



Human bones tell the story of atmospheric mercury and lead exposure at the edge of Roman World

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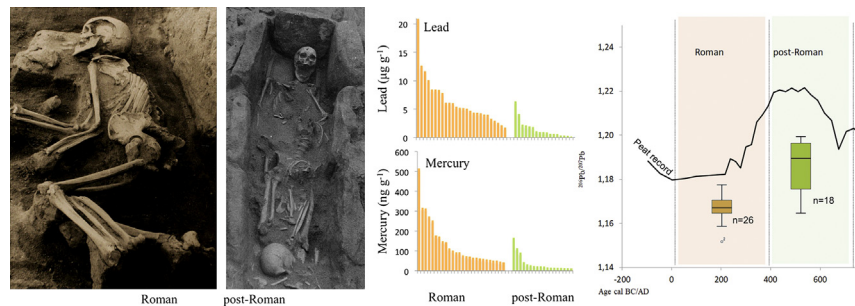
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HIGHLIGHTS

- The novelty of our work relies on combining [Hg], [Pb] and Pb isotopes in human bone.
- [Hg] was on average 3.5-fold greater in Roman compare to post-Roman inhabitants.
- Atmospheric Pb pollution contributed ~57% in Roman and ~24% in post-Roman skeletons.
- Lead bone records mirror atmospheric Pb pollution recorded in a local peat archive.
- Bones can serve as archives of metal pollution and complement other natural archives.

GRAPHICAL ABSTRACT



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ABSTRACT

Atmospheric metal pollution is a major health concern whose roots pre-date industrialization. This study pertains the analyses of ancient human skeletons and compares them with natural archives to trace historical environmental exposure at the edge of the Roman Empire in NW Iberia. The novelty of our approach relies on the combination of mercury, lead and lead isotopes. We found over a 700-year period that rural Romans incorporated two times more mercury and lead into their bones than post-Romans inhabiting the same site, independent of sex or age. Atmospheric pollution sources contributed on average 57% (peaking at 85%) of the total lead incorporated into the bones in Roman times, which decreased to 24% after the decline of Rome. These values and accompanying changes in lead isotopic composition mirror changes in atmospheric Pb deposition recorded in local peatlands. Thus, skeletons are a time-transgressive archive reflecting contaminant exposure.

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1. Introduction

Lead and mercury are among the trace metals whose natural cycles have been most heavily affected by human activities, dramatically increasing their emission to the atmosphere, soils and water bodies since prehistory (Nriagu, 1979). Both metals can produce harmful effects on ecosystems and human health. Exposure, even at low levels, may cause complex and serious health effects, including developmental delays, cancer, brain and kidney impairment, cardiovascular functioning and even death (Järup, 2003; WHO, 2004; WHO, 2008). Infants born or breastfed to mothers with high lead or mercury levels are the most susceptible to develop multi-organ complications (Dorea, 2004; Gulson et al., 2003). Lead accumulates in the skeleton (Schroede and Tipton, 1968) and is particularly problematic when released during times of physiological stress and bone turnover (Silbergeld, 1991). Mercury targets the central nervous system (Holmes et al., 2009). Synergistic effects of both mercury and lead appear even with low exposure, e.g., exacerbating hypertension, pulse pressure and neurodegenerative diseases, by becoming cardiovascular and neurological toxicants (Monnet-Tschudi et al., 2006; Wildemann et al., 2015). Today, lead and mercury toxicity is of global public concern and in consequence, their environmental and physiological levels are monitored in different populations by international bodies (WHO, 2004; WHO, 2008).

Studies from environmental archives, such as polar ice, lake sediments and peat deposits (Hong et al., 1994; Murozumi et al., 1969; Renberg et al., 1994; Shotyky et al., 1998), place past human activities into a temporal perspective and provide evidence for widespread preindustrial atmospheric metal pollution. In Western Europe, the earliest evidence dates back to the very beginning of mining and metallurgy ca. 3000 BCE, as found in peat records from NW Spain (Martínez Cortizas et al., 2016). The common pattern of pollution observed in much of Europe shows a climax during the Roman period before a dramatic decrease by the 5th century CE only to resume centuries later during Medieval times (Kylander et al., 2005; McConnell et al., 2018; Mighall et al., 2002; Renberg et al., 1994; Shotyky et al., 1998; Weiss et al., 1997). Iberia is well known as an important centre of mining during Roman rule, producing approximately 40% of the total Empire lead production - ~50,000 tons (Nriagu, 1983). Early mercury pollution released to the atmosphere during the Late Iron Age (Martínez Cortizas et al., 1999) and drained into coastal waters (e.g. Serrano et al., 2013), has also been detected in Iberia. Thus, lead and mercury were widely and deliberately exploited by humans in preindustrial times, unaware of their hazardous nature, as tools, in kitchen goods and as medicines. This was particularly true in Roman times when, for example, lead pipes may have compromised urban public health such as in Imperial Rome (Delile et al., 2014).

