which suggests that the paleosol was possibly truncated to some extent.

The local increase in $\delta^{15}N$ values found in the upper part of 4A and the abrupt change in $\delta^{13}C$ (Fig. 4) suggest a deforestation event followed by a change in vegetation with respect to the underlying samples. Accordingly, recent palaeoenvironmental investigations suggested a critical change in the exploitation of natural resources in NW Iberia in this period (MARTÍNEZ CORTIZAS *et al.* 2009). It is not clear which deforestation technique was used since both clear-cutting and burning would lead to a similar isotopic soil signature. However, the use of fire as a clearance tool is supported by the results of Verde *et al.* (2008) who found a SOM molecular composition coherent with repeated fire episodes in the paleosol samples of the Monte Gaiás terraces system. Anthropogenic fires have been documented in Galicia from 8000-6000 years ago (KAAL *et al.* 2011, 2013) as a practice commonly used to induce changes in the vegetation cover, favouring the sprouting of lignified vegetation and the liming of the soils with the ashes. Still, a shift in soil use to cattle grazing could also help

to punctual increases of the $\delta^{15}N$ by animal dung deposition and to $\delta^{13}C$ decrease as explained above, and would also be coherent with the increase in P contents recorded in these samples.
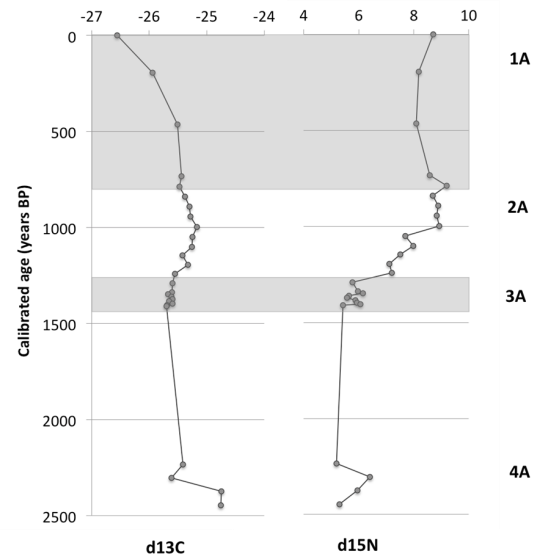

Fig. 4. Variation of $\delta^{13}C$ and $\delta^{15}N$ with interpolated age.
Fig 4. Variación de $\delta^{13}C$ y $\delta^{15}N$ con la edad interpolada.

Both, fire and grazing, are in agreement with a non-intensive use of the slope soil before the construction of the terraces. A similar situation was found in other European locations. For the Late Iron Age and the Roman Period in southern England, Rippon (2000) and Fyfe *et al.* (2004) reported a land management characterized by pastoral activities with no evidence of arable cultivation. However, this is in contrast with the findings for other Galician sites where a more intensive activity was reported for the Iron Age in comparison to that of Monte Gaiás (see, e.g. TERESO *et al.* 2013, who found evidence of massive wheat and millet storage in southern NW Spain).

The radiocarbon dating of the 3A layer indicates that it was in use during 6th-7th centuries (Fig. 4), when the Early Middle Ages Cold Period started (c. 1700 BP, MARTÍNEZ CORTIZAS *et al.* 1999). In the NW of the Iberian Peninsula, this period is characterized by climate deterioration with a decrease in global temperature that would have caused abiotic stress (such as decreased temperature, drought or low nutrients availability) and could have led to decreased $\delta^{13}C$ and $\delta^{15}N$ by decreasing SOM mineralization (Fry 1991; MARRIOTT *et al.* 1997; AUSTIN & VITOUSEK 1998). However, previous studies have shown that an effective increase in SOM decomposition indeed occurred after the construction of the terrace (FERRO-VÁZQUEZ *et al.* 2014). Therefore, a contribution of some ^{13}C and ^{15}N depleted material that lowered the soil $\delta^{13}C$ and $\delta^{15}N$ seems to have occurred. The more likely explanation is the applica-

tion of *Ulex* spp. shrubs as fertilizer. Gorse (*Ulex europeaus*) in particular, which is an abundant shrub throughout NW Iberia, would provide nitrogen and phosphorous, both limiting nutrients in soils with high anion retention capacity as CC1. The use of this species as fertility amendment in the traditional agricultural practices has been reported in previous investigations (BOUHIER 1979; CRIADO-BOADO 1989; KAAL *et al.* 2011) and Ballesteros-Arias *et al.* (2011) considered that gorse is indeed the basis for the exploitation of the soil in the traditional Galician agriculture.

