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# **DEATH METAL: Characterising the effects of environmental lead pollution on mobility and childhood health within the Roman Empire.**

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## ABSTRACT

The use of lead was ubiquitous throughout the Roman Empire, including as material for water pipes, eating vessels and as a sweetener for wine. Children are particularly susceptible to the effects of lead and it is likely that the widespread use of this deadly metal amongst Roman populations led to a range of adverse health effects. Indeed, lead poisoning has even been implicated in the downfall of the Roman Empire. This research examines the direct effect of lead poisoning on the inhabitants of the Empire, and for the first time introduces a bioarchaeological perspective to how lead exposure affected health during the Roman period. The results provide strong evidence that Roman lead pollution contributed to the high prevalence of metabolic diseases during childhood and implicates elevated lead burdens in the high prevalence of infant remains in Roman skeletal assemblages.

This study has also shown the effectiveness of lead isotope analysis as a tool in archaeological migration studies. The successful establishment of baseline lead isotope ranges in previously unstudied regions of the Roman Empire has greatly enhanced our ability to identify the potential origins of isotopic outliers. Although this study has shown that anthropogenic lead isotope ratios are not country specific, the results have demonstrated that lead isotope ratios can differentiate between populations based on the orogenic age of the region in which an individual spent their childhood. This has improved our understanding of how anthropogenic lead isotope ratios are capable of discriminating between geographical regions of origin when other isotope system are not.

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"The copyright of this thesis rests with the author. No quotation from it should be published without the author's prior written consent and information derived from it should be acknowledged." For Mum,

Helping me achieve my dreams since 1985

# CHAPTER ONE

## Introduction

### **1.1 Research context**

#### **1.1.1 Using lead isotopes to explore migration.**

Lead is ubiquitous within the environment, found within most rocks, soils, and water systems. It has both common (<sup>204</sup>Pb) and radiogenic (<sup>206</sup>Pb, <sup>207</sup>Pb and <sup>208</sup>Pb) sources that combine to create specific isotope characteristics, the variability of which reflects the underlying geology of the local area (Faure, 1986). Humans incorporate lead into their body through dietary consumption and inhalation, where it is predominantly stored within the mineral matrices of teeth and bones (Gulson et al., 1997a; Waldron, 1983). The interpretation of lead isotope ratios in archaeological skeletal remains for the purpose of investigating mobility and migration has been developed predominantly from research carried out on British populations (Montgomery, 2002; Montgomery et al., 2014, 2010, 2005; Shaw et al., 2016). Although the use of lead isotope ratios to answer questions surrounding geographic origins is beginning to gain momentum in European and North American studies (Keller et al., 2016; Price et al., 2017a, 2017b, 2017c; Sharpe et al., 2016).

One of the first applications of lead isotope ratios in archaeological human remains was carried out using four bone samples from the Roman site of Poundbury Camp, Dorset (Molleson et al., 1986). However, it was not until the work of Montgomery (2002) that real strides in the advancement of lead isotope analysis of archaeological human remains were made. Montgomery (2002) developed a methodology that minimised the risk of contamination from the burial environment, providing a means of confidently assessing *in vivo* lead characteristics. Through diachronic analysis of tooth enamel samples spanning from the Neolithic to the Medieval period, Montgomery (2002) established the baseline for human lead isotope ratios in England and Wales. This research also demonstrated for the first time how local lead isotope ratios altered depending upon the type of exposure (geogenic or anthropogenic) dominating an individual's environment. In prehistoric societies lead incorporated into bodily tissues tends to reflect natural, geogenic lead concentrations, allowing for regionally specific signatures to be used as tracers in archaeological migration studies (Montgomery, 2002; Montgomery et al., 2000). However, with the advent of metallurgical technologies (mining, smelting etc.) and the subsequent increases in environmental pollution and use of lead compounds, human lead isotope ratios alter to reflect exploited lead ore sources (Montgomery, 2002; Montgomery et al., 2010).

It has been shown that lead ore isotope ratios vary throughout the world because they are formed at different times (McCrady, 1952; Sangster et al., 2000). As Roman human lead isotope ratios are dominated by lead ore isotope ratios it is probable that human lead isotope ratios also differ by geographic region. Montgomery et al., (2002, 2010) deomonstrated that archaeological British populations have lead isotope ratios consistent with British lead ore, and collated data from archaeological populations from across the world deomonstrate that human lead isotope ratios differ on a continental scale (Åberg et al., 1998; Montgomery, 2005; Turner, 2009; Bower et al., 2007; Valentine, 2008). However, little has been done to assess how, or if lead isotope ratios differ significantly within a single continent.

Research focusing on the Roman period has demonstrated that these anthropogenic lead isotope ratios can be used to successfully identify migrants in culturally mixed populations (Montgomery, 2002; Montgomery et al., 2010; Shaw et al., 2016). However, despite being a population well-known for the extensive movement of people and a popular target for migration studies, there is a notable lack of comparable human lead isotope data from regions of the Empire outside of Britain. As a result, studies have had to use lead ore datasets as proxies for human isotope ratios when attempting to establish local ranges and identify migrants in Roman populations (Montgomery, 2002; Montgomery et al., 2010; Shaw et al., 2016). This research aims to address this issue by establishing the lead isotope ratios in skeletal material excavated from sites across Europe to ascertain how they differ with socio-cultural and geographic variation and provide an initial baseline in human lead isotope ratios for different regions of the Roman Empire.

#### **1.1.2 Investigating lead poisoning within the Roman Empire**

Few theories evoke more fervent debate than what might have brought about the fall of the Roman Empire. For centuries scholars have put forth arguments for a plethora of singular causes for its decline, positing everything from the conversion to Christianity to overexpansion (Gilfillan, 1990). It is, however, the notion that lead poisoning was a key contributing factor behind its decline that has captured the interest of scholars and general enthusiasts alike. The urban myth-like quality that this theory has taken on has ensured its endurance. Historical texts describe a range of maladies associated with lead poisoning, affirming that Roman populations did indeed suffer the deleterious effects of lead toxicity (Lessler, 1988; Needleman, 2009; Retief and Cilliers, 2006; Waldron, 1973). It was Nriagu's (1983a) use of this historical literature to demonstrate the endemic nature of lead poisoning and its consequences that gave support to the role that lead played in the downfall of the Empire. Despite how mainstream the theory of endemic lead poisoning became, many scholars refuted the claims. Some openly questioned the validity of the translations of the ancient texts and suggested that lead pollution during the Roman period was not significant enough to have resulted in the Empire's decline (Cilliers and Retief, 2014; Drasch, 1982; Gaebel, 1983; Needleman and Needleman, 1985; Scarborough, 1984). Although it may never be possible to truly ascertain the role, if any, that lead poisoning played in the fall of Rome, the effect it had on childhood health and mortality throughout the Empire can be explored directly via skeletal analysis.

Studies that have analysed lead concentrations in Roman skeletal material reveal lead burdens up to three times higher than what is today considered 'severely toxic' (Montgomery et al., 2010). Therefore, it could be surmised that lead was deleterious to Roman health, especially in children, who are more susceptible to lead poisoning than adults (Needleman, 2004). The demographic profiles of Romano-British skeletal populations attest to the fragility of childhood health during this period, especially within the first year of life (Carroll, 2014). It is therefore surprising that so little research exists on childhood lead burdens and their effects on Roman non-adult health and mortality. This research will explore whether the extensive use of lead in the Roman Empire contributed to the high infant mortality rates evident in Roman skeletal populations. Offering new insights into the impact of anthropogenic lead exploitation on child health within the Roman Empire and how this may have differed according to geographic and socio-cultural variations.

### **1.2 Research aims**

The overarching aim of this project is to explore how exposure to anthropogenic lead pollution during the Roman period impacted upon childhood health, and what this exposure can tell us about geographic mobility within the Empire. It focuses upon the use of lead isotopes as a discriminant in migration studies and attempts to determine the extent of variation in lead isotope ratios between modern countries via a highly polluted archaeological population. This study also aims to determine how lead pollution impacted upon the health and mortality of children within the Roman Empire, and examine which, if any, skeletal markers of disease can be used to help identify individuals suffering from lead poisoning.

#### 1.2.1 Objectives

- Investigate the impact lead burdens had upon the health of Roman children throughout the Empire via paired analysis of tooth enamel lead concentrations and osteological data from non-adult skeletal remains.
- Determine whether variations in socio-cultural and geographic origins influence childhood lead burdens by interpreting contextual information alongside lead concentration data.
- Ascertain whether anthropogenic lead pollution could have contributed to the high infant mortality rates observed in Roman skeletal populations through the comparison of lead concentrations and age-at-death.

- Establish how skeletal lead isotope ratios vary between different regions of the Roman Empire by analysing tooth enamel lead isotope ratios in Roman skeletal populations from different regions of the empire.
- Explore the usefulness of lead isotope ratios in human tooth enamel in identifying migrants in culturally mixed Roman skeletal populations by combining lead isotope data with contextual information.

### 1.3 Period of study

In order to determine how anthropogenic lead isotope ratios can be used to establish geographic origins from archaeological skeletal material, a population known for its use of lead and lead products is essential. In this respect the Romans provide a perfect study population. The use of lead was ubiquitous throughout the Roman Empire. This versatile metal was included in everything from water pipes, building materials and eating vessels, to medicine, make-up and food sweeteners (Gilfillan, 1990). The significant increase in the bioavailability of lead throughout this period resulted in widespread exposure to unprecedented levels of the toxic metal (Needleman, 1991; Nriagu, 1983). The *in vivo* anthropogenic lead isotope ratios this type of exposure creates, makes Roman skeletal assemblages ideal study populations. Not only for determining the efficacy of anthropogenic lead isotope ratios in archaeological migration studies but also for assessing how leads exposure impacted upon health.

The Roman Empire (27 BC - 476 AD) was amongst the most powerful economic, cultural, political and military forces in the world at this time. At the peak of its power it covered 5 million square kilometres, ruling over an estimated 60 - 70 million people across parts of Europe, North Africa and Western Asia (Taagepera, 1979; Turchin et al.,

2006). Approximately 21% of the world's population during this period, lived within the Empire's borders, ensuring its place as one of the largest empires in world history (Potter, 2004, p. 17). An integral part of the development, expansion and maintenance of this vast territory was the movement of people to and from all regions of the Roman Empire (Hin, 2013; Killgrove, 2013; Scheidel, 2001). Migration was not limited to those of a low socioeconomic status looking to improve their livelihoods elsewhere. But included people from all levels of society hoping to better their life station or take on administrative or entrepreneurial roles in newly acquired lands (Sweetman, 2011; Tacoma, 2016; Woolf, 2013). Whatever the reason for migration within the empire, it is clear that levels of migration were both high and multidirectional (Killgrove, 2014). With their inclination for migration and unprecedented levels of lead exposure, Roman skeletal populations offer the chance to assess whether variations in human lead isotope ratios are sensitive enough to identify outliers in what are assumed to be culturally mixed skeletal populations.

### **1.4 Sample population**

The analysis of lead isotope ratios from different regions of the Roman Empire provides a means of assessing how well lead isotope ratios can discriminate between contemporaneous individuals from different countries. An approach that is, to date, unique in bioarchaeological studies. This study incorporates eight Roman populations (1st to 4th centuries AD), from seven different regions of the Roman Empire (see Fig. 1.1). Sites were chosen based on their location within the Empire. Different regions that encompassed a large proportion of the Roman Empire were needed to allow visualisation of how lead isotopes within a highly polluted and mobile population varied according to geographic region. To that end, skeletal assemblages from Scotland, England, Spain, France, Slovenia, Romania and Lebanon were included in this study. In doing so, an expanse of the Roman Empire spanning from its most north-westerly outpost to its most easterly province was covered.

The dispersed locations of sites provided the opportunity to assess the discriminant resolution offered by human lead isotope ratios in contemporaneous populations across a continent, plus Lebanon. The widespread geographical locations of samples incorporated into this study also facilitated a means of acquiring and establishing human lead isotope ranges from regions of the Roman Empire where there is currently no comparative data. Although the anthropogenic lead isotope range for humans exposed to English and Welsh lead ore has been well established (Budd et al., 2004; Montgomery, 2002; Montgomery et al., 2010), eight Roman individuals from Britain were also analysed in this current study. This small group of individuals were included due to their unusual burial rites. Six of these individuals were excavated in Musselburgh, Scotland near a fort on the Antonine wall, all six individuals exhibited skeletal evidence for decapitation and trauma. The remaining two individuals were from lead coffin burials excavated in York and Ilchester, England. Both are uncommon burial rites in Roman Britain, and have previously been shown to be associated with migrants (Montgomery et al., 2010; Müldner et al., 2011). These two case studies (Musselburgh and Lead coffin burials) also provide a means of testing the usefulness of lead isotope ratios in discerning an individual's geographic origins using the data obtained in this study.



Figure 1.1 – Map showing the location of the nine sites used in this study. Musselburgh, Scotland (n = 6), York, England (n = 1), Ilchester, England (n = 1), Tarragona, Spain (n = 27), Barcelona, Spain (n = 34), Caen, France (n = 37), Ljubljana, Slovenia (n = 8), Alba Iulia, Romania (n = 37) and Beirut, Lebanon (n = 40).

### **1.5 Structure of the thesis**

The following four chapters discuss the relevant background literature that forms the basis for the current study. Chapter Two introduces lead and the premise behind using lead isotope analysis to investigate the geographical origins of people from archaeological populations. Chapter Three discusses current isotope systems used to identify migrants in archaeological populations and focuses on the type of populations suited to anthropogenic lead isotope studies. This chapter also summarises how the Romans, a population well-known for their extensive exploitation of lead and propensity for migration, make an ideal study population. Chapter Four explores the biochemical interactions of lead poisoning within the human body to inform our understanding of the way in which lead poisoning may manifest in skeletal remains, how lead burdens can be quantified, and how this can be used to inform our

interpretations of lead poisoning in archaeological populations. Chapter Five reviews literature relating to health and mortality during the Roman period, outlining current reasoning for the poor health and high infant mortality rates during this time and positing the role that lead poisoning may have contributed to this.

Chapter Six focuses on the methodology employed in this study. It presents the archaeological sites and the sample population included in this study, and details the sampling strategy, sample preparation method and analytical techniques used to obtain and interpret the data collected. The results are presented and discussed in Chapters Seven and Eight. Chapter Seven draws together the results of the trace element analysis and palaeopathological data obtained from osteological analysis to offer a comprehensive overview of how lead burdens impacted upon health and mortality in the past. While Chapter Eight presents the results of the lead isotope analysis, with discussions focusing on determining the resolution at which geographical origins can be determined within the Roman Empire. The efficacy of lead isotope ratios in identifying migrants from culturally mixed, polluted populations is also discussed here. The concluding chapter provides a brief synopsis of the thesis followed by the overarching findings of the research and suggestions for further work. All of the isotope and trace element data is tabulated in the appendices.

# CHAPTER TWO

# Lead Analysis in Bioarchaeology

### 2.1 Introduction

One of the fundamental questions often posed in bioarchaeology pertains to human migration and mobility. Do the individuals within a given skeletal population represent individuals from the same or varied biological and socio-cultural groups? Traditionally, anthroposcopic and anthropometric analysis techniques were the predominant methodological approaches used to investigate migration within a skeletal population. However, with advancements in chemical analyses, isotopic studies are coming to the forefront in addressing questions pertaining to cultural affiliation (Bentley, 2006; Katzenberg, 2008; Nehlich, 2015; Montgomery et al., 2010). It has been well documented that various isotope systems (e.g. C, N, O, Sr, S, H, Pb), have the potential to be powerful discriminants in origin studies, and are therefore increasingly used to investigate the mobility of archaeological populations (Åberg et al., 1998; Beard and Johnson, 2000; Bentley, 2006; Chenery et al., 2012; Sealy et al., 1995). However, lead (Pb) despite its potential, has received comparatively less attention than other isotope systems such as strontium (Sr) and oxygen (O).

Lead is widely dispersed within the environment from both common and radiogenic sources, which combine to create geologically specific isotope characteristics. When viewed as isotope ratios these characteristics can be used as isotopic signatures or fingerprints for identifying geogenic and anthropogenic lead sources in provenance studies, whether they be human, artefact or otherwise (Wilson et al., 2006). A wealth of provenance studies, especially those focusing on archaeological artefacts, has proven lead to be an effective discriminatory tool. However, from a bioarchaeological perspective lead, despite its potential to discriminate between exposures to different ore sources, has been under utilised when compared to the use of other isotope systems in mobility studies. This is somewhat surprising as lead offers several advantages over lighter isotopes, and those with only one radiogenic parent, such as strontium (Gulson, 1986). Due to its high atomic weight, lead does not fractionate in the low temperature processes that alter the composition of lighter elements, and its relative rarity compared to elements such as carbon, nitrogen or hydrogen mean that there is less scope for mixing from innumerable source (Gulson, 1986).

The interpretation of lead isotopes in archaeological skeletal remains for the purpose of investigating mobility and migration has been developed predominantly from research carried out on British populations (Montgomery, 2002; Montgomery et al., 2010, 2014, 2005; Shaw et al., 2016). There appears to be an increasing use of lead isotope ratios in Roman migration studies. Focus on this period derives mainly from the extensive exploitation of lead throughout this time, and the resultant environmental pollution that created elevated human lead burdens, coupled with homogenised lead isotope ratios. These isotope ratios are thought to represent the dominant anthropogenic ore source utilised by a particular population. As such, migrant individuals who spent their childhood in a different country or region to that in which they were interred would have been exposed to different anthropogenic ore sources to those considered local to their interment area. Therefore, they would exhibit lead isotope ratios that are distinct from those of the local population, making the identification of migrants in a culturally mixed skeletal population relatively easy. A number of bioarchaeological studies have shown that the lead isotope ratios in the majority of Romano-British skeletal remains

are congruent with the isotopic range expected from exposure to British lead ore sources, and that those with isotope ratios inconsistent with this British range are easily identified (Millard et al., 2014; Montgomery et al., 2010; Shaw et al., 2016). However, there is a notable lack of comparative data for human skeletal material excavated from Roman period sites throughout the rest of Europe. This limits any possible interpretation of non-British human lead isotope ratios identified in British populations, as there is little to no reference data available for direct comparison. Instead lead isotope ratios obtained from artefacts of known provenance have to be used as a proxy. This research will address this by determining the lead isotope ratios in Roman individuals from several countries from within what was the Roman Empire, establishing if and how human lead isotope ratios vary in Roman individuals from different geographical regions across Europe and to what extent they can be used to investigate mobility within the Roman Empire. This chapter introduces lead and the premise behind the use of lead isotope analysis to investigate the geographical origins of people from archaeological skeletal populations.

### **2.2 Geochemistry**

#### 2.2.1 Lead

Lead is a dense, yet soft, ductile metal with a low melting point (327°C) and high resistance to corrosion. Because of these properties it has been utilised in the manufacture of a wide range of products for millennia (Settle and Patterson, 1980; Sherwood Lollar, 2005, p. 71). It is classified as a post-transitional heavy metal with chalcophile tendencies, meaning that it has a higher affinity for sulphur than oxygen and therefore forms dense sulphides that remain predominantly within the upper levels of

the Earth's crust. Naturally occurring sources of lead are most commonly found in minerals such as galena (PbS), anglesite (PbSO<sub>4</sub>), Cerussite (PbCO<sub>3</sub>) and minium (Pb<sub>3</sub>O<sub>4</sub>). However, trace amounts also occur in numerous other minerals such as K-feldspar, zircon, micas and magnetite. Due to the intermediate size of Pb<sup>2+</sup> ions (1.19 Å) it can readily replace Ca<sup>2+</sup> (1.14 Å) and K<sup>+</sup> (1.52 Å) ions in several minerals, which is why lead is enriched in felsic rocks such as granite (Mielke, 1979). The distribution of lead within sedimentary rocks is governed by the lead content of the detrital minerals and organic matter present at the time of its formation (Heinrichs et al., 1980). Due to lead's affinity for organic material, sedimentary rocks such as shale and greywackes contain the highest concentrations of the metal. As soils are a major contributor to human lead burdens details such as these are important because soil lead composition and concentrations are largely dependent upon the lead within the underlying rocks that formed it.

Lead ions within soils are relatively immobile as they readily form secondary minerals with low solubility or ion complexes with manganese or iron. However, soils with low calcium concentrations or high pH values exhibit increased lead solubility that facilitates the aqueous movement of lead in the environment (Hem, 1976; Martínez and Motto, 2000; Zimdahl and Skogerboe, 1977). Natural levels of lead within soil have been estimated to be no more than 25 mg/kg<sup>-1</sup> unless anthropogenic contributions have polluted it (Kabata-Pendias, 2010, pp. 338–349). As mentioned above, anthropogenic activities such as metalliferous mining and the manufacture and use of lead or lead containing products (e.g. pipes, paints, glazes, petrol etc.) causes a significant increase in environmental lead concentrations (Patterson, 1965). These increased lead concentrations are deleterious to health as lead is highly toxic to all living organisms.

#### 2.2.2 Lead isotopes

There are four naturally occurring lead isotopes, three of which are radiogenic daughters of thorium and uranium decay (<sup>206</sup>Pb, <sup>207</sup>Pb and <sup>208</sup>Pb), and one primeval non-radiogenic isotope (<sup>204</sup>Pb). <sup>204</sup>Pb is considered a stable reference isotope, the abundance (1.4%) of which has remained constant since the formation of Earth.

<sup>238</sup>U 
$$\rightarrow \underline{8\alpha}_{6\beta} \rightarrow {}^{206}\text{Pb} (t_{1/2} = 4.467 \text{x} 10^9) \text{ [abundance 24.1\%]}$$
  
<sup>235</sup>U  $\rightarrow \underline{7\alpha}_{6\beta} \rightarrow {}^{207}\text{Pb} (t_{1/2} = 0.704 \text{x} 10^9) \text{ [abundance 22.1\%]}$   
<sup>232</sup>Th  $\rightarrow \underline{6\alpha}_{4\beta} \rightarrow {}^{208}\text{Pb} (t_{1/2} = 14.010 \text{x} 10^9) \text{ [abundance 52.4\%]}$ 

(Baskaran, 2011; McSween et al., 2003)

There is extensive natural variation in the isotopic composition of lead in geological bodies, ranging from highly radiogenic lead in old uranium/thorium rich minerals such as granite, to minerals low in uranium/thorium lead but high in common lead such as K-feldspar. These differences in isotopic composition reflect the different chemical environments in which the lead originated (e.g. crustal rock, mantle rock, ore bodies etc.). Additionally, the mixing of one or more lead sources with disparate compositions may further modify the isotopic composition of any given sample. This complex aggregation of multiple lead sources results in divergent isotope compositions that are specific to their geological environment, making them useful provenance tools (Evans et al., 2015; Faure, 1986). As different geological regions across the world are composed of varying types of rocks of different ages, each geographical region produces lead isotope ratios uniquely characteristic of its underlying geology.

Lead isotopes are determined as ratios of one isotope to another rather than a direct measurement of abundance (e.g. <sup>206</sup>Pb/<sup>204</sup>Pb, <sup>207</sup>Pb/<sup>204</sup>Pb etc.). Conventional plots used to present lead data include <sup>207</sup>Pb or <sup>208</sup>Pb as ratios with the invariant <sup>204</sup>Pb plotted against <sup>206</sup>Pb/<sup>204</sup>Pb. A bivariate plot using <sup>207</sup>Pb/<sup>204</sup>Pb against <sup>206</sup>Pb/<sup>204</sup>Pb allows visualisation of any changes in the uranium-lead systematics of the samples as it only compares the isotope ratios of uranium derived lead (uranogenic lead). When <sup>208</sup>Pb/<sup>204</sup>Pb is plotted against <sup>206</sup>Pb/<sup>204</sup>Pb, any changes in both thorium-lead (thorogenic) and uranogenic lead processes can be visualised. However, due to its naturally low abundance (~1.4%) accurate measurement of <sup>204</sup>Pb has, until recently, been problematic (Baskaran, 2011). To avoid the higher uncertainties surrounding <sup>204</sup>Pb measurements <sup>208</sup>Pb/<sup>206</sup>Pb against <sup>207</sup>Pb/<sup>208</sup>Pb plots are often used. These plots benefit from higher precision, but compress the data fields. Therefore, the subtler variations that frequently differentiate one lead field from another may, unlike in conventional bivariate plots using <sup>204</sup>Pb, be lost.

