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Geochemical fingerprint of agricultural liming as a regular management practice in Modern-period Basque farming

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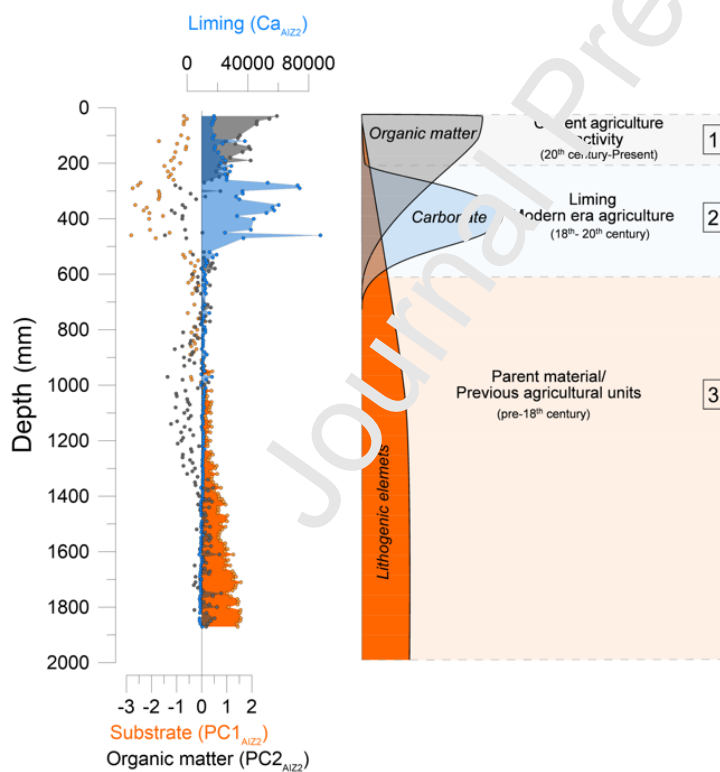
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Abstract

The Modern period in Europe is marked by the introduction of deep agricultural changes. In the Basque Country (northern Spain), the implantation of an intensive crop rotation was made possible by the expansion of agricultural liming, although the extent and implications of this practice have not been previously explored in depth. The present paper proposes a multidisciplinary approach to this question, based on the combined analysis of archival sources, toponymy, visual prospection focused on the presence of limekilns, and agricultural

soil coring in four local contexts of the Atlantic Basque Country. The results show, for the first time, evidence of concurrent and widespread liming in this territory at the edge of the 18th century, with strong implications for the model of agricultural management in the communities involved. The spreading of mineral fertilisation reflects an intensification in the forms of agricultural management, in the framework of a new relationship between land and labour that emerged after the introduction of American crops. Continuous liming for more than 200 years exerted a deep impact in the analysed soils, with interesting socio-economic and ecological implications that are representative of the potential short-term effects that changing relationship between humans and their socio-ecological environment may produce in agricultural soils.

Graphical Abstract



Keywords

Liming; agroecosystems; agricultural intensification; soil geochemistry; archaeomagnetism.

1. Introduction

The Modern period (16th-19th centuries) is defined by the rise of traffics that linked both

edges of the Atlantic Ocean into an increasingly tight exchange of peoples, ideas, and goods on the basis of a colonial relationship. As a result of this contact, the face of the Americas was transformed forever, but the repercussions for European societies were equally drastic. The activation of transatlantic trades not only permitted an increasing concentration of commercial capitals in the urban centres of many Atlantic regions, but also enhanced a complete reorganisation of the socio-economic, ecological, political and cultural structure of their respective hinterlands (Crosby, 2003). This was primarily driven by deep transformations in agricultural management practices, including the introduction and consolidation of new crops, like maize (*Zea mays*), potato (*Solanum tuberosum*) or common beans (*Vicia faba*) (e.g., Fassina, 1982; Cazzola, 1991, Fornasin, 1999; Contis, 2008); a progressive enclosure of common lands (e.g., Yelling, 1977; Wordie, 1983); the colonisation of wastelands and marshes (e.g., Van de Ven, 1994; Morera, 2010); or the introduction of technical innovations and new agricultural implements (e.g., Delleaux, 2005; Barnebeck Andersen, *et al.* 2013). Parallely, a theoretical and ideological superstructure was developed by an incipient modern agronomy from early empiricists like Olivier de Serres to classic authors like J. Tull or A. Young, in Britain, or the physiocrats, among others, in France. Eventually, these works set the intellectual basis for the development of public projects aimed at promoting agrarian improvement, encouraged by enlightened governors in different European states with unequal success (e.g., Hubatsch, 1975; Mattolini, 1981; Lluch & Argemí, 1985).

Regardless of the controversy on the 'revolutionary' character, or not, of such transformations from a socio-economic point of view (e.g., Morineau, 1971; Moriceau, 1994), it is clear, at least in some areas, that their concurrence permitted the implementation of more intensive agricultural systems. One of the key factors for this was the experimentation and massive use of soil amendments, especially lime, whose benefits include

balancing soil pH and restoring nutrient losses, mainly calcium and magnesium, in acidic soils (Kirk *et al.*, 2010; Fageria & Nascente, 2014; Kunhikrishnan *et al.*, 2016; Holland *et al.*, 2018), as well as improving soil structure, increasing plant productivity and stimulating soil microbial activity (Chang & Sung, 2004; Paradelo *et al.*, 2015). Although the use of lime to increase soil fertility was already a common practice in Roman times (e.g., Vanwalleghem *et al.*, 2004; Connor *et al.*, 2011), from the 18th century onwards, lime was widely applied with the purpose of counteracting soil acidification and improving soil quality in a number of European regions, with long-lasting effects on the local systems of agricultural management (e.g., Walsh *et al.*, 1957; Clout & Phillips, 1972).

Still, the long-term effects of historical use of lime as a regular farming strategy remain largely unexplored. This gap may be partially fulfilled by applying the theoretical framework developed within the last few decades by the Agrarian Archaeology, which focuses on the relevance of past agricultural management practices as a primary factor of landscape modelling and, eventually, of environmental change (e.g., Guilaine, 1991; Marcus & Stanish, 2006; Denham *et al.*, 2009; Kirchner, 2010; Quirós-Castillo, 2014; Narbarte-Hernandez *et al.*, 2019; 2020). To this purpose, the present paper aims to address the environmental impact of past agricultural liming in several local contexts of the Atlantic Valleys of the Basque Country, combining archaeological survey, documentary sources and geoarchaeological core analyses.

2. Area of study

The Atlantic Valleys of the Basque Country are located in the easternmost sector of the Cantabrian coast, roughly corresponding to the Historic Territories of Biscay and Gipuzkoa, and the Bidasoa Valley in Navarre (Fig. 1). This region consists of a narrow strip of land, with a set of accidented fluvial valleys running parallel from south to north. Among them, mountains, mostly of Cretaceous calcareous and marly rocks, progressively grow in height

from the coast towards the south, reaching their maximum in a set of aligned peaks (Gorbea, 1482 m asl; Aizkorri, 1523 m asl; Aralar, 1431 m asl) that define the limit between the Atlantic and Mediterranean watersheds of the northern Iberian Peninsula. Such geomorphological features, as well as a mild, humid Atlantic climate, define an erosive topography where human settlement is generally placed in the lower slopes of the mountains, or in the bottom of the valleys, around flood plains and meadows. The region is nowadays densely populated, with an uninterrupted demographic growth since the 16th century having sharply accentuated after the second half of the 19th as a consequence of the Industrial Revolution (Hernández-Marco & Piquero-Zarauz, 1988).



Figure 1. Location of the four study areas within the Atlantic Valleys of the Basque Country.

Spatial data source: GeoEuskadi.

Four locations from three different river valleys were selected in this area for fieldwork: 1) the village of Aizarna and the neighbour hamlet of Akoa, both included in the municipality of Zestoa (Urola valley); 2) the neighbourhood of Ibiri, included in the municipality of Mutriku (Deba valley); 3) the village of Alkiza (Oria valley); and 4) the village of Zizurkil (Oria valley) (Fig. 1). All of them are representative of the traditional rural landscape of the region,

organised as a complex integration of agro-silvo-pastoral resources distributed around the institution of *baserría* — individual farmsteads with an autonomous landholding, managed by a family group. The presence of such farmsteads is well attested since, at least, the Late Middle Ages, with detailed typo-chronological series having been developed from an architectural-archaeological point of view (e.g., Ibáñez-Etxeberria & Agirre-Mauleón, 1998; Santana, 2001; Susperregi *et al.*, 2017). The achievement of this model seems to have occurred during the Modern period, when the economic and demographic expansion associated to the rise of Atlantic traffics encouraged the enclosure of common lands with speculative purposes (e.g., Aragón Ruano, 2015; Narbarte-Hernandez, 2020). Parallely, the introduction of American crops like maize (*Zea mays*) in the 16th century, and their generalisation over the 17th, enhanced this process permitting the cultivation of lands that, so far, had been unpracticable due to their unfavourable topography, altitude, or soil features. As a result, many common lands — mainly extensive pasture or forest areas — were enclosed and ploughed, giving origin to an increasingly dispersed settlement pattern with agricultural decision-making being progressively decentralised to a domestic scale (Narbarte-Hernandez, 2020).

