Journal of Archaeological Science 67 (2016) 43–51

Contents lists available at ScienceDirect

Journal of Archaeological Science

journal homepage: http://www.elsevier.com/locate/jas

Chemical compositional changes in archaeological human bones due to diagenesis: Type of bone vs soil environment

Olalla López-Costas [a, b, c, *], Óscar Lantes-Suárez [d], Antonio Martínez Cortizas [e]

[a] Group Earth System Sciences, Department of Geography. Fac. Xeografía e Historia, Universidade de Santiago de Compostela, Santiago de Compostela, 15782, Spain
[b] Archaeological Research Laboratory, Stockholm University, Wallenberglaboratoriet, SE-10691 Stockholm, Sweden
[c] Laboratory of Anthropology, Department of Legal Medicine, Toxicology and Physical Anthropology, Faculty of Medicine, University of Granada, Granada 18012, Spain
[d] Unidade de Arqueometría, RIAIDT, Universidad de Santiago de Compostela, Spain
[e] Departamento de Edafoloxía e Química Agrícola, Fac. Bioloxía, Universidade de Santiago de Compostela, Campus Vida, 15782 Santiago de Compostela, Spain

ARTICLE INFO

Article history:
Received 20 April 2015
Received in revised form
18 January 2016
Accepted 6 February 2016
Available online xxx

Keywords:
Elemental composition
Lead
Post-mortem
Taphonomy
Roman
post-Roman
Principal component analysis

ABSTRACT

Diagenesis in human remains is a subject of growing interest due to the increase in bone chemical studies to reconstruct *pre-* and *post-mortem* features in archaeological and forensic sciences. The efforts made during the last decades have solidified our understanding of diagenetic processes; however, their high complexity demands more research to address them empirically, specifically considering factors such as types of soil substratum and skeletal element. In this work, a geochemical study of human remains from the archaeological site of A Lanzada (NW Spain) is performed to understand diagenesis (i.e. chemical alteration) and life environmental exposure. Three types of bone (thoracic, long and cranial) from 30 skeletons of two periods (9 Roman, 21 post-Roman) were analysed by X-ray fluorescence. Bones were recovered from burials located in slightly alkaline (Haplic Arenosol (calcaric)) and acidic (Cambic Umbrisol (humic)) soils. Principal components analysis was applied to extract the main chemical signatures, and analysis of variance to determine the influence of different factors.

Bone composition was characterized by four chemical signals related to: i) alteration of bone bioapatite; ii) metal sorption from the soil solution; iii) presence of fine (silt–clay) soil particles; and iv) lead incorporation. Thoracic bones were found to be more sensitive to diagenesis and the burial environment; long bones and crania presented a similar response. Skeletons buried in the acidic soil were significantly poorly preserved. Lead content was higher in bones of the Roman period, which seems to be related to *pre-mortem* conditions. Previous investigations on palaeopollution in NW Spain enable us to hypothesize that Roman individuals may have been subjected to a high exposure of Pb due to elevated atmospheric metal contamination.

1. Introduction

Most osteological studies in both archaeological and forensic contexts concern skeletons that have been part of the soil for a variable period of time. The composition of buried bones is the result of changes that happened before (*pre-mortem* conformation) and after (diagenesis, *post-mortem* degradation) the inhumation took place. Defined as the *post-mortem* bio-chemical alterations of bones, diagenetic processes are highly heterogeneous due to post-depositional time, taphonomy and burial environment (Hedges, 2002). The study of diagenesis has been a popular topic since the 1980s and an impressive number of investigations has been performed from a theoretical, actualistic and empirical point of view, including a EU project on bone deterioration (ENV4-CT98-0712). This research demonstrated that diagenesis can provide valuable information on the taphonomic history of bones (e.g. Bocherens et al., 2008; Maurer et al., 2014), the characteristics of the grave (e.g. Salesse et al., 2014; Müller et al., 2011), the soil microorganisms (Maurer et al., 2014, e.g. Salesse et al., 2014; Jans et al., 2004;

* Corresponding author. Group Earth System Sciences, Department of Geography. Fac. Xeografía e Historia, Universidade de Santiago de Compostela, Santiago de Compostela, 15782, Spain.
E-mail addresses: olallalc@gmail.com, olalla.lopez@usc.es (O. López-Costas), oscar.lantes@usc.es (Ó. Lantes-Suárez), antonio.martinez.cortizas@usc.es (A. Martínez Cortizas).

http://dx.doi.org/10.1016/j.jas.2016.02.001
0305-4403/

Collins et al., 2002), and it has also been used in forensic cases to provide clues about burial location (González-Rodríguez and Fowler, 2013). Geochemical studies focused on *pre-mortem* conditions, e.g. diet or mobility, likewise consider the diagenetic signals in order to discriminate them from the pre-depositional ones (among others Lambert et al., 1985; Price et al., 1992; Bentley, 2006; Nelson et al., 1986). Indeed, the recent advances on studies that recover the life characteristics from the bone tissues (see an overview in Ambrose and Krigbaum, 2003) have increased the interest in recognizing and comprehending the diagenesis of bones.

In both *pre*-and *post-mortem* phases, bone composition changes by incorporating and releasing elements. The skeleton of a living individual is in constant turnover adding new elements until the individual's demise. Food and drink present a geochemical signal that is transferred to the human body, with appropriate fractionations (DeNiro and Epstein, 1978; Schwarcz, 1991). Additionally, humans incorporate elements during life by exposure, i.e. inhalation. The *post-mortem* chemical alterations start soon after death, stimulated by bacterial action (Pfretzschner, 2006). Once the individual is buried, it is necessary to consider the interaction between skeleton and soil or diagenesis, which is influenced by local geology (Maurer et al., 2014; Nielsen-Marsh et al., 2007; Nielsen-Marsh and Hedges, 2000; Hedges et al., 1995) since the body incorporates elements by sorption or particle-attachment, and also releases them by bone alteration. The diagenetic processes are deeply complex, involving a good number of physical, chemical, histological and mechanical changes at different scales (Hedges, 2002; Stathopoulou et al., 2008).