### 2.3 Lead in the environment

#### 2.3.1 Geographical variation of lead isotopes

Geogenic lead isotope ratios within soils tend to be a homogenous representation of the isotopic composition of the underlying local geology. Therefore, any geographical variations within geogenic lead isotope ratios stem from the mixing of lead sources with disparate isotope compositions. These can either be from inter-ore spatial variations or preferential weathering and mechanical degradation of heterogeneous rocks (Erel et al., 1994, p. 5565; Giacalone et al., 2005). Lead within soils is largely immobile, staying predominantly within the organic fraction due to its affinity for forming complex

sulphide species (Adriano, 1986; Giacalone et al., 2005; Sheppard and Thibault, 1992). As such, anthropogenic lead contributions to soil composition also stay within the organic fraction, altering the isotopic composition of the upper soil fractions to reflect that of the dominant pollutant source rather than local geology. Lead isotope ratios do, however, gradually conform to those exhibited by the underlying geology as they approach the bedrock (Bacon et al., 1996, p. 2516).

As with most geochemical processes, the mobility of lead within the organic fraction can be increased when subject to decreases in soil pH or environments rich in soluble organic matter (Giacalone et al., 2005; MacKenzie et al., 1998; Stewart and Fergusson, 1994). Although metallurgical activities such as mining increases the lead concentration of soil, its low solubility ensures that it is not a major contributor to human lead burdens (Cotter-Howells and Thornton, 1991). It is the use of lead artefacts and lead compounds rather than lead in soils that dominates human lead burdens in metallurgical societies, as the direct ingestion of lead and its products circumvents normal biopurification of the toxic metal (Elias et al., 1982; Katzenberg and Grauer, 2018, p. 508). Therefore, it is the isotope ratios of potential sources of anthropogenic lead pollution (e.g. lead ores) that will provide the most useful comparisons for human migration studies involving societies with metallurgical capabilities, such as the Romans (McBride et al., 2014; Weiss et al., 1999; Wuana and Okieimen, 2011).

It has been shown that lead ore isotope ratios vary throughout the world as they were formed at different geological times (McCrady, 1952; Sangster et al., 2000). Data such as that published by Sangster et al., (2000), demonstrate how the world's most important lead ore deposits exhibit varying <sup>206</sup>Pb/<sup>207</sup>Pb isotope ratios, ranging from 0.98 to 1.41. For example, one of the world's major producers of lead is the Broken Hill ore

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deposit in Australia which has characteristically low <sup>206</sup>Pb/<sup>207</sup>Pb isotope ratio of 1.04, starkly contrasts with the world's largest lead ore deposit, the Mississippi Valley Type deposit in North America which is younger than the Broken Hill deposit and has a higher <sup>206</sup>Pb/<sup>207</sup>Pb isotope ratio of 1.40. Yet, despite the multidisciplinary applications of lead isotope analysis for source tracing in both modern and archaeological contexts, large-scale isoscapes representing the distribution of lead and lead isotope ratios are scarce.

#### 2.3.2 Lead isoscapes

A European study conducted by Reimann et al. (2012), was one of the first to address the need for lead isoscapes on a continental scale. Their analysis of European lead concentrations in agricultural soils revealed a distinct boundary in concentration levels between North East and South West Europe; with Northern Europe exhibiting lower lead concentrations than Southern Europe (see Fig. 2.1). This boundary coincides with one of Europe's major tectonic borders (the Trans-European Suture Zone) and extends from the agricultural (A-horizon) soils down into the deep C-horizon soils. These regions of high lead concentrations, especially in Britain, appear to correlate with major lead mining zones and industrial areas. This North/South divide in lead concentrations is also echoed in the <sup>206</sup>Pb/<sup>207</sup>Pb isotope ratios presented in Reimann et al.'s (2012) study.



Figure 2.1 – Map of lead concentrations in European agricultural soils. Black line represents the approximate location of the Trans-European Suture Zone (adapted from Reimann et al., 2012, p.234)



Figure 2.2 – Map of lead isotope ratios in European agricultural soils. Black line represents the approximate location of the Trans-European Suture Zone (adapted from Reimann et al., 2012, p237-238)
The spatial distribution of <sup>206</sup>Pb/<sup>207</sup>Pb and <sup>207</sup>Pb/<sup>208</sup>Pb isotope ratios can be seen in Figures 2.2a and 2.2b respectively. Generally, Northern and Eastern European soils (Sweden, Finland Ukraine etc.), predominantly developed on Precambrian igneous rocks have low <sup>207</sup>Pb/<sup>208</sup>Pb and high <sup>206</sup>Pb/<sup>207</sup>Pb isotope ratios, while the inverse is seen in South Western Europe. There are, however a few localised exceptions with high <sup>207</sup>Pb/<sup>208</sup>Pb values that correlate with significant lead deposits in the area (Reimann et al., 2012). A common geological feature, the Trans-European Suture Zone (TESZ), dominates all three maps. This region represents the geological border between the old Precambrian craton in the North-East of Europe and the younger Palaeozoic platform in Western Europe (McCann, 2008, p. 358). The fact that geological and isotope systems share a common border of change stands to further highlight the intrinsic relationship between a region's lead concentrations and geogenic lead isotope ratios with the underlying geology and prevailing soils. However, these isoscapes are modelled using modern agricultural soil samples. While this type of data is clearly a valid and useful tool for the tracing of modern human origins in continental-wide, forensic contexts (Boyd, Jr. and Boyd, 2017; Kamenov et al., 2014; Kamenov and Curtis, 2017; Keller et al., 2016), both modern and historical anthropogenic lead pollution has resulted in their efficacy for tracing mobility in archaeological populations being questioned.

Studies have shown that environmental lead isotope ratios are significantly altered by anthropogenic contributions to the atmospheric deposition of lead (Farmer et al., 2002, 1999, 1997). Through the analysis of historical peat bogs, lake sediments and archival herbage, spatiotemporal changes in <sup>206</sup>Pb/<sup>207</sup>Pb isotope ratios have been identified, and these changes have been linked to human activities such as the burning of fossil fuels and the introduction of leaded petrol (Boutron et al., 1994; Mil-Homens et al., 2013; Renberg et al., 2000; Shotyk et al., 1996). Therefore, if lead isotope ratios in organic

materials are to be used as indicators of geographical location there is a real need to ensure that the samples are contemporaneous with those of the skeletal population being analysed. Keller et al., (2016) is one of the first studies to explore this with their production and application of spatiotemporal lead isoscapes using 19<sup>th</sup> and 20<sup>th</sup> century North American samples. This study found that lead isotope ratios from a combination of dated sediment, peat bog, coral, tree ring and lichen samples provided isotope ratios sufficiently similar to those in contemporaneous human remains for the purpose of origin identification. Keller et al., (2016) conclude that spatiotemporal isoscapes correlate more closely to human tooth enamel values than isoscapes produced using non-contemporaneous soil samples. Despite the apparent success of this study, very few others have attempted to make spatiotemporally specific lead isoscapes. Instead, it has remained more common practice to use lead isotope datasets produced from the analysis of artefacts of known provenance as proxies for the palaeo-pollution that would have contributed to anthropogenic archaeological human lead isotope ratios.

#### 2.3.3 Anthropogenic environmental lead pollution

Human activities have always had an impact upon the environment, and the mining and smelting of lead is no exception. Before the advent of metallurgical activities atmospheric lead levels remained consistently low (Bindler et al., 2008). However, palaeo-pollution studies have shown how human activities have caused significant fluctuations in these natural background concentrations (Bränvall et al., 2001). Numerous studies have used Icelandic salt marshes, Greenland ice cores, Scandinavian lake sediments, European peat bogs and historical herbage to characterise palaeopollution over time (Brännvall et al., 1997; Hong et al., 1994; Komárek et al., 2008; Marshall et al., 2009; Rosman et al., 1997). These studies show a low natural baseline level of atmospheric lead concentration in pre-history that steadily rises during the onset of metallurgical technologies around 500 BC. During the Roman period these levels dramatically rise, yet after the decline of the Roman Empire, during the  $5^{th} - 11^{th}$ centuries AD, palaeo-pollution records demonstrate a drop in atmospheric lead levels. However, a second surge in lead production during the industrial revolution in the  $18^{th}$ century induced a second increase in atmospheric lead concentrations, bringing about unprecedentedly high lead levels within the environmental record. Interestingly, the same temporal fluctuations in atmospheric lead pollution can be seen in the lead burdens of archaeological human remains.

This is best visualised by comparing the archaeological human data presented by Montgomery et al., (2010) with the atmospheric pollution data published by Settle and Patterson (1980) (see Fig. 2.3 and 2.4 respectively). Montgomery et al.'s (2010) data demonstrates how the median lead concentrations in human tooth enamel from British skeletal populations spanning from the Neolithic to the Late Medieval period have concentration fluctuations congruent with the fluctuations seen in historic lead production. As metallurgical activities increased, so did the lead concentrations in human tooth enamel. From as early as the Iron Age, tooth enamel has shown elevated lead concentrations ( $0.02 - 30.1 \text{ mg kg}^{-1}$ ) compared to those seen in pre-historic samples ( $0.003 - 0.68 \text{ mg kg}^{-1}$ ). This correlation between anthropogenic lead production and human lead burden illustrates how anthropogenic lead pollution is linked to human lead burdens, thus, providing a unique tool for assessing how technological advancements affected the health and environments of past populations.

# IMAGE REMOVED BY AUTHOR FOR COPYRIGHT REASONS

Figure 2.3 – Median lead concentrations from human skeletal material dating from the Neolithic to the late Medieval period (Source: Montgomery et al., 2010)



Figure 2.4 – Diachronic atmospheric lead pollution data (Source: Settle and Pattterson, 1980)

## 2.4 Uses in bioarchaeology

The use of lead isotope analysis in archaeology has primarily been developed as a discriminatory tool in artefact provenance studies, addressing questions about trade, movement of goods and the geological and geographical origins of manufacturing materials (Boni et al., 2000; Durali-Mueller et al., 2007; Henderson et al., 2005; Renson et al., 2013; Stos-Gale et al., 1997). Its development was founded on the premise that different lead ore sources have isotopic compositions sufficiently different from one another to allow differentiation between ores mined in different countries (Brill and Wampler, 1967). Comparison of lead isotope ratios from archaeological artefacts with the isotopic signature from ore deposits has been shown to successfully facilitate the identification of possible geographic origins of the metals used to make the artefacts (Albarède et al., 2012; Balliana et al., 2013; Delile et al., 2014; Desaulty et al., 2011; Niederschlag et al., 2003). The use of lead isotope analysis in bioarchaeology is based upon a similar assumption, mainly that the lead sequestered in skeletal material reflects the lead source a person was exposed to at the time of tissue mineralisation. Although this technique is not as widely used as other isotope systems in bioarchaeology, mostly due to cost and complexity of interpretation, a growing number of studies are demonstrating the efficacy of the technique in identifying migrants, especially in British and North American populations (Dudás et al., 2016; Keller et al., 2016; Millard et al., 2014; Montgomery et al., 2010; Shaw et al., 2016).

## 2.4.1 Lead in skeletal tissues

The weathering and dissolution of rocks facilitates the movement of lead into the surrounding soils. From here it can enter the food chain via incorporation into local water systems, plants and animals. With regards to humans, this means that the lead

sequestered within skeletal material represents an average of the isotopic composition of the food and water ingested during life (see Chapter 4 for more details). Due to their high mass, lead isotopes do not fractionate during the low temperature reactions associated with geological and biological processes, therefore lead isotope ratios in skeletal tissues tend to reflect those of the local geology or at least an integrated homogenous representation of the underlying geology (Erel et al., 1994; Komárek et al., 2008). This is particularly true for people from pre-metallurgical societies where lead burdens are consistently low (<1 ppm), and acquired almost entirely from naturally occurring, geogenic lead in the environment (Millard et al., 2014; Montgomery et al., 2010; Shaw et al., 2016). Subsequent societies exhibit increased lead burdens as a result of widespread environmental lead pollution created through the use of mining and metallurgical technologies. This anthropogenic lead pollution swamps the natural geogenic lead in an individual's local environment, resulting in lead isotope ratios reflecting those of the lead ore sources utilised by that particular society (Montgomery, 2002; Montgomery et al., 2010, 2014). Of course, in reality a society is likely to be exposed to more than one ore source, in which case human lead isotope ratios would reflect an average of the isotope ratios from all sources, essentially a mixed or homogenised isotope ratio. These homogenised isotope ratios are likely to show slight variations between individuals within a single population, but the variation will inevitably be smaller than the variation between the different ore sources (end members) contributing to the exposure (Wilson and Pollard, 2001). No matter what type of exposure (natural or polluted) the strong environmental links for both geogenic and anthropogenic lead burdens enables the interpretation of lead isotope ratios in conjunction with lead concentration data to be a powerful tool in human mobility studies.

#### 2.4.2 Variations in human lead isotope ratios

Human teeth and bones accumulate trace amounts of lead from the local environment as they grow and remodel. As lead isotopes do not fractionate as they move through the biogeosphere, the incorporation of these potentially geologically unique lead isotope compositions into mineral matrices provide isotopic compositions that are indicative of the geographical region resided in at the time of incorporation. Numerous studies have found that well preserved skeletal remains, especially teeth, are excellent archives for biogenic lead and have proven that lead isotope analysis of skeletal remains can be a useful tracer for sources of lead to which a population was exposed (Budd et al., 2004; Gulson et al., 1997; Kowal et al., 1991; Molleson et al., 1986; Montgomery et al., 2010, 2005).

The potential for using lead isotope ratios to discriminate between different cultural groups is demonstrated in Figure 2.5. Here, previously published lead isotope ratios from archaeological human samples are grouped by continent. It is evident that although each continent contains values that overlap with each other, the data points cluster in four distinct groups based on geographic location. The European samples show the tightest grouping, which is most likely due to the cultural focusing induced by the extensive mining, diverse use and reuse of lead throughout the Roman Empire and to some extent the Medieval period. These European values cluster around  $^{206}Pb/^{204}Pb = 18.44$ ,  $^{207}Pb/^{204}Pb = 15.61$  and  $^{208}Pb/^{204}Pb = 38.40$  which are congruent with the lead isotope ratios found in British lead ores. The samples from North America and South America plot much higher than the European samples, especially those from North America which exhibit the highest lead isotope ratios with averages of  $^{206}Pb/^{204}Pb = 19.3$ ,  $^{207}Pb/^{204}Pb = 15.70$  and  $^{208}Pb/^{204}Pb = 39.20$ , thereby making them easily distinguishable from the European, South America and Asian samples.

Although some of the South American samples overlap with the European field, the majority of the South American isotope ratios are higher than those from Europe and plot above  ${}^{206}Pb/{}^{204}Pb = 18.60$ . The samples from Asia show the greatest degree of overlap with the other continents. However, when all of the lead isotope ratios are considered, the Asian samples can be distinguished on the basis of their <sup>208</sup>Pb/<sup>204</sup>Pb isotope ratios. The average <sup>208</sup>Pb/<sup>204</sup>Pb isotope ratio for the Asian sample was 38.48, which is higher than the average for the European and South American samples (38.40 and 38.38 respectively) but considerably lower than the North American average of 39.20. This highlights the importance of analysing all four lead isotopes when using lead as a source-tracer in mobility studies, as it is not always a single value that proves to be sufficiently discriminant but rather a combination of the isotope ratio possibilities. From this it is evident that the lead isotope ratios incorporated into archaeological human teeth and to a lesser extent bones (see section 2.4.4), can reflect the local environment of the individual and can be a useful discriminant in mobility studies. While it is clear that archaeological individuals from different regions of the world are relatively easily distinguishable from one-another, especially North Americans from Europeans, there is still a lack of comparable data for countries within the same continent. Therefore, the resolution at which mobility can be identified via lead isotope analysis has yet to be determined.



**Figure 2.5** – **Continental variation in lead isotope ratios from archaeological human remains (Source: Author)**. Data published by Europe:(Åberg et al., 1998; Budd et al., 2004, 2000; Millard et al., 2014; Montgomery et al., 2005; Montgomery et al., 2010; Shaw et al., 2016). South America: (Turner et al., 2009). North America: (Bower et al., 2007). Asia: (Valentine et al., 2008).

It is also important to consider lead concentrations in conjunction with lead isotope ratios as they can often inform the interpretation of results. Low lead concentrations are usually accompanied by variable lead isotope ratios, which represent the natural lead acquired through diet and therefore reflects geographical origins. This trend is typical of prehistoric populations before the advent of metallurgical technologies (Montgomery et al., 2010). Therefore, source-tracing applications of prehistoric human remains can utilise lead isotope ratios in the same way as strontium isotopes to ascertain geological origins. However, anthropogenic activities such as mining and smelting have resulted in widespread lead pollution that has altered the natural lead isotope background of the environment, and this change in both isotope ratio and concentration is reflected in historical teeth (Kamenov and Gulson, 2014). Thus, samples with high lead

concentrations consistently exhibit narrow lead isotope ranges centred round the isotope ratios of the dominant lead ore source utilised by the population. High lead concentrations are predominantly found in societies with the technology to exploit mineral ore sources (Carlson, 1996; Montgomery et al., 2005). As a result, lead isotope ratios in metallurgical societies become less affiliated with natural baseline levels characteristic of geological origin and more indicative of exposure to pollutants (i.e. access to lead and its products). This clustering of lead isotope ratios as a result of anthropogenic lead exposure has been termed 'cultural focusing' (Montgomery, 2002).

### 2.4.3 Cultural focusing

The term 'cultural focusing' refers to the homogenisation of lead isotope ratios within an archaeological population, and usually occurs in conjunction with elevated lead concentrations (Millard et al., 2014; Montgomery, 2002; Montgomery et al., 2005). As these homogenised isotope ratios tend to have ranges congruent with those from local lead ore bodies, elevated lead concentrations and their resultant homogenised isotope ratios are thought to be induced by anthropogenic environmental lead pollution. This 'cultural focusing' alters the type of information that can be obtained from lead isotope analysis of skeletal remains. Shifting the lead isotope ratios in skeletal remains away from isotope ratios congruent with geographical provenance, towards isotope ratios much more indicative of a socio-cultural provenance (Carlson, 1996; Montgomery et al., 2005). This means that the metallurgically induced influx of anthropogenic lead into the environment effectively severs the link between skeletal lead isotope ratios and geographic origin and replaces it with isotope ratios converging around the predominant ore sources utilised in a particular cultural sphere. Despite the fact that cultural focusing potentially reduces the resolution of geographic variability of lead isotopes, these homogenised anthropogenic skeletal lead isotope ratios can still be effective in differentiating between cultural groups within skeletal populations. They are still capable of discriminating between individuals exposed to different sources of lead. Therefore, while the identification of movement within a specific country may not be possible, individuals exposed to foreign lead sources should stand out from those exposed to local lead, making migrants from other countries relatively easily identifiable in culturally mixed skeletal populations. The data published by Montgomery et al., (2010), and presented in Figure 2.6 best demonstrates this. As can be seen, when lead concentrations remain below 0.5 mg kg<sup>-1</sup> a divergent spread of lead isotope ratios is exhibited. However, when lead concentrations rise above this divergent threshold (dotted line on the graph) isotope ratio begin to cluster around a narrow range, in this case <sup>207</sup>Pb/<sup>206</sup>Pb isotope ratios of 0.845 to 0.849, which is in line with British lead ore isotope signatures. The outlier in this dataset (Spitalfields 4<sup>th</sup> century AD) exhibits a high lead concentration with isotope ratios inconsistent with British lead ore, suggesting that this individual had access to foreign sources of anthropogenic lead during childhood and therefore originates from somewhere outside of Britain. In fact, after comparison with the lead and strontium isotope ratios of three individuals from Rome, Montgomery et al. (2010) conclude that the Spitalfields outlier has isotope ratios consistent with a childhood origins in the Mediterranean.

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Figure 2.6 – Data from archaeological human remains demonstrating cultural focusing of isotope ratios wit increased lead concentrations (Source: Montgomery et al., 2010)

A more recent study by Millard et al., (2014) also found that increased lead concentrations resulted in a decreased spread of lead isotope ratios. Examining British post-medieval populations from London, Millard et al., (2014) found that the majority of individuals with high lead concentrations clustered within the expected lead isotope fields for British lead ores. However, this study determined the point at which human lead isotope ratios become dominated by anthropogenic contributions (culturally focused) to be 0.87 ppm, slightly higher than that suggested in the initial study by Montgomery et al., (2010). As yet, the current literature offers no explanations as to why different populations have different thresholds for the onset of cultural focused lead isotope ratios. Although, as environmental lead concentrations have shown to vary geographically (Reimann et al., 2012), it may simply be that the threshold for cultural focusing is also population/geographically specific.

#### 2.4.4 Contamination from the burial environment

As with all chemical analyses involving archaeological remains, the question of contamination from the burial environment is a major concern, especially in migration studies. The success of isotope analysis as a means of reconstructing residency and migration hinges upon the assumption that the analytically targeted elements represent those incorporated during life, not those acquired after death (Montgomery et al., 2000). The majority of archaeological remains are buried and therefore contact with possible contaminants is largely unavoidable. However, the extent to which diagenetic alterations impact upon chemical analyses can be controlled for by careful consideration of the sample material.

Certain intrinsic features of bone, such as porosity, density and surface area render bone highly reactive with its surroundings, readily exchanging mineral and organic elements with its burial environment (Hedges et al., 1995; Nielsen-Marsh and Hedges, 2000). Tooth enamel on the other hand is almost completely composed of mineral, effectively fossilised before burial. Therefore, enamel is far more kinetically stable than bone or dentine. Its dense structure and low porosity leaves limited opportunities for mineral infiltration and ion exchange between the enamel and its burial environment (Montgomery et al., 2000; Neuman and Neuman, 1953). Therefore, enamel is considered relatively stable and resistant to the diagenetic structural and chemical changes common in other skeletal tissues (Budd et al., 2000; Hoppe et al., 2003; Montgomery et al., 2007). As such, tooth enamel offers the best sample medium for chemical analyses as it greatly reduces the risk of post-mortem contamination, giving the highest likelihood of obtaining *in vivo* isotope ratios incorporated into the tissues (Ericson, 1993; Montgomery, 2002; Montgomery et al., 2010).

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#### 2.4.5 Lead concentrations

Lead concentrations in tooth enamel provide a measure of childhood exposure to environmental lead pollution, and can offer information beyond simply the extent of an individual's lead burden. As briefly discussed in section 2.4.3, when combined with lead isotope ratios, lead concentration analysis can facilitate differentiation between geogenic and anthropogenic lead exposure. Being able to distinguish between geogenic and anthropogenic lead exposure in this way offers insights into the technological capabilities and/or technologies actively utilised by a particular population. It is likely that populations exhibiting predominantly geogenic lead isotope ratios do not use lead or lead products to the extent necessary for the acquisition of high, anthropogenic lead concentrations. This is touched upon in the works by Montgomery et al. (2010), who demonstrated that prehistoric populations with limited metallurgical technologies have the lowest mean lead concentrations while retaining their divergent geogenic lead isotope ratios. Yet with the introduction of large-scale lead use, human lead concentrations increase, producing anthropogenic lead isotope ratios in technologically advanced societies. While little work has been done to thoroughly explore the extent to which this 'rule of thumb' holds true, to date studies utilising lead isotopes have shown that human lead burdens below 1 ppm in conjunction with non-ore lead isotope ratios appear to represent geogenic lead exposure and values over this are considered anthropogenic (Millard et al., 2014; Montgomery et al., 2010; Shaw et al., 2016).

Within polluted populations lead concentrations can also offer a means of differentiating between broad settlement types, such as rural or urban environments. The premise behind this is that rural environments are generally less polluted than urban environments, therefore individuals from rural settlements are likely to have had lower

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lead exposure and as a result, lower lead burdens than their urban counterparts (Lepow et al., 1975). Bioarchaeological investigations exploring how lead exposure differed between rural and urban environments are scarce. However, studies that have assessed lead concentrations between different environments have found that people from urban environments tended to have higher lead concentrations than people who lived in rural environments (Drasch, 1982; Millard et al., 2014). The same trend has been seen in Roman and pre-Roman societies, with individuals from Roman societies having higer lead concentrations than individuals from pre-Roman societies (Montgomery, 2002; Montgomey et al., 2010; Beherec et al., 2015).

The toxic nature of lead also means that lead concentration analysis can be used to explore the effects of lead exposure on the health of people from past populations. However, because tooth enamel is the only skeletal material to provide reliable *in vivo* lead concentrations, research focused on lead poisoning is limited to studies on childhood health. As teeth mineralise during childhood, the lead concentrations obtained from tooth enamel pertain to that period in life. Therefore, to accurately assess how these concentrations impacted upon health they can only be used in conjunction with palaeopathological alterations on non-adult skeletal material as both datasets are most likely to overlap with the time of lead exposure.