In this context, agricultural liming appears as one of the most relevant features of Modern agriculture in the region. The oldest mentions to its use date from 1704-1705 in the Bidasoa Valley, Navarre (Caro-Baroja, 1973; Mikelarena-Peña, 1988), and is described as being widespread in the valleys of Gipuzkoa by the mid of the 18th century:

Fields tend to weaken within a few years. To address this, every nine years the fields are amended with lime, and for this reason almost every household has its own limekiln, used to produce lime with much work and much expenditure of wood (M. Larramendi, 1756).

The practice of agricultural liming is still widely reported by ethnographic literature during

the first half of the 20th century, when the traditional system of domestic agricultural production was still current (e.g., Laffite, 1919; Lefebvre, 1933; Caro-Baroja, 1944; 1969; Douglass, 1975; Greenwood, 1976). In contrast, its use is virtually absent in the present-day landscape, though not from the local memory. In fact, the rests of old limekilns are still visible in many villages (e.g., Del Barrio & Zaldua, 1992; Agirre-Mauleón, 1995; Olañeta & Urkiola, 1998; Burgi *et al.*, 2008; Fernández-Carvajal, 2013; Martínez-Montecelo, 2014; Sánchez-Zufiaurre, 2014), albeit most of them very eroded, invaded by vegetation and, occasionally, partly or totally torn down.

3. Materials and methods

The study areas were selected in the framework of a regional-scale study of the evolution of rural landscapes, according to three criteria: 1) relatively good conservation of the ‘traditional’ landscape layout, with limited impact of modern urbanisation and/or industrialisation; 2) homogeneous distribution across the region — Alkiza and Zizurkil being located in the Oria hydrographic basin, while Aizarna and Akoa in the Urola basin and Ibirri in the Deba basin —; and 3) representativeness of the regional diversity in terms of altitude, geomorphological features, and socio-economic aspects. The occurrence of Modern-period agricultural liming in each study area was assessed through a combination of different proxies.

3.1. Documentary sources, ethnography, and toponymy

Documentary records on Modern-period agricultural practices were consulted in local and regional archives, both public and private. Special attention was set on mentions to the following aspects: 1) agricultural liming, its frequency and intensity; 2) the construction, maintenance and management of limekilns; 3) general management problems linked to agricultural intensification and/or soil erosion; 4) historical cartography.

Ethnographic fieldwork was also a relevant source of information concerning traditional

resource management and knowledge, as well as the symbolic value attributed to some spaces and practices. Local informants were regularly consulted on these questions in each focus area, following a methodology of informal conversation.

Finally, toponymy is a very relevant source of information for the investigation of past agricultural management, particularly in currently inhabited contexts where it is still performative and articulates complex systems of perception and appropriation of the space. The Municipal Toponymic Maps of Alkiza (2005), Mutriku (1999; 2010), Zestoa (2010) and Zizurkil (2016) were used to identify micro-toponyms referring to functional or symbolic aspects of specific spaces in each local context.

3.2. Core sampling

Traces of past agriculture were documented through soil core sampling, carried out between 2016 and 2018 along the fields of each area of study. This method is non-invasive and allows the recovery of complete soil archives, from which particular aspects of land use and agrarian management practices can be inferred. The samples were collected using a *Van Walt/Eijkelkamp* mechanic percussion portable corer, then sealed and stored at 3-4°C.

Eleven cores were considered in total: 5 cores in Aizarna/Akoa (Zestoa) (AIZ/1, AIZ/2 & AIZ/3 & AKU/1 & AKU/2), 1 in Alkiza (ALK/1), 1 in Ibirri (Mutriku) (IBI/1) and 4 in Zizurkil (ZIZ/1, ZIZ/2, ZIZ/4 & ZIZ/5) (Fig. 2). Sampling points were selected in areas where sedimentary records were more likely to be preserved, seeking to cover the widest range of possible situations regarding agricultural management. All the cores were obtained from Plio-Quaternary fluviokarstic siliciclastic sedimentary records infilling karstic depressions (dolines and uvalas) (Fig. 2) except ALK/1 core, which was performed on the current floodplain of the Mandabe river (Alkiza) (Fig. 2). Three cores are representative of small plots aimed to intensive vegetable and/or fruit cultivation close to the habitations themselves: Core AIZ/3 was gathered in the garden of the rectory house of Aizarna, Core

IBI/1 in a pasture plot next to the farmhouse Ibiritxo (Ibiri), and Core ZIZ/2 in an orchard close to the farmhouse Aretagoikoa (Zizurkil). The rest of cores correspond to cereal fields or grasslands interspersed within the general spatial layout of the settlements: Cores AIZ/1 and AIZ/2 were sampled near the farmhouses Potzueta and Aranguren (Aizarna), Cores AKU/1 and AKU/2 near the farmhouses Potzueta and Aranguren (Aizarna), Cores AKU/1 and AKU/2 near the farmhouses Akoabarrena and Akoa (Akoa), Core ALK/1 near the farmhouse Intxaurreandiaga (Alkiza), and Cores ZIZ/1, ZIZ/4 and ZIZ/5 near the farmhouses Azkarate, Kamioaundi and San Millan (Zizurkil). The depth of the cores varied from 1 to 5 m; the first 2 meters of the longer cores are considered in this work.

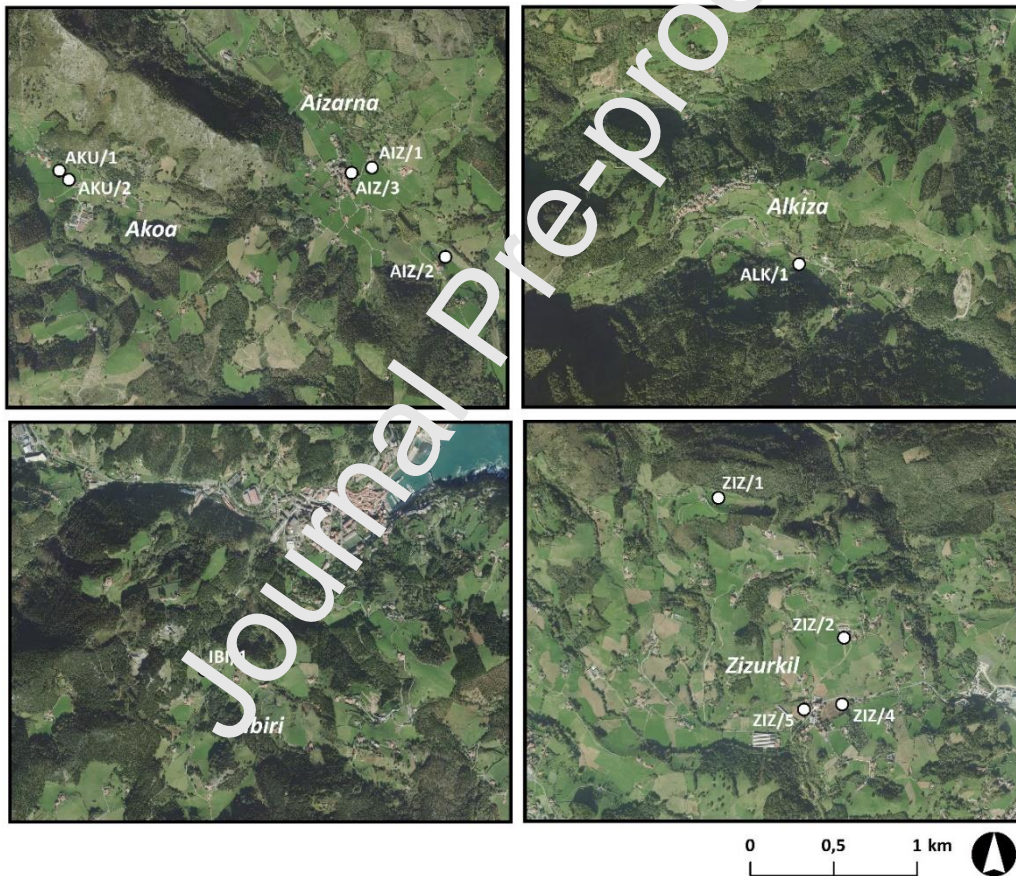




Figure 2. Points of core sampling in the four focus areas. (a) Aizarna and Akoa. (b) Alkiza. (c) Ibiri. (d) Zizurkil. Spatial data source: GeoEuskadi. (e) Core sampling (ZIZ/1) in a field by the farmhouse Azkarate, in Zizurkil. Photo: Oihane Mendizabal Sardonís.

3.2.1. X-Ray Fluorescence (XRF)

Cores AIZ/1, AIZ/2, AIZ/3, AKU/1, AKU/2, ALK/1, ZIZ/1, ZIZ/2, ZIZ/4 and ZIZ/5 were analysed every 1 cm, using an *Avaatech* XRF core-scanner, at the Corelab Laboratory (University of Barcelona). The analysis was performed under two different working conditions: 1) with an x-ray current of 800 μA , at 10 s count time and 10 kV x-ray voltage for the measurement of Al, Si, P, S, Cl, Ar, K, Ca, Ti, V, Rh, Cr, Mn and Fe; 2) with an x-ray current of 2000 μA , at 25 s count time, 30 kV x-ray voltage and using a Pd filter, for the measurement of Ni, Cu, Zn, Ga, Ge, As, Br, Rb, Sr, Y, Zr, Nb and Pb. This method allowed a semi-quantitative analysis of the elemental chemical composition from Al to U, based on the proportion of counts per second (cps) for each element compared to the rest. The most

abundant and significant 13 elements (well above 600 counts) present in all cores were selected for statistical analysis: Al, Si, K, Ca, Fe, Ti, Rb, Sr, Y, Zr, Mn, Zn and Pb. Before the PCA analysis, all unreliable measurements, due to the presence of small gravels and disaggregated sediment intervals, were removed from the final results, so as not to obscure the statistical treatment of the data. XRF geochemical data were first transformed using centred log-ratio transformation (Weltje *et al.*, 2008; 2015) by means of *CoDaPack* software (Comas-Cufí *et al.*, 2011), and then processed with multivariate statistics, Principal Component Analysis (PCA), for each core and all cores together, using the program *SPSS 23.0*. The goal of these operations was to reduce the number of variables and to define the main phases and processes involved in the formation of each core record. PCA was performed at correlation mode. Rotated (Varimax) and non-rotated solutions were evaluated, and the most suitable one to geochemical data variance was selected. Finally, factor scores were calculated for each measurement.