Many studies have analysed the lead content of human skeletal remains (e.g. Aufderheide et al., 1992; Drasch, 1982; González-Reimers et al., 2003; Grandjean and Patterson, 1988; Jarcho, 1964; Martínez-García et al., 2005; Millard et al., 2014; Molleson et al., 1986; Montgomery et al., 2010), which have clearly established that individuals from prehistoric (Bronze Age, Iron Age) and Early Medieval populations typically had very low metal contents, while Roman, later Medieval and post-Medieval populations had much higher concentrations. Only a handful of these studies used lead isotopes (Bower et al., 2005; Budd et al., 2004; Chiaradia et al., 2003; Melton et al., 2010; Millard et al., 2014; Molleson et al., 1986; Montgomery et al., 2010) and focused mainly on *pre-mortem* mobility or as a way to distinguish local and non-local individuals. In terms of mercury, the number of studies on archaeological human remains is surprisingly low (Emslie et al., 2019; Emslie et al., 2015; Rasmussen et al., 2008; Rasmussen et al., 2017; Rasmussen et al., 2015; Stadlbauer et al., 2007; Yamada et al., 1995), and they focused upon medication using mercury-containing remedies (Rasmussen et al., 2008; Stadlbauer et al., 2007), postmortem burial decoration of the body (Yamada et al., 1995) or cinnabar exposure (Emslie et al., 2019; Emslie et al., 2015). Regardless of

the metal under study, elevated total contents of lead and mercury were mainly attributed to high metal exposure in life (i.e., cultural factors), while variations in isotopic signals were related mostly to local geology (i.e. mobility). However, there is an ongoing, unresolved debate regarding the exclusive dependence of the isotopic signals on local geology (e.g. Montgomery et al., 2010); that is, would a cohort of individuals who lived in the same area but were exposed to different levels of pollution present similar isotopic signals or not?

1.1. Lead and mercury in A Lanzada bones

Our research concerns a skeletal collection recovered at the archaeological necropolis of A Lanzada (NW Spain, 42°25'46"N 8°52'25"W, Fig. SM_1), which presents two consecutive funerary areas: a Roman (AD 1st to 5th centuries - Fig. 1) and an Early Medieval/post-Roman (AD 5th to 7th centuries - Fig. 1) (López-Costas, 2015; López-Costas and Müldner, 2016). All well-preserved individuals from both sexes and different ages at death were included in the present study, 26 from Roman and 18 from post-Roman times (Table S1). We sampled non-pathological long bone diaphysis (mainly femur; cortical bone) for our study.

This cemetery provided us with an extraordinary opportunity to examine metal exposure in a local community: a 700-year timeframe that portrays a rural population, isolated from luxury items containing lead or mercury, whose individuals probably lived and were buried in the same place. Local soil contains very low lead and mercury concentrations ($<10 \mu\text{g g}^{-1}$ and $<5 \text{ ng g}^{-1}$, respectively). The necropolis is remote from any urban areas ($<100 \text{ km}$) that had a significant tradition of lead and mercury-made items and, indeed, no such items have been found at the site. As a result, A Lanzada's citizens are assumed to have had a similar exposure to natural lead sources, but not to natural geogenic mercury because no mercury-bearing mineral phases have been found in local soils and rocks. We previously established that both Roman and post-Roman skeletons experienced similar intensity in bone diagenesis with no significant differences in their elemental composition regarding this period (except for lead) (López-Costas et al., 2016). They also had the same preservation of the organic bone fraction (i.e. collagen; see Kaal et al., 2016; López-Costas and Müldner, 2016). This is important, as poor collagen preservation could cause spurious increases on lead content, because lead is mainly associated to bioapatite. The degree of collagen preservation in our samples indicates that lead incorporation into bones responded to a *pre-mortem* cause rather than diagenesis (this is also supported by the fact that they were buried at the same site, see Fig. 1). Bacterial activity could have remodeled bone bioapatite, but these processes would have affected skeletons from both periods equally. The latter is constrained here by combining the two metals (lead and mercury), which have different sources and atmospheric cycles, and by the lead isotopic composition, which enables us to estimate the contribution of pollution-derived lead to the total lead content (using isotopic mixing models). In addition, we compare the bone data with a detailed regional (NW Spain) atmospheric lead isotope curve (Kylander et al., 2005) covering the last 8000 years, which provides a context for the trends shown by the human remains.