# 2.5 Summary

Natural geographic variations in lead isotope ratios make them an ideal tool for investigating the geographic origins of people in the past. The availability of three isotope ratios rather than the single isotope ratio available in other commonly used isotope systems (e.g. strontium), allows greater scope for differentiation in migration studies. Previous studies have demonstrated the potential for lead isotope ratios in bioarchaeological research and highlighted the need for contemporary comparative data for source identification. The paltry amount of published lead isotope data from archaeological human remains not only from the Roman period but from all time periods is a major limiting factor in the use of lead isotopes in human migration studies. Without datasets from archaeological populations from different countries there is no way to directly compare isotope ratios with human baselines in other countries. This forces the use of proxies such as artefacts or lead ores in their stead. While these have shown to be useful in the identification of approximate geographical origins of individuals with isotope ratios that do not conform to the expected isotope ratios of their interment region, their accuracy should be questioned, as the method does not account for the mixing of numerous lead sources. This is an inevitable component of human lead isotope ratios, especially in populations that exploited the use of the metal in all aspects of daily life. Therefore, human lead isotope ratios are not likely to reflect the isotope ratios of a single source such as local lead ore or artefacts of known provenance. Thus, the first steps in advancing the efficacy of lead isotope ratios in Roman migration studies would be the compilation of human lead isotope ratio datasets for various regions of the Roman Empire. This is an objective that this study will contribute to by analysing the lead isotope ratios in Roman skeletal remains from four countries from different regions of the Roman Empire.

# CHAPTER THREE

# Isotope Analysis of Roman Populations

# **3.1 Introduction**

In order to explore how environmental lead pollution impacted upon the health of past populations, as well as how anthropogenically induced changes to environmental isotopic ratios influence how these ratios can be used in mobility research the study population is required to fulfil two criteria. Firstly, the population must have engaged in metallurgical activities to elicit such a change in their environment, or have inhabited a previously polluted environment. Secondly, the population would ideally have been prone to migratory events as to allow for any potential differences between the isotopic composition of an individual's tooth enamel, which would reflect their former place of residence and the isotopic composition of the region in which they died.

This chapter discusses migration in the Roman Empire, highlighting the extent of movement underway during this time, and touching upon the motivations for such large-scale mobility. The ubiquitous use of lead during the Roman period will also be explored, with focus on how lead was used and how this may have impacted upon health. In doing so this chapter sets out to demonstrate how the Romans, who were somewhat characterised by their proclivity for widespread migration and large-scale lead mining throughout the Empire, provide the perfect target population for this study.

### **3.2 People on the move**

The Roman Empire was a vast suzerainty with large territorial holdings in Europe, Western-Asia and Northern-Africa, a contiguous expanse encompassing the entire Mediterranean Sea (Garnsey and Saller, 2014). It was one of the largest empires in history, spanning approximately 5 million km<sup>2</sup> (Taagepera, 1979), the limits of which are summarised rather poetically by historian Christopher Kelly:

"The Empire stretched from Hadrian's Wall in drizzle-soaked northern England to the sun-baked banks of the Euphrates in Syria; from the great Rhine-Danube river system, which snaked across the fertile, flat lands of Europe from the low country to the Black Sea, to the rich plains of the North African coast and the luxuriant gash of the Nile Valley in Egypt. The Empire completely circled the Mediterranean."

(Kelly, 2006, p. 1)

The Roman Empire was renowned for the widespread movement of people both within and between the varied geographic and cultural spheres it controlled (Killgrove, 2010). The movement of people both voluntary and forced via migration, military invasions, slavery or trade was a defining characteristic of the Roman Empire (Braudel, 1995; Horden and Purcell, 2000; Scheidel, 2004) and integral to its development and expansion (Birley, 1979; Hin, 2013; Scheidel, 2001a).

The relatively peaceful period during the period of the Roman Empire known as the *Pax Romana*, facilitated the movement of people across large expanses of the Empire with comparative ease (Moatti, 2006). People with the financial means to move could do so freely as a result of the contiguous geopolitical expanse and the well-established road and seafaring infrastructures the Empire provided (Killgrove, 2010, p. 27). Documentary evidence demonstrates that slaves, soldiers and civilians were migrating

from all regions of the Empire, with examples of migrants from France, Germany, Hungary, Spain, Syria and Egypt among others, commemorated on funerary monuments within Rome (Noy, 2000; Stark, 2017). It was not only those with the financial means to travel who moved freely throughout the Empire, migration is known to have been a part of all social strata with people of lower social status also migrating in search of better lives, (Sweetman, 2011; Tacoma, 2016, 2014; Woolf, 2013). The movement of people within this culturally, ethnically and geographically diverse population has been broadly separated into two categories, voluntary migration (e.g. free citizens) and compulsory/forced migration (e.g. military service or slavery).

#### **3.2.1 Voluntary migration**

Voluntary migration refers to people who have made a conscious decision to move, either short distances within their home province or long distances to a different region of the Empire. Although most likely biased towards wealthy, literate individuals from higher social classes, epigraphical evidence suggests that motivations for voluntary migration were overwhelmingly driven by either push or pull factors (Noy, 2000). Push factors relate to the region of origin and usually compel people to leave their homes in search of a better life in a different region of the Empire, a problem pushing them to move. Pull factors on the other hand are factors connected to the destination, something that entices an individual to make it their new home; a benefit pulling them to move (Killgrove, 2010; Noy, 2000; Stark, 2017). The motivations behind voluntary migration during the Roman period are poorly recorded, but it is unlikely that the decision to move was made on account of a singular reason. Undoubtedly a multitude of reasons played a part in compelling a person or family to uproot their lives and start afresh in a new region of the Empire of their own volition (Noy, 2000).

#### **3.2.2 Compulsory migration**

Compulsory migration refers to people who were forced to move, with no choice as to when or where they were sent. It is widely accepted that compulsory migration accounted for a large proportion of the movement of people within the Roman Empire, with members of the Roman army, slaves and their families relocated involuntarily through expulsion from their conquered homeland, enslavement or military deployment (Jongman, 2003; Scheidel, 2005; Tacoma, 2014). Slaves were integral to both the social and economic structure of the Empire, performing a diverse range of jobs encompassing everything from domestic services to highly skilled professions such as accountant or physician (Bradley, 1994, p. 2; Scheidel, 2010a). It has been estimated that from 50 BC to AD 150 the Roman Empire required over 500,000 new slaves every year (Bradley, 1994, p. 32). The sheer volume of slaves existing within its provinces has led to the Empire being termed a slave society or slave economy (Scheidel, 2010a). As Roman slaves were often captives of war they represent a population with vastly diverse origins. Historical evidence at Rome for example documents that the Parthian war supplied approximately 100,000 slaves to Rome from Iran, while the Punic wars provided the Empire's capital with upwards of 75,000 slaves from North Africa (Bradley, 1994, pp. 33–40). Not all slaves were captive prisoners of war, slaves were also acquired through piracy in the Mediterranean or traded or bought from other slave societies such as Sub-Saharan Africa or Egypt (Bradley, 1994, pp. 33–39). Infants were also often enslaved, either by being born to a slave mother (verna) in which case the infant belonged to the mothers' master, or infants turned out through exposure could be taken and raised in a life of slavery. However, the number of slaves acquired through exposure diminished during the 4<sup>th</sup> century AD after the sale of infants was authorised by Constantine (Harris, 1994).

Soldiers and their families also make up a large proportion of people considered as compulsory migrants. Recruitment of Roman military forces occasionally occurred en masse from a particular region of the Empire where soldiers would find themselves serving with people of a similar cultural background (Haynes, 1999). However, a multitude of conscripts and volunteers were often taken from the nearest convenient source, both within and beyond the Empires frontiers (James, 1999). As such, the ranks of the Roman army were rarely culturally homogenous. Evidence for the cosmopolitan nature of the Roman army is demonstrated on various military monuments such as the Tropaeum Traiani in Civitas Tropaensium, Dacia (Adamclisi, Romania) and the Birrens altars in Scotland, which attest to the diverse ethnic groups within individual regiments (Haynes, 1999; "RIB Online," 2014). The Roman army created large groups of highly mobile, multicultural people, and although military forces were largely concentrated at frontiers or regions of the Empire within which peace was not always guaranteed, they could be sent anywhere within the Empire at anytime (Noy, 2000).

#### 3.3 Finding the foreigners

In the 1990s David Anthony (1990) pointed out the value of mobility studies in fully understanding the movement of people in the past and urged archaeologists to utilise all available sources of evidence (material culture, historical records, skeletal remains etc.) to reconstruct patterns of human mobility. In the decades that followed, much of the work on migration had been predominantly theoretical, focusing on the large-scale movements of people from either prehistoric or post-Roman periods (Anthony, 1990; Burmeister et al., 2000; Champion, 1990; Härke, 1998). Advancements in bioarchaeological and biogeoarchaeological methods have facilitated a robust scientific approach that has rejuvenated interest in mobility studies (Bentley, 2006; Eckardt, 2010). Through the analysis of the physical, chemical and molecular features of skeletal remains, archaeologists can now provide empirical evidence for migration rather than simply offering theoretical arguments for the movements of people in the past (van Dommelen, 2014).

The analysis of the funerary practices (material culture, monuments, epitaphs, grave type, skeletal remains etc.) of archaeological civilisations is the most accessible way of assessing migration and mobility within past populations. Historical documents have recorded many migratory events in tomes such as Bede's Ecclesiastical History of the English People and the Anglo-Saxon Chronicles (Colgrave and Mynors, 1993; Points, 2013). These types of sources are often written decades if not centuries after the initial event and are therefore not always entirely accurate. As a result of this, current archaeological studies tend to focus more on where and how individuals were buried, piecing together biographical information gleaned from the evidence of funerary practices surviving in the archaeological record (Pearce, 2000).

## **3.3.1 Burial rites**

Many archaeological studies on migration within the Roman world have attempted to differentiate between migrant new comers and the native population from a given site by interpreting changes in local burial practices as manifestations of Romanisation. However, such deductions from these so-called 'intrusive' burial practices have been criticised for assuming a direct link between burial practice and individual ethnicity (Halsall, 2011, 2010; Willems, 1978). Burial rituals throughout the Roman Empire have been shown to vary spatially and temporally at both local and provincial levels (Pearce, 2010, 2000). For example, during the early Roman period furnished cremations were the predominant rite in western provinces, while inhumation dominated in the east and

as the Empire transitioned into the late Roman period the frequency in the deposition of grave goods decreased significantly (Pearce, 2010, 2000). While there is clearly value in considering this type of evidence when assessing human mobility in order to successfully characterise a burial as indicative of a migrant a secure understanding of the predominant preceding burial rite is crucial, and as the evidence for late Iron Age funerary practices is sporadic (e.g. plentiful in Moselle, yet scarce in Bavaria) current knowledge of the 'indigenous' burial rites for much of the Empire is limited (Fasold, 2000; Fitzpatrick, 2000).

Additionally, recent diaspora studies have highlighted that an individuals' culture is not static, rather it has an intrinsic fluidity, and can be altered by interactions with other cultures. This creolisation results in a 'blending' of cultures creating societies with a new culture distinct from both the host population and their original homeland, where identities are formed around commonalities such as profession or status rather than ethnicity (Eckardt, 2010). Essentially, the diasporic nature of the Roman Empire has blurred the lines between what has traditionally been seen in the archaeological record as indicative of foreign in origin and what is actually an expression of a complex integrated identity. Therefore, areas of study that exploit more direct links between what survives in the archaeological record and an individual's geographic origin are coming to the forefront in mobility studies (Fitzhugh, et al., 2019).

### **3.3.2** Isotope analyses

For decades non-destructive methods of osteological analysis have been the standard way of identifying groups of affiliated individuals, working on the premise that populations displaying the most similarity are the most closely related (White and Folkens, 2005). However, with advancements in analytical techniques a new wave of bioarchaeological studies of migration are harnessing the discriminatory powers of stable and radiogenic isotope systems extracted from archaeological teeth and bones to answer questions about geographic origins at an individual and population level. It has been well established that the isotopic composition of archaeological teeth and bone reflects the isotopic composition of the food and drink consumed at the time of new bone formation, which in turn reflect the region in which they were sourced (Katzenberg, 2008; Kohn, 1999; Richards et al., 2006; Van der Merwe and Vogel, 1978). Therefore the isotopic characteristics of an individual represent a weighted average of the isotope compositions they have ingested from their local environment. Advancements in stable and radiogenic isotope analysis methods have exploited this concept and taken it beyond the reconstruction of palaeo-diets, significantly transforming the way in which mobility studies can establish patterns of migration in the past.

To date, the isotopic systems of several elements, most notably carbon, nitrogen, oxygen, sulphur, strontium and lead, have been developed as possible indicators of movement in past populations (Lightfoot and O'Connell, 2016; Montgomery et al., 2010; Nehlich, 2015; Slovak and Paytan, 2012). Mobility studies using these isotope systems are reliant upon regionally distinct isotopic compositions that allow differentiations to be made between childhoods spent in local or foreign locations (Katzenberg, 2008). One of the major constraints of using isotope systems in mobility studies is no one region produces a unique signature. Often a number of geographical areas with similar geology and climate overlap in their isotopic characteristics, rendering the inference of geographical origins difficult. As such, isotope characteristics can be more suited to excluding regions of origin, rather than definitively assigning a

specific geographical origin (Bruun, 2010; Knudson and Price, 2007; Prowse et al., 2007).

## 3.3.2.1 Stable light isotope systems

Stable light isotope analysis of carbon ( $\delta^{13}$ C) and nitrogen ( $\delta^{15}$ N) is a well-established technique for the reconstruction of palaeo-diets using archaeological teeth and bones. Within these two tissue types, collagen is the predominant protein present and provides a rich source of averaged  $\delta^{13}$ C and  $\delta^{15}$ N values. The composition of these  $\delta^{13}$ C and  $\delta^{15}$ N values reflect the isotopic composition of the foods consumed during the tissues formation (Kohn, 1999; Richards et al., 2006; Schoeninger et al., 1983; Van der Merwe and Vogel, 1978). This is due to the metabolism and reuse of dietary proteins acquired from the plant and animal products consumed during life in collagen synthesis. Therefore, once metabolic fractionation has been accounted for, these carbon and nitrogen isotope ratios can be used to garner insights into not only the types and quantities of food resources utilised by a population, but also the socioeconomic and cultural influences surrounding the dietary practices of groups as well as individuals (Dietler, 2007).

The variations that arise in  $\delta^{13}$ C values result from differences in ecosystems (marine vs. terrestrial) and the photosynthetic pathways (C<sub>3</sub> and C<sub>4</sub>) used by plants in their manufacture of carbohydrates. As such, variations in carbon values allows differentiation between the relative contribution of C<sub>3</sub> or C<sub>4</sub> plants and the animal products based on these plants, to diet (Ambrose et al., 1997; Beaumont et al., 2013; Camin et al., 2008). In temperate regions, such as Britain, plants tend to use the C<sub>3</sub> pathway. Therefore, isotopic evidence of dietary proteins based on C<sub>4</sub> plants (e.g. maize or millet) not native to Britain, would suggest a foreign influence on diet within a

British population. Variability in nitrogen isotope values reflects the balance between biological nitrogen fixation, biosphere recycling and nitrogen release (Robinson, 2001). This variability facilitates the visualisation of terrestrial and marine food source input into diet as marine-based food sources tend to be more enriched in <sup>15</sup>N than land based food sources (Liu and Kaplan, 1989). In addition to this, nitrogen levels also vary with trophic level, as a result of metabolic fractionation, creating a 2-6 ‰ enrichment with every trophic level shift (Schoeninger and DeNiro, 1984). This shift is most noticeable in marine food consumers as aquatic food sources have high  $\delta^{15}N$  values owing to the relatively long food chains compared to those observed in terrestrial food sources (Tykot, 2004). As such, combining the analysis of  $\delta^{13}C$  and  $\delta^{15}N$  values can facilitate the interpretation of plant, animal and marine protein contributions to palaeo-diets, allowing for distinctions to be made between broad categories of food groups, such as herbivore vs. carnivore or marine vs. terrestrial input and any potentially non-local influence on diet (Ambrose and Katzenberg, 2006), and published studies of Roman populations have demonstrated this (Chenery et al., 2011; Müldner et al., 2011; Pollard et al., 2011a)

Oxygen incorporated into hydroxyapatite is predominantly derived from ingested fluids, the isotopic composition of which fluctuates due to climatic and environmental variables such as temperature, rainfall, altitude and latitude (Darling and Talbot, 2003). Therefore, oxygen isotopes ( $\delta^{18}$ O) measured in human tissues are an indirect reflection of the local meteoric water composition (Kohn, 1996). As with carbon and nitrogen, oxygen also undergoes metabolic fractionation once ingested. Therefore, regression formulae must be applied to allow comparison with modern drinking water values. These calculated  $\delta^{18}O_{dw}$  values reflect the composition of the local meteoric water (Kohn, 1996). Thus, analysis of  $\delta^{18}O$  from tooth enamel can provide information pertaining to childhood origin and palaeo-climate ( Chenery et al., 2012; Fricke et al., 1995). However,  $\delta^{18}$ O values can be influenced by culturally mediated behaviour. If a significant proportion of the individual's drinking water was processed before ingestion (boiled, brewed etc.) a higher  $\delta^{18}$ O value than expected for the geographical region in question would be obtained (Brettell et al., 2012). One of the first studies to apply oxygen isotopes in human mobility studies was carried out on archaeological populations in Mexico (White et al., 1998). Following this, the use of oxygen isotope analysis became popular means of assessing geographic origins. It has been widely used in archaeological populations from all periods and diverse geographical locations (Budd et al., 2003; Buzon and Bowen, 2010; Dupras and Schwarcz, 2001a; Emery et al., 2017; Hoogewerff et al., 2001; Keenleyside et al., 2011; Mitchell and Millard, 2009; Pearson et al., 2016; Prowse et al., 2007; Schroeder et al., 2009; Turner et al., 2009).

## 3.3.2.2 Radiogenic isotope systems

A more direct link between migrant and geographic origin can be obtained through the analysis of strontium (Bentley, 2006; Montgomery, 2010; Montgomery et al., 2007; Price et al., 2002, 1994) and lead (Carlson, 1996; Gulson et al., 1997; Montgomery, 2002; Montgomery et al., 2010). Strontium has four stable isotopes, <sup>88</sup>Sr, <sup>87</sup>Sr, <sup>86</sup>Sr and <sup>84</sup>Sr, all of which occur naturally in a large variety of rocks and mineral deposits (Dickin, 2005; Faure, 1986; Nakano, 2016). The amount of <sup>88</sup>Sr, <sup>86</sup>Sr and <sup>84</sup>Sr within a geological deposit remains constant from the time of its formation. However, as <sup>87</sup>Sr is the daughter isotope of rubidium (<sup>87</sup>Rb) the abundance of <sup>87</sup>Sr increases over time (Bentley, 2006; Capo et al., 1998; Faure, 1986). Therefore, the strontium isotope ratio obtained from any given geological deposit will vary depending on the age of the deposit and the relative abundance of Rb/Sr it contained at the time of its formation.

(Bentley, 2006). For example old, silica-rich granites provide higher <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios than younger, low Rb/Sr basalts (Faure and Powell, 2012; Rogers and Hawkesworth, 1989). As different geographical regions are composed of varying types of rocks of differing ages, each geographical region produces elemental isotope ratios characteristic of its underlying geology. This in effect creates an isotopic fingerprint that allows strontium to be used as source tracer in mobility studies.

Erosion of rocks and minerals facilitates the movement of strontium into the surrounding soils where they can be incorporated into local water systems, food chains and ultimately human skeletal tissues. Assuming that the majority of a populations food and drink is sourced locally, the strontium isotope composition in human skeletal material should then reflect the bioavailable strontium isotopes in their region of origin (Ericson, 1985; Price et al., 1994b; Sealy et al., 1991; Sealy et al., 1995). The first applications of strontium isotope analysis to assess human childhood origins was carried out by Ericson (1985, 1989) and subsequently by Sealy (1989) on prehistoric Californian and South African populations respectively to identify individuals with coastal versus inland childhood origins respectively. This work was subsequently built upon with a number of studies investigating migration within prehistoric populations in both North and South America (Ezzo et al., 1997; Price et al., 1994b, 2000), Britain (Montgomery et al. 2000) and Germany (Grupe et al., 1997; Price et al., 1998, 1994a; Schweissing and Grupe, 2000). This extensive body of work has affirmed that strontium isotope analysis is an effective tool for investigating human migration within archaeological populations, especially when used in conjunction with contextual information. The advancements made in strontium isotope analysis by Price et al., (2002) and Montgomery (2002) has led to its continued use in a large number of mobility studies ranging from pre-Hispanic populations in the Andes (Knudson et

al. 2005, 2004), and Norse settlements in Scotland (Montgomery et al. 2003) to Viking Iceland (Price and Gestsdóttir, 2006) and various Mediterranean settlements (Nafplioti 2008, 2012; Haverkort et al. 2008; Lê 2006). The efficacy of strontium isotope analysis alongside contextual information has led to its use as one of the primary methods to explore human mobility in archaeological studies (Knudson and Price, 2007b, p. 25).

As with strontium, lead isotope ratios also vary with respect to regional geology, and are incorporated into human tissues via diet in preindustrial societies. However, populations that exploited lead, such as the Romans, tend to have anthropogenic lead isotope ratios that reflect the ore sources being utilised in their cultural sphere (Montgomery 2002; Montgomery et al., 2005). As discussed in chapter 2, anthropogenic lead exposure tends to be accompanied by a homogenisation of a given societies lead isotope ratios. Therefore, any discordant isotope ratios standout from what would be considered the norm for the population, allowing for relatively easy identification of migrants in polluted populations. As such, lead isotope ratios in metalusing societies are well suited for exploring questions of long-distance migration, whereas the efficicy of strontium isotope ratios entirely depends upon local geology, but can be especially useful when assessing short-distance migration in geologically heterogeneous area. Despite their usefulness in identifying migrants in culturally mixed skeletal populations (Montgomery et al., 2005, 2010; Montgomery, 2002; Shaw 2016), lead isotopes have not been widely used in archaeological mobility studies.

#### 3.3.2.3 Multi-isotope approach

The progression of isotope analyses in migration studies has led to an emphasis on employing multiple isotope systems in attempts to obtain a higher resolution in the possible geographical origins of the individuals analysed. Over the past decade the growing number of studies applying a multi-isotope approach has demonstrated its improved ability for identify non-locals in archaeological populations (Jay et al., 2013; Laffoon et al., 2017; Lamb et al., 2012; Müldner et al., 2011, 2009; Oelze et al., 2012; Sehrawat and Kaur, 2017). Typically these studies have used various combinations of  $\delta^{13}$ C,  $\delta^{15}$ N,  $\delta^{18}$ O and  ${}^{87}$ Sr/ ${}^{86}$ Sr analyses. However, the incorporation of lead isotope analysis in mobility studies is becoming increasingly popular (Montgomery, 2002; Montgomery et al., 2010, 2005, 2000; Shaw et al., 2016). What all of these studies have demonstrated is that where one isotope system tends to identify multiple geographic areas of possible origin, the utilisation of additional isotopes systems can greatly narrow down the possibilities. This is because each isotope system reflects a particular variable, whether that be dietary, climatic or geological, and the combined geographical differences in these variables can facilitate greater regional discrimination (Knudson and Price, 2007). While there are many examples of where a multi-isotope approach has been advantageous in refining the geographic origins of archaeological populations (White et al., 2007), it does not provide a perfect means of alleviating the difficulties in isolating geographic origins. Some studies have shown that despite employing a multitude of isotope systems the geographic origins of some individuals can still remain ambiguous, especially where migration was taking place between regions with similar environments and cultural practices (Knudson and Price, 2007).

#### **3.3.3 Isotopic evidence for Roman migration**

Isotope studies have provided strong evidence for high levels of migration within the Roman Empire (Millett et al., 2016, p. 209; Prowse et al., 2007; Schweissing and Grupe, 2003). The introduction of isotopic analyses to this area of research has broadened the

scope of mobility studies, and facilitated the answering of more nuanced questions. No longer limited by the innate biases associated with historical texts and epigraphical evidence, which favour society's elite, a more inclusive understanding of migration during this period can be obtained. A good example of this is the age-related migration study by Prowse et al (2007), demonstrating through the use of oxygen isotopes that entire families were on the move rather than the assumed theory that migration was predominantly undertaken by adult males.