Due to its drier (harder) character, Core IBI/1 was analysed every 6 cm through standard XRF analysis. The equipment used for XRF analysis is a *Thermo Electron Corporation ARL ADVAT XP Sequential XRF* (Technical Scientific Park of the University of Burgos). The preparation of the samples consisted on the production of glass beads by borate fusion mixing 0-5 gr of sample powder with metaborate/tetraborate (50:50). The obtained spectra were interpreted using the *WinXRF.ADVANT 3.2.1* software. Finally, the obtained results were analysed to determine the semi-quantitative concentration of the elements with the *UNIQUANT 5.47* software, the results being expressed in percentage (%) in the form of oxides.

3.2.2. C and N analyses

The total C and N content of the samples was determined at the Laboratory of Soil Science and Agrochemistry of the University of Burgos. Subsamples of 0.2 g were collected from

Cores AIZ/2, AKU/2 and IBI/1 every 2 cm, dried and milled using an agate mortar, then processed in an automated combustion analyser LECO TruSpec.

Additionally, the organic carbon present in the sediments was measured through thermal oxidation method or *Loss on Ignition* (LOI) in Core IBI/1. 0.7-0.8 grams of milled and homogenised sample were heated to 110 °C for 14 hours inside the muffle (Heron HD-230 PAD) and then to 550° for 5 hours. The weight of the samples was measured after each heating using an analytical balance and the lost organic C calculated according Santisteban *et al.* (2004).

3.2.3. Soil pH

Soil pH was measured in cores AIZ/2 and AKU/2 since they showed the most representative soil records, with relevant liming signal and radiocarbon dates. The pH analysis was carried out at the Laboratory of Soil Science and Agrochemistry of the University of Burgos. Soil samples from the top 1 m of Cores AIZ/2 and AKU/2 were taken every 4 cm, dried at ambient temperature, and sieved at 2 mm. Soil pH was determined in an aqueous suspension 1:2.5 (w/v) using a Crison GLP21 pH meter.

3.2.4. Chronology

Radiocarbon dating was practiced on charcoal or bulk sediment samples from the most significant stratigraphic units identified in each core. Samples from Cores AIZ/2, AKU/2 and ZIZ/4 were prepared at the Centre for Isotopic Research on the Cultural and Environmental heritage (University of Campania ‘Luigi Vanvitelli’) and AMS measurement was finally done at the Laboratory of Nuclear Techniques for the Environment and the Cultural Heritage (National Institute for Nuclear Physics; Florence, Italy). In the case of Core IBI/1, radiocarbon dating was performed at the Poznan Radiocarbon Laboratory (Poland).

3.3. Limekiln inventory

Field survey was driven in each local context to detect the number, location, and present-day

situation of limekilns.

3.4. Limekiln archaeomagnetic dating

An archaeomagnetic study was carried out on one of the limekilns in the village of Aizarna, with the purpose of determining the date of its last use and evaluating the suitability of these limekilns for archaeomagnetic dating. 12 oriented samples (14 specimens) were extracted with a water-cooled electric drill which incorporates a 2.5 cm-diameter diamond-coated bit. The samples were magnetically oriented with a Brunton compass and an inclinometer. The measurement of the natural remanent magnetization (NRM) and its progressive demagnetization was carried out with a 2G cryogenic magnetometer in the Paleomagnetic Laboratory at the University of Burgos (Spain). The NRM stability was analysed by stepwise alternating field (AF) and thermal demagnetization. AF demagnetization was applied up to a peak field of 120 mT with the demagnetization unit attached to the 2G magnetometer. Thermal demagnetization was performed with a TD-48 oven up to 600 °C. Additionally, bulk sample was collected to study the magnetic properties with the aid of a variable field translation balance. The structure of remanence components and the direction of the characteristic remnant magnetization (ChRM) was determined by principal component analyses (Kirschvink 1980), using the Remasoft software (Chadima & Hrouda, 2006). The mean archaeomagnetic direction was calculated using Fisher (1953) statistics.

4. Results

4.1. Archival and ethnographic work

The study of documentary, cartographic, ethnographic, and oral sources provided a broad overview on the chronology, extent and relevance of liming as a regular practice of agricultural management in the different focus areas. The volume, quality and conservation of these information varied greatly from one local context to another and was also thematically and chronologically limited by the nature of the records themselves. The most relevant

records were detected in the Municipal Archive of Zestoa. As a matter of fact, a private familiar archive from Akoa also provided very interesting records.

4.2. Core pedostratigraphy

The sedimentary records were similar in all cores. Most of the cored areas were karstic depression (dolines and uvalas) fillings consisting of silt and clays (Plio-Pleistocene to Holocene fluviokarstic sediments and slope deposits), where the organic content increased towards the top (modern agricultural soil). No aggrading paleosoil cycles were observed in the core records, since most of them corresponded to anthropically altered cambisols affecting previous fluviokarstic sediments with additional Medieval to Modern Age terracing sediments. Only in AIZ/2 core, an early medieval paleosoil related to fire-induced forest clearance was observed (Narbarte-Hernández *et al.*, 2019). Agricultural terrace fillings could be recognised in the upper halves of most of the cores. They consisted of clayey anthropic fillings, sometimes slightly pedogenised Ap horizons, including abundant anthropic material (ceramic, lime remains, charcoal, etc.), root bioturbation and ploughing traces.

4.2.1. Geochemistry and chemostratigraphy

The results of the PCA of XRF-CS geochemical data from all the analysed cores are summarised in Table I and Supplementary Information. Overall, the elemental geochemical composition and chemostratigraphy was very similar in all cores, with three similar principal components being repeated in all of them (Fig. 3 & Suppl. Inf.). These components are well recognized in both the general PCA analysis (Table I & Suppl. Inf.) and the individual PCAs realised for each core (Fig. 3 & Suppl. Inf.).

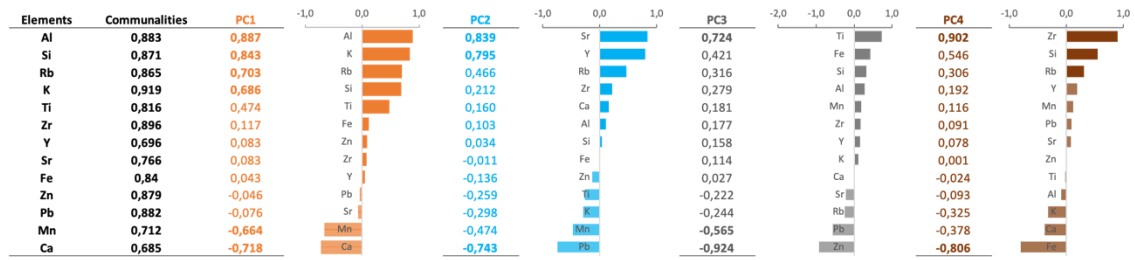


Table I. Results from PCA of the XRF-CS geochemical data from all the studied cores. Communalities and Factor loads of the selected 13 elements are presented. Bold values indicate the maximum explained variance (factor loading) for each element.

Variation in the elemental composition of the analysed cores is summarised in four principal components (PC1 to PC4) (Table I and Fig. 3), which explain 82.4% of the total variance. PC1 explains 28.3% of the variance. Al, Si and Rb show high positive factor loadings (> 0.7) and K has a moderately positive loading (> 0.5) (Table I). Ca has a high negative factor loading (-0.72). The factor scores of the elements with positive loadings included in PC1 (Al, Si, Rb and K) predominate in the deeper areas of the cores. The element with negative loadings (Ca) is present in the uppermost 1 to 0.5 metres (Fig. 3a and Suppl. Inf.).

PC2 explains 19.9% of the variance. Sr and Y show high positive loading (> 0.7) and Pb shows high negative loading (-0.74) (Table I). Factor scores of elements included in PC2 do not show clear and common trends in all the analysed cores, in general their values are quite homogeneous except some positive maxima coinciding with PC1 negative peaks (Ca-rich intervals) (Table I and Suppl. Inf.).

PC3 explains 17.4% of the variance. Ti shows a high positive factor loading (0.72) and Zn and Pb have a high (-0.9) and moderate (-0.57) negative loading respectively (Table I). Factor scores of the elements involved in PC3 (Pb and Zn) content show a marked transition to negative values, in upper 0.2 to 1 m of the studied cores (Table I, Fig.3b and Suppl. Inf.).

PC4 explains 16.7% of the variance. Zr and Si show high (0.9) and moderate (0.55) positive

factor loadings respectively, and Fe shows high negative loading (-0.8) (Table I). The factor scores of PC4 show quite constant negative values (around -2) in all analysed cores, significantly IBI, ZIZ/2 and ALK/1 cores show more negative values (-3 to -4) pointing to higher Fe content.