2. Methods

Lead concentrations and lead isotopic composition were determined by ICP-MS, and MC-ICP-MS, respectively, while mercury concentrations were determined using a Milestone DMA-80 analyzer.

2.1. Mercury analysis

Samples (cortical bone) were dried at room temperature (25°) until constant weight, milled and homogenized. The mercury content in bone and associated sediment samples was determined by means of cold-vapour atomic absorption spectroscopy (CV-AAS)

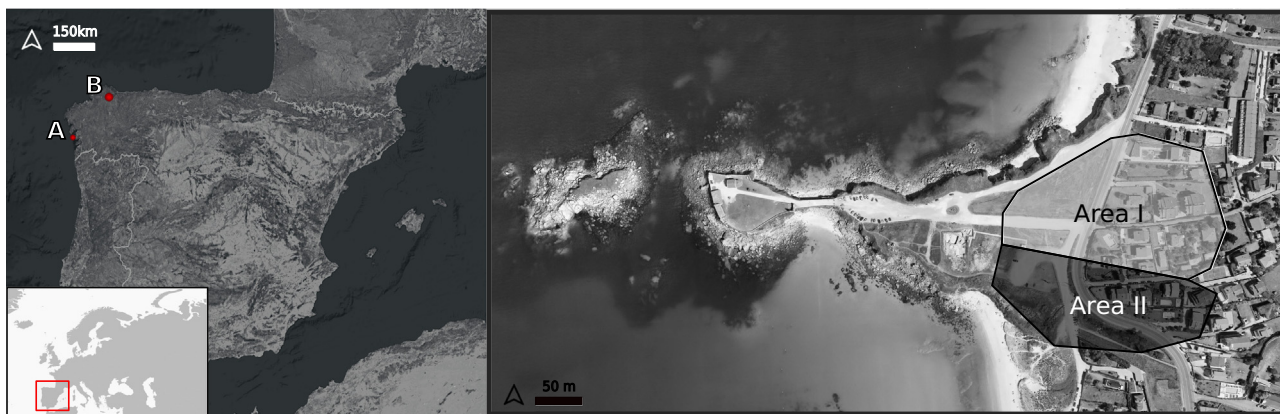


Fig. 1. Map of the site showing the location of A Lanzada site (red square; left) and the area of the peat records including Penido Vello record (red circle; left), the two funerary phases of A Lanzada necropolis are also shown (right); obtained from Lopez-Costas (2015). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

after thermal decomposition and pre-concentration of mercury by amalgamation on a gold trap using a DMA-80 direct mercury analyzer (Milestone, USA) at laboratory of Ecology, Faculty of Biology, Universidade de Santiago de Compostela. Four certified reference materials M2 ($58 \pm 5 \text{ ng g}^{-1}$) and M3 ($35 \pm 4 \text{ ng g}^{-1}$) (Steinnes et al., 1997), NIST 1486 ($2.3 \pm 1.4 \text{ ng g}^{-1}$), GBW07601a ($670 \pm 100 \text{ ng g}^{-1}$) blanks and replicates (two in every seven samples) were used to ensure analytical quality. The mean recoveries for the reference materials were between 88% and 116%. The median difference between duplicates was 5.4%.