A large proportion of multi-isotope studies exploring migration during the Roman period have been carried out on Romano-British populations (Chenery et al., 2010, 2011; Eckardt et al., 2009; Evans et al., 2006; Hughes et al., 2014; Leach et al., 2010, 2009; Montgomery et al., 2011; Müldner et al., 2011; Pollard et al., 2011). The majority of these studies have been carried out at major urban or military settlements, such as York and London, and many of them have identified migrants with diverse and farreaching origins, demonstrating the diverse nature of the Roman Empire. Leach et al., (2009) used FORDISC alongside strontium and oxygen isotope analysis to verify the previous craniometric identification of North African and Middle Eastern individuals at Trentholme Drive and The Railway, York (Warwick, 1968). This multi-analytical technique was unable to identify any Middle Eastern origins but did find craniometric evidence for individuals consistent with North African origins. From their isotope data Leach et al., (2009) also identified four outliers with isotope characteristics indicative of origins in North Africa, Western Europe or the Mediterranean, but could not narrow down the geographic origins any further. A more recent study, building on earlier work at Driffield Terrace (Montgomery et al., 2011, 2010; Müldner et al., 2011), combining genome and multi-isotope data provided compelling evidence for an individual buried at Driffield Terrace, York (3DRIF-26) with childhood origins in the Middle East (Martiniano et al., 2016). A multi-isotope study into population diversity of a Roman Gloucester population usitilising  $\delta^{13}$ C,  $\delta^{15}$ N,  $\delta^{18}$ O and  ${}^{87}$ Sr/ ${}^{86}$ Sr was carried out by Chenery et al. (2010). The authors found the carbon, nitrogen and strontium results to be inconclusive with regards to identifying possible migrants. However, the oxygen values obtained from the population indicated that at least six individuals were nonlocals with possible origins in regions of the empire with a warmer climate than Britain (Chenery et al., 2010). Redfern et al., (2016) conducted a study on 22 individuals from Lant Street, London to investigate the geographic origins of the group using macromorphoscopics, carbon, nitrogen and oxygen isotopes. From this small population skeletal morphometrics identified four individuals with possible African ancestry and two with possible origins in Asia, while the isotope results suggest a further five individuals with origins within the circum-Mediterranean (Redfern et al., 2016). The authors conclude that their study adds to the growing wealth of isotopic evidence for Roman migration but that the addition of strontium and lead isotopes would help refine the identification of places of origin and strengthen their conclusions. Of course, Britain was not the only region of the Empire home to a diverse, multi-cultural population. Multi-isotope studies have also identified non-locals within Roman populations in Italy (Killgrove, 2013; Killgrove and Montgomery, 2016; Montgomery et al., 2010; Prowse et al., 2007, 2004), Egypt (Dupras and Schwarcz, 2001) and Bavaria (Schweissing and Grupe, 2003).

What is clear from these examples of Roman migration studies is that the majority of them use the conventional combination of oxygen and strontium analyses to identify possible migrants. Very few studies use lead as a discriminant despite its potential for refining regions of geographic origin. Those that have included lead in their interpretations have found it useful in further constraining possible places of origin (Montgomery et al., 2010; Shaw et al., 2016). The first application of lead isotopes to answer question of Roman migration was carried out by Molleson et al., (1986) on bone samples from individuals from Poundbury Camp, Dorset. From the four individuals analysed a child was identified as a possible non-local with lead isotope ratios inconsistent with British lead ore, from which it was concluded that the child was a migrant to England from the Laurion region of Greece (Molleson et al., 1986). However, Montgomery (2002), in the first lead isotope study on enamel from Roman-period individuals, points out that this 'Greek' isotope ratio actually sits on a mixing line between the local Cretaceous chalk in Dorset and English lead ore. As such it is likely that the highly porous nature of the non-adult bones facilitated diagenetic ion incorporation directly from the burial environment that resulted in a natural geological isotope ratio rather than one deriving from anthropogenic exposure to English ore lead (Montgomery, 2002).

Tooth enamel has shown to be largely resistant to diagenetic alteration and has therefore been the target sample material for more recent lead isotope studies. One such study by Shaw et al., (2016), analysed strontium and lead isotope ratios to assess geographic origins in a Roman London population. This study identified three non-locals, one child with possible origins in the Rhine Valley and two adults with isotope characteristics consistent with Mediterranean origins. Shaw et al., (2016) conclude that lead isotopes were valuable in terms of refining potential areas of childhood residency and significantly aided in the interpretation of geographic origins where homogenous strontium isotope ratios was inconclusive. Another example of the efficacy of lead isotopes is the study of an elaborate lead coffin burial from Spitalfields, London (Montgomery et al., 2010). Again, the strontium isotope result was relatively undiagnostic due to the widespread occurrence of the Mesozoic terrains with which the isotope ratio is associated. However, the lead isotopes obtained from the individual fell within the field of silver dienarii from Roman mints as well as contemporaneous individuals from Rome and suggests that the female originated from Italy, possibly even Rome (Montgomery et al., 2010).

# **3.4 Lead as a commodity**

Despite there being evidence for the use of lead in the form of lead glazed pottery and lead figurines from as early as 4000 BC (Rich, 2014, p. 4), it is the Romans over 3000 years later that come to the forefront of any discussions pertaining to the ancient exploitation of the metal. This is likely due to the magnitude of lead mining underway during this period. Unlike during earlier periods, the Romans engaged in large-scale lead mining for the extraction of the metal in its own right, rather than acquiring it as a by-product from the mining of more precious metals such as silver (Bayley, 1992; Tylecote, 1992).

Lead was used heavily in the municipal water systems throughout the Roman Empire. Water storage and distribution systems such as cisterns and aqueducts were lined with vast numbers of lead sheets. For example, it is reported that 12 tonnes of lead was used in the construction of just one of the pumps at an aqueduct in Lyons (Waldron et al., 1979). Lead pipes and lead seals were also extensively used in plumbing systems distributing water to a range of buildings, including public baths, fountains, stadiums, theatres, temples, and private housing (Ortloff and Crouch, 2001). Drinking water was not the only potential source of lead contamination for Romans, tableware and cookware also contained a high proportion of lead (Hornborg et al., 2007, p. 37). Pewter, a lead and tin alloy, was often used to make dining and kitchen utensils (Needleman and Needleman, 1985; Waldron, 1973). It was also popular to line pots and pans made of

bronze or copper with lead to prevent food taking on an undesirable metallic taste from the cookware (Retief and Cilliers, 2006).

Besides its use in Roman water systems, the lead content of wine is possibly one of the most well documented ways in which the Romans utilised lead. Wine was often sweetened with *sapa*, *defrutum* or *caroenum*, which were preservative syrups made by boiling down grape must in lead-lined kettles (Eisinger, 1982). Wine was not the only consumable regularly tainted with the toxic metal, sugar of lead (lead acetate) was a popular additive in many recipes to add sweetness to the dishes. Translations of the only surviving recipe book from the period demonstrate how commonplace sweetening foodstuffs with lead was, with one fifth of Apicius' 450 recipes calling for the inclusion of sugar of lead (Vehling, 2012).

The Romans also made use of lead in cosmetics and medicines. Lead compounds such as white lead (lead carbonate) and red lead (lead tetraoxide) were commonly used in face powders to lighten the skin and as rouge respectively. Kohl, made from finely ground galena (lead sulphide), was also a popular eye makeup during the period (Bergman, 2017; Needleman and Needleman, 1985; Retief and Cilliers, 2006). With regards to medicines, there is documentary evidence describing the use of white lead in salves and eye ointments (Needleman and Needleman, 1985; Westhrop, 2011) and the inclusion of yellow lead (lead oxide) as an ingredient in many topical treatments for dermatological diseases and anti–wrinkle creams (Bergman, 2017; Gilfillan, 1965; Retief and Cilliers, 2006). White, yellow and red lead were also used to make pigments and paints (Gooch, 2014, p. 15). Finally, children's toys such as dolls, figurines and *crepundia* (necklaces incorporating rattles or trinkets designed to keep children quiet, especially during teething) were often made from pewter or lead (Gilfillan, 1965, p. 58;

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Roberts, 2009, p. 49). Hand to mouth activity is a common route of lead acquisition, especially in children (Baghurst et al., 1992; Bellinger et al., 1986; Bornschein et al., 1985; Freeman et al., 2001; Lanphear et al., 1996; Muller et al., 2018). Therefore, the use of lead in toys, especially those made purposefully for teething infants who are the most susceptible to lead poisoning was a particularly hazardous use of the metal. While lead was by no means limited to these uses, the above examples demonstrate the multifarious ways in which Roman individuals of all ages could come into contact with lead in a manner that would have resulted in bioaccumulation of the toxic metal.

# 3.5 Unwittingly poisoned

As outlined above (section 3.4), the Romans used lead for a multitude of social and industrial practices. Consequently, the Romans were the first to significantly contribute to widespread environmental lead pollution and as a result also unintentionally increased human lead burdens (Montgomery, 2002; Montgomery et al., 2010; Settle and Patterson, 1980). It is this previously unprecedented accessibility to the metal in all social strata that makes the idea of widespread lead poisoning throughout the Empire a popular topic of debate. Scholars have argued both for and against lead playing a significant role in the fall of the Roman Empire (Cilliers and Retief, 2014; Gilfillan, 1965; Nriagu, 1983b; Retief and Cilliers, 2006; Scarborough, 1984). A more recent approach to this issue does not set out to prove or disprove the role of lead in the downfall of Rome, but rather attempts to quantify the extent of lead exposure in Roman provinces and assess the impact it may have had on health within these regions (Delile et al., 2014; Le Roux et al., 2005; Mackie et al., 1975; Molleson et al., 1986; Véron et al., 2006; Waldron et al., 1976; Whittaker and Stack, 1984).
The preference for making wine in lead vessels is well documented, with Roman writers such as Pliny the Elder, Columella and Cato all advocating for its use (Columella, 1989; Eisinger, 1982; Mould, 1996; Waldron, 1973). Putting these ancient methods into practice, a study by Hofmann (1883) found that boiling grape must in lead lined kettles produced a sapa with a lead concentration of 237 µg/L. A further study in which Hofmann (1918, p. 638) followed the preparation instructions for two types of wine and grape must by the Roman writer Columella, produced liquids that contained between  $390 - 788 \ \mu g/L$  of lead (Mould, 1996, p. 67). From these results it is clear that the Roman methods for fermenting, sweetening and preserving wines created products containing harmful concentrations of lead. Boeckx (1986) has estimated that drinking wine prepared in this way could have led to the average Roman ingesting between 35 -320  $\mu$ g/Pb per day, and members of the aristocracy ingesting as much as 160 – 1520 µg/Pb per day. This toxic preparation technique seems to have been the norm as Pliny the Elder wrote that wine contained lead more often than not (Westhrop, 2011). Adults were not the only people effected by the lead in wine, there is documentary evidence that children were also consuming the beverage (Laes, 2011, p. 81). The physician Soranus advocated for the use of breadcrumbs soaked in wine as a weaning food (Soranus: Translated by Temkin, 1956), and the wirings of Galen, Aristotle and Hippocrates suggest that allowing children to drink wine was commonplace (Garnsey, 1999, p. 107; Laes, 2011, p. 81). As it was commonly consumed daily and accessible to everyone, wine is likely to have been a major route of lead exposure throughout the Empire, especially if the concentrations above are to be believed. This provides compelling evidence for the possibility that varying degrees of lead poisoning were being suffered throughout the Empire, especially when considering that wine was not the only route of exposure.

The extensive use of lead pipes in the Roman Empire is often suggested as a significant source of lead exposure throughout the period (Gilfillan, 1990; Hodge, 1981; Nriagu, 1983b; Retief and Cilliers, 2006). It appears that the Romans were also aware of the hazards of its use in water systems. Vitruvius, a prominent Roman architect and engineer was critical of the use of lead in water systems, and explicitly warned against its use (Eisinger, 1982; Hodge, 1981). Instead he advocated for the use of clay or terracotta pipes to distribute water (Eisinger, 1982; Hodge, 1982; Hodge, 1981).

'Water is much more wholesome from earthenware than from lead pipes. For it seems to be made injurious by lead because ceruse is produced by it; and this is said to be harmful to the human body ... Therefore, it seems that water should not be brought in lead pipes if we desire to have it wholesome.'

#### (Vitruvius in Hodge, 1981)

Despite these warnings lead piping was used to distribute water over long distances, from aqueducts and *castellum aquae* into people's homes and other municipal buildings (Waldron, 1973). Kobert (1909) reasoned that evidence of poisoning from 18<sup>th</sup>-century lead pipes, which ran from inside the home to the main cast iron pipes in the street was proof enough that Romans using lead piping over much longer distances would have also suffered its ill effects. Aside from Koberts (1909) bold declaration, there is little other research that explicitly states that lead pipes caused widespread lead poisoning. Rather it appears that until recently it was simply accepted that lead poisoning was widespread and that drinking water could be attributable to the cause (Gilfillan, 1990; Waldron, 1973).

A more recent study employing lead isotope analysis attempts to quantify the level of lead exposure from Roman water systems. A study of sediment core samples from the Tiber River and Trajanic Harbour by Delile et al., (2014) suggests that drinking water distributed using Roman leaded water systems would have contained up to 100 times more lead than water sourced from local springs. Even lead seals on clay and terracotta pipes have been shown to release between  $210 - 390 \,\mu\text{g/L}$  of lead into the water passing through them (Cosgrove et al., 1989). Modern guidelines for water quality recommend that lead concentrations do not exceed 5 µg/L in drinking water (WHO, 2011). Therefore these studies certainly suggest that Roman populations were exposed to harmful levels of lead in their drinking water. Arguments against Roman leaded water systems posing a significant risk to health propose that the build-up of sinter (calcium carbonate) deposits inside water pipes, cisterns and aqueducts, especially in areas of hard water, would have created a protective lining separating drinking water from the toxic metal (DWI, 2013). In fact, historical documents show that sinter build-up was so prevalent that its removal was a fulltime occupation, carried out predominantly by slaves (Hughes, 2011), with build-up rates estimated to be up to six inches per year if left to accumulate naturally (Hodge, 2013, p. 290, 2002, p. 8). Together with the biologically antagonistic nature that exists between lead and calcium, sinter would have presumably provided a buffer against the bioaccumulation of lead from drinking water (Rosborg, 2016, p. 126). However, studies by the World Health Organisation have shown that the flaking of sinter into a water supply can contaminate the water, significantly increasing its lead concentration even when the water is no longer plumbosolvent (WHO, 2011).

It is evident that lead, whether ingested intentionally (e.g. wine) or unwittingly (e.g. water), was highly accessible to everyone within the Roman Empire, and exceeded lead concentrations that are considered safe today. However, it is uncertain to what extent these sources of lead impacted upon health. There is no doubt that the Romans knew of

leads toxicity, with many of the authors of the time writing of its poisonous nature. Pliny the Elder noted that both red lead and lead acetate were deadly poisons and should not be used medicinally, while Celsus, a  $2^{nd}$  century philosopher, wrote of an antidote for white lead poisoning (Celsus, 1989, p. 125; Hodge, 1981). However, there appears to have been a cognitive dissonance between the acknowledgment of lead's toxic nature and its use in food and drink. The most compelling evidence for lead poisoning in the Roman Empire comes from Paulus Ægineta, a Greek physician who is credited as describing the first account of a lead poisoning pandemic (Ægineta, 1847; Waldron, 1973).

"... the colic affliction which now prevails is occasioned by such humours; having taken its rise in the country of Italy, but raging also in many other regions of the Roman Empire, like a pestilential contagion, which in many cases terminates in epilepsy, but in others in paralysis of the extremities."

(Ægineta, 1847, p. 534)

Roman texts often mention the symptoms of lead poisoning, especially colic and constipation, both of which are concomitant with the affliction, but do not attribute their causes to lead (Lessler, 1988; Needleman, 2009; Waldron, 1973). Therefore, it is modern interpretations of Roman texts that form the basis of any speculations on the clinical effects of lead poisoning experienced by Roman populations. For example, the madness of Emperors Caligula and Nero may be attributable to the central nervous system effects of lead poisoning (Gilfillan, 1965; Nriagu, 1983b); gout among the aristocracy could have been caused by lead-induced kidney damage (Gaebel, 1983; Nriagu, 1983b) and lead poisoning could even be implicated in the supposed decrease in the aristocratic class due to its infertility effects (Gilfillan, 1990). As specific accounts

of lead poisoning are scare in Roman texts it is becoming increasingly popular to search for more direct evidence of lead poisoning through lead concentration analysis of human remains from the period.

Some of the first studies to analyse lead concentrations in Roman skeletal material were carried out on Romano-British populations using atomic absorption spectroscopy. The Romano-British sites at Poundbury Camp, Dorset, Henley Wood, Somerset and Trentholme Drive, York recorded unprecedentedly high bone lead concentrations in the skeletal material recovered there (Mackie et al., 1975; Molleson et al., 1986; Waldron et al., 1976; Whittaker and Stack, 1984). While little attempt was made to link these high concentrations with lead poisoning, the exceptionally high lead concentrations in the foetal remains were suggested as possible cause of death for the infants as high lead levels can induce spontaneous abortion and stillbirth (Waldron et al., 1979). In these studies all lead concentrations between the bone samples, and while there were no correlations in lead concentrations between the bone and soil samples collected at the sites, the authors acknowledged that the porous nature of the samples used may have facilitated some degree of ion exchange in the burial environment (Molleson et al., 1976; Waldron et al., 1976; Waldron et al., 1979).

It was not until the work of Montgomery (2002) that real strides in the advancement of lead isotope analysis of archaeological human remains were made. Montgomery (2002) developed a methodology that minimised the risk of contamination from the burial environment, providing a means of confidently assessing *in vivo* lead characteristics (see Chapter 2). This method utilised tooth enamel instead of bone, and although this limited investigations to childhood lead burdens, Montgomery et al. (2002, 2010) found that Roman childhood lead burdens were up to three times higher than what is

considered severely toxic today (Montgomery et al., 2010). Despite Montgomery's ground-breaking research, until recently there have been very few bioarchaeological studies assessing the impact of environmental lead pollution on Roman populations. A joint study by McMaster University and the University of Sheffield, entitled Deadly Lead: How lead poisoning affected the Roman Empire (Prowse and Carroll, 2017), is one of the first studies since the work of Montgomery et al. (2010) to explore lead concentrations and health in a Roman population. In a similar vein to this current study, Prowse and Carroll (2017) hope to garner new insights into the physiological effects of lead pollution on a Roman population.

#### **3.6 Summary**

It is widely accepted that there was extensive movement of people throughout the Roman Empire (Braudel, 1995; Horden and Purcell, 2000; Scheidel, 2004). In fact the term cosmopolitan has become quite a popular adjective when describing Roman populations (Chenery et al., 2011; Gray, 1958; Leach et al., 2010; Martiniano et al., 2016; Montgomery et al., 2011; Turner, 2002; Wright, 2002, p. 98), which is testament to the cultural diversity thought to characterise Roman societies. This propensity for migration together with the substantial environmental lead pollution of the period makes the Romans an ideal study population for the further development of lead isotope ratios as a discriminatory tool in mobility studies.

It has been shown that difficulties often arise when attempting to identify non-locals using traditional methods such as epigraphy, grave goods or burial rites, which are largely dependent upon what has been consciously chosen to be recorded or included (Pearce, 2000). The relatively new application of multi-isotope techniques to assess geographic origins overcomes some of these issues by facilitating a more inclusive analysis of all individuals irrespective of their funerary rite. The majority of mobility studies have used various combinations of carbon, nitrogen, oxygen, and strontium isotope analyses in attempts to identify migrants. However, over the last 20 years there has been an increase in the number of studies employing lead isotope ratios to address archaeological questions of geographic origins (Budd et al., 2004; Montgomery, 2002; Montgomery et al., 2014, 2010, 2005; Shaw et al., 2016). The majority of Roman lead isotope studies have been done using British populations, and while they are beginning to further our understanding of how lead isotope systems can benefit mobility studies, they are limited in their geographical scope. The lack of human lead isotope ratio baselines for different regions of the Roman Empire means that when individuals inconsistent with the local population are identified it is difficult to place their actual childhood origins. Therefore it is clear that more human reference datasets are needed from all areas of the Roman Empire to establish the normal variation expected in Roman populations as well as to assess temporal-spatial isotope variation throughout the Empire.

As previously discussed, there is a correlation between increased environmental lead concentrations and increased human lead concentrations during the Roman period (see Chapter 2). Studies have also shown that during this period there was an increased prevalence of metabolic diseases that can be associated with lead poisoning; this is especially well documented within Romano-British populations (Roberts and Cox, 2003). Historical literature also describes maladies consistent with lead poisoning, and acknowledges the toxic nature of the metal (Lessler, 1988; Needleman, 2009; Waldron, 1973). This will be discussed further in Chapters Four and Five, but what is important to note here is that there are both literary and bioarchaeological evidence to support the notion that people experienced both environmental and physical consequences of lead

pollution during this period (Jonasson and Afshari, 2017). These increased lead burdens make the Romans an ideal population for investigating how anthropogenically induced environmental changes impacted upon heath and mortality.

## CHAPTER FOUR

## The Impact of Lead on Human Health

#### 4.1 Introduction

Human exploitation of the versatile physical and chemical properties of lead has an enduring history that stretches into antiquity, a history that is intimately intertwined with the insidious nature of the prized metal. For as long as people have utilised lead they have suffered the deleterious effects of lead poisoning (plumbism) on their health. These effects are usually systemic and manifest as metabolic diseases, neurological deficits and a failure to thrive in infants (Needleman 2004). Due to the widespread accessibility of lead to Roman populations it is likely that they too suffered the ill effects of lead toxicity. Historical literature describes maladies that, under modern interpretation, strongly suggest that lead poisoning was a cause for concern. However, despite bioarchaeological evidence for high lead concentrations in technologically advanced past populations, and historical documentation describing the negative impact lead had on health, very little research has been done to assess this impact archaeologically. This chapter explores the biochemical interactions of lead poisoning within the human body to inform our understanding of how lead poisoning may manifest in skeletal remains, how lead burdens can be quantified and how this can be used to inform our interpretations of lead poisoning in past populations.

#### 4.2 Metabolism and toxicokinetics of lead

#### 4.2.1 Exposure, absorption and storage

Lead is a non-essential trace element that is ubiquitous within the environment. It acts as a cumulative poison when incorporated into biological systems, causing a myriad of biological, physiological and behavioural dysfunctions (Casas and Sordo, 2011). The predominant route for human acquisition of lead is usually via inhalation or ingestion of inorganic lead salts, which then slowly accumulate in soft tissues and bone. Organic lead compounds are generally more toxic than their inorganic counterparts due to their lipid solubility, facilitating a greater adsorption rate especially through direct contact with skin. Therefore a more rapid bioaccumulation of lead is seen in soft tissues with organic lead exposure than inorganic lead exposure (Hathcock, 2012; Mahaffey, 1978).

Airborne lead can either be in the form of aerosols (dust, smoke, fog etc.) or vapours (free molecules/gas). Inhaled lead vapours can quickly infiltrate the entire pulmonary system and have an exceedingly high absorption rate of approximately 99% with only 1% being exhaled during normal respiratory function (Booker et al., 1969). However, lead vapours have an extremely short lifespan as they are rapidly condensed into smoke, incorporated onto the surface of dust particles or readily react with oxygen to form clusters of lead oxide particles (Castellino and Castellino, 1995). The extent to which aerosolised lead is absorbed into the bloodstream is dependent upon the size of the particulates inhaled, airborne concentrations and the individuals' ventilation rate (Chamberlain, 1983). Approximately 40 - 50% of inhaled lead with a particulate diameter of <1 µm can penetrate deep into the respiratory tract and be absorbed through the alveoli into the bloodstream. The remaining fraction of inhaled lead is confined

within the upper respiratory tract where it is removed by mucociliary mechanisms, enabling it to be either swallowed or exhaled (Needleman, 1991). The absorption percentage of ingested lead differs considerably to that of inhaled lead. Adults tend to absorb approximately 5 – 15% of ingested lead into the bloodstream (Alexander et al., 1974; Ziegler et al., 1978), while in children, absorption rates can be as high as 50% (Hursh and Suomela, 1968; Rabinowitz et al., 1976). Although the reasons behind this age related decrease in absorption are still unclear, studies have shown that dietary deficiencies and excesses can significantly alter the efficiency of lead absorption (Mahaffey, 1981). A low dietary intake of iron (Fe), zinc (Zn), and calcium (Ca), for example, have all been associated with an increased absorption of lead. Conversely diets high in fibre and phytate tend to reduce the amount of lead absorbed through the gastrointestinal tract (Ahamed and Siddiqui, 2007; Needleman, 1991).