When PCA is performed individually for each core, each of them shows 3 PCs except for ZIZ/1 and ZIZ/4, which show 4 PCs (see Suppl. Inf.). PCs obtained for all cores show similar element distribution and factor scores as the ones of the general PCA (Table I and Suppl. Inf.). The results are very robust in all cores and PC1 includes the same elements (Al, Si, Rb and K) as general PCA's PC1 (Fig. 3a). Chemical elements with negative loadings (Pb and Zn) included in PC3 of the general PCA are also mostly present in PC1 (negative loading) of nearly all cores (Fig. 3), except for AIZ/2 (included in PC2 with negative loading) and ZIZ/4 (included in PC2 with positive loading) (Fig. 3b). Finally, Ca is normally included in PC1, with negative loading in all cores but AIZ/2, ALK/1, ZIZ/1 and IBI/1 (included in PC2's negative loading) (see Suppl. Inf.). Nevertheless, the relative Ca content is better observed representing the normalized Ca values obtained in XRF-CS analysis (Fig. 3C).

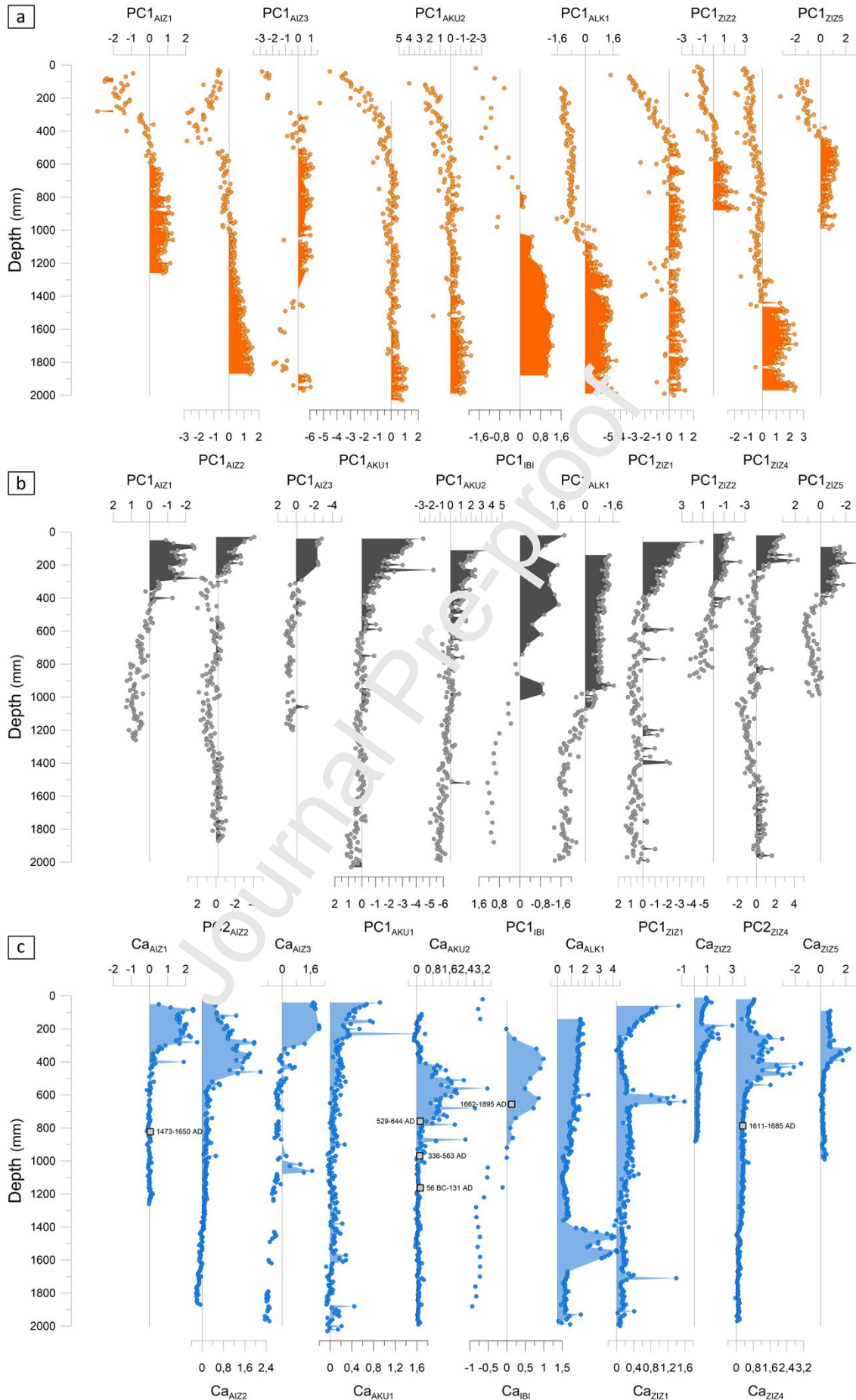


Figure 3. Synthesis of the XRF-CS geochemical composition and individual PCA results of studied cores. (a) Lithogenic element-related PCs' factor score graphs of the studied cores. (b) Organically-bound element-related PCs' factor score graphs of the studied cores. (c) Calcium content graphs from studied cores. Geochemical data are normalized using centred log-ratio (CLR) transformation except for IBI core (Ca_{IBI}) where it is expressed in weight percentage (wt%) of CaO. Most significant radiocarbon dates (yr cal AD) are shown (see 4.2.4. *Dating* section).

4.2.2. Carbon and Nitrogen analyses

Both C and N content were analysed in the most representative and well-dated cores, AIZ/2 and AKU/2 (Fig. 4). Both showed similar total carbon (C_t) values varying from 0.76 to 5.56 weight %, in AIZ/2 core and from 0.61 to 3.95 weight % in AKU/2 core. The same occurs for the nitrogen (N_t), values range from 0.1 to 0.5 weight % in AIZ/2 core and from 0.07 to 3.95 in AKU/2 core (see Fig. 4 and Suppl. Inf.). C:N ratio trends and variations suggest that they have recorded similar stratigraphy and agricultural practices in the analysed cores (Fig. 4). In both cases, soil C_t , N_t and C:N ratio are much higher at the surface (0–5 cm). The C:N ratio tended to decline with depth, parallel to the relative increase in soil clay content and more decomposed organic matter with lower C:N ratio (Diekow *et al.*, 2005; Ouédraogo *et al.*, 2006; Yamashita *et al.*, 2006).

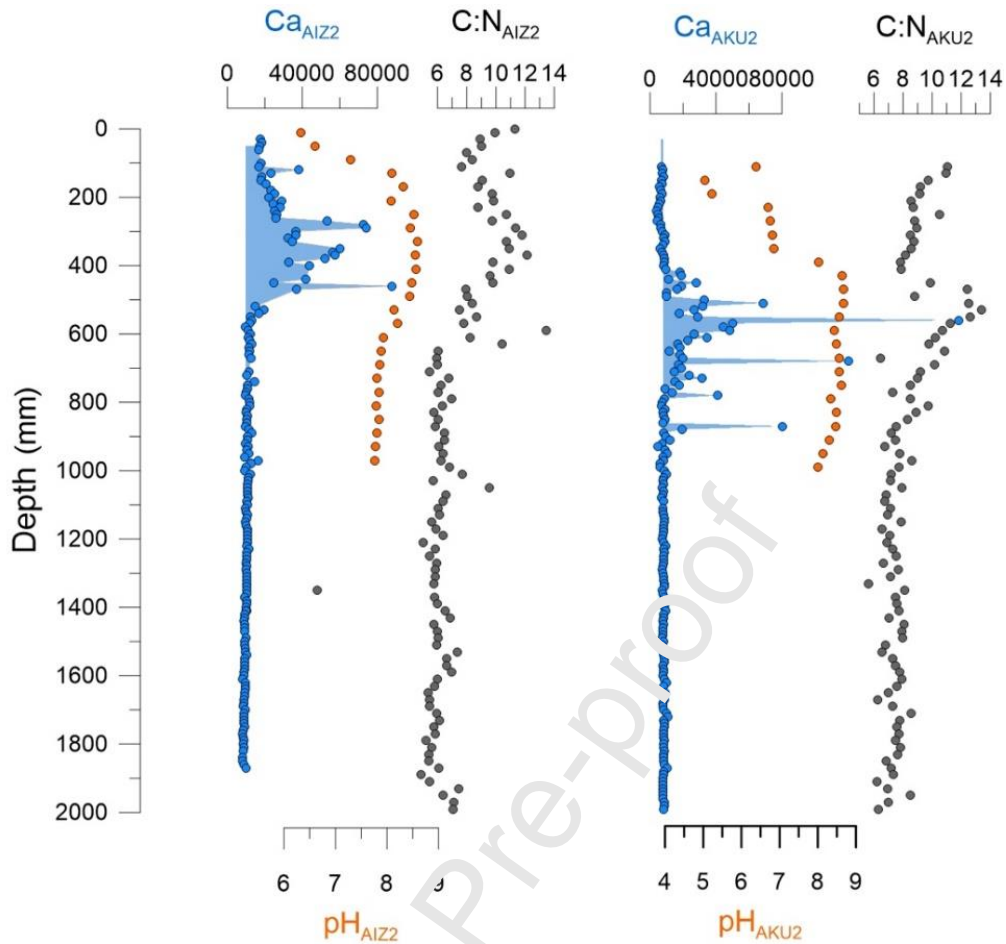


Figure 4. C:N ratio, Ca content (in cps) and pH values for AIZ2 and AKU2 cores. Note the pH and C:N ratio increase and maximum values in the Ca rich interval.