The ^{15}N signature of horizon 2A points to an intensification of land use and a shift to fertilization with animal manures in the period of use of

this layer (8th to 11th centuries, Fig. 4 and 5). This is in agreement with the hypothesis of Criado Boado (1989) and Bauer (2005) about the use of manure-enriched shrubs after being used as livestock bedding, which is a common practice in the traditional agricultural system. The $\delta^{13}C$ increase in this layer indicates that other possible changes have taken place such as an increase in temperature during the Warm Medieval Period (MARTÍNEZ CORTIZAS *et al.* 1999), or the introduction of C4 crops - e.g. millet, *Panicum miliaceum L.*, whose use has been documented as early as the 6th century BC in Galicia (AIRA *et al.* 1990) and has been a major crop in the traditional agricultural system (VÁZQUEZ-VARELA 1994).
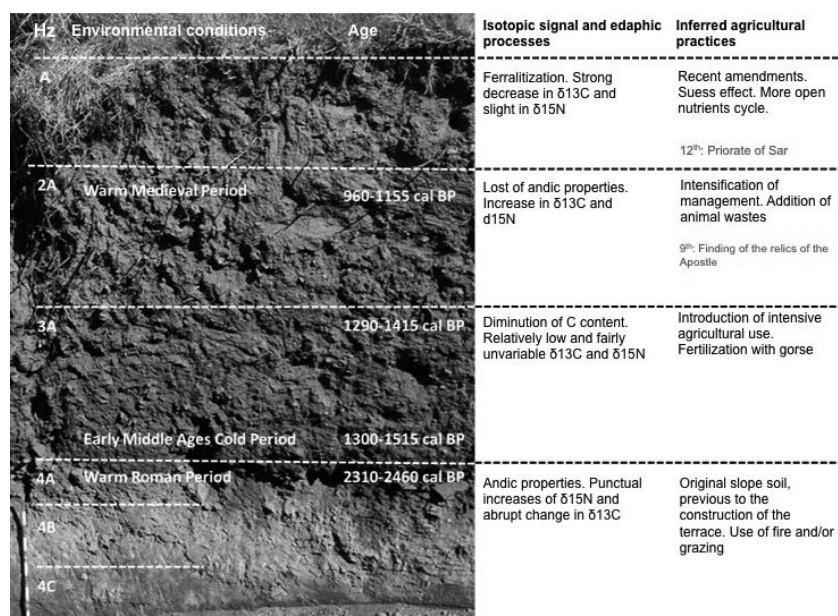
Fig. 5. Chronology, environmental conditions, synthesis of the isotopic signal and soil formation processes and inferred agricultural management for each soil horizon (Hz).
Fig. 5. Cronología, condiciones ambientales, síntesis de las señales isotópicas y de los procesos edáficos, y manejo agrícola inferido para cada horizonte del suelo (Hz).

Two different episodes are suggested in this period (roughly 8th to 9th century and 10th to 11th century) related to an intensification of the cultivation: a different composition or a change in the frequency of addition of amendments, and/or a crop change. In this sense, after the finding of the relics of the Apostle Saint James the Great in the 9th century AD started a social and economic development that led to the transformation of the small town of Santiago into an emerging demographic, administrative and trade centre, resulting in a major urban development in the 11th and 12th centuries AD (CAAMAÑO & SUÁREZ 2003; BALLESTEROS-ARIAS 2006a). This transformation required a large structure to provide livelihood and probably had an effect on the intensification of the cultivation.

The isotopic signature of 1A (built during 12th-13th centuries) reflects the effects of an intensive use from the Late Middle Ages to present. Other recent archaeological research elsewhere in Santiago de Compostela provided further evidence on intense agricultural activity during 12th and 13th centuries (TEIRA BRIÓN *et al.* 2010). These authors found a large abundance of Poaceae pollen in sealed storage structures dating to the 12-13th centuries. This intensive use is in agreement with FERRO-VÁZQUEZ *et al.* (2014) who suggested and increase in soil degradation due to the agricultural intensification and recent additions of lime in this soil horizon. Manuring practices were reported for 11th and 12th centuries in other locations of the Iberian Peninsula, consisting in the addition of domestic wastes as fertility

amendments (QUIRÓS CASTILLO *et al.* 2014). However, no systematic research has been done on this topic so far, rather manuring has been suggested by the lack of proper middens or dump heaps and from surface scatters of domestic residues.

4. CONCLUDING REMARKS

Agricultural terraced soils are suitable to be used as environmental archives. Their multiproxy study offers a great potential for identifying and interpreting the impact of past agricultural activities, providing key information when archaeological artifacts are missing.

The study of the changes in the $\delta^{13}C$ and $\delta^{15}N$ isotopic fingerprint provided information on the evolution of the fertilization practices in Monte Gaiás terraced soils. Evidence was found of extensive land management previous to the construction of the terrace, with the utilization of fire as liming and clearance tool. The addition of plant remains from N_2 fixing shrubs is proposed as fertilization technique in the Late Antiquity and Early Medieval Ages. For the Late Middle Ages the introduction of the use of animal wastes as a means of maintain or increase soil fertility to address the growing productivity needs, was also detected.

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