Once absorbed, lead circulates around the body in the bloodstream. Up to 99% of absorbed lead is bound to erythrocytes (red blood cells), with the remaining 1% associated with the plasma fraction of blood. As lead circulates it is either deposited in soft tissues and bone or excreted with other waste products. The lead stored in the body is known as the lead body burden and levels depend on the level and duration of exposure, bone metabolism rates and rate of excretion. Approximately 90-95% of the body's lead burden is stored in the skeletal tissues (Berman, 1966; Raj, 2010), with the remaining 5% distributed in the soft tissues (predominantly the liver, kidney and brain). The preferential storage in skeletal tissues, alongside the supposed substitution of  $Ca^{2+}$  for Pb<sup>2+</sup> in bone mineral matrices has prompted lead to be termed a 'bone-seeking' element. However, there is little evidence to indicate that the mechanism behind the incorporation of lead into bone is a simple substitution of  $Ca^{2+}$  for Pb<sup>2+</sup> (Lockitch, 1993; Neuman and Neuman, 1953). Lead stored in the body is not homogenously distributed

throughout the skeleton; there are considerable variations in the concentration of lead between different skeletal elements (rib, femur etc.) as well as between different types of bone (cortical, cancellous) (Aufderheide, 1989; Barry, 1978; Drasch, 1982; Erkkilä et al., 1992).

#### 4.2.2 Mechanisms of toxicity

The basic principle behind lead toxicity is its ability to impair the normal function of metabolic pathways. Due to their electropositive state, lead ions have an affinity for sulfhydryl and amide groups, which are ubiquitous in biological molecules. When bound to these groups, lead weakens the structural and functional integrity of innumerable biologically important molecules (Flora et al., 2012). The various cellular, intracellular and molecular interactions that underpin the toxicological effects of lead to biological systems are complex, but are predominantly the result of ionic and oxidative stress mechanisms.

Lead toxicity induces oxidative stress in cells by stimulating the generation of reactive oxygen species (ROS) such as hydrogen peroxide ( $H_2O_2$ ) and superoxide ( $O_2^{*}$ ). ROS are reactive molecules and free radicals that derive from molecular oxygen. Despite their ability to cause numerous deleterious events, when closely regulated these molecules act as inter- and intracellular messengers that stimulate metabolic pathways that regulate gene expression, cell signalling cascades and apoptosis (Hancock et al., 2001). The disruption to ROS regulation by lead toxicity is twofold, as in addition to catalysing the increased production of ROS it also diminishes the availability and activity of important antioxidant molecules responsible for neutralising damaging ROS molecules (Flora et al., 2012; Flora, 2002).

Lead also disrupts normal cellular processes by substituting for biologically important cations, such as Na<sup>+</sup>, Ca<sup>2+</sup>, Fe<sup>2+</sup> and Mg<sup>2+</sup> (Lidsky and Schneider, 2003). The substitution of Pb<sup>2+</sup> for these cations interferes with numerous cellular processes including apoptosis, protein folding, ionic transportation and the regulation of neurotransmitters and enzyme activity (Garza et al., 2006). The consequences of these ionic substitution are primarily responsible for the neurotoxicity associated with lead poisoning, especially its substitution for Ca<sup>2+</sup> and Na<sup>+</sup> cations (Bressler et al., 1999).

#### 4.3 Clinical manifestations of lead poisoning

#### 4.3.1 Acute poisoning

Acute lead poisoning is usually related to occupational exposure, during which an individual absorbs a high concentration of lead within a short period of time, and is an uncommon occurrence in modern societies. The effects of acute lead toxicity are found at blood lead (B-Pb) levels above 80 µg/dL, and the most likely route of intoxication is accidental ingestion of a large quantity of lead salts or inhalation of lead vapours (Tsuchiya, 1986). The most common clinical symptoms that accompany acute expose include intestinal colic, anorexia, nausea, polydipsia, hypertension, bradycardia and constipation. If the concentration of absorbed lead is high enough, peripheral paraesthesia, muscle pain and weakness, severe anaemia, renal failure and encephalopathy may ensue, with death following within days (Cilliers and Retief, 2014).

#### 4.3.2 Chronic poisoning

Chronic lead poisoning is much more insidious than its acute counterpart. Long-term exposure to small amounts of lead facilitates the gradual accumulation of the toxin in

bodily tissues. Due to the systemic nature of lead toxicity the clinical manifestations of chronic poisoning are varied and complex. With its propensity to disrupt metabolic pathways, it is unsurprising that chronic lead poisoning has been associated with a number of metabolic diseases (Landrigan, 1989). Of these, anaemia is one of the most common presentations, resulting from lead's ability to inhibit haem synthesis and induce haemolysis of red blood cells (Gossel and Bricker, 2001, p. 192; Papanikolaou et al., 2005; Piomelli, 2002). Rickets is another metabolic disorder often seen in conjunction with chronic lead poisoning and is likely due to the metal's inhibition of 25-hydroxyvitimin-D-1α-hydroxylase, the enzyme responsible for converting vitamin D into its active form (Alasia, 2010; Logham-Adham, 1997). Renal failure is a common complication of lead poisoning and the resultant hyperuricemia is the main cause of gout (Alasia, 2010; Lin et al., 2002, 1999). Saturnine gout is a secondary rheumatic arthropathy and this excruciatingly painful condition has been associated with lead poisoning since antiquity (Bennett, 1985). While hyperuricemia is the same cause of primary gout, saturnine gout is somewhat distinct in its predilection for manifesting in the knee rather than the toe (Dalvi and Pillinger, 2013; Taylor and Grainger, 2011, pp. 105–120).

The neurological symptoms associated with lead toxicity are particularly devastating, especially in children (Bellinger, 2004), causing disruptions to neuromuscular and neurobehavioural functions, inducing encephalopathies. The manifestations of these encephalopathies are diverse in their severity, ranging from headaches and delirium, to seizures and cerebral oedema (Holtzman et al., 1984). The neuromuscular dysfunctions associated with lead poisoning are characterised by paralysis, particularly intestinal smooth muscle causing intestinal colic and peripheral paralysis in the extremities. The radial nerve is most commonly affected, resulting in the classic wrist drop manifestation

of chronic poisoning (Geraldine and Venkatesh, 2007; Needleman, 2004; Pearce, 2007). The neurobehavioural effects of lead poisoning are also numerous and well documented. Correlations between lead levels and reduced IQ are particularly well established, with an estimated half an IQ point lost with every 1  $\mu$ g/dL increase in blood lead levels (Lanphear et al., 2005; Pocock et al., 1994; Schwartz, 1994). Other neurobehavioural impairments include memory loss, attention deficit disorder (ADD), reduced impulse control and delinquent behaviour, of which increased aggression is particularly common (Dietrich et al., 2001; Pabello and Bolivar, 2005; Wright et al., 2008).

#### 4.4 Skeletal manifestations of lead poisoning

With the exception of metaphyseal lead lines visible on radiographs (Gandhi et al., 2003; Needleman, 2004), no specific skeletal lesions have been associated with lead poisoning. This is most likely due to the toxicodynamics of absorbed lead culminating in clinical manifestations common to many other disease processes. However, using modern clinical literature and the known biochemical pathogenesis of lead toxicity it is possible to postulate the types of skeletal pathological alterations that may be common among individuals exhibiting high lead concentrations.

#### 4.4.1 Carious lesions

Dental caries, or tooth decay, is a disease characterised by the localised demineralisation of tooth enamel (see Fig. 4.1). It is a chronic disease that can be seen in the crown (coronal caries) and root (root caries) portions of both the deciduous and permanent teeth (Irish and Scott, 2015). It is a multifactorial disease catalysed by a microbiological shift within the complex biofilm on the tooth surface, and can be affected by salivary flow and composition, diet and exposure to heavy metals (Selwitz

et al., 2007). There was an increase in prevalence rates of carious lesions in European populations during the Roman period, likely as a result of changes to diet and increased exposure to environmental heavy metal pollution (Touger-Decker and Van Loveren, 2003). Although Touger-Decker and Van Loveren (2003) suggest that increased heavy metal burdens may have contributed to the increase in carious lesions during the Roman period, it is likely that multiple factors contributed to this observed increase in prevalence rates.

The negative effects of lead poisoning on dental health has been studied since the mid-19<sup>th</sup> century (Des Planches, 1839), and corroborated by modern epidemiological studies (Bartsiokas and Day, 1993; Brudevold et al., 1977; Gil et al., 1996; Moss et al., 1999; Nriagu et al., 2006). However, some studies have found that there is only a weak association between elevated lead burdens and the presence of carious lesions and that the development of these lesions is probably population specific (Campbell et al., 2000; Gemmel et al., 2002). In all studies, children of a lower socioeconomic status and those living in urban environments have been identified as being the most at risk of developing carious lesions and acquiring high lead burdens. To date the biochemical mechanisms behind the cariogenic effects of lead are unknown (Nriagu et al., 2006). However, theories include lead damage to acinar cells in the parotid gland, which alters the secretion of proteins, lysosomal enzymes and calcium (Abdollahi et al., 1997). Impaired salivary secretion causing dry mouth (xerostomia), a common symptom of lead poisoning (Watson et al., 1997), and an overall reduced quality of saliva impeding its natural disease preventative functions (Mandel, 1993) have also been suggested as causes. As previous studies have shown, the relationship between lead levels and carious lesions is clearly a complex one and likely multifactorial. However, despite the

uncertainties there is clearly a correlation between increased lead levels and prevalence of carious lesions.



Figure 4.1 – Carious lesion on the mesial surface of a mandibular first molar (Source Klingner, 2013)

#### 4.4.2 Enamel hypoplasia

Enamel hypoplasia results from a break in the continuity of enamel synthesis, which reduces the number of layers of enamel, creating grooves or depressions on the surface of the tooth crown (see Fig. 4.2) (Umapathy et al., 2013). The most common causes of disruption to the genetic and environmental factors that tightly regulate the synthesis of enamel are vitamin deficiency, systemic illness, and environmental pollution (Fagrell et al., 2011). While no studies have actively sought to verify a link between lead burdens and the presence of enamel hypoplasia, the general stress induced by the systemic disruption to metabolic pathways caused by lead poisoning may be sufficiently deleterious to health as to result in hypoplastic change.

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Figure 4.2 – Linear enamel hypoplasia in both the mandibular and maxillary dentition (Source Henderson, 2012)

#### 4.4.3 Anaemia

Anaemia results from the inability of erythrocytes to transport sufficient amounts of oxygen around the body, and can either be congenital (e.g. sickle cell anaemia) or acquired (e.g. deficiencies in iron or vitamin  $B_{12}$ ) in its aetiology (Ortner, 2003). Elevated lead burdens can result in an acquired anaemia. At very low levels ( $\geq$ 5 µg/dL) lead has been shown to inhibit the activity of key enzymes in the haemopoietic pathway ( $\delta$ -aminolevulinic acid dehyratase (ALA-D) and ferrochelatase), which decreases the formation of haem in erythrocyte precursor cells (normoblasts) (Gossel and Bricker, 2001, p. 192; Papanikolaou et al., 2005; Piomelli, 2002). Lead also weakens the cell membrane of erythrocytes, making them prone to haemolysis and thereby shortening their normal lifespan. The overall effect of lead poisoning on the haem synthesis pathway is microcytic hypochromic anaemia, which is characterised by immature erythrocytes with low levels of haemoglobin and basophilic stippling (Bain, 2014).

The most common skeletal lesions associated with anaemia are porotic hyperostosis and cribra orbitalia (see Fig. 4.3). Both are descriptive terms used to describe abnormal pitting and porosity on the external surface of the cranial vault and orbital roofs respectively (Stuart-Macadam, 1987, 1989, 1992; Walker et al., 2009). Anaemia is so pervasive in clinical literature associated with lead poisoning that the skeletal manifestations of the disorder are a popular target in bioarchaeological studies exploring lead exposure in past populations. A review of recent literature shows that cribra orbitalia is the most commonly used pathological alteration in bioarchaeological studies attempting to correlate high lead burdens with skeletal evidence of lead poisoning (Facchini et al., 2004; Gleń-Haduch et al., 1997; Millard et al., 2014; Zarifa et al., 2016). However, while cribra orbitalia is undoubtedly an indication of stress and has strong clinical links to haemopoietic stress, not all types of anaemia are thought to cause this particular osseous response. Until relatively recently, iron-deficiency anaemia was thought to be the predominant cause of cribra orbitalia (Ponka, 1997; Stodder, 2006), and so popular was the theory that the presence of the lesion became synonymous with the presence of the disorder. However, recent research suggests that iron-deficiency anaemia does not elicit the marrow hypertrophy necessary to induce the bony changes of cribra orbitalia (Wapler et al., 2004). The recent works of Walker et al. (2009) puts forward a strong argument for megaloblastic and haemolytic anaemias being a more likely cause for the lesion. However, Oxenham and Cavill (2010) contest Walker et al's., (2009) claims that iron-deficient anaemia cannot cause cribra orbitalia, stating that although there is a reduction in the production of fully matured erythrocytes there is a large increase in intramedullary erythropoiesis, which would cause marrow hyperplasia (Oxenham and Cavill, 2010). Although like iron-deficiency anaemia, lead poisoning causes microcytic hypochromic anaemia, lead inhibits rather than stimulates

erythropoiesis (Kwong et al., 2004), and as such is unlikely to induce the physiological responses necessary to produce cribrous changes (Lewis, 2017, p. 200). This study will compare the presence of cribra orbitalia and lead concentrations to test the hypothesis that cribra orbitalia cannot be used as a skeletal marker suggestive of lead poisoning.

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Figure 4.3 – Cribra orbitalia in the orbital roof and porotic hyperostosis on the cranial vault (Source: Krenz-Niedbała, 2017)

#### 4.4.4 Rickets

Rickets is caused by a deficiency in the pro-hormone vitamin D (1,25dihydroxyvitimin-D), which plays an important role in the regulation of calcium and phosphorous levels in the body. It is synthesised in the kidneys and is involved in the stimulation of alkaline phosphatase, osteocalcin and osteopontin synthesis, all of which are important for the normal mineralisation of newly formed bone osteoid (Brickley and Ives 2009; Favis 1999; Pitt 1988; Heaney 1997; Holick and Adams 1998). A deficiency in vitamin D results in a shortage of these essential ions and proteins, preventing osteoid mineralisation and ultimately weakening bone mineral matrices (Arnaud and Glorieux 1997; Holick 2003). Chronic exposure to lead often leads to nephropathy (renal damage), and if allowed to persist, renal failure ensues (Alasia, 2010; Lin et al., 2002, 1999). Prolonged lead toxicity can inhibit the activity of 25-hydroxyvitimin-D-1 $\alpha$ -hydroxylase, which is the enzyme responsible for converting 25-hydroxyvitimin-D into its active form 1,25-dihydroxyvitimin-D (Rosen et al., 1980). The inhibition of this enzyme has been recorded with blood lead levels as low as 10 µg/dL, and results in increased demineralisation of osseous tissues that can manifest as rickets or osteomalacia in children and adults respectively (Alasia, 2010; Chisholm Jr. et al., 1955; Logham-Adham, 1997).

The most characteristic skeletal manifestations of rickets are pathological bowing of long bones (see Fig. 4.4). Porosity, fraying and flaring of metaphyses, especially of the long bones and sternal rib-ends are common pathological alterations associated with rickets (Lewis, 2017, p. 210; Waldron, 2009, p. 129). Abnormal porosity on the cranium (cribra orbitalia and porotic hyperostosis) and mandibular ramus deformities are also commonly seen in rachitic skeletons (Brickley et al., 2005, 2018; Mays et al., 2006; Ortner and Mays, 1998; Swinson et al., 2010; Watts and Valme, 2018) Although the skeletal manifestations of rickets are varied and can have numerous causes, the co-occurrence of multiple pathological alterations provides strong support for a diagnosis of rickets.

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Figure 4.4 – Abnormal bowing and metaphyseal flaring of the lower limbs associated with rickets (Source: Watts and Valme, 2018)

#### 4.4.5 Scurvy

Scurvy is a nutritional deficiency disease resulting from a lack of dietary vitamin C (ascorbic acid). This deficiency impedes the activity of proline hydroxylase (a vitamin C dependent enzyme), an enzyme important to the hydroxylation process involved in collagen synthesis. This results in the production of weakened collagen structures that culminates in defective, reduced or arrested osteoid formation and fragile blood vessels prone to haemorrhage (Ortner et al., 2001). Like proline hydroxylase, lysyl oxidase (a copper dependent enzyme) is responsible for creating stabilising cross-links between lysine and hydroxyproline amino acids in collagen fibrils (Dollwet and Sorenson, 1988). Reduced activity of this enzyme results in the synthesis of weakened collagen. The manifestations of copper deficiency has been termed pseudo scurvy due to its similarities with the scorbutic disease (Hoyle et al., 1999; Hurwitz et al., 2004; Nguyen

and Kerner, Jr., 2007). Lead is a copper antagonist, and therefore inhibits the activity of copper dependent enzymes, mimicking a copper deficiency in individuals suffering from lead poisoning. Cases of lead-induced scurvy or pseudo scurvy are documented in the clinical literature (Ramel and Schenk, 1942).

The pathological skeletal alterations associated with a copper deficiency are the same as those associated with scurvy, as both disorders result in weakened collagen prone to tearing (Allen et al., 1982). The skeletal manifestations of scurvy are subtle and common to many other disease processes, added to this the often fragmentary nature of archaeological remains makes assigning a definitive diagnosis of the disease difficult (Armelagos et al., 2014; Geber and Murphy, 2012). Thus, it is distinctive lesions described in works such as the 'Ortner Criteria' that prove vital to the successful identification of scorbutic skeletal remains (Crandall and Klaus, 2014; Zuckerman et al., 2014). These lesions primarily consist of porotic hyperostosis, cribra orbitalia and abnormal porosity (often with periosteal new bone formation) on the scapulae, long bone metaphyses, and mandible (see Fig. 4.5) (Brickley and Ives, 2006; Klaus, 2017; Moore and Koon, 2017; Ortner, 2003; Resnick and Niwayama, 1988; Stark, 2014). These lesions also tend to manifest bilaterally and are thought to be caused by chronic, low-grade haemorrhage of weakened blood vessels, predominantly at muscle attachment sites, which stimulates an inflammatory response (Ortner et al., 2001, 1999; Ortner and Ericksen, 1997).

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Figure 4.5 – Abnormal porosity and new bone formation associated with scurvy (Source Bourbou, 2014)

#### 4.4.6 Gout

Gout is a type of inflammatory arthritis characterised by the deposition of monosodium urate crystals in joints and soft tissues. Although any joint can be affected, the first metatarsophalangeal joint (big toe) is usually the primary site of inflammation (Dalbeth et al., 2016). Lead toxicity can cause secondary gout known as saturnine gout, due to lead-induced hyperuricemia, which is the primary cause of gout (Aşkin et al., 2015; Baki et al., 2016; Brewster and Perazella, 2004; Dalvi and Pillinger, 2013). The Romans were notorious for their consumption of wine sweetened with defrutum and sapa (lead acetate), which likely led to the development of gout amongst those who drank considerable amounts of wine. It has been estimated that members of the Roman aristocracy drank upwards of 2 litres of wine a day, containing lead concentrations in the region of 90  $\mu$ g/L (Nriagu, 1983b). However, there was a preference to drink the wine diluted (Guy, 1981), which would have likely reduced the amount of lead ingested

via wine consumption. In their bioarchaeological review of health in prehistoric and historic Britain, Roberts and Cox, (2003, p. 389) note that gout first appeared in Britain during the Roman period. Although the change in diet that came with the Roman occupation of Britain likely influenced the prevalence of the disease, the consumption of lead-sweetened wine may have also been a contributing factor to the emergence of gout during this time.

Lead poisoning was first linked to gout in the  $18^{th}$  century with the import of Portuguese Port and Madeira wine sweetened with lead acetate (Green, 1985). Chemical analysis of these wines revealed their high lead content, with concentrations ranging between 320 -1900 µg/L (Halla and Ball, 1982). By the  $19^{th}$  century a firm link had been established between lead poisoning and gout, with fortified wines implicated as the primary source of intoxication (Healey, 1975; Porter and Rousseau, 2000; Storey, 2001).

"... no truth in medicine is better established than the fact that the use of fermented ... liquors is the most powerful of all the predisposing causes of gout."

#### -Alfred Baring Garrod, (1859)

The skeletal manifestations of gout are primarily those of an erosive arthropathy, and tend to involve the head of the first metatarsal (see Fig. 4.6). Features that support a diagnosis of gout is a punched out and undercut appearance to the lesion, manifestation in a para-articular position and extension onto the bone diaphysis (Resnick and Niwayama, 1988). Unfortunately, the reliance upon the presence of para-articular lesions to diagnose gout means that only cases severe enough to cause erosive tophaceous deposits can be identified palaeopathologocally (Swinson et al., 2010).

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Figure 4.6 – Tophaceous, punched out lesion with Martel's hook on the distal para-articular surface of the first metatarsal, indicative of gout (Source: BARC)

#### 4.4.7 Lead lines

Lead lines are the only skeletal lesion specifically associated with lead poisoning. They are visible on radiographs and present as dense transverse bands in the metaphyses of long bones and along the margins of flat bones, such as the iliac crest (see Fig. 4.7). A common misconception is that these lead lines represent areas of lead deposition in the cancellous bone (Papanikolaou et al., 2005). However, studies have demonstrated that there are no significant increases in lead concentrations in these areas (Papanikolaou et al., 2005; Patrick, 2006). The increased density is actually attributable to an increase in calcium deposition in these areas. Lead inhibits osteoclastic activity in zones of provisional calcification, causing increased calcium deposition in the areas which present as dense bands in radiographs (Chew, 2012). These dense bands of bone are best visualised in bone where areas of rapid growth occur, such as the proximal tibia or distal radius (Chew, 2012, p. 293; Woolf et al., 1990).



Figure 4.7 – Radiograph showing bilateral lead lines (opaque bands) in the femora, tibiae and fibulae metaphyses at the knee (Source: LearningRadiology.com accessed 2017).

#### 4.5 Lead body burden

The term lead burden refers to the total amount or concentration of lead contained within the human body, and represents the difference between absolute lifetime cumulative absorption and aggregated excretion (Landrigan and Todd, 1994). However, as lead is heterogeneously distributed in various bodily tissues that differ in both their capacity and accessibility for lead accumulation, the assessment of total lead burden is not straightforward.

#### 4.5.1 Estimating lead body burden

Clinical studies dominate the literature on lead burdens and health. Studies into the effectiveness of different tissues for estimating lead body burdens focus on living patients. Therefore methods tend to use minimally invasive and easily collectable sample types such as blood, urine, saliva, hair, and nails (Barbosa Jr et al., 2005).

Although occasionally hair and nails can survive in the burial environment, especially if the remains are mummified, in archaeological contexts the majority of surviving material is the mineralized tissues. Therefore, this section will focus on the use of bones and teeth in the estimation of lead body burdens.

#### 4.5.1.1 Bones

One of the most striking aspects of lead in the human body is its predilection for bioaccumulation in osseous tissues, with approximately 90-95% of the bodies lead burden stored in the skeleton (Berman, 1966; Raj, 2010). Within the skeleton, lead is not homogenously distributed. There are considerable variations in the concentration of lead between different skeletal elements and even between different types of bone (Aufderheide, 1989; Barry, 1978; Drasch, 1982; Erkkilä et al., 1992). Cortical bone has a slower turnover rate than cancellous bone. Therefore, skeletal elements comprised predominantly of cancellous bone (e.g. patellae and calcanei) contain a higher amount of biologically available lead than skeletal elements comprised predominantly of cortical bone (e.g. tibiae and phalanges) (Ambrose et al., 2000; Fleming et al., 1999; O'Flaherty, 1995; Roy et al., 1997; Tsaih et al., 1999). This non-uniform distribution of lead within bones appears to be governed by the volume of blood delivered to the bone. A study by Todd et al., (2001) reported that regions of lower lead concentration contained a lower percentage of cancellous bone and a higher percentage of cortical bone than regions exhibiting high lead concentrations. It is suggested that the larger surface area, higher volume of blood flow to Haversian canals and a higher concentration of active osteons in cancellous bone facilitates a higher rate of lead incorporation into the bone matrix than cortical bone (Todd et al., 2001). Although the residency time of lead in bone is estimated to be between 15-30 years (Rabinowitz et al., 1991), the dynamic nature of bone results in the release of previously incorporated lead and the incorporation of new lead. This results in lead isotope ratios and lead concentrations that gradually alter over time as bone turnover proceeds (Priest and Van de Vyver, 1990).