4.2.3. Soil pH

The pH measurement was also focused on cores AIZ/2 and AKU/2. Both records provided similar results, again suggesting similar uses and sedimentological and pedogenetic processes. In both cases, basal sediments showed pH values of about 7.8-8, which increased to values close to 8.5 at 30-60 cm depth in Core AIZ/2, and at 40-70 cm depth in Core AKU/2. In contrast, the surficial horizons of both records show much more acid pH, with values ranging from acid (pH 5.2) to slightly alkaline (pH 7.3) (Fig. 4). To infer the possible correlation between pH changes and Ca and/or C_t content, a PCA was performed for C_t , N, Ca and pH data of AIZ/2 and AKU/2 cores (see Suppl. Info.).

4.2.4. Dating

Twelve samples from different depths of Cores AIZ/2, AKU/2, IBI/1 and ZIZ/4 were radiocarbon dated in total. The results of these measurements are summarised in Table II. These results encompass radiocarbon ages spanning from the Iron Age until present. The best represented phases are the Middle Ages (4 samples) and the Modern period (5), the last one recurrently related to very characteristic geochemical features, i.e. the presence of high contents of organic matter and of anthropic additions of CaO.

Table II. Radiocarbon dated samples.

COR E	DEPT H (cm)	SIMPL E TYPE	LAB COD E	¹⁴C CON. (pMC)	t_r (YEAR ± BP)	CAL. AGE (YEARS - 1s)	CAL. AGE (YEARS - 2 s)
AIZ/ 2	47	sedimen t	Fi355 6	96.57 ± 0.56	800 ± 40	[1211–1270 AD]	[1166–1278 AD]
AIZ/ 2	82	sedimen t	Fi349 5	96.16 ± 0.45	314 ± 38	[1516-1597 AD] [1618-1643 AD]	[1473-1650 AD]
AIZ/ 2	97	sedimen t	Fi349 4	95.19 ± 0.48	396 ± 41	[1443-1514 AD] [1600-1617 AD]	[1433-1528 AD] [1553-1634 AD]
AIZ/ 2	142	sedimen t	Fi355 7	82.91 ± 0.56	1545 ± 45	[535–614 AD] [435–448	[506–641 AD] [428–497 AD]

						AD] [472-487 AD]	
AKU /2	77	sedimen t	Fi374 1	82.98 ± 0.41	1499 ± 40	[536-622 AD] [477-483 AD]	[529-644 AD] [429-494 AD] [510-518 AD]
AKU /2	97	sedimen t	Fi367 3	81.83 ± 0.50	1611 ± 50	[486-535 AD] [395-433 AD] [413-473 AD]	[336-563 AD]
AKU /2	117	sedimen t	Fi367 4	78.29 ± 0.39	1956 ± 46	[2 BC - 77 AD] [36-31 BC] [21-11 BC]	[56 BC - 131 AD] [88-76 BC]
AKU /2	137	sedimen t	Fi367 5	71.61 ± 0.53	2682 ± 50	[854-803 BC] [895-868 BC]	[930-791 BC]
ZIZ/ 4	36	Charcoa l	Fi423 0	99.51 ± 0.61	Modern	Modern	Modern
ZIZ/ 4	76	charcoal	Fi423 1	73.28 ± 0.51	2498 ± 56	[774-727 BC] [718-705 BC] [695-541 BC]	[794-430 BC]
ZIZ/ 4	79	charcoal	Fi423 3	96.93 ± 0.57	251 ± 47	[1523-1572 AD] [1630-1676 AD] [1769-1771	[1490-1603 AD] [1611-1685 AD] [1732-1808

						AD] [1941-... AD]	AD] [1928-... AD]
IBI/1	61	Charcoa 1	Poz- 5656 4	98.03 ± 0.49	160 ± 40	[1669-1695 AD] [1725-1781 AD] [1797-1812 AD] [1839-1845 AD] [1852-1877 AD] [1916-... AD]	[1662-1895 AD] [1902-... AD]

4.3. Limekiln inventory

The specific survey aimed at quantifying and mapping the rests of old limekilns has revealed the existence of at least 50 units in the different studied areas: 18 in Aizarna, 1 in Akoa, 8 in Alkiza, 6 in Ibiri and 16 in Zizurkil (see Supplementary Materials).¹ Their conservation is generally precarious, with most items being partially or totally eroded; in some cases, only a ground alteration was documented in the place where toponymy or oral memory indicated the existence of a former limekiln. In three cases from Aizarna, Alkiza and Zizurkil, the limekilns have been restored over the last years with the impulse of local heritage associations.

¹ In Zizurkil, this work has been driven by the local association “Hernandorena” in collaboration with the municipality (Arzamendi-Berraondo, 2004; www.hernandorena.eus [consulted: 22-06-2018]).

Limekilns are typically placed close or annexed to the farmsteads, with some exceptions of units located in more peripheral positions nearby limestone outcrops. A great typological homogeneity has been observed along all three case studies (Fig. 5).

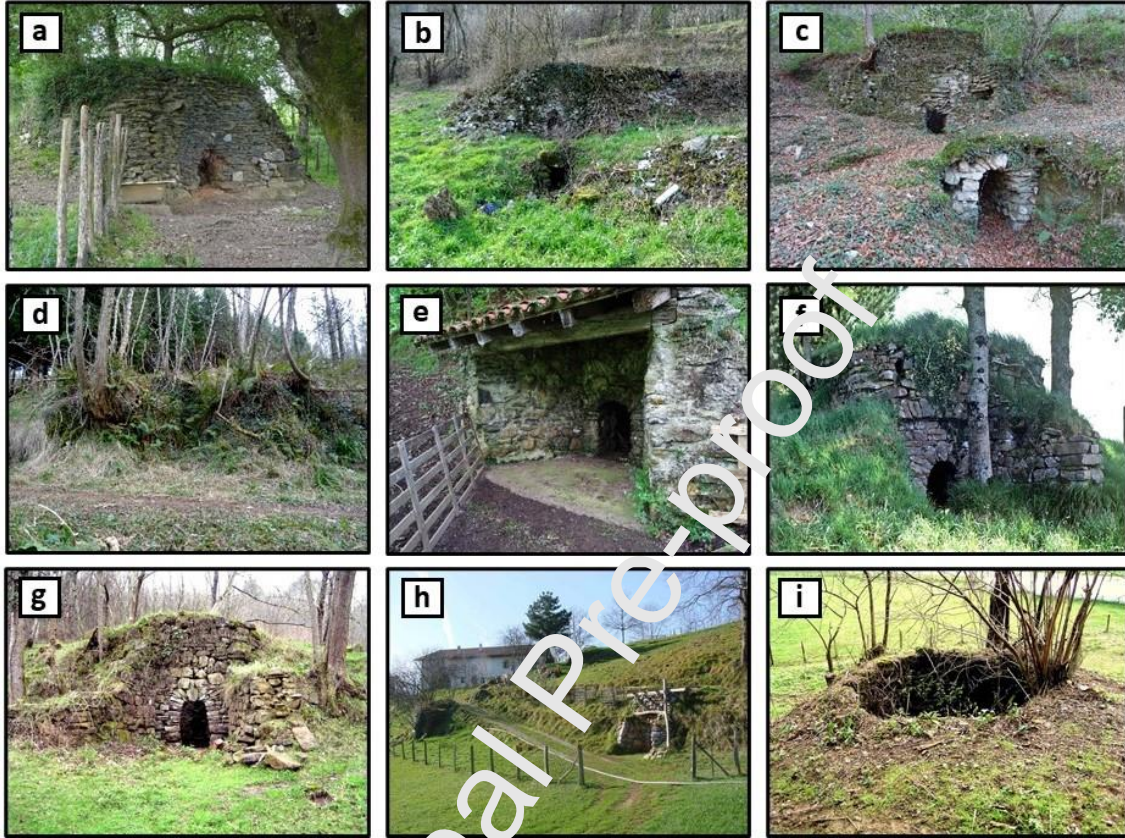


Figure 5. Examples of limekilns found in the studied villages. (a) Ezenarro-azpi, Aizarna. (b) Ibarre, Aizarna. (c) Karobietan, Aizarna. (d) San Pelaio, Aizarna. (e) Intxaurrendieta, Alkiza. (f) Ipidegi, Zizurkil*. (g) Lizardi, Zizurkil*. (h) Andrezketa, Zizurkil*. (i) Luzuriaga, Zizurkil*. *Photo credits: P. Otegi & J. Azkue – hernandorena.eus [consulted: 22-06-2018].

4.4. Limekiln archaeomagnetic dating

The NRM orthogonal demagnetization diagrams are defined by highly magnetic, univectorial normal polarity samples (Fig. 6). After removing a secondary viscous component up to 10-15 mT or 200-250 °C, the ChRM direction is univectorially defined towards the origin. Two specimens broke during the thermal demagnetization and were discarded. According to the

IRM acquisition curves and thermomagnetic curves, the main remanence carrier is magnetite with Curie temperatures around 580 °C and some high-coercivity mineral contribution (see Suppl. Info).