2.2. Lead concentration and lead isotopes

Lead isotopic ratios were determined on a Nu Plasma II HR-MC-ICP-MS at Laboratoire G-Time from ULB. Prior to the isotope measurements, about 50 mg of the individual bone samples were processed through a specific chemical treatment after the samples were dried in an oven at 35° until constant weight. The first step involved dry ashing of the bulk samples (550°C for 12 h) and an acid dissolution using concentrated HF:HNO₃ (1:3) followed by concentrated HCl. After a careful check for the absence of solid residues, the sample solutions were loaded on the AG1-X8 exchange resin for the Pb separation and purification as described in Weis et al. (2006). Two successive passages on the same columns (after a careful rinsing of the resins) were applied to ensure the removing of matrix elements and Pb isolation efficiency. At the beginning of an analytical session, Pb elution fractions from each sample were re-dissolved in 20 μL of concentrated HNO₃, then evaporated and finally re-dissolved in 2 mL 0.05 M HNO₃. A thallium standard solution was added to samples and NBS981 Pb standard solutions to reach 125 ppb Pb and 25 ppb Tl. A sequence of two samples bracketed by the NBS981 standard solution was repeated and included duplicates and replicates. All Pb analyses were conducted in wet plasma mode with a minimum intensity for the ²⁰⁴Pb signal of 130 mV and a total beam of $\sim 10 \text{ V}$ ($= 80 \text{ V/ppm}$). Mass bias was corrected by external normalisation through Tl doping (Barling and Weis, 2008) and Standard Sample Bracketing (SSB) techniques (Mason et al., 2004). The ²⁰²Hg beam intensity, consistently below 0.15 mV was carefully monitored to prevent isobaric interferences of ²⁰⁴Hg with ²⁰⁴Pb. Repeated analyses of NBS981 provided an average value for ²⁰⁶Pb/²⁰⁴Pb of 16.9343 ± 48 (2SD) ($n = 46$), in line with the values reported in the literature (Weis et al., 2006). SSB correction of the sample data was conducted using the values from Abouchami et al. (2000) and Galer and Abouchami (1998).

2.3. Statistical analyses

Data analysis include Anova, Mann Whitney *U* test. (MWtest) and Kruskal-Wallis (K—W test) and were performed using IBM SPSS Statistics version 24. *P*-value <0.01.

3. Results

Lead and mercury concentrations in the bones varied from 0.1 to 20.9 $\mu\text{g g}^{-1}$ and 12 to 516 ng g^{-1} , respectively (Fig. 2A and B). Differences in metal concentrations between the two populations were found to be significant ($P < 0.01$), with individuals from Roman times showing higher values than those from post-Roman times (Table 1). No significant differences were found for sex and age or correlation with collagen yield. Significant differences ($P < 0.01$) were found for all stable lead isotope ratios, with Roman individuals showing lower ²⁰⁶Pb/²⁰⁴Pb, ²⁰⁷Pb/²⁰⁴Pb, ²⁰⁸Pb/²⁰⁴Pb, and ²⁰⁶Pb/²⁰⁷Pb and higher ²⁰⁸Pb/²⁰⁶Pb ratios (Fig. 3) than those from post-Roman times (Table 1). Despite these differences, both populations showed a certain degree of overlap in their elemental and isotopic compositions, as would be expected from a continuous occupation of A Lanzada for seven centuries (Figs. 2, 3). The (logged-) concentrations of the two studied metals were found to be highly correlated ($r = 0.82$ and 0.77, respectively) to the lead isotope ratios (see SM Fig. S1). This result suggests the mixing of, at least, two sources with different isotopic composition. This is consistent with the trend shown by the ²⁰⁸Pb/²⁰⁶Pb versus ²⁰⁶Pb/²⁰⁷Pb plot (Figs. 2D and 3), commonly used in environmental chemistry to trace lead sources. Samples follow a typical pattern of a two-component mixing, with one source having low concentrations of lead, lower ²⁰⁸Pb/²⁰⁶Pb and higher ²⁰⁶Pb/²⁰⁷Pb ratios and another one with higher lead concentrations, higher ²⁰⁸Pb/²⁰⁶Pb and lower ²⁰⁶Pb/²⁰⁷Pb ratios.

4. Discussion

4.1. Human skeletons and natural archives

Isotopic research using environmental archives has already shown that the Roman period represented a pre-industrial zenith in atmospheric metal pollution, recognizable to the point that the “Roman Pb Peak” was proposed as a chronological marker in Europe (Renberg et al., 2001). Peat records from NW Spain (that record atmospheric signals) (Kylander et al., 2005; Martínez Cortizas et al., 2002) show the same trend (Fig. 2C, D, Fig. SM2): high ²⁰⁶Pb/²⁰⁷Pb ratios (1.24–1.26),

typical of local geogenic lead before 1000–800 BCE followed by a decreasing trend from the local Early Iron Age (800 BCE), to reach a minimum (1.18) by the 1st–2nd centuries CE, reflecting the addition of lead due to increased mining and metallurgy. The peat records indicate atmospheric lead deposition at this time was ~25 times higher than natural background rates. With the decline of the Roman Empire by the 5th century CE, the isotopic ratios return to values similar to those seen in the Early Iron Age, pointing to a decrease in atmospheric metal pollution, although it remained ~5 times higher compared to the natural background. The $^{206}\text{Pb}/^{207}\text{Pb}$ ratios remained stable for nearly two centuries and then began to decrease again as the mining and metallurgy resumed in the Early Middle Ages. The isotopic composition of A Lanzada bone samples follow the same mixing line as the peat records from NW Spain and the values are consistent with the reconstructed pace of atmospheric metal pollution in the region (as seen in Fig. 2D). The data for the eight available radiocarbon-dated individuals also support this interpretation as, for example, the $^{206}\text{Pb}/^{207}\text{Pb}$ ratios in bone are highly correlated (r 0.85; $P < 0.01$) to the average ratios in peat samples covering the 2-sigma interval of the bone-calibrated ages (data in SM Table S2).

One difference between the bone and peat records is that the former shows lower $^{206}\text{Pb}/^{207}\text{Pb}$ values. This is explained by the fact that the individuals living at A Lanzada have incorporated any background lead from a local geogenic source (soils and bedrock) that has a slightly lower Pb isotope signature than the geogenic sources contributing to the studied peatlands. The correlation plots of lead and mercury concentrations (Fig. 3) point to a geogenic $^{206}\text{Pb}/^{207}\text{Pb}$ value of ~1.21 at the study area. Bones with the lowest lead ($<1 \mu\text{g g}^{-1}$) and mercury ($<20 \text{ng g}^{-1}$) concentrations have $^{206}\text{Pb}/^{207}\text{Pb}$ between 1.19 and 1.21 (Fig. 3). These values are in agreement with the isotopic signature of

soils from NW Spain developed on geological materials similar to those of A Lanzada (Fig. 2D) (Kylander et al., 2005). If we assume that the local geogenic lead in A Lanzada area has a $^{206}\text{Pb}/^{207}\text{Pb}$ ratio value of 1.21 and pollution lead a value of 1.15 (in agreement with the values typical of NW Spain ores; Kylander et al., 2005), a simple mixing model can be used to estimate the percentage of atmospherically-derived pollution lead (Pb_{Poll}) incorporated into the skeletons of people when alive. This approximation indicates that Pb_{Poll} in Roman individuals was on average $57 \pm 10\%$ (range 41–85%), which is significantly ($P < 0.01$) higher than the estimated values ($24 \pm 21\%$; range 2–63%) in post-Roman individuals. If we used the isotopic signature of ores from SW Spain the pollution contribution would be even greater. Individuals with over half of their total lead coming from Pb_{Poll} have $^{206}\text{Pb}/^{207}\text{Pb}$ ratios between 1.158 and 1.178 and Pb concentrations $>2 \mu\text{g g}^{-1}$. These isotope ratios are lower and the lead concentrations higher than those reported for indigenous, polluted ($>1 \mu\text{g g}^{-1}$ in tooth enamel) people living in pre-medieval Britain (Montgomery et al., 2010). In contrast, individuals with lower ($<50\%$) Pb_{Poll} have $^{206}\text{Pb}/^{207}\text{Pb}$ ratios ranging from 1.178 to 1.206. Taken together, our data show that the stable lead isotopic composition of bones tracks environmental exposure to contaminants rather than representing exclusively an individual's geography of origin, as has been suggested elsewhere (Montgomery et al., 2010). The use of lead isotopes for mobility studies may only be applicable when concentrations are low, or when the regional environmental pollution trend is known, and departures from the cultural-mediated effect can be identified.

Investigations of environmental archives in NW Spain have shown that the proportion of atmospheric Pb_{Poll} was 70–80% during the Roman period, while in post-Roman times it dropped to 25–30% (Kylander et al., 2005; Martínez Cortizas et al., 2013). Although the

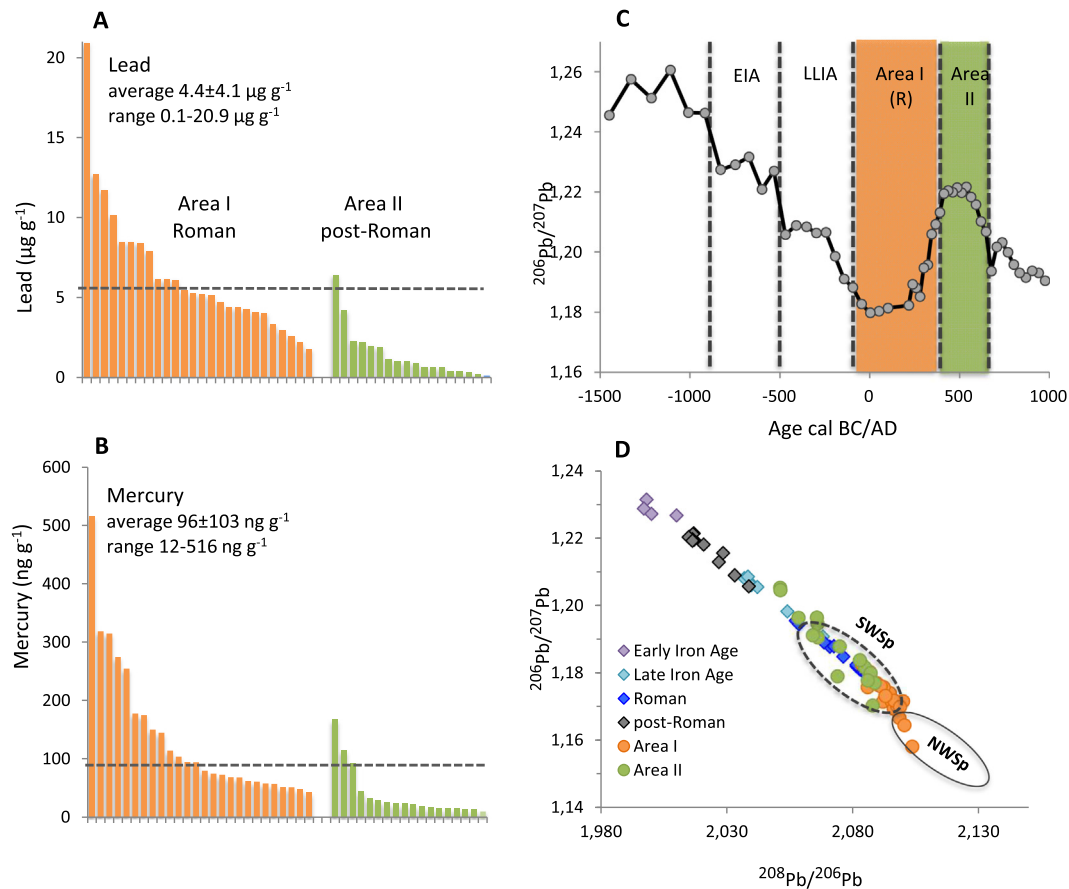


Fig. 2. Lead (A) and mercury (B) concentrations in human bones from Roman (Area I) and post-Roman (Area II) from A Lanzada site. (C) Chronology of $^{206}\text{Pb}/^{207}\text{Pb}$ values in a peat record from NW Spain for the period 1500 BCE to 1000 CE (Kylander et al., 2005) (EIA local Early Iron Age; LLIA local Late Iron Age). (D) Isotope ratios for NW Iberia peat samples (diamonds) and A Lanzada human bones (circles) and the isotopic fields for ores from SW and NW Spain.

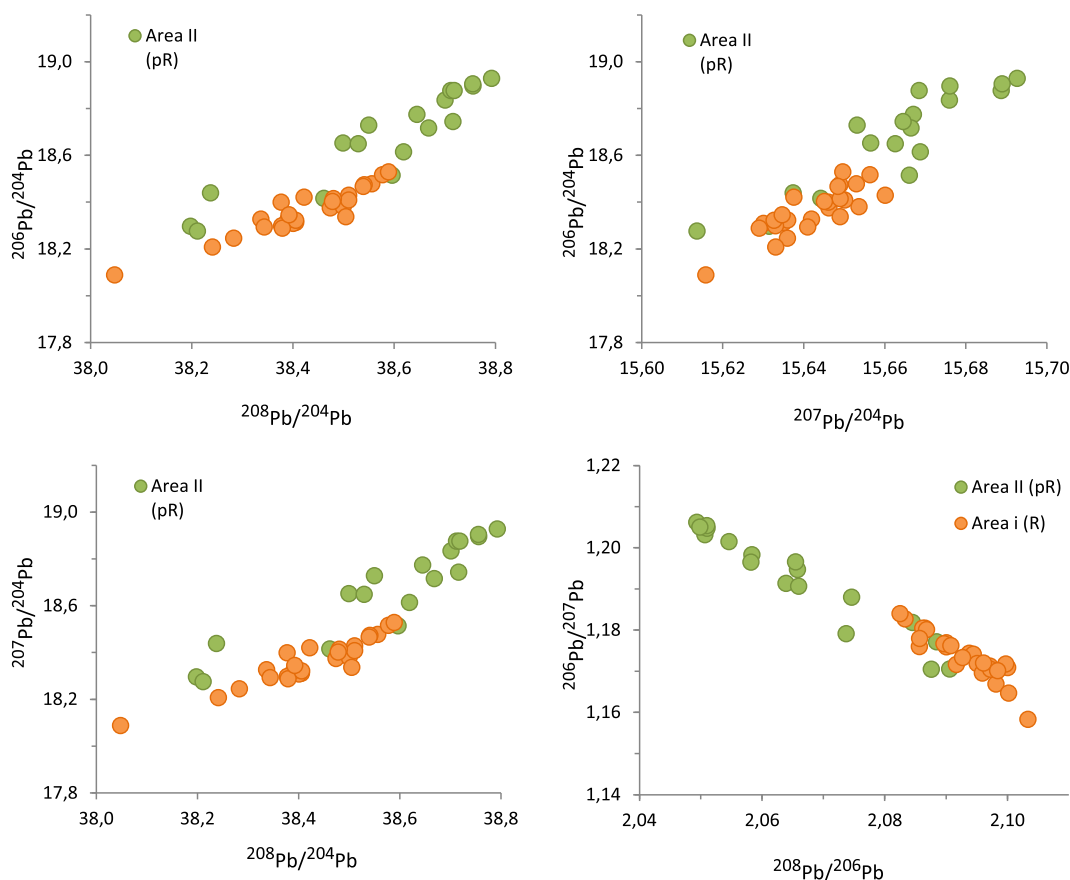


Fig. 3. Lead isotope ratios of the analysed bone samples from Roman (R; Area I) and post-Roman (pR; Area II) individuals from A Lanzada site.

average proportion of Pb_{poll} found in individuals from the Roman period is lower than that for the atmospherically deposited lead, the most polluted individuals for this period show values on the same order (70–85%). The average estimated proportion of Pb_{poll} in post-Roman individuals fits well with that of the atmospherically deposited lead. Thus, despite the differences in the natural background isotope ratios, the estimated proportions of pollution lead in bones from A Lanzada individuals are consistent with the values obtained using other environmental archives.

4.2. Atmospheric incorporation

Whilst higher metal content can be related to different sources, the evidence presented here strongly suggests both mercury and lead were incorporated atmospherically. Higher metal contents in an ancient human population may have come from a number of sources, including work exposure (i.e., metallurgists or miners), the intake of contaminated food and drink, migration from a region with a higher geological background, or even living in a more polluted environment.

Table 1

Average (avg) and standard deviation (std) for lead (Pb; $\mu\text{g g}^{-1}$) and mercury (Hg; ng g^{-1}) concentrations, lead isotopic composition and the estimated pollution lead content (Pb_{poll} ; %) of human bones from A Lanzada.

	Pb	Hg	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{207}\text{Pb}$	Pb_{poll} %	
Area I (R)	avg	6.4	136	18.36	15.64	38.42	2.093	1.174	57
<i>n</i> = 26	std	4.2	114	0.09	0.01	0.12	0.005	0.005	10
Area II (pR)	avg	1.5	39	18.68	15.66	38.58	2.066	1.191	24
<i>n</i> = 18	std	1.6	43	0.21	0.02	0.19	0.014	0.012	21
All	avg	4.4	96	18.49	15.65	38.49	2.082	1.181	44
<i>n</i> = 44	std	4.1	103	0.22	0.02	0.17	0.017	0.013	23

Occupational exposure and/or direct ingestion from metal artefacts (e.g., lead-glazed ceramics, pewter vessels, etc.) (Millard et al., 2014) were probably less likely at A Lanzada because both lead and mercury show a similar behaviour (Table 2) and correlate with the lead isotopic composition in the analysed individuals, despite the different geochemical cycles for these two metals. The mercury cycle is dominated by deposition of atmospheric species (Selin, 2009) and no mercury minerals are present in the study area. Thus, the parallel increase suggests that both metals were atmospheric in origin. Additionally, with A Lanzada being a rural settlement, the long-term metal exposure was probably much lower than in urban areas, as suggested by other researchers (Millard et al., 2014; Montgomery et al., 2010). Populations relying on subsistence in rural areas may have escaped intense exposure to culturally mediated pollution (i.e., plumbing, household items, medicines, etc.) (Montgomery et al., 2010). The lack of differences according to sex and age, and the consistent isotopic signal during each period, point to sources that affected the entire population, thus making migration a less realistic explanation in our case.

Our data support the hypothesis that Roman individuals were exposed to larger environmental loadings of metals than those of the following Post-Roman period, even at the edges of the Empire. Inhalation of ambient pollution lead and mercury is the most suitable explanation for the observed results. This was suggested for samples from southern

Table 2

Correlation between the (logged) lead (Pb) and mercury (Hg) concentrations and lead isotopic ratios determined in the bone samples.

	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{206}\text{Pb}/^{207}\text{Pb}$
Pb (<i>n</i> = 44)	−0.82	−0.69	−0.54	0.90	−0.82
Hg (<i>n</i> = 44)	−0.76	−0.67	−0.64	0.76	−0.77

Britain (1st century CE), which showed no agreement between enamel and regional geogenic lead isotope ratios (Montgomery et al., 2010); although the ratios were consistent with those of ore sources, a unique relationship was not established. However, for A Lanzada, a clear relationship with other environmental archives exists, and it can be demonstrated for the 700 years of continuous necropolis occupation. Here, both human bones and peatlands (Kylander et al., 2005; Martínez Cortizas et al., 2002; Martínez Cortizas et al., 2013; Martínez Cortizas et al., 1999) show higher atmospheric lead and mercury concentrations and lower Pb isotope ($^{206}\text{Pb}/^{207}\text{Pb}$) values during Roman times than in post-Roman times.

Lead and mercury can be absorbed by humans through their lungs and gastrointestinal tract, and less likely through the skin (Agency for Toxic Substances and Disease Registry ATSDR, 1999, 2019; Brodtkin et al., 2007). In areas with no direct exposure to metals, such as A Lanzada, oral intake – through water and food – is the primary pathway for lead with some additional contribution from inhalation (Agency for Toxic Substances and Disease Registry ATSDR, 2019); while mercury intake most likely occurred via the inhalation of vapours and contaminated food (Clarkson et al., 2007). Atmospheric lead and mercury are largely the main sources of the deposition on plants (Agency for Toxic Substances and Disease Registry ATSDR, 1999, 2019); therefore, atmospheric pollution produces contaminated vegetal foodstuffs and consequently contaminated animals. Geogenic lead in soil can also get into the food chain through plant roots and drinking water, but it has a different isotopic signature (e.g. higher in $^{206}\text{Pb}/^{207}\text{Pb}$) than atmospheric pollution lead (e.g. lower in $^{206}\text{Pb}/^{207}\text{Pb}$) (see Fig. 2D). In consequence, an admixture of pollution lead and mercury and geogenic lead was likely incorporated into the human bodies from A Lanzada's people directly through inhalation and indirectly through food and water.

Once absorbed by the body, both metals are incorporated into soft tissues and bone through blood circulation (Brodtkin et al., 2007). Lead is incorporated in the mineral fraction of the bone content (Silbergeld, 1991), while mercury is more likely incorporated into the thiol groups of the amino acid cysteine of collagen (Clarkson et al., 2007). Our study was conducted on bulk cortical bone, which does not allow us to distinguish the molecules to which lead and mercury are bonded. The lack of correlation with collagen preservation and elements constituent of the mineral part of bone (i.e. Ca, P, Sr) (López-Costas et al., 2016; López-Costas and Müldner, 2016), the absence of mercury in local geologic sources and the lead isotopic signatures (low values for $^{206}\text{Pb}/^{207}\text{Pb}$) suggest that the intensity of diagenesis is not a key factor for lead and mercury content in our case. However, more studies applied on bioapatite and collagen separately will help to get a better understanding about how bone forms an archive of changing metal pollution.

5. Conclusions

Our study shows that atmospheric metal pollution affected the metal content (lead and mercury) and lead isotopic composition of human bones in living individuals in the past. The data from the A Lanzada necropolis indicate that archaeological human bones can be used as an environmental archive of past atmospheric metal pollution. We recommend that further studies of metal contamination in bone from archaeological contexts should also include mercury. The findings presented here clearly open up a new approach - the analysis of metals preserved in human remains - to document the pollution history of our ancestors.

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2019.136319>.

Declaration of competing interest

Authors declare no competing interests.

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Author contributions

- Olalla López-Costas: Conceptualization, Formal analysis, Investigation, Methodology, Resources, Supervision, Writing – original draft, Writing – review & editing
- Malin Kylander: Conceptualization, Writing – original draft, Writing – review & editing
- Nadine Mattielli: Investigation, Writing – original draft, Writing – review & editing
- Noemi Álvarez-Fernández: Investigation, Visualization, Writing – review & editing
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- Tim Mighall: Conceptualization, Writing – review & editing
- Richard Binder: Conceptualization, Visualization, Writing – review & editing
- Antonio Martínez-Cortizas: Conceptualization, Formal analysis, Investigation, Methodology, Funding acquisition, Supervision, Visualization, Writing – original draft, Writing – review & editing

Data and materials availability

All data is available in the main text or the supplementary materials.

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