#### 4.5.1.2 Teeth

The proclivity lead exhibits for mineralised tissues means that in addition to bone, lead also accumulates in the dentition. However, the way in which lead is acquired and retained in different tooth portions (e.g. enamel, dentine etc.) means that teeth provide a means of reconstructing an individual's exposure history (Budd et al., 1998). Lead is incorporated into enamel matrices during the mineralisation of the tissue. As enamel does not remodel or have an active blood supply, its lead composition is considered a reliable indicator of lead exposure at the time of enamel mineralisation (Brudevold et al., 1977; Gulson, 1996; Gulson and Wilson, 1994; Veis, 1989; Wieser et al., 1996). Conversely, lead in dentine is known to be more dynamic, continuously accumulating lead from its blood supply (Gulson, 2008; Shapiro et al., 1975). This is corroborated by a study carried out by Gulson et al. (1997a) in which lead concentrations and lead isotope ratios in paired dentine and enamel samples from European immigrants to Australia were analysed. The results showed that there was no change to the lead in enamel, while lead in the dentine samples taken from the apical section of permanent tooth roots slowly altered to reflect the isotopic composition of the new environment, with an exchange rate of approximately  $1 \pm 0.3$  % per year (Gulson et al., 1997a; Gulson, 2008). As such, the lead contained within dentine can be considered an averaged representation of lifetime accumulation (Budd et al., 1998). This temporal

difference in lead acquisition between enamel and dentine potentially facilitates the comparison of lifetime changes in lead exposure (e.g. childhood vs. adulthood).

#### 4.5.2 Dynamic lead burdens

Lead burdens are far from static; they are influenced by both exogenous and endogenous factors and thus are prone to fluctuation. Living and working in less polluted, usually rural environments with little industrial activity will result in lower lead burdens. Conversely, living in more heavily polluted environment such as urban, industrial areas poses a much greater exposure risk. As expected, exposure to higher concentrations of lead in an individual's home or occupational environment will increase lead burdens. Exogenous factors such as these are the most obvious variables that directly influence individuals lead burdens. However, even remaining in the same environment with a consistent level of pollution does not ensure a stable lead burden, as endogenous factors also play a significant role in lead burden fluctuations.

Factors that influence an individual's physiological equilibrium, such as disease, pregnancy, lactation and menopause, have all been shown to alter lead burdens (Cory-Slechta, 1996; Gulson et al., 2003a; Silbergeld, 1991; Silbergeld et al., 1988; Symanski and Hertz-Picciotto, 1995). This is, in the most part, due to the remobilisation of lead that has been sequestered in mineralised tissues. While it was thought that this lead was stored indefinitely within mineral matrices, subsequent studies have shown that physiological stress, which can influence bone turnover rates, results in the release or remobilisation of this stored lead (Gulson et al., 2004, 2003, 1999, 1997; Spivey, 2007). The repercussions of this remobilisation of endogenous lead is particularly significant for pregnant and breastfeeding women as the toxic effects of lead are most potent in developing foetuses and young children.

Some types of physiological stress can create a higher demand for calcium (Clarke, 2008; Kovacs, 2015; Ross et al., 2011), and this increased demand has been shown to have a significant effect on blood lead levels. It has been suggested that this is because the resorption rate of skeletal tissues increases to release the needed calcium in individuals with an inadequate dietary intake (Gulson et al., 1997; 2004, 2003). Bone turnover of this nature has been shown to be most prolific in pregnant and breastfeeding women (Gulson et al., 2016), and is likely due to calcium requirements increasing from 1000 mg to 1300 mg/day during gestation and lactation (Prentice, 2000). A study by Gulson et al., (2004) demonstrated that the isotopic composition of maternal blood lead was identical to foetal cord blood, thereby confirming that there is placental transfer of lead during pregnancy. Numerous studies have also shown a strong correlation between maternal and foetal blood lead concentrations (Furman and Laleli, 2001; Klein et al., 1994; Moura and Valente, 2002; Navarrete-Espinosa et al., 2000; Wan et al., 1996), with an estimated 79% of maternal blood lead transferred to the foetus (Gulson et al., 2003). Other studies have also demonstrated that lead is transferred through breast milk (Rabinowitz et al., 1985; Ettinges et al., 2004), with this sourcecontributing to between 36 – 80% of an infant's blood lead level (Gulson et al., 1998).

Using the same mixing relationship as applied in isotopic geochemistry, Gulson et al., (2003) showed that this increase in blood lead concentration is derived predominantly from bone lead stores. By determining the isotopic composition of blood lead before, during and after pregnancy in migrants to Australia, Gulson et al. demonstrated that between 41% - 73% (mean 31%) of the lead responsible for this increase was remobilised bone lead (Gulson et al., 1997b; Gulson et al., 1998, 1995). Supporting evidence for the remobilisation of lead from bone during pregnancy has also been demonstrated in studies examining pregnant women from different populations across

the world (Gulson et al., 1997b, 1999; Klein et al., 1994; Lagerkvist et al., 1996; Rothenberg et al., 1994, 2000). These studies all reported that blood lead concentrations increased on average by  $\sim 20\%$  during pregnancy. Gulson et al., (1998) also showed that these concentrations were even higher in postnatal mothers. Lactation has been posited as the cause for this post pregnancy increase, as it increases the bodies' calcium requirements. As such, if the mothers' calcium intake and absorption rates fall short of the physiological need, bone resorption and the subsequent release of calcium is the likely mechanism responsible, in an attempt to correct the deficit (Gulson et al., 1998). The breastfeeding and dietary intake histories collected during Gulson et al's., (1998) study seems to corroborate this hypothesis. Their findings showed that women that breastfed for longer, and therefore experienced prolonged physiological stress relative to the other participants, had the highest blood lead concentrations. While women taking calcium supplements, and therefore ensured the recommended daily requirement intake, exhibited the lowest blood lead concentrations. This has been corroborated in other studies that have also reported the protective qualities of calcium against increased lead levels during pregnancy (Farias et al., 1996; Hernandez-Avila et al., 1996; Johnson, 2001; Moura and Valente, 2002).

This predisposition of pregnant women and new mothers to higher than normal lead burdens, may offer insights into the high infant mortality rates or failure to thrive seen in past populations. Especially those that have been shown to have high levels of environmental pollution, as lead levels as low as 5  $\mu$ g/dL have been shown to increase the risk of spontaneous abortion, premature delivery, low birth weight and stillbirths (Borja-Aburto et al., 1999; Edwards, 2013; Flora et al., 2012; Gilfillan, 1965; Hamilton and Hardy, 1974; Troesken, 2008; Zhu et al., 2010). Further implications of these dynamic lead burdens, in an archaeological context, are the uncertainties it creates surrounding the type (chronic vs. acute) and duration of exposure. Especially when attempting to infer the impact of lead on health from a single bone or dentine measurement. Enamel lead concentrations may provide a way forward in this respect, as enamel lead concentrations do not alter once mineralisation of the tissue is complete and therefore reflect an averaged exposure for a fixed period of time.

#### 4.5.3 Estimating blood to mineral lead ratios

The nature of archaeological remains limits the assessment of lead burdens to the measurement of lead concentrations in mineralised (skeletal and dental) tissues. Lead sequestered within these mineralised tissues can be considered inert because of its inability to interfere with biochemical processes in soft tissues. As such, lead within mineralised tissues does not directly reflect the lead burden responsible for causing the disease processes associated with lead toxicity. It is therefore important to understand how the lead levels observed in mineralised tissues reflect the amount of lead that would have been circulating within blood and soft tissues.

Clinical literature pertaining to lead poisoning is centred on measurements of blood lead concentrations, with few studies exploring how blood lead concentrations relate to lead concentrations within teeth or bones. Thus, archaeological inferences about lead poisoning are difficult, as the accuracy of the extrapolated effects of blood lead concentrations to those in mineralised tissues is poorly understood. Studies that have explored this relationship have done so using different portions of exfoliated deciduous teeth, and blood collected at different periods in the individual's life, making results difficult to interpret as the samples used (blood, dentine, enamel etc.) relate to different periods of life. As lead stored in tooth enamel represents the lead sequestered during a definable period of time (during mineralisation), only studies that include blood samples

collected during this period of mineralisation offer any insight into the blood lead to enamel lead relationship. Determining this in bone samples is much harder as lead in bone does not simply accumulate over time, becoming stored within the mineral matrix indefinitely. Instead bone lead concentrations fluctuate; sequestering and releasing lead at different rates depending on an individual's age and physiological status (see section 4.5). As lead from enamel samples is used in this study only the relationship between blood lead concentrations and teeth will be considered further.

Few studies have examined how childhood tooth lead concentrations correlate with the lead concentrations in blood, and those that have, reveal a complex and poorly understood relationship (Rabinowitz et al., 1993). Despite this, a small number of studies on modern populations have reported a correlation between enamel lead concentrations (taken using acid etch biopsies) and blood lead concentrations that were collected during the period of tooth mineralisation (Brudevold et al., 1977; Cleymaet et al., 1991; Robbins et al., 2010). However, the lead concentrations within tooth enamel are not homogenously distributed. Polido Kaneshiro Olympoi et al., (2010) demonstrated that lead concentration in two samples taken from the same tooth, but at depths that differed by 0.5 µm, showed a 50% decrease in lead concentration in the deeper layer of enamel. Several studies have demonstrated that lead concentrations decrease sharply as sampling moves away from surface enamel towards core enamel samples (Budd et al., 1998; Fergusson and Purchase, 1987; Purchase and Fergusson, 1986; Robbins et al., 2010). This is important to consider when interpreting lead burdens in archaeological remains as it is unclear whether this concentration gradient within tooth enamel is derived from endogenous processes (during mineralisation), exogenous interactions with surface enamel, or indeed a mixture of both. As such, core enamel is the preferential sample type when assessing lead concentrations in

archaeological human remains, as it not only ensures contamination from the burial environment is minimal but also removes the surface enamel variable and ensures all measurements represent lead burdens derived from blood lead.

One of the few studies that goes further than simply reporting a correlation between blood lead and enamel lead levels was conducted by Grobler et al., (2000). In this study lead concentrations in enamel, dentine, circumpulpal dentine and blood samples from 48 South African children were determined. Median concentrations were then expressed as ratios to blood lead. The results show that mineralised tissues all contained higher lead concentrations than blood, with an enamel to blood lead ratio of 10:1. If this is an accurate representation of the concentration difference between tooth enamel and contemporary blood samples then reducing the lead concentrations recorded in archaeological enamel samples by a factor of 10 should provide approximate blood lead levels and allow comparison with clinical literature pertaining to lead poisoning.

#### 4.6 Evidence of lead poisoning in the Roman Empire

#### 4.6.1 Lead poisoning in antiquity

The earliest known documentation alluding to the toxicity of lead is found in ancient Egyptian papyrus scrolls, in which the potential use of lead as a murderous toxin is referenced (Hernberg, 2000; Lessler, 1988). It is therefore apparent that humans have been aware of lead and its diverse applications, as well as its inherent toxicity for millennia. With the large quantities of lead mined and utilised throughout the Roman Empire it is unsurprising that there is historical evidence for lead poisoning in Roman populations and bioarchaeological evidence of toxically high lead body burdens. Although the Romans were generally unaware of the exact aetiology that underpinned

many of the afflictions they described, today we recognise the symptoms outlined in these ancient texts as indicative of lead poisoning.

The Greek physician Hippocrates may well have been the first to describe the effects of lead poisoning in his 3rd century BC account of intestinal colic and recognition of the relationship between gout and wine consumption. However, it is the 2<sup>nd</sup> century BC poet and physician Nicander of Colophon who is widely credited as being the first scholar to describe lead poisoning through his description of lead-induced palsy and anaemia (Lessler, 1988; Needleman, 2009; Waldron, 1973). Indeed, even authors such as Dioscordes (*De Materia Metica*), Vitruvius (*De Architectura* viii.6.10 and 11) and Pliny the Elder (*Historia Naturalis* xxxiv.50.167) recognised the risks associated with ingesting lead (F. P. Retief and Cilliers, 2006). The following translation from Pliny the Elder's work *Historia Naturalis* demonstrates the known dangers of drinking wine adulterated with lead sweeteners and the commonality with which it was done. The description, '*dangling, paralytic wrists*' is clearly a reference to what is today known as 'wrist drop' or radial nerve palsy, in which paralysis of the extensor muscles in the upper limbs inhibits the extension of the arm and wrist.

#### "... genuine, unadulterated wine is not to be had now, not even by nobility ... From the excessive use of such wines arise dangling, paralytic hands."

#### – Pliny the Elder

It is evident that lead, whether ingested intentionally (e.g. sapa) or unwittingly (e.g. contamination from water pipes), was highly accessible to everyone within the Roman Empire. Scholars have therefore claimed that lead poisoning must have been a common, even endemic affliction throughout the Empire. Despite this, lead poisoning is poorly
documented in Roman texts (Hernberg, 2000). However, this has not deterred some scholars from suggesting that lead poisoning was responsible for the preponderance of stillbirths, deformities and cases of brain damage in Roman infants (Gilfillan, 1965; Nriagu, 1983; Woolley, 1984); some even go as far as to hypothesise that lead poisoning played an important role in the eventual downfall of the Roman Empire (Gilfillan, 1990; Nriagu, 1983). This viewpoint has been vehemently opposed by a number of authors, who see the impact of anthropogenic lead pollution during the Roman period as much less significant (Drasch, 1982; Gaebel, 1983; Needleman and Needleman, 1985; Scarborough, 1984).

## 4.6.2 Assessing lead poisoning in archaeological remains

With a shortage of surviving literary evidence to support or indeed refute the degree to which lead impacted upon the morbidity and mortality of Roman populations, attention must be directed to the skeletal remains from the period. Human skeletal remains offer a direct link to the past, providing a rich source of information pertaining to the lives and living conditions of past populations (Scott, 2013). However, a shortage of literature exists here too. Despite the known toxic effects of lead, and the unprecedentedly high lead concentrations seen in Roman skeletal material, little bioarchaeological research has been conducted to investigate how lead burdens may have impacted upon health throughout the empire. This makes it difficult to establish how the Romans profuse use of lead impacted upon the lives of people within the Roman Empire, and how this may have differed according to geographic and socio-cultural variations. In fact, there are very few examples of bioarchaeological investigations that explore lead poisoning in skeletal populations from any time period. Those that have, simply compared skeletal lead concentrations with the lead concentrations reported in clinical literature (Griffin,

2015; Facchini et al., 2004; Waldron et al., 1982). This is problematic as the majority of clinical studies use blood to calculate lead burdens, forcing comparisons between different sample types. Therefore, while identifying exposure to high lead concentrations is relatively simple within bioarchaeological contexts, determining how these high concentrations relate to morbidity and mortality has proven to be much more challenging.

Some of the first bioarchaeological investigations into the lead burdens of past populations were conducted in the late 1970s. The Romano-British sites at Poundbury Camp, Dorset and Trentholme Drive, York recorded, unprecedentedly high bone lead concentrations in the skeletal material recovered there (Mackie et al., 1975; Waldron et al., 1976; Whittaker and Stack, 1984). While little attempt was made to link these high concentrations with lead poisoning, the exceptionally high lead concentrations in the foetal remains were suggested as possible cause of death for the infants as high lead levels can induce spontaneous abortion and stillbirth (Waldron et al., 1979b). In these studies all lead concentrations were obtained from bone samples, and while there were no correlations in lead concentrations between the bone soil samples collected at the sites, the authors acknowledged that the porous nature of the samples (particularly the higher porosity of the foetal remains) used may have facilitated some degree of ion exchange in the burial environment (Molleson et al., 1986; Waldron et al., 1979b).

The Franklin expedition is probably one of the most recognised examples of bioarchaeological investigations into lead poisoning to date. Bioarchaeological analysis of the frozen remains of known crew members, William Braine, John Hartnell and John Torrington, discovered on Bleechey Island (Amy et al., 1986; Beattie and Geiger, 2017;

Notman et al., 1987) and the disarticulated remains of at least 20 individuals on King William Island (Beattie, 1983; Beattie and Savelle, 1983; Keenleyside et al., 1997) were conducted in the 1980s and 1990s. Using bone and hair samples these studies found lead concentrations ranging from 49 ppm to 204 ppm (mean 103 ppm). These results were interpreted as evidence for lead poisoning among the crew as they were equal to or greater than those recorded in modern clinical literature documenting lead poisoning (Keenleyside et al., 1996; Kowal et al., 1991). From the analysis of these remains it was posited that lead poisoning was a major contributing factor to the 19<sup>th</sup>-century disaster, with improperly soldered canned goods suggested as a significant contributor to the toxic lead burdens observed (Beattie, 1985; Kowal et al., 1991, 1989). Several scholars have challenged this hypothesis, suggesting that contamination of food from solder occurs at such low concentrations that it would not be significantly detrimental to health (Farrer, 1993, 1989; Trafton, 1989). These studies conclude that while the observed lead concentrations were undoubtedly high, the long residency time of lead in bone together with the high 19<sup>th</sup>-century baseline lead burdens, instil significant uncertainty surrounding how long the lead had been in the bone and how much of it was acquired during the expedition (Farrer, 1993, 1989; Trafton, 1989).

A more recent study by Millard et al. (2014) analysed the lead concentrations in 18<sup>th</sup> – 19<sup>th</sup> century London populations in an attempt to correlate the high lead burdens associated with post-medieval populations with evidence of disease and infertility. This study found tooth enamel lead concentrations ranging from 0.47 ppm to 99.2 ppm (mean 22.22 ppm). These results demonstrate high levels of pollutant exposure, with lead concentrations peaking at values three times higher than those recorded in Roman skeletal material (Millard et al., 2014; Montgomery et al., 2010). Using the Stuart-Macadam (1991) scoring system, the presence and severity of cribra orbitalia was used

as an indicator of anaemia. Surprisingly, despite the plethora of clinical literature discussing the almost diagnostic quality of anaemia in relation to lead poisoning, Millard et al. (2014) found no significant correlation between cribra orbitalia score and lead concentrations. However, this may be related more to the uncertainty surrounding the exact aetiology of cribra orbitalia than the possibility of successfully correlating skeletal lesions with elevated lead concentrations. In addition to this, Millard et al., (2014) also compared childhood lead concentrations accumulated between the ages of 2.5 to 6.5 years, with pathological lesions on adult skeletal remains. This may have contributed to the difficulty in establishing how lead impacted upon health as, while a specific period of accumulation can be assigned to the lead concentrations used, the same cannot be done for the pathological lesions, which could have developed during any time prior to death.

What these studies demonstrate is the difficultly with single lead concentration measurements for determining the presence of lead poisoning. Thus, other evidence should be sought in conjunction with lead concentrations. A logical starting point here would be the inclusion of skeletal evidence of disease, specifically those known to be associated with lead poisoning, (see section 4.4). As such, this research will combine trace element analysis with palaeopathological data from Roman skeletal remains to contribute to our understanding of the impact anthropogenic lead pollution on the health of populations throughout the Roman Empire. This process is not without its own obstacles, the majority of which stem from the dynamic nature of living bone and the elements stored within it. As a living tissue, bone undergoes modelling (new bone formation) and remodelling (replacement of weakened bone) throughout life, and this continuous turnover alters the lead profile of skeletal tissues. Consequently, a single bone lead measurement does not provide an accurate representation of the lead burden experienced by an individual. Due to this dynamic relationship, the analysis of bone lead cannot currently differentiate between a period of acute high-level lead exposure that would have resulted in lead-induced pathologies or an extended period of low-level exposure resulting in a high, cumulative lead burden that is likely to have remained asymptomatic (Montgomery et al., 2010). This makes the assessment of lead poisoning in archaeological populations problematic, as discerning the type of exposure bone lead concentrations represent, and how they relate to observable palaeopathological lesions is problematic.

An alternative indicator of lead burdens, that would eliminate a number of variables that make bone lead concentrations so dynamic, is tooth enamel. However, this would limit bioarchaeological investigations to non-adult exposure. As previously discussed in section 4.5, lead is incorporated into core tooth enamel as the tissue mineralises during childhood and it does not accumulate lead thereafter. This effectively provides a snapshot of an individual's lead exposure within a definable period of time during an individual's life, and allows the comparison of lead exposure at specific ages between individuals that died at different ages. Additionally tooth enamel lead concentrations are generally unaffected by many of the age and health related factors that influence the incorporation of lead into bone mineral. Therefore, the paired analysis of tooth enamel lead concentrations with the palaeopathological lesions on non-adult skeletal remains facilitates the comparison of childhood lead burdens with disease processes that occurred at a similar time to lead acquisition. This gives the best chronological correlation between lead concentrations and their contribution to the manifestation of certain pathological lesions.

# 4.7 Summary

The World Health Organisation recently reported that there is no known 'safe' blood lead concentration (WHO, 2011), stating that lead is toxic even at sub-clinical levels under 5  $\mu$ g/L and it would appear that for as long as people have been utilising lead, they have been aware of its pernicious qualities. The Romans were no exception, with historical documents clearly detailing maladies associated with lead poisoning and references to the dangers of its ubiquitous use. The bone seeking quality of lead lends itself to archaeological studies into how environmental pollution impacted upon health. However, the small numbers of bioarchaeological studies that have attempted to identify lead poisoning in archaeological remains have had some difficulty in determining what constitutes *in vivo* lead concentrations and what has been acquired from the burial environment. The limitations identified by these early studies can help shape the way future works approach the problem. It is clear that the use of bone, an innately porous material, is unlikely to yield accurate results as it readily exchanges ions with its burial environment. Therefore tooth enamel, which is a much more diagenetically stable tissue, would provide a better sample medium.

While all of the mechanisms behind lead's biochemical interactions are not fully understood it is clear that lead poisoning can result in the manifestation of numerous metabolic disorders. As some metabolic diseases are identifiable on skeletal remains there is a real potential for the identification of individuals suffering from lead poisoning in archaeological populations. The paired analysis of palaeopathological and tooth enamel lead concentration data provides the most robust strategy when attempting to identify individuals that suffered from lead poisoning. This does however, restrict bioarchaeological studies of lead poisoning to childhood as this is when both datasets are most likely to overlap with the time of lead exposure.

# CHAPTER FIVE

# Roman Health and Mortality

# **5.1 Introduction**

Due to the continuous movement of people and conquest of new lands, living in the Roman Empire meant living in a world of change and diversity, and this included changes to the health and mortality of the population (Redfern et al., 2018). Numerous studies have shown how changes to living environments (urbanisation), diet and population diversity can negatively impact upon health (Larsen and Milner, 1994). The high infant mortality rates and increased prevalence of infectious and metabolic diseases evident in skeletal material demonstrates how the Roman Empire was no exception. This chapter provides a brief overview of health and childhood mortality within the Roman world. Important parallels are drawn between the emergence of diseases known to be associated with lead poisoning and the dramatic increase in lead pollution seen during this period, and the concept that lead poisoning may have contributed to the high infant mortality rates evident throughout the Roman Empire is also introduced.

# 5.2 Roman health

Historical evidence for health and disease within the Roman Empire is relatively scarce, and is largely anecdotal, focusing predominantly on Mediterranean provinces such as Italy (Pitts and Griffin, 2012). However, there appears to be a general consensus that urbanisation, ethnicity and inequalities in wealth and social status were important factors influencing the health of populations throughout the Empire (LaVeist and Isaac, 2012). Although some studies have shown that living in rural environments came with its own negative effects on health (Lewis, 2010; Rohnbogner, 2017; Rohnbogner and Lewis, 2017), it was urban living that proved more deleterious to health (Redfern et al., 2015). The development of road networks and overcrowding that came with Roman urbanisation likely facilitated the easy transmission of disease within and between communities (Roberts and Cox, 2003, p. 389). Urban centres had close living quarters, inadequate waste disposal infrastructures, increased exposure to migrants and higher levels of smoke and lead pollution (Morley, 2004; Roberts and Cox, 2003, p. 389; Scobie, 1986). These characteristics of urban living likely influenced the high prevalence of infectious diseases such as malaria and tuberculosis in towns and cities compared with life in the Empire's rural settlements (Griffin et al., 2011; Morley, 2002; Redfern and Roberts, 2005; Redfern et al., 2015; Sallares, 2002; Scheidel, 2003). Even Roman culture often promoted the benefits of rural living, suggesting that people at the time were aware of the disparity in health associated with urban versus rural living (Baker, 2018; Eaton, 2014, p. 89). However, the impact of these variables on health cannot be fully understood solely from documentary evidence, which tends to be biased towards those of high status and social influence. Here osteological evidence of disease provides useful insights into the wellbeing of wider Roman populations (Gowland, 2017).

A number of studies have explored the health of Roman populations through the analysis of skeletal remains. The majority of these studies have been conducted on British and Italian populations (Bonfiglioli et al., 2003; Bonsall, 2013; Cucina et al., 2006; FitzGerald et al., 2006; Gowland and Redfern, 2010; Minozzi et al., 2012; Redfern, 2008; Redfern et al., 2015; Rohnbogner, 2017; Rohnbogner and Lewis, 2017),

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and very few of these have directly compared urban and rural populations with contemporaneous sites from other regions of the Empire. Although relatively few in number, these bioarchaeological studies concur with textual sources, demonstrating that there are higher levels of disease and childhood mortality at urban locations (Jongman et al., 2019; Redfern and Roberts, 2005). Despite the increasing wealth of evidence supporting the notion of poorer health in urban environments not all studies have observed the same pattern. In their analysis of four 4<sup>th</sup> century AD sites in Croatia, Šlaus et al. (2004) found no difference in health between settlement types, indicating that there is local variability in the impact that socioeconomic factors have upon health throughout the Roman Empire. In Romano-British contexts, a comparison of the urban site of Poundbury Camp, Dorset with contemporaneous rural sites also found that rural poverty affected the health of those growing up in the countryside to a similar extent as living in urban environments affected city dwellers (Rohnbogner and Lewis, 2017). From their analysis of British and Italian populations, Gowland and Redfern (2010) point out that Roman health patterns are complex, not simply a function of toxic environments, and that local variability in weaning practices, migration levels and living environments play an important role in the health of a population.

Using the presence or absence of grave goods to infer the socioeconomic status of individuals a number of studies have also attempted to correlate poor health with socioeconomic status. Jenny (2011) found more non-adults buried with grave goods had skeletal markers of stress than non-adults buried in unfurnished graves, Griffin et al (2011) found the same correlation at Baldock, Hertfordshire. Conversely, Redfern and DeWitte (2011) found that higher status individuals had lower mortality risks than lower status groups. However, as Gowland (2016) states, the interpretation of an individual's or population's health status is not straightforward and it is important not to

over simplify any correlations made between osteological evidence of poor health and perceived indicators of status from the burial environment.

It is clear that the factors affecting Roman health across the Empire were multifarious, and the extent of local variations in health and their relationship with socioeconomic inequality in this period requires further investigation. However, an avenue of research that has been relatively overlooked in Roman health studies is the impact that environmental lead pollution had upon populations adapting to Roman rule. From studies examining differences in health during the late Iron Age and Roman period in Britain it is apparent that the changes that came with Roman occupation had a generally negative impact upon health (Redfern, 2008; Roberts and Cox, 2003). The majority of these changes have interesting parallels with pathological alterations consistent with lead poisoning (see Chapter 4). As environmental lead pollution increased during the Roman period compared with the Iron Age, there was a concurrent increase in prevalence rates of dental disease and osteoporosis (Roberts and Cox, 2003; Touger-Decker and Van Loveren, 2003b), it was also the first time that metabolic diseases such as gout, rickets, osteomalacia and scurvy were seen in British populations (Roberts and Cox, 2003). Skeletal evidence from Britain, Italy and Gaul also reveals that there was a reduction in average adult stature compared to Iron Age populations (Giannecchini and Moggi-Cecchi, 2008; Gowland, 2017; Gowland and Walther, 2018; Redfern and DeWitte, 2011; Roberts and Cox, 2003; Scheidel, 2010b). Significantly, during the Anglo-Saxon period, a time when environmental lead pollution and human lead burdens decreased (Montgomery, 2002; Montgomery et al., 2010; Settle and Patterson, 1980), the average stature of British populations increases alongside a reduction in skeletal evidence of these diseases (Gowland, 2017; Roberts and Cox, 2003).

With regards to childhood health much of attention has been paid to the Romano-British site Poundbury Camp, Dorset (Farwell and Molleson, 1993; Lewis, 2010; Molleson, 1992, 1989; Molleson and Cox, 1988; Redfern, 2007; Redfern et al., 2012; Rohnbogner and Lewis, 2017). This site offers a unique opportunity to explore childhood health as the site contained an unusually large proportion of non-adult individuals (n = 364, <18years of age), 75 of which have been categorised as perinatal infants (Molleson, 1989), the age group most susceptible to the deleterious effects of lead. In the initial evaluation of the skeletal remains from this site, Molleson (1989) noted that a high proportion of the infants exhibited evidence of metabolic disease. A re-evaluation of the site two decades later by Lewis's (2010) corroborates these findings, with results indicating that over 30% of infants exhibited pathological alterations consistent with rickets (vitamin D deficiency) and scurvy (vitamin C deficiency). Although both Molleson (1989) and Lewis (2010) suggest that the high prevalence of metabolic disease is a result of early weaning, Molleson also goes on to posit that lead contamination of weaning foods and water may have contributed to the poor health of this vulnerable group of individuals. High rates of infant mortality and metabolic disease is not unique to Poundbury Camp, analyses of other Romano-British sites such as Butt Road, Colchester (Crummy and Crossan, 1993), Cannington, Somerset (Rahtz et al., 2000), Winchester (Ottaway et al., 2012) and Mays et al's (2018) study of 15 Mediterranean Roman 1<sup>st</sup> to 6<sup>th</sup> century sites have revealed high numbers of non-adults with evidence of poor health.

The overarching observation that can be made here is that Roman health was generally poor across the Empire, with individuals from all site types affected by metabolic diseases and infections (Rohnbogner and Lewis, 2017). Of course no singular cause can be ascribed to the decline of Roman health compared with that of previous populations, as socioeconomic inequalities, population growth and the implementation of Roman hierarchical systems will have all impacted upon a populations health (Gowland, 2017). However, there appears to be compelling evidence for the inclusion of environmental lead pollution as a contributing factor in the contextualisation of a population's health within their sociocultural sphere.

# **5.3 Childhood mortality**

Life expectancy during the Roman period has long been an area of interest within Roman research, and the basis of much of this interest stems from the wealth of age-atdeath data surviving in the form of inscriptions, census records and skeletal evidence (Hope, 2009, p. 42). However, this data is not always reliable or representative of the population as a whole. For example, ages inscribed on funerary monuments were not always precise and appear to have been either rounded up or down (Hope, 2009, p. 42). Additionally infants, children and women tend to be underrepresented epigraphically, with a distinct bias towards the commemoration of males in funerary monuments (Carroll, 2011). To compensate for the uncertainty surrounding this method, estimating life expectancy at birth and infant mortality rates has also been attempted via the comparison of Roman populations with modern life tables. These calculations suggest that the average life expectancy was 40 - 50 years of age if adulthood was reached, but that the chances of surviving past childhood were low (Hope, 2009, p. 43). However, critics of this method argue that life tables overlook important spatial and temporal variances in climatic and epidemiological conditions and likely provide an inaccurate proxy of Roman demographics (Pilkington, 2013; Scheidel, 2001b).

Whichever way the demography of Roman populations is assessed, it is clear that child mortality was high, with a large proportion of children failing to survive into adulthood (Carroll, 2006, p. 176; Garnsey, 1991, p. 51). As mortality is influenced by the living environment (urban versus rural) and the socioeconomic status into which a child is born, child mortality rates are likely to have varied considerably throughout the Empire (Gowland et al., 2014). However, there tends to be a general consensus that up to 50% of children died before the age of 10 years, with 20 - 40% of these dying in their first year of life (Carroll, 2014; Harlow and Laurence, 2002, p. 8; Parkin, 1992, p. 92; Rawson, 2003, p. 104; Saller, 1997, p. 12). Childbirth was extremely risky for both the mother and the infant, resulting in the frequent deaths of both (Jackson, 1988, p. 86), and although it will never be possible to determine exactly what percentage Roman children died in infancy or early childhood, there is a significant amount of textual evidence indicating that a large number died within the first few days of life (Bradley, 2005, p. 92; Dasen and Späth, 2010; Garnsey, 1991, p. 57; Shaw, 2001, p. 97). For example, it is reported that Cornelia, the mother of politicians Tiberius and Gaius Gracchus, gave birth to a total of 12 children, of which only three survived into adulthood (Hope, 2007, p. 10). Furthermore, texts by Hippocrates and Celsus both note that the first and seventh months were the times when an infant was most at risk of dying (Adams, 1849), while Plutarch identified the first week as the time in which infants were most vulnerable (Rose, 1974). Plutarch's identification of the seventh day as the most dangerous for newborns coincides with the deadline for fathers to decide whether they were going to rear their child, before the naming day on either the eighth (for girls) or ninth day (for boys) after birth (Harlow and Laurence, 2002, p. 39; Rawson, 2003, p. 105).

There is no doubt that childhood was a perilous stage of life during the Roman period, with higher risk of mortality evidenced by high numbers of infant remains (Gilfillan, 1965). Some of the reasons put forward for the high infant mortality rates seen throughout the Roman Empire include malnutrition, disease, poor medical knowledge and infanticide (Gowland et al., 2014; Mays, 1993; Pilkington, 2013). However, it is unlikely that there is a singular underlying reason for the high number of infant deaths, and rather it is a combination of sociocultural and epidemiological factors (Molleson, 1989). Nonetheless the high levels of environmental lead pollution seen throughout the Empire may have exacerbated the problem (Aneni, 2007). Infants and young children are more susceptible to lead poisoning than adults as their developing bodies are prone to absorbing higher quantities of ingested lead (see Chapter 4). It has also long been known that a mother's lead burden can cross the placental barrier as well as collate in breast milk, therefore not only is the developing foetus at risk to succumbing to the toxic effects of lead poisoning but so too are breastfeeding infants (Gulson et al., 2003). Despite this, and modern documentary evidence of lead poisoning being responsible for stillbirths, spontaneous abortion, congenital deformities and metabolic disease in infants (Hertz-Picciotto, 2000; Nriagu, 1983; Wibberley et al., 1977; Woolley, 1984), little has been done to explore any link between childhood lead exposure and high infant mortality rates within the Roman world.

## 5.4 Infanticide and exposure

Infanticide is defined as the intentional killing of children under the age of 12 months (Garner, 2001, p. 442), and historical and ethnographical evidence demonstrates that past and present populations have had a long history of this practice (Bonsall, 2013; Langer, 1974; Scrimshaw, 1984). In past societies, the practice of infanticide appears to have been relatively common, with a multitude of societies on every continent and from all levels of cultural complexity openly accepting it with little to no moral stigma (Mays, 2000; Montague, 1989; Warren, 1985; Williamson, 1978). Although there are a

multitude of socioeconomic, cultural and religious pressures that can result in increased levels of infanticide (Kelly, 1992; Kilday and Watson, 2008; Leboutte, 1991), a detachment towards the killing of new-borns is predominantly prevalent in societies that do not consider children as fully-fledged social beings (Harlow and Laurence, 2002). This view was held by Roman societies who considered childhood to constitute a separate stage of a person's life course before transitioning into adulthood (Harlow and Laurence, 2002; Hrdy, 1992).

In part due to the abundance of textual evidence documenting its practice during this period, infanticide has received a lot of scholarly attention (Bennett, 1923; Engels, 1980; Harris, 1994). Infanticide was thought to have been permitted by Roman law, however, the interpretation of this textual evidence has been questioned, with some scholars highlighting that these texts refer to exposure or abandonment of infants, particularly those that appear to have a physical disability, not the direct killing of newborns (Gowland et al., 2014). There is no certainty surrounding the understanding of the motives behind abandonment or exposure (Grubbs, 2013; Harris, 1994), however, it is thought that a father could dispose of a child on the basis of disability, illegitimacy, poverty and sex (Scott, 2001). This abandonment of infants was carried out with the assumption that the child would be recovered and cared for by another, even if as a slave (Amundsen, 1987, p. 6; Hope, 2007, p. 13; Patterson, 1985, p. 105). Therefore, the legality of abandonment cannot be equated with infanticide, as there was no direct intent to kill the child (Krause, 2011, p. 636). Infanticide was only condoned in cases of severe physical deformity (Laes, 2008, p. 95; Southwell-Wright, 2013, p. 80; Stahl, 2011, p. 721).

Archaeologically, infanticide was initially inferred from the widespread exclusion of infants from formal cemeteries and their high occurrence in excavated villa and settlement sites (Gowland et al., 2014). This burial practice has been interpreted as an indication of unequivocal emotional detachment to infant mortality, resulting in unceremonious deposition of infant remains (Cocks, 1921, p. 150; Frere, 1987; Molleson, 1999). However, this interpretation is conjectural, as ethnographic evidence has shown that many societies afforded infants different burial rites to their adult counterparts (Craig-Atkins, 2012; Millett and Gowland, 2015). As such, the preponderance of Roman infant burials in domestic sites may simply be an indication of different burial rites for infants, not necessarily infanticide (Harris, 1982; Moore, 2009; Pearce, 2000; Scott, 2001; Ucko, 1969).

More recently, archaeological infanticide studies have focused on the age-at-death distribution of perinatal deaths, particularly within Romano-British populations. The pronounced peak observed around the age of 38 - 41 gestational weeks has often being interpreted as evidence of widespread infanticide (Mays, 2000, 1993; Mays and Eyers, 2011; Mays and Faerman, 2001). This view has generated considerable debate from a number of authors who question both the validity of the scientific methods employed and the interpretation of the textual evidence used (Gowland, 2001; Gowland et al., 2014; Gowland and Chamberlain, 2002; Millett and Gowland, 2015). Rather it is suggested that the age distribution of infants is consistent with natural mortality rates, and that the 'unusual' burial locations within domestic environments is a specific funerary rite associated with this age group, a careful choice with links to ritual and beliefs, not a random disposal of unwanted children (Moore, 2009). Therefore, although the archaeological record leaves no doubt that Roman populations endured high infant

mortality rates, there appears to be little evidence that infanticide significantly contributed to them.

## **5.5 Summary**

Life was fragile in the Roman world and death, especially of the very young, was a frequent occurrence. In an ever-expanding Empire it was a time of socioeconomic and sociocultural change, and this change appears to have brought about an increase in disease prevalence and high infant mortality rates. Although it has been established that poor health often has multifaceted aetiologies, the correlation between the emergence of metabolic diseases and shortened stature, both of which can be caused by lead poisoning, in a time when lead pollution peaked opens up intriguing questions as to the role lead played in the health and mortality of Roman children.

The high lead burdens that have been observed in Roman populations indicates that foetuses and new-borns would have been at risk of acquiring lead from both placental transfer and breast milk, the effects of which could result in full-term still births, spontaneous abortion and infant death. Thus, there is compelling evidence that lead poisoning may have been a contributing factor to the high infant mortality rates seen in Roman skeletal populations. As such, it is surprising that so little bioarchaeological research has been done to establish whether lead could have contributed to the high mortality rates seen in so many Roman populations. However, this research aims to help fill this void by exploring whether lead exposure contributed to the high infant mortality rates evident in Roman populations.

# CHAPTER SIX

# Materials and Methods

# 6.1 Introduction

This chapter provides synopses of the sites and skeletal samples used in this study. It also describes the skeletal analyses and data collection methods employed, alongside the sampling strategy, processing and analysis methods used. The statistical analyses applied to the data are also presented at the end of the chapter.

# 6.2 Methodology outline

The methodological process followed during this study is briefly outlined below:

- 1. Secure access to Roman skeletal material in a number of European countries.
- 2. Create a sampling strategy, skeletal recording forms and database to record the osteological and contextual information from each site.
- 3. Carry out osteological analysis of skeletal material to determine age, sex (where possible) and presence of disease.
- Collect tooth samples from 12 adults (six females and six males) and 20 nonadults (10 with skeletal evidence of metabolic disease and 10 without) from each site.
- 5. Where information is available, collect, translate and record contextual data (grave goods, grave type etc.).
- 6. Remove and clean tooth enamel samples from all collected teeth at Durham University Isotope Laboratory.

- Process and analyse enamel samples using MC-ICP-MS and ICP-MS for lead isotope and trace element analysis respectively at NERC Isotope Geoscience Laboratory (NIGL), Keyworth.
- 8. Combine isotope data with osteological and contextual information.
- 9. Compare results with previously published lead isotope data.
- 10. Run statistical analyses to determine the significance of the results.

# 6.3 Sites

As this study aimed to go some way to bridging the gap in human lead isotope data from Roman provinces outside of Britain, the majority of sites were spread across mainland Europe (see Fig. 6.1). Samples from Lebanon were included as it represents the most easterly border of the Empire. Additionally, although human lead isotope ratios are well established in Britain, a small number of individuals excavated in Scotland and England were included due to their unusual burial rites. This section introduces the sites included in this study, using both published articles and unpublished reports provided by the curating institutions. Any documents not written in English were translated using Google Translate. All Spanish and Arabic translations were provided by Laura Castells-Navarro (University of Bradford) and Vana Kalendrian (University of Groningen) respectively.



**Figure 6.1** – **Map showing the location of the nine sites used in this study.** Musselburgh, Scotland; York, England; Ilchester, England; Tarragona, Spain; Barcelona, Spain; Caen, France; Ljubljana, Slovenia; Alba Iulia, Romania; Beirut, Lebanon.

### 6.3.1 Dealul Furcilor - Alba Iulia, Romania

Dealul Furcilor (Pitchfork Hill) is a large Roman hill necropolis located next to the urban centre Apulum in Alba Iulia, and was excavated by D. Protase in 1956-58, 1970-71 and again in 2006 by G. Bounegru and R. Ota. From artefactual (most notably bronze coins) and stratigraphic evidence, the site is thought to have been in use during the 2<sup>nd</sup> to 4<sup>th</sup> centuries AD. The necropolis was a mixed rite cemetery with cremations and inhumations present, which was a normal practice for Dacia and other Northern provinces of the Roman Empire at the time. A total of 227 burials were excavated, and although variation in burial orientation was displayed the majority of inhumations were aligned east-west or west-east. Both simple oval 'boat' pits and more elaborate stone sarcophagi were used for inhumation burials and coins, textiles, ceramics, jewellery and

animal bone were recovered from both types of inhumation (Gligor et al., 2010; Ota, 2009).



Figure 6.2 – Plan of the Dealul Furcilor excavation site (Source: Ota, 2009)

## 6.3.2 Beirut, Lebanon

*ASH 002* – A team led by Dr. Assaad Seif (DGA) excavated this site in 2007, and dating based on stratigraphy, grave goods, and pottery sherds suggest the site was in use during the 1<sup>st</sup> to 4<sup>th</sup> centuries AD. Inhumation was the only burial rite practised at ASH 002 and the 122 individuals recovered were buried in north-south and east-west orientations. Grave types consisted of simple pits, wooden coffins, terracotta sarcophagi, stone sarcophagi, stone cists, and one masonry tomb with multiple cells. Grave goods found at the site include jewellery, glass vessels, coins, metal objects, gold leaves and shells.

*ASH* 163 – Recent rescue excavations led by Dr. Georges Abou Diwan (Lebanese University) in 2015 at the ASH 163 site revealed 20 graves, all of which contained inhumation burials. Dating based on stratigraphy; grave goods and pottery sherds determined that the site was in use between the 1<sup>st</sup> century BC and the 2<sup>nd</sup> century AD. The type of graves included simple pits, pits with stone capping, wooden coffins, and terracotta sarcophagi. The majority of individuals were interred in an east-west orientation, with only two individuals placed on the north-south axis. Grave goods were plentiful at the site, with many graves containing items such as ceramic unguentaria, glass vessels, coins, jewellery, bone objects, metal objects.

*BCH* 740 – Site BCH 740 is currently being excavated by Dr. Georges Abou Diwan (Lebanese University). To date approximately 200 individuals have been recovered and while inhumation is the dominant burial rite practised at the site cremation rites are also represented. Types of graves included masonry tombs, simple pits, terracotta sarcophagi and wooden coffins, and preliminary dating using grave goods suggest that the site was in use during the  $1^{st} - 4^{th}$  centuries AD.

*MDWR 466 and MDWR 468* – Assaid Seif undertook excavations of sites MDWR 466 and MDWR 268 in 2009 and 2011 and using stratigraphic evidence and artefacts determined that the site was in use during the  $1^{st} - 4^{th}$  century AD. The sites were located adjacent to one another and are thought to constitute part of the same cemetery. A total of 41 individuals were recovered from the excavations and at both sites inhumation was the only burial rite present. Single burials representing the majority of inhumations, however five graves did containe multiple burials. The inhumation rite showed some variation across both sites with a combination of simple pit burials, simple pits with stone cappings, stone cists, wooden coffins, terracotta and stone sarcophagi being used. The MDWR sites were rich in grave goods with items such as glass vessels, spindles and spindle whorls, jewellery, coins, gold leaves, hobnails and shells included in many of the burials.

*MDWR 02* – The recent rescue excavation at MDWR 02 in 2011 by Assaad Seif resulted in the recovery of 89 individuals, all but one of which were inhumations interred in north–south and east–west orientations. The only cremation was interred in a lead urn. Grave types at the site varied considerably, and included simple pits, wooden coffins, terracotta sarcophagi, burial jars, a masonry tomb and a masonry tomb with multiple cells. Grave goods such coins, pottery vessels, glass vessels, jewellery, spindle whorls and metal objects were included in many of the burials. From these artefacts, pottery sherds and stratigraphy, the site was thought to be in use during the 1<sup>st</sup> century AD.

*RML 2385* – Dr. Assaad Seif (DGA) excavated Site RML 2385 in 2009. Dating of the site based on stratigraphic evidence, grave goods and pottery sherds suggest the site was in use from early 1<sup>st</sup> century BC to the 1<sup>st</sup> century AD. The human remains at RML 2385 consisted of 25 inhumations interred in an east-west orientation, one cremation and 8 disarticulated contexts, truncated by later activity at the site. The majority of burials consist of simple pits, but there were also a small number of wooden coffins, a single lead urn, one hypogeum and four masonry tombs. Grave goods from the site include pottery and glass vessels, jewellery, coins, gold leaves and bone objects

## 6.3.3 Michelet - Caen, France

Michelet was a necropolis in Northern France and was excavated in 1990-93 as a result of building works commissioned by the municipality of Lisieux. Stratigraphic, artefactual and documentary evidence indicate that the cemetery was in use from the late 3<sup>rd</sup> to 9<sup>th</sup> centuries AD. Inhumation was the predominant burial rite and 970 individuals were recovered, of these 575 were in wooden coffins while eight, 3<sup>rd</sup> to 4<sup>th</sup> century AD individuals were interred in lead coffins, an uncommon occurrence for the region. Wooden underground burial chambers were also found during the excavations as well as a singular limestone sarcophagus used in an infant burial. Grave goods such as jewellery, hobnails, belt buckles, ceramics, glassware, wooden chests and leather bags were plentiful in the necropolis despite evidence for looting in some of the Roman graves (Paillard and Alduc-Le Bagousse, 2012).



Figure 6.3 – Plan of the Michelet excavation site (Source: Paillard and Alduc-Le Bagousse, 2012).

## 6.3.4 Santa Caterina - Barcelona, Spain

The Santa Caterina necropolis was located just outside the city walls of the urban centre Barcino (Barcelona). Miró, Oliver and Grandos carried out initial rescue excavations in 1984 and 1986, but it was in 1999 – 2002 that full excavation of the site was undertaken by Bordas and Torres to understand the evolution of the sites continuous occupation from the Bronze Age through to the mid 13<sup>th</sup> century AD. From stratigraphic evidence the Roman necropolis is thought to have been in use during the 4<sup>th</sup> to 6<sup>th</sup> centuries AD, and was delineated by walls made with stone, mortar and ceramics. A total of 130 individuals were recovered during the excavations, all of which were inhumation burials interred in southwest– northeast or northwest–southeast orientations. Despite the varied types of burials present (simple pits, coffins, amphorae, mausolea and tile burials), there was conformity in the position of the body. All adults were interred in extended and supine positions with their arms crossed over the pelvis or waist, and all of the infants were placed in a flexed position within amphorae (Aguelo et al., 2001; Arroyo et al., 2005).



Figure 6.4 – Plan of the Santa Caterina excavation site (Source: Arroyo et al., 2005).

## 6.3.5 PERI 2 - Tarragona, Spain

The PERI 2 Roman necropolis was located just outside the city walls of Tarraco (Tarragona), a major port city on the northeast coast of Spain, and was excavated in 1979, 1994-96, and 2001 by the Museu Nacional Arqueològia de Tarragona (MNAT). Through the use of artefacts (pottery, coins etc.) and stratigraphic evidence the site is thought to have been in use during the 3<sup>rd</sup> to 4<sup>th</sup> centuries AD. A small number of cremations were recovered at the site, though inhumation was the predominant burial rite practiced at the PERI 2 necropolis, with a total of 431 individuals recovered during the excavations. The inhumation rite showed some variation across the site with a combination of amphora, wooden coffin, sarcophagi, double sloped tile and flat roof tile burials present. Despite this variability the majority of individuals were interred in a supine position with an east-west orientation. The PERI 2 necropolis was rich in grave goods, with items such as jewellery, ceramics, hobnails, buckles, coins and small mirrors included in many of the inhumation burials (i Prast, 2011).



Figure 6.5 – Plan of the PERI 2 excavation site (Source: i Prast, 2011)

### 6.3.6 Western Emonske Necropolis - Ljubljana, Slovenia

The Western Emonske Necropolis was uncovered during excavations by B. Hofman at the site of a  $19^{\text{th}}$  century tobacco factory in 2009. The  $1^{\text{st}} - 4^{\text{th}}$  century necropolis was located outside the city walls of Emona (Ljubljana), alongside the Trieste road leading from the western side of the city. The necropolis was a mixed rite cemetery with cremations and inhumations present. The majority of cremation burials were in amphora or glass urns, with only a small number deposited directly into the ground. All inhumation burials were in rectangular tile graves and bone preservation was poor. Grave goods at the site included ceramic oil lamps and table vessels (Hofman, 2011; Hvalec et al., 2011).

## 6.3.7 Primary Health Care Centre - Musselburgh, Scotland

Excavations carried out by CFA Archaeology Ltd at Musselburgh in 2010 revealed archaeological features indicative of multi-phase activity. Four burial pits were identified as Iron Age, one of which was a stone-lined cist. All of the Iron Age pits contained fragmentary human remains, amounting to a minimum of six individuals (two pits contained double inhumations). A small number of grave goods were also recovered, including a brooch from one of the single burials. A further six inhumation burials, all without grave goods, were identified and radiocarbon dates suggest that they date from the Roman Period. Two skeletons displayed sharp force trauma to their cervical vertebrae and their skulls were displaced. Although the vertebrae of two other individuals were too fragmentary to observe any evidence of trauma, their skulls were also displaced indicating that these four individuals had been decapitated. The custom of decapitated burials is known from a number of Roman sites in England, most notably York (Tucker et al., 2014), but this site would appear to be the first time that this custom has been identified in Scotland (Kirby, 2016, 2011).



Figure 6.6 – Plan of the Musselburgh excavation site (Source: Kirby, 2016 in press. © CFA Archaeology Ltd)

## 6.3.8 Lead coffin burials - York and Ilchester, England

After accidental discovery by a local farmer while ploughing his land, a single Roman lead coffin burial was unearthed in York, England (Wilson, pers. comm.). The burial was subsequently excavated by Yorkshire Archaeological Trust in 2008, and analysed at the University of York. Osteological analysis suggests that the individual was an adult male over 45 years old (Wilson, pers. comm.). More recently a Roman  $3^{rd} - 4^{th}$  century lead coffin burial was unearthed by a metal detectorist in Ilchester, England and excavated by South West Heritage in 2013 (Hopkinson, 2013). Analyses of the remains were carried out at the Biological Anthropology Research Centre (BARC), University of Bradford. Osteological analysis indicated that the human remains were those of a

young adult female, approximately 1.55m tall, with a well-healed rib fracture. Carbon and nitrogen isotope analysis revealed the individual to have had a terrestrial C<sub>3</sub> based diet typical of Roman Britain (Hopkinson, 2013).

Lead coffin burials are an uncommon burial rite in Roman-Britain and have previously been shown to be associated with migrants (Montgomery et al., 2010; Müldner et al., 2011). These two burials provide a rare opportunity to explore the geographic origins of individuals with intrusive burial rites. Naturally, with any lead coffin burial there is always a concern regarding contamination of the human remains from the burial environment. However, a study using a Roman lead coffin burial from Spitalfields, London has shown that human tooth enamel samples can be successfully analysed from these types of burials without contamination masking *in vivo* isotope ratios (Montgomery et al., 2010). However, to ensure the validity of the results a small sample from both lead coffins were also analysed for comparison.

# 6.4 Osteological analyses

Biological identity typically refers to the age and sex of an individual. To establish the biological identity of the individuals used in this study, standard osteological methods of assessment were used (Brickley and McKinley, 2004; Buikstra and Ubelaker, 1994). The level of preservation and completeness of the skeletal remains often dictates which methods can be applied during analysis. The burial environment (soil type, pH, moisture levels etc.) and subsequent post depositional activity (animal activity, land reuse etc.) can significantly influence the level of taphonomic damage to the bones (Walker, 1995, pp. 35–36), affecting which osteological methods can be applied to the skeletal remains. Therefore, not every osteological method was applied to every individual studied, instead the analysis was adapted to incorporate the methods most

appropriate for the individual being analysed. The methods used in this study are separated according to their purpose (i.e. assessing sex or estimating age-at-death) and are discussed below.

#### 6.4.1 Sex assessment

Observations of the morphological differences in the ossa coxae offer the highest accuracy levels for sex assessment, with reported confidence levels of 90–95% (Işcan and Derrick, 1984; Phenice, 1969). Morphological observations of the skull have also demonstrated high levels of accuracy for sex assessment; Walker's (1995) method provides accuracy of  $\geq$ 90% (Buikstra and Ubelaker, 1994; Loth and Henneberg, 1996). Other studies do not corroborate these results, reporting accuracy levels via skull morphology of 62-68% (Donnelly et al., 1998; Haun, 2000; Hill, 2000). However, removing subjectivity with metric analysis proved to be less reliable than the objective methods, yielding accuracy levels of 80-88% (Bongiovanni and Spradley, 2012; Giles and Elliot, 1963; Kocak et al., 2003; Meindl et al., 1985). Therefore, morphological differences were used for the purpose of this study. However, it is important to note that these morphological changes are most accurate once the individual reaches puberty, when skeletal material differentiates sufficiently for reliable sex assessment (White and Folkens, 2005).

#### 6.4.1.1 Adult sex assessment

#### **6.4.1.1.1 Sexually dimorphic pelvic traits**

The pelvis exhibits the highest level of sexual dimorphism in the human skeleton, due to the tight genetic controls surrounding its development. This makes it the most accurate and reliable indicator of adult skeletal sex. Generally, the male pelvis is larger and more robust than the female pelvis. However, the female pelvis tends to be wider, with larger superior and inferior apertures to facilitate childbirth. As the morphological features of the pelvis used to assess sex are exhibited on a sliding scale between males and females, methods that provide a scale to assess sex allow for the natural variations seen between individuals of both sexes. One such method is Walker's (2005) five-point scale for assessing the sexual dimorphism of the greater sciatic notch (1 = female, 5 = male), which tends to be wider in females than males. The most reliable features of the pelvis for sex assessment are thought to be the Phenice traits on the pubic bone, which assesses the morphology of the ischio-pubic ramus, presence or absence of the ventral arc and the level of sub-pubic concavity (Phenice, 1969). As sexually dimorphic traits of the pelvis are thought to be more accurate than those of the skull, where possible pelvic traits (see Table 6.1) were used to assess the sex of each skeleton.

 Table 6.1 - Sexually dimorphic pelvic traits used for sex assessment

| Pelvic traits         | Male                   | Female               |  |  |
|-----------------------|------------------------|----------------------|--|--|
| Acetabulum            | Large                  | Small                |  |  |
| Auricular surface     | Large / Flat           | Small / Elevated     |  |  |
| Greater sciatic notch | Narrow                 | Wide                 |  |  |
| Ilium                 | Z shaped crest / High  | S shaped crest / Low |  |  |
| Ischial tuberosity    | Large                  | Small                |  |  |
| Obiturator foramen    | Large / Ovoid          | Small / Triangular   |  |  |
| Pelvic inlet          | Heart shaped           | Eliptical            |  |  |
| Pelvic outlet         | Narrow                 | Wide                 |  |  |
| Pre-auricular sulcus  | Absent                 | Present              |  |  |
| Pubic bone length     | Short                  | Long                 |  |  |
| Sacrum                | Long / Narrow / Curved | Short / Broad        |  |  |
| Ventral arc           | Absent                 | Present              |  |  |

(after Ferembach et al., 1979; Phenice, 1969; Walker, 2005)

## 6.4.1.1.2 Sexually dimorphic cranial traits

Although not as accurate as the pelvis due to the higher variability between individuals and populations (Mays and Cox, 2000; Meindl and Lovejoy, 1985), skull morphology can be useful in assessing the sex of an individual. As with the pelvis, differences in the morphological features of the skull are exhibited on a sliding scale between males and females, although male skulls tend to be larger and more robust than female skulls. Again, Walker (2005) took into account this natural variation in male and female sexual dimorphism and developed a five-point scale for sex assessment using select features of the cranium and mandible. Other cranial traits such as the size of the orbits and mastoid processes or the prominence of muscle attachment sites have been suggested as sexually dimorphic (Buikstra and Ubelaker, 1994; Mays and Cox, 2000; Meindl and Lovejoy, 1985; Rogers, 2005). Where possible all sexually dimorphic traits of the skull (see Table 6.2) were used to assess sex.

| Skull traits         | Male               | Female             |  |
|----------------------|--------------------|--------------------|--|
| Dental arcade        | Large / U-shaped   | Small / Parabolic  |  |
| Frontal eminence     | Small              | Pronounced         |  |
| Glabella             | Pronounced         | Faint              |  |
| Gonial angle         | Approx. 90°        | > 90°              |  |
| Gonial flaring       | Pronounced         | Slight             |  |
| Mastoid process      | Large              | Small              |  |
| Mental eminence      | Pronounced         | Indistinct         |  |
| Nuchal crest         | Pronounced         | Smooth             |  |
| Occipital area       | Robust             | Gracile            |  |
| Occipital condyles   | Large              | Small              |  |
| Orbits               | Rounded margins    | Sharpe margins     |  |
| Parietal eminence    | Small              | Pronounced         |  |
| Supra orbital margin | Rounded            | Sharp              |  |
| Supra orbital ridge  | Pronounced         | Faint              |  |
| Zygomatic bone       | Large / Arched     | Small / Compressed |  |
| Zygomatic arch       | Extends past E A M | Short              |  |

Table 6.2 - Sexually dimorphic traits of the skull used for sex assessment

(after Walker, 2005)

#### 6.4.1.1.3 Metric sex assessment

Metric analysis removes the innate subjectivity associated with macroscopic methods, and therefore has lower inter- and intra-observer errors than the macroscopic sex assessment methods described above (Adams and Byrd, 2002; Moore et al., 2016; Spradley and Jantz, 2011). Despite this, sex assessment using the morphology of the pelvis or skull is more accurate than metrical analysis. However, it does rely upon the skeletal elements being relatively well preserved. Therefore, postcranial metrics can offer a useful additional method for the sex assessment of incomplete of fragmentary remains (Acsádi and Nemeskéri, 1970; Rogers, 1999). The measurements used for sex assessment are summarised in Table 6.3.

| Skeletal element | Measurement            | Male (mm) | Female (mm) |
|------------------|------------------------|-----------|-------------|
| Clavicle         | Maximum length         | >150      | < 138       |
| Scapula          | Glenoid cavity width   | > 29      | < 26        |
| Humerus          | Epicondylar breadth    | > 60.1    | < 60.1      |
|                  | Vertical head diameter | > 47      | < 44.9      |
| Femur            | Maximum head diameter  | >48       | < 43        |
|                  | Epicondylar breadth    | >76       | < 74        |

Table 6.3 - Skeletal elements used for metric sex assessment

(after Bass, 2005)

### 6.4.1.2 Non-adult sex assessment

Many studies have attempted to develop techniques for juvenile sex assessment, yielding promising accuracy levels that ranged between 70-92% (Fazekas and Kósa, 1978; Loth and Henneberg, 2001; Schutkowski, 1993; Weaver, 1980). Tests of these methods have not been able to reproduce these results, achieving accuracy levels below 70% (Loth and Henneberg, 1996; Scheuer, 2002; Schutkowski, 1987). It is evident that population variation is a major limiting factor for non-adult sex assessment. Until geographically diverse studies are conducted to provide population specific standards, that are accurate and reliable (>90% confidence level), non-adult sex assessment is not possible (Veroni et al., 2010). While differences obviously exist they are as yet, not measurable; therefore non-adult sex assessment was not carried out during this study.

### 6.4.1.3 Application of sex assessment methods

Sex assessment was only carried out on adult (18+ years) individuals, using the methods outlined above (section 6.3.1). As certain skeletal elements provide higher accuracy when assessing sex those with stronger morphological associations with sexual

dimorphism were considered most indicative of sex. The results of the assessments were combined and used to place each individual into one of five categories (see Table 6.4), allowing for a degree of uncertainty that is often encountered when assessing fragmentary and incomplete remains. All non-adult individuals were categorised as indeterminate.

| Sex categories  |  |  |
|-----------------|--|--|
| Male            |  |  |
| Female          |  |  |
| Probable male   |  |  |
| Probable female |  |  |
| Indeterminate   |  |  |

 Table 6.4 - Sex categories used

#### 6.4.2 Age-at-death estimation

An individual has two ages, a chronological age that refers to the time an individual has been alive, and a biological age, which refers to how old an individual looks physically. Although it is always desirable that biological age is consistent with chronological age, the biological age of an individual is often influenced by factors such as environment, genetics, disease and activity. Age estimations within adult skeletal remains (>18 years) are based upon age related degenerative changes observable in the joint surfaces and dentition; however some epiphyses do fuse during early adulthood (c. 18-29 years). Within adult remains, various skeletal elements exhibit age related changes at differing rates in different individuals. Adding observer error to this creates substantial differences in estimates from actual chronological age. Therefore, current methods used to obtain age-at-death estimations tend to produce large age ranges (Buckberry, 2015; Buikstra and Konigsberg, 1985; Meindl et al., 1985). A

multifactorial approach to age-at-death estimation allows the comparison of average ages, providing estimates that correlate more accurately with chronological age than any singular ageing method (Mays, 2010).

With regards to non-adults, skeletal maturation can significantly vary between individuals in response to environment, genetics, disease and secular changes. However, dental development is thought to continue in its regimented developmental patterns regardless of most external stressors (Lewis, 2007). As the remains of non-adults in archaeological contexts are usually, by definition, not healthy individuals it is reasonable to suggest that age-at-death estimation via skeletal maturation is likely to be inaccurate. Therefore, dental age-at-death estimation methods, which provide well-documented and consistent stages of development, should be the primary method for non-adult age-at-death estimation (AlQahtani et al., 2010; Hillson, 1996). Long bone length and epiphyseal fusion can also provide accurate non-adult age-at-death estimates (Krogman and Işcan, 1986; Stewart, 1976; Ubelaker, 1987) and as with adults, a multifactorial approach is preferred, therefore they are also described below.

### 6.4.2.1 Adult age-at-death estimation

## 6.4.2.1.1 Dental attrition

Dental attrition refers to wear patterns on teeth and can be used to estimate age-at-death by assessing the degree of attrition on the occlusal surface of the permanent molars (Brothwell, 1981; Brothwell and Powers, 1967; Mays, 2002). Studies have shown dental attrition to offer accurate age-at-death estimates that correlate well with ages obtained using the pubic symphysis (Buikstra and Ubelaker, 1994; Hillson, 1996; Lovejoy, 1985; Miles, 1962). As dentition is frequently recovered from the burial environment and resilient to diagenetic alteration it is a useful age-at-death estimation
method, although it is population specific. In this study Brothwell's (1981) attrition method was used to estimate age-at-death of adult individuals.

## 6.4.2.1.2 Pubic symphysis

Todd (1920) pioneered the use of the pubic symphysis for age estimation, recording changes to the symphyseal surface (the anterior joint between the pubic bones of the pelvis). Tests of this method found it to significantly overestimate age after 40 years (Brooks, 1955), yet attempts to improve the method were unsuccessful (Gilbert and McKern, 1973; Katz and Suchey, 1986; McKern and Stewart, 1957; Meindl et al., 1985). However, Brooks and Suchey (1990) devised the Suchey-Brooks 6 phase method, which built upon Todd's (1920) work, improving the accuracy to a 95% confidence level. Although this method is sex specific, the phases can be combined to produce wide age ranges for those of indeterminate sex (Brooks and Suchey, 1990). The pubic bones are often absent or poorly preserved in archaeological remains, however due to the perceived high level of accuracy provided by the method it was applied when preservation and completeness allowed.

## 6.4.2.1.3 Auricular surface

The auricular surface is the joint between the ilia and sacrum, and is a popular target for age-at-death estimation methods because its robusticity results in high levels of preservation in the burial environment. Lovejoy et al. (1985) originally developed the use of the auricular surface (the joint between the ilia and sacrum) for age estimation. However, tests of this method have demonstrated inconsistent ageing accuracies and high levels of intra-observer error (Murray and Murray, 1991; Saunders et al., 1992). Buckberry and Chamberlain (2002) revised this method, proposing a technique that used a more objective scoring system, reduced inter- and intra-observer errors, was

independent of sex and had a better correlation with age than the Suchey-Brooks pubic symphysis method. Due to these improvements and the relative ease of its application, the Buckberry and Chamberlain (2002) method was used.

## 6.4.2.1.4 Cranial suture closure

At birth the bones of the skull are unfused and as individuals age the sutures (joints) between these bones gradually begin to fuse together, becoming less defined with increasing age. Cranial suture closure had been considered unreliable and inaccurate for decades (Brooks, 1955; McKern and Stewart, 1957), until Meindl and Lovejoy (1985) proposed a new method for observing the progression of suture closure at designated points on each of the cranial sutures. This new method reduced standard deviations and improved accuracy. Although they are not as reliable as other skeletal elements for ageing, they can contribute valuable information when used in conjunction with other methods (White and Folkens, 2005).

#### **6.4.2.1.5** Late fusing epiphyses

Epiphyses are the articular ends of bones, at birth these are separated from the main body of the bone by cartilage and through gradual ossification with increasing age they eventually unite (fuse) with the rest of the bone. Observing the level of ossification at the epiphyses is generally used to estimate non-adult age-at-death. However, a small number of epiphyses, such as the sternal end of the clavicle, vertebral annular rings, S1 to S2 in the sacrum, the iliac crest and the rib heads (Black and Scheuer, 1996; Scheuer and Black, 2000; Webb and Suchey, 1985) do not fully fuse until early adulthood. These provide an additional method of age-at-death estimation that is particularly useful in the assessment of incomplete or poorly preserved skeletons.

## 6.4.2.2 Non-adult age-at-death estimation

#### 6.4.2.2.1 Dental development and eruption

It has been well established that endogenous and exogenous stress can have significant effects on skeletal maturation, with the exception of dental development, which remains consistent in its regimented development. To take advantage of this age related constant, multiple methods have been developed in an attempt to accurately assess tooth formation, root resorption and eruption patterns in non-adults as a method of age estimation (AlQahtani et al., 2010; Demirjain et al., 1973; Moorrees et al., 1963; Schour and Massler, 1941). However, Ubelaker (1987) highlights that different methods can often produce varying results from the same material due to population specific standards. As AlQahtani et al.'s (2010) dental atlas is not sex specific and visually easy to use, it was the method used to estimate non-adult age-at-death.

### 6.4.2.2.2 Long bone length

Long bone length as an indicator of age-at-death becomes less accurate with increasing age due to individual and population variation and sex differences (Ubelaker, 2005). Therefore, this method is most accurate in foetal and infant (under 1 year) remains (Ubelaker, 1987; Ubelaker, 1988). Both foetal long bone length (Scheuer et al., 1980) and non-adult diaphyseal length (Scheuer and Black, 2000) were used to estimate age-at-death were preservation allowed.

## 6.4.2.2.3 Epiphyseal fusion

The gradual fusion of skeletal epiphyses predominantly occurs during early childhood and puberty. The first to ossify are the bones of the skull and vertebrae, usually fusing during early childhood, while long bone epiphyses and those of the pelvis tend to continue fusing throughout adolescence (Scheuer and Black, 2000). Although the age ranges associated with epiphyseal fusion is sex specific, they can be combined to produce wider age ranges that can be applied to individuals of indeterminate sex. As sex assessment of non-adults is currently difficult to conduct with an acceptable level of accuracy, a combined age range is the most appropriate range to be applied. As with late fusing epiphyses in young adults, observing the level of fusion in non-adults is a useful method for age-at-death estimates in incomplete or poorly reserved immature skeletons.

## 6.4.2.3 Application of age-at-death estimation methods

A multifactorial approach to estimating age-at-death was carried out using the methods outlined above. Using multiple methods on each individual produced a number of age ranges, which were combined to provide a single, overall age range for each individual. To facilitate the collation and comparison of age related data, ordinal age groups were used to create broad age categories (see Table 6.5). The ranges at which the age-at-death estimation methods overlapped were used to determine which age category an individual was placed into. Obtaining an age range for any given individual is dependent upon the level of preservation and completeness of the skeleton. In instances where only a minimum age could be estimated the individual was categorised as either adult or non-adult, and in those where estimating age-at-death was not possible an indeterminate category was included.

| Age category   | Age range          |
|----------------|--------------------|
| Foetal         | <40 weeks in utero |
| Infant         | 0 - 1 year         |
| Young juvenile | 2 - 6 years        |
| Older juvenile | 7 - 12 years       |
| Adolescent     | 13 - 18 years      |
| Young adult    | 19 - 25 years      |
| Middle adult   | 26 - 44 years      |
| Mature adult   | 45+ years          |
| Adult          | 18+ years          |
| Non-adult      | <18 years          |
| Indeterminate  | -                  |

Table 6.5 - Age categories used

### 6.4.3 Palaeopathological analysis

In skeletal populations, disease is identified through the observation of pathological alterations on the skeleton (Siek, 2013). However, those that die shortly after contracting an illness will show no skeletal indicators of disease, while those that survive longer with an illness will develop observable pathological lesions (Siek, 2013). This osteological paradox is well documented (DeWitte and Stojanowski, 2015; Wood et al., 1992), and although non-adult individuals in skeletal populations are not healthy irrespective of the presence or absence of skleletal lesions, for the purpose of this study the category 'without disease' has been used where no visible skeletal alterations are present. However, it is acknowledged that these individuals do not represent healthy children.

Due to the clinical manifestations of lead poisoning (see Chapter 4), palaeopathological analysis of the non-adult individuals focused on the identification of metabolic diseases. Although all pathological alterations present were recorded (i.e. non-specific infection, congenital abnormalities) their inclusion was outside the scope of this thesis. Therefore, the following section only outlines the palaeopathological features of the metabolic diseases often associated with lead poisoning and the parameters used to diagnose these diseases within the skeletal assemblages. No radiographs were available for any of the study sites; therefore diagnoses were made solely from macroscopic examination of the skeletal material.

## 6.4.3.1 Carious lesions

Carious lesions were macroscopically identified as the localised destruction of dental tissues (Hillson, 1996; Waldron, 2009, pp. 237–8), and were recorded as present or absent and by tooth position. Lesion size was estimated as small, medium, large, or total destruction (complete loss of the crown). The position on the tooth (crown, cemento-enamel junction, mesial, buccal etc) was also recorded.

# 6.4.3.2 Enamel hypoplasia

Enamel hypoplasia was identified as linear furrows, pitting, or grooves in the enamel surface of teeth (Ortner, 2003; Waldron, 2009, p. 244). The presence and tooth position of the defect was recorded and categorised in accordance with the guidelines outlined by Ogden et al., (2007) (see Fig. 6.7).

# IMAGE REMOVED BY AUTHOR FOR COPYRIGHT REASONS

Figure 6.7 – Categories of enamel hypoplasia (Source: Ogden et al., 2007).

## 6.4.3.3 Cribra orbitalia and porotic hyperostosis

Cribra orbitalia was identified as abnormal pitting or porosity of the orbital roofs and recorded for each orbit using the Stuart-Macadam (1991) grading system (see Table 6.6). Distinctions were made between headed/inactive and active cribra orbitalia based on the appearance of the lesions. Inactive cribra orbitalia was identified from the presence of smooth-edged lesions, while active lesions were identified from the presence of sharp edges (Mensforth et al., 1978, p. 23). Porotic hyperostosis was identified as pitting and/or porosity on the ectocranial surfaces of the skull and recorded as either present or absent (Mann and Hunt, 2013, p. 28; Waldron, 2009, p. 137). The presence or absence of marrow hyperplasia (thickening) of the cranial vault bones was also recorded.

Table 6.6 – Codes and pathological descriptions for presence and severity of cribra orbitalia

| Score | Description   |
|-------|---|
| 0     | No change to the bone surface                             |
| 1     | Capillary like impressions on the bone                    |
| 2     | Scattered foramina  |
| 3     | Large and small isolated foramina                         |
| 4     | Foramina have linked into a trabecular structure          |
| 5     | Outgrowth in trabecular form from the outer table surface |

(after Stuart-Macadam 1991: 109)

### **6.4.3.4 Rickets**

A multitude of developmental and pathological processes can cause