The archaeomagnetic direction obtained is shown in Fig. 6 (N = 12; Dec. = 356.0°; Inc. = 60.8°; $k = 138.9$; $\alpha_{95} = 3.7^\circ$). The archaeomagnetic dating was performed using a paleosecular variation curve between 1590 and 2015 AD developed with the *gufm1* (Jackson *et al.* 2000) and *igrf12* geomagnetic models (Thébaud *et al.* 2015). Archaeomagnetic dating was carried out with the Matlab dating tool of Pavón-Carrasco *et al.* (2011) at site coordinates. The probability density functions of possible dates obtained for both directional parameters (declination and inclination) at the 95% confidence level are shown in Fig. S2 in Supplementary Materials. A single age interval for the kiln's use has been obtained between 1943 and 2015 A.D. Casas and Tema (2019) indicate that adding archaeointensity cannot further constrain the directional dating results for ages from the mid-16th century onwards, which is interesting given that those analyses are time-consuming with often low success rate.

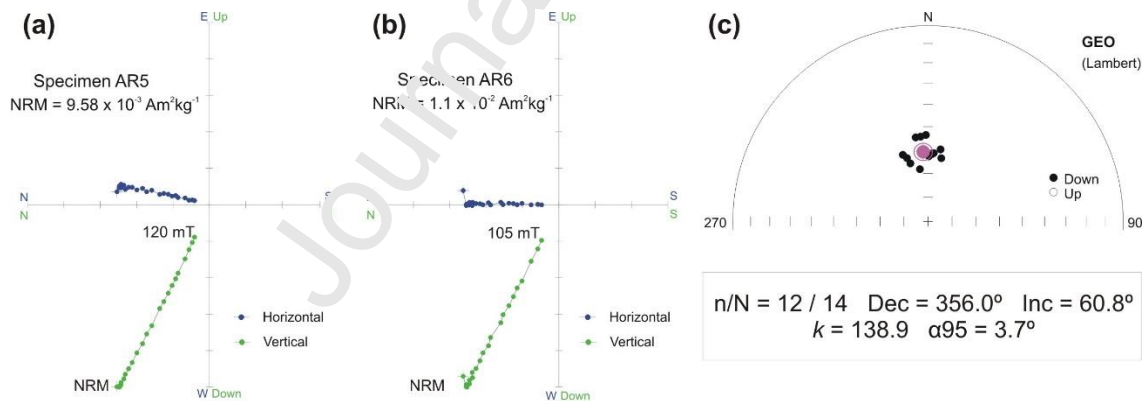


Figure 6. ((a-b) Representative orthogonal NRM demagnetization plots from Aranguren limekiln. Green (blue) symbols represent the vertical (horizontal) projections of vector endpoints. The specimen code, intensity (NRM) and main demagnetization steps are indicated. (c) Equal-area projection of all ChRM directions, with the mean direction calculated at sample level and the α_{95} confidence circle. n/N = number of specimens considered/number of specimens analysed; Dec. = declination; Inc. = inclination; k =

precision parameter; α_{95} = semi-angle of confidence.

5. Discussion

The multi-proxy study presented in this paper has explored, for the first time in the Atlantic Valleys of the Basque Country, the extent and legacy of soil liming during the Modern-period. The great visibility of past agricultural liming traces in the present-day landscape suggests that this was, to a higher or lesser extent, a common practice in all four locations here studied, reaching a surprising level of penetration for a peasant society that was, in theory, operating to a domestic level-based model of production. Three considerations are worth to be highlighted. First, the presence of Ca-enriched deposits can be regarded as a characteristic stratigraphic marker of anthropic liming on the soil records of the Atlantic Valleys of the Basque Country. Second, the chronology of these deposits induces to interpret them as a material reflection of the agricultural changes that affected the region during the Modern period. Third, these changes can be related to a wider social and economic transformation at the regional scale. In the following sections, these aspects will be discussed upon a combined consideration of the different records analysed.

5.1. Regional-scale agricultural liming

The eleven core records analysed in this paper reveal the recurrence of lime-enriched deposits in the agricultural soil records over the Atlantic valleys of the Basque Country. A clearly distinct stratigraphic unit, consisting of a Ca-rich interval related to the presence of carbonated lime remains ($\text{CaO (lime)} + \text{H}_2\text{O} + \text{CO}_2 \rightarrow \text{CaCO}_3$), has been detected in all the cores analysed, retrieved in either intensively cultivated gardens and orchards, or in cereal fields (Fig. 3c). These Ca-rich agricultural soils are stratigraphically located above the basal parent materials (fluviokarstic sediments and agricultural terrace sediments) characterised by low C_t content, acidic pH (Figs. 3, 7 and Table II) and higher relative content of lithogenic elements

(e.g., Si, Al, K, Ti and Rb, included in PC1), normally present in fine-grained siliciclastic sediments composed of clay minerals and quartz (e.g., Das & Haake, 2003; Koinig *et al.*, 2003; Jin *et al.*, 2006); Zr and Si are generally linked to coarser silt and sand size fractions (Kylander *et al.*, 2011). Atop of these Ca-rich intermediate units, there is the present-day topsoil, a dark humic horizon of variable thickness (5 to 40 cm) related to its use as artificial grassland that is geochemically characterized by a high C:N ratio and C_t content, low pH values and the presence of Pb, Zn and Br (englobed in PC1) (Figs. 3, 7 and Table II). All these elements are usually incorporated to soil organic matter (i.e., are organically bound): Pb binds to organic matter forming metal-OM complexes and Br is incorporated to OM by enzymatic bromination; Zn can be biophilic (Alloway, 1990; Huang & Jin, 2008; Atafar *et al.*, 2010). Hence, their presence reflects the humified, active-binding organic matter present in the soil. The Ca-rich horizons sometimes show traces of earlier cultivation characterized by relatively high organic content (high C_t content and C:N ratios) (Narbarte-Hernández *et al.*, 2019; 2020) and, in some cores (e.g., AIZ/1, AIZ/3 and AKU/1), it is mixed, amalgamated, with the modern organic topsoil (Fig. 3).

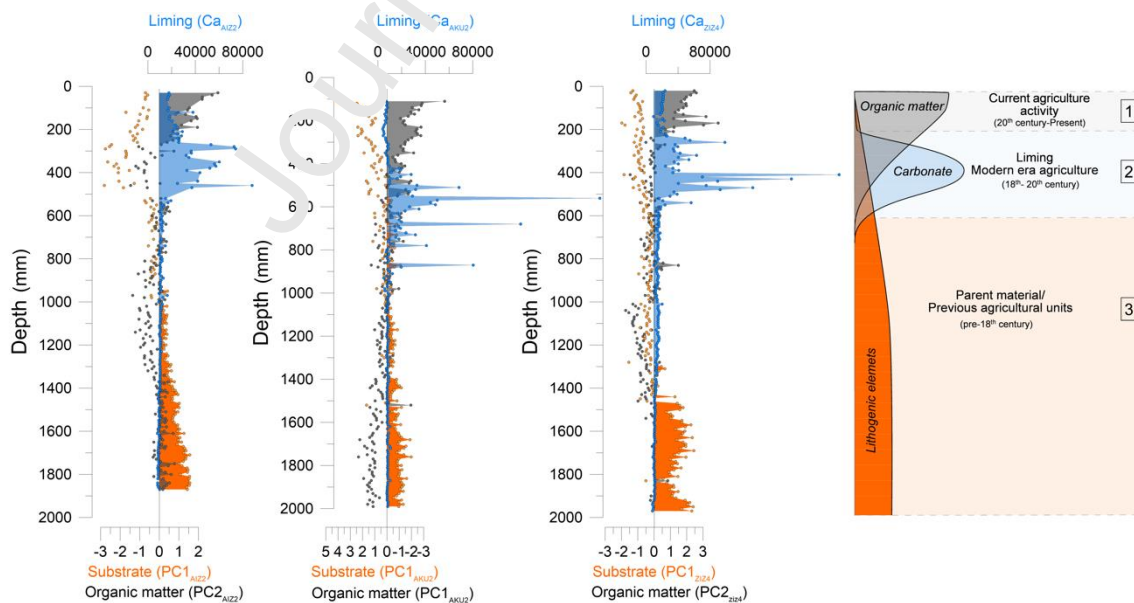


Figure 7. Summary of the main processes inferred from the geochemical analyses of Cores AKU/2, AIZ/2 and ZIZ/5.

High Ca content is interpreted as an evidence of liming. In the case of XRF analyses, the high content of Ca in the intermediate stratigraphic unit ($\geq 100,000$ cps) (Fig. 3c) contrasts with the absence or very low Ca content in the basal parent sediments and the surficial organic-rich topsoil (8,000-10,000 cps). In all cases, the incursion in Ca content in these units coincides with an increase of the organically-bound element content, while the concentration of lithogenic element content tends to decrease (Fig. 7). The increase of organically-bound element content, along with the presence of pottery fragments scattered within the Ca-rich stratigraphic unit, is an indicator for some form of additional organic fertilisation or manuring. Oral memory of the use of manure is present across the Atlantic Valleys of the Basque Country, where ferns and gorse were mixed with animal dung and then left to ferment for a variable period of time, before being spread on the fields. In a context of dispersed settlement, the presence of such extra input is a strong indicator for an intensive agricultural management, favoured by the plots' proximity to the settlements (Van der Veen, 2005). This fact is corroborated by the C and N analyses of cores AIZ/2 and AKU/2, where both C_t and C:N ratio tend to increase in the Ca-enriched levels (Fig. 7). In the absence of carbonate minerals, like in the basal sediments, measured C_t could be a good indication of total organic carbon (TOC) content; but, in the lime-enriched deposits, the C_t values should be regarded as encompassing both organic C and carbonates (e.g., Khan *et al.*, 2015; Contreras *et al.*, 2018). The results of pH analyses further confirm the impact of agricultural liming on soil acidity. The lime-enriched deposits show clearly alkaline pH levels — up to 8.59 in Core AIZ/2 and 8.68 in Core AKU/2 —, in contrast with the basal sediments, which are neutral or only slightly alkaline, 7.77 in Core AIZ/2 and 8.01 in Core AKU/2 (Fig. 7). In any case, both basal sediments and lime-enriched levels differ clearly from the present-day topsoil, whose pH levels are much more acid —6.33 in Core AIZ/2 and 5.22 in Core AKU/2—, surely due to

the massive addition of slurry related to present-day husbandry. To infer the possible correlation between pH changes and Ca and/or C_t content, a PCA was performed for C_t , N, Ca and pH data of AIZ2 and AKU2 cores (see Suppl. Info.). The results indicate that pH changes are mainly controlled by Ca presence (liming), since in PC1 (53,8% of the variance) pH (-0,62) and Ca (-0.74) vary together against C (0.97) and N (0.88). Moreover, PC2 (31,6% of the variance) points out the inverse relationship of sediment pH (0.58) and Ca content (-0.65) content. These data confirm the beneficial effect of liming in agricultural soils enhancing the alkalinity and the organic content (measured C_t) of acidic soils (e.g., Holland et al., 2018).

5.2. When and why?

Radiocarbon dating of the lime-enriched units proved highly problematic (Table I). In Core AKU/2, samples from 137, 117, 97 and 77 cm depth were dated in 2682 ± 50 BP, 1966 ± 46 BP, 1611 ± 50 BP and 1499 ± 40 BP, respectively, following a stratigraphic logic that supports the reliability of the results; all of them correspond to sediments stratigraphically located below the lime-enriched level (Fig. 3C). In Core AIZ/2, samples from 142, 97 and 82 cm depth were dated in 1545 ± 45 BP, 396 ± 41 BP and 314 ± 38 BP, respectively; the fourth sample, gathered at 47 cm depth in the lime-enriched deposit, broke this sequence with a date in 800 ± 40 BP. In Core AIZ/4, samples from 79, 76, 36 cm depth were dated as 251 ± 47 BP, 2498 ± 56 BP and “modern”. In this case, the first date indicates the age of the last sediment deposited before the lime-enriched level; the second sample was gathered in the lime-enriched deposit itself, and is again incoherent with the sequence; the last sample, gathered in the modern soil located above the lime-enriched deposit, suggests a very young age for this level. Finally, in Core IBI/1, the lime-containing unit at 61 cm depth yielded an age of 160 ± 40 BP.

With these results, radiocarbon ages from lime-enriched deposits must be considered very

cautiously and are sometimes unreliable, probably due to modern organic matter mixing and incipient Suess effect during the 18th-19th centuries. Hence, only an approximation can be done to the age of these deposits, based on the radiocarbon ages of the lime-enriched or their underlying deposits; Core AKU/2 points to a vague postclassical age, while Cores AIZ/2 and ZIZ/4 indicate that liming may have begun *after* the 16th or the 17th centuries; finally, in Core IBI/1 the lime-containing unit is dated in the 18th century. Therefore, the 18th century seems the most likely date for the beginning of liming. Regarding the date of end of this practice, the modern age provided by the uppermost sample from ZIZ/4 core suggests that it did not end before 1950.

These dates are overall coherent with the results of the archaeological excavation driven in the emplacement of the core AIZ/3, which has brought to light abundant fragments of pottery included in the lime-enriched level — interpreted as the rests of domestic waste used as manure in the rectory house's garden of Airarna. The typological analysis of those materials suggests a period of use shifting between the late-17th and the mid-20th centuries (Narbarte-Hernandez *et al.* 2019).

The analysis of the core records therefore points to a change in the systems of agricultural production of the Atlantic Valleys of the Basque Country having occurred during the Modern period — 17th or, more likely, 18th to 20th centuries. The introduction of American crops, such as maize (*Zea mays*), in this period might have permitted an expansion of the cultivated surface, but also an intensification of the production. This was operated by the concentration of soil nutrients in the agricultural fields, by transferring increasing amounts of biomass from shrublands — ferns and bracken being used as a bedding for the fabrication of manure — and balancing the resulting soil acidification with the regular addition of lime (Olarieta *et al.*, 2019).

5.3. Liming for an intensive polyculture

The introduction of American crops occurred in a short period of time and led to a radical transformation in the physiognomy of these villages (e.g., Narbarte-Hernandez, 2020). This change was coetaneous with a sustained demographic progression; in Aizarna and Akoa, the number of households increased from 53 in 1543 (ZUA/1) to 82 in 1776 (ZUA/2), resulting in a subdivision of the local space into smaller landholdings. This increase of the anthropic pressure on the land might have enhanced a trend towards intensive polyculture, with multiple crops being alternated in the same plots, allowing an increase in yield per unit of area but also requiring large inputs of manure and labour to balance the lack of fallow.

There is archaeological and documentary evidence that supports this interpretation. On the one hand, a new model of intensive biennial rotation on agricultural fields, formed by a combination of wheat (*Triticum aestivum*), maize (*Zea mays*), and forages or tubers, is described from the mid-17th century onwards in the whole regional context:

They say that, when a field has provided wheat & once they have collected it, they sow maize at the end of August, which they harvest shortly after (Bertaut F., lord of Fréauville, 1669 [BnF/1])

The cycle began in spring, with maize being sown in combination with another crop of American origin, common beans (*Phaseolus vulgaris*). Once these products were harvested in autumn, wheat was sown in the same plots, then harvested in summer. The last products of the cycle were forages — *Trifolium incarnatum* or *Trigonella foenum-graecum* being the most commonly cited — and tubers like turnip (*Brassica rapa*) or, later, potato (*Solanum tuberosum*). These labours were generally used to feed the livestock necessary as a track force and to produce manure (Caro-Baroja, 1973). By the end of the 18th century, this system seems to have been clearly widespread across the Atlantic Valleys of the Basque Country:

The order seems to be maize, wheat, turnip, maize, etc. [...] Not even a vineyard or a meadow; turnip and maize, and nothing else (Jovellanos, G.M., 1791)

[BA/1]).

This is, in fact, the kind of agricultural landscape described by the Geographical-Historical Dictionary of Spain for the Atlantic Valleys of the Basque Country, including the locations here addressed (Real Academia de la Historia, 1802 [KM/5]). The confection of the dictionary was based on direct local information. For instance, the report prepared to this purpose by Ignacio de Errasti, secretary of the municipality of Zestoa, is conserved in the local archive and describes a diversified agroecosystem where wheat and maize were the main products, complemented with “all kinds of legumes and vegetables”, as well as fruits and animal products (ZUA/6).

On the other hand, the onset of these new model of crop rotation coincides with the oldest documentary records of agricultural liming. In Aizarna and Akoa, several limekilns are mentioned in archival sources, as early as 1705 (ZUA/2; ZUA/3; ZUA/4; ZUA/5), as relevant elements of the local landscape both acting as visual landmarks and delimiting the plots of different owners. Similar dates are suggested by the agronomical reports prepared by local notables like J.I. de Armasa, priest of Beintza-Labaien in the Bidasoa Valley, Navarre (Caro-Baroja, 1973: 35-36), and also by the enlightened — and aristocratic — Royal Basque Society of Friends of the Country (RBSFC) in the second half of the 18th century:

[Basque peasants] started to use lime as a fertiliser only one hundred years ago, but the success of those first experiments has contributed to expand it across the whole Country (Real Sociedad Bascongada de Amigos del País, 1768 [KM/1]).

Agricultural liming therefore was introduced in the Basque Country in relation to the development of a new crop cycle, and expanded rapidly over the 18th century — which is contemporary with other Atlantic regions of northern Spain, like Asturias (García López del Vallado, 2009).

Most local informants refer that the domestic production and use of agricultural lime was

abandoned between the 1930s and the 1960s and replaced with industrial fertilisers, which is overall coherent with the radiocarbon age of the modern topsoil of Core ZIZ/4. Further evidence about this is provided by the archaeomagnetic study carried out on a limekiln belonging to the farmhouse Aranguren, in Aizarna. The archaeomagnetic dating indicates that its last use took place between 1943 and 2015 AD at the 95% confidence level (Fig. S2 in Supplementary Materials), in line with the youngest available radiocarbon ages and oral sources. Interestingly, this study also provides evidence that archaeomagnetism can be systematically applied to date the last use of these local limekilns.

Yet the historical process by which agricultural liming became such an important management practice in Basque farming in the 18th century remains unclear. Even if this was a period in which many of the modern agronomic innovations that were being developed in different European countries were discussed by intellectual societies like the RBSFC, their introduction was not properly promoted on the basis of a formal scientific program (Berriochoa-Azcárate, 2014). Indeed, the reports prepared by the RBSFC in these decades typically describe the agrarian landscapes of the Basque Country as a “garden”, developed by the local peasants’ “care and experience”, where domestic agricultural production was to a high extent founded on the empiric knowledge of the producers themselves (Real Sociedad Bascongada de Amigos del País, 1777-1779 [KM/2]; 1780-1782 [KM/3]; 1791 [KM/4]).

The widespread presence of old limekilns in the landscapes of Aizarna and Akoa, Alkiza, Ibiri and Zizurkil supports the idea of production, management and application of agricultural lime being performed on a domestic basis. Most limekilns are placed close or even annexed to the farmsteads, being one of their main productive structures and their importance is highlighted by the existence of numerous micro-toponyms referring to limekilns (*karobi* in Basque) in different points of the local landscapes: Ka(ro)bialde and Ka(ro)bieta in Aizarna, Karobialdea in both Ibiri and Zizurkil — all of them still recognisable by local informants.

Typologically, most of these limekilns correspond to the “intermittent” calcination typology that was widespread in most European regions prior to the invention of the continuous burning-kilns (cf. Diderot & D’Alembert, 1751-1772 [BnF/2]; Fourcroy de Ramecourt, 1761 [BnF/3]; Vicat, 1828 [BnF/4]). However, the great typological homogeneity observed across the different focus locations suggests a specialised fabrication, following a pre-established — maybe imported? — pattern, although there is no direct evidence of this.

Hence, the domestic production and use of agricultural lime needs to be explained emphasising the local factors; first of all, by the availability of raw materials. Considering that limestone constitutes the substrate of much of the Atlantic Valleys of the Basque Country (EVE, 2018), this can be considered a factor that encouraged the adoption of liming in the region, the low costs of extraction and transport making it simple to manage at the domestic or local scale. In fact, Ignacio Iztueta’s manuscript report from 1875 stated, about Zizurkil, that

limestone is so easy to find in the mountains of this village that, anywhere, a limekiln can be excavated close to a field so as to burn as much lime as necessary to fertilise the soils (Iztueta, 1875; KM/6).

Regarding fuels, wood is the most commonly cited by local informants, in particular the least profitable parts of the beech (*Fagus sylvatica*), like branches and knots. Other elements were used as well, especially small trees like hazel (*Corylus avellana*) (Garmendia-Larrañaga, 2007), or shrubs like gorse (*Ulex europaeus*) and heather (*Calluna vulgaris*) (Abella, 2016; Olarieta *et al.*, 2019).

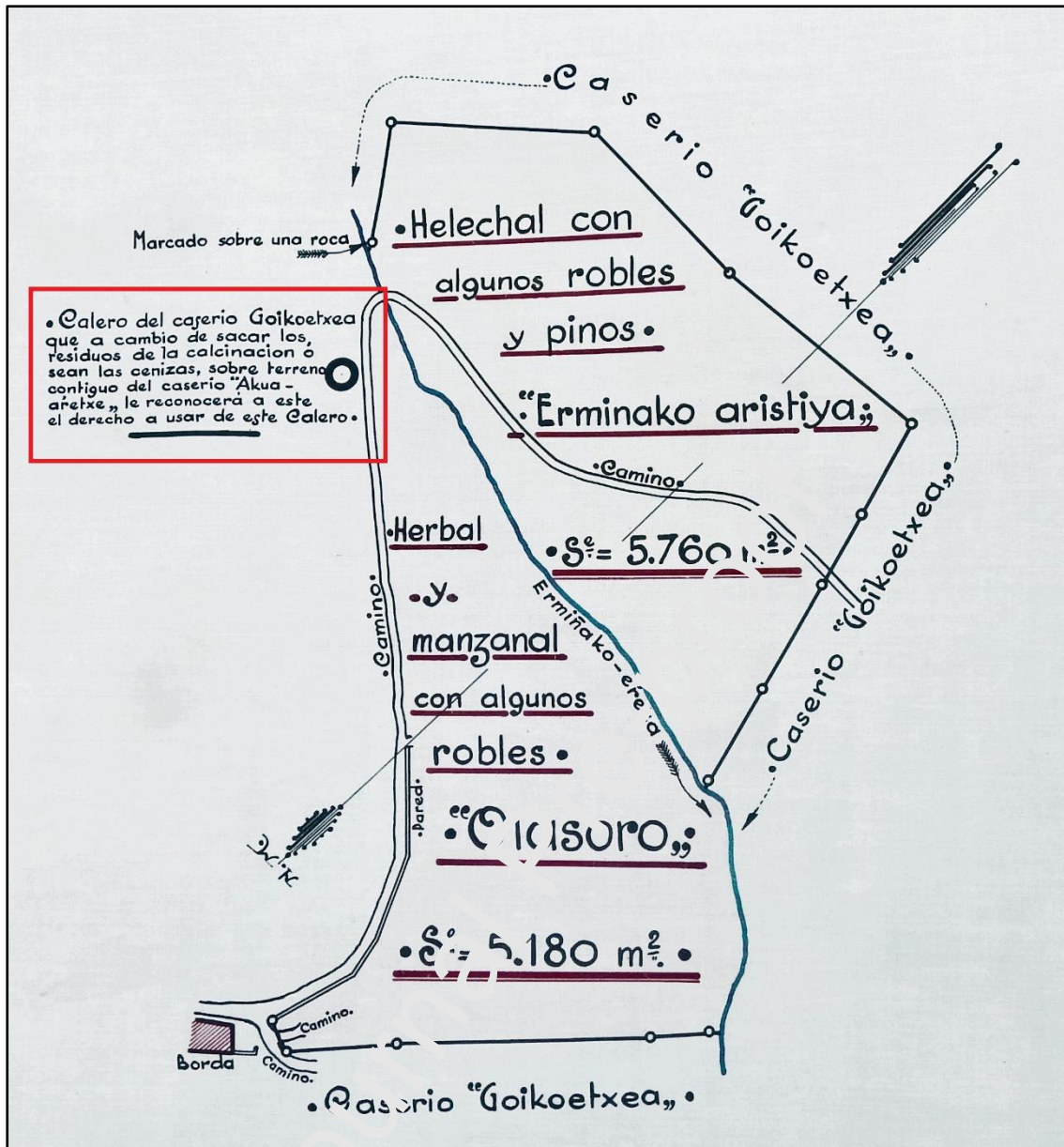


Figure 8. Detail of a land demarcation in 1934, with an arrangement between the proprietaries of two farmstead for the management of a limekiln: "calero del caserío Goikoetxea". The document belongs to the proprietary of the house Akoarretxe (Akoa).

The amount and intensity of the labour required to burn a limekiln for several days was another limiting factor for this practice. Larramendi (1882) mentions nine years as the common interval for limekiln burning in 18th-century Gipuzkoa, while Olazabal (1857) speaks of two or three years in 19th-century Biscay; but there are records of yearly

frequencies in the early 19th-century Bidasoa Valley as well (Caro-Baroja, 1944). In any case, aged informants from Aizarna and Alkiza consulted for this work, who remembered burning lime in their farmsteads during their infancy, agreed that this was quite an exceptional event that would take place no more often than once every few years, involving some form of cooperation with individuals from outside the domestic unit. As stated also by ethnographic literature, this collective action was commonly performed on the basis of relativeness or neighbourhood (Lizarralde, 1927; Llanos, 2009). For example, the proprietary of the house Akoarretxe in Akoa conserves an agreement for the demarcation of some plots, signed in 1934, where the limekiln of the house Goikoetxea is depicted stating that

the ashes resulting from its calcination will be discarded through the adjacent plot, which belongs to the neighbour house Akoarretxe; in exchange, the latter will have the right to use this limekiln (fig. 2)

Lime was therefore a valuable resource of the modern-period Basque domestic economies. In fact, the value of lime production could eventually go far beyond its agricultural application, especially when the impact of industrialisation enhanced an increase of demand for lime during the 19th century. In the case of Aizarna and Akoa, for example, this demand was canalised since 1838 through the creation of a flourishing industry of hydraulic lime, the so-called “Zumaya cement”: limestone continued to be burnt in the local limekilns and used with agricultural purposes, but one (increasing) part of the production could also be destined to be processed in the forges of Iraeta, Lili and Altzolarats — soon replaced by modern industrial facilities in the nearby towns of Zumaia and Zestoa (Varas *et al.*, 2007; Urdangarin Altuna & Izaga Renier, 2020; ZIIZ, 2020).

Summarising, the spreading of liming as a regular practice of agricultural management can be regarded as a reflection of the social and economic transformations that took place in the Atlantic valleys of the Basque Country during the Modern period. As emerges from the

research presented in this paper, this practice was key to the establishment of a domestic-based model of intensive polyculture, which gave origin to the ‘traditional’ peasant society of the region and eventually laid the foundations of the rural landscapes that have survived until present. This evidence provides new insights on the complexity of the agricultural changes that defined Modern-period Europe, permitting to overcome monocausal explanations and opening new paths of research on their long-term social and environmental effects.

6. Conclusion

The widespread use of lime as a regular soil amendment appears to be one of the most relevant material expressions of Modern-period agricultural change in the Atlantic Valleys of the Basque Country. This practice is attested since the early 18th century in documentary records, a date confirmed by the systematic presence of lime-enriched deposits of the same age in agricultural soil records. The spreading of this kind of practice reflects an intensification in the forms of agricultural management, in the framework of a new relationship between land and labour that emerged, as in many other Atlantic regions of Europe, after the introduction of American crops such, as maize, and the establishment of an intensive polyculture. Lime was produced and applied on a domestic basis, as witnessed by the many limekilns interspersed within the local rural landscapes, as well as the oral memory that is still conserved in the local communities. Continuous liming for more than 200 years exerted a deep and durable impact in the soils, which is clearly recognisable in a characteristic lime-enriched stratum that the different records analysed show to be coherent over several different local contexts, and can therefore be considered as a diagnostic chronological marker in the agricultural soils of this region.

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Declaration of competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Highlights

- Agricultural liming was a common practice in traditional Basque farming.
- Multidisciplinary research was used to address the expansion of liming.
- Results provide new insights on the complexity of Modern agricultural changes.
- Liming was a key factor in the implantation of ‘traditional’ polyculture.
- Effects of agricultural liming may be longstanding and affect present-day soils.

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