Bone composition has been widely explored in Anthropology to address different characteristics of bones from archaeological sites (see a summary in Burton, 2008). Specific elements such as Sr, Br, Ba or Zn were used as indicators of ancient diet (among others Arnay-de-la-Rosa et al., 2009; Price et al., 1985; Brown, 1974; Szostek et al., 2009; Velasco-Vázquez et al., 1997; Dolphin et al., 2013). *Post-mortem* alteration within the soil environment is also well explored (among others Jans et al., 2004; Nielsen-Marsh et al., 2007; Nielsen-Marsh and Hedges, 2000; Stathopoulou et al., 2008; Zapata et al., 2006; Kyle, 1986). Many studies about bone composition compare affected with less affected ones in order to assess the importance of diagenesis. As early as 1993, Edward and Benfer (1993) indicated that one of the most obvious ways to proceed is by comparing types of bones more and less susceptible to alteration (e.g. long bones vs. ribs) (among others Carvalho and Marques, 2008; Edward and Benfer, 1993; Lambert et al., 1982). In contrast, other works prefer to focus in just one more (e.g. vertebra, János et al., 2011) or less (e.g. femur, Buikstra, et al., 1989; Zapata et al., 2006) fragile type of element, which limits the possible intra-skeletal comparisons. Despite this relative abundance of studies, many works are focused on correlating or calculating ratios (e.g. Sr/Ca or Ca/P) between chemical elements in order to identify *pre-mortem* dietary signals from the *post-mortem* soil "contamination"; although they rarely include multielement statistical analyses or discuss the diagenetic processes in detail. Previous research, like the one developed by Buikstra et al. (1989), wisely approached the study of bone chemical composition using multivariate analysis (PCA on the correlation matrix and varimax rotation). As indicated by these authors, this type of analysis enables a much better detection of the chemical bone signature and the interpretation of effects of *pre-* and *post-mortem* processes. However, they include little discussion about how the diagenetic processes took place.

Several authors (among others Burton, 2008; Zapata et al., 2006; Wilson and Pollard, 2002) have recently pointed out the need to understand how diagenesis works and how different environments operate, in order to reconstruct bone composition. To address this topic, we applied a non-destructive technique, X-ray fluorescence spectrometry (XRF), and multivariate statistics (PCA) to analyze and process the multielemental signals together including samples from different types of bones, chronological-cultural periods and two different soil environments.

In this study, the geochemical analysis of an ancient human skeletal collection, with estimated age and sex, has been performed. There are two objectives: first, to ascertain the variability in the intensity of chemical diagenetic change (sorption/release of elements), according to different types of bone; second, to comprehend the influence of soil environment in bone chemical changes and discuss the *post-mortem* composition in order to differentiate the sources related with *pre-mortem* incorporation.

2. Material and methods

The study was carried out on human bones from, relatively well preserved, skeletons found in the site of A Lanzada (Noalla, Sanxenxo) in the Spanish province of Pontevedra (42°25′46N; 8°52′25W). A Lanzada is a well-known archaeological site, which has been excavated since 1949 in several campaigns during the 1960s, 1970s and 2010 (among others Blanco Freijeiro et al., 1961, Fariña Busto, 1975). The works have discovered a wide variety of archaeological remains from the Bronze Age onwards, including an extensive necropolis with two funerary areas, one from Roman times (2nd to 4th centuries AD) and the other from post-Roman times or Late Antiquity (4th to 6th century AD) (López-Costas, 2015). The type of burials, the presence/absence of grave goods, the grave orientation and the body position were characteristically different between the two funerary areas (López-Costas, 2015; López-Costas, 2012). Three AMS radiocarbon datings support this division (López-Costas, 2012).

A total of 59 inhumation and two cremation burials have been described (López-Costas, 2015). The cremations were not included in this study, since no osteological material was collected from them (Blanco Freijeiro et al., 1961). The collection of the buried skeletal assemblage was more thorough, with a total of 84 identified skeletons, 38 Roman, 40 post-Roman and 6 undetermined. A complete osteological and paleopathological analysis of the remains was performed and published elsewhere (López-Costas, 2012). Although a large number of Roman burials were discovered during the 1960s campaigns, the skeletons were only partially collected (long bones and crania), which limits the available sample -particularly for thoracic bones- for this period. A selection of this assemblage was sampled, comprising 30 skeletons (79 bones): 9 from Roman and 21 from post-Roman period (Table 1). One of the samples (245), which may be Early Medieval, has been included in

Table 1
Distribution of the 79 samples (30 individuals) according to the type of bone, buried substratum, period, sex and age of the skeleton. Bone type: 1, thoracic bone; 2, long bone; 3, cranial bone. Age group key: Subadult (13–20 years), young adult (20–35 years); middle adult (35–50 years); old adult (50 + years).

	Type of skeletal element		
	Type 1	Type 2	Type 3
Nº id	24	30	25
Umbrisol	5	7	4
Arenosol	19	23	21
Roman	3	9	6
post-Roman	21	21	19
Male	9	12	9
Female	7	10	8
Subadult	8	8	8
Young adult	8	11	9
Middle adult	6	7	6
Old adult	2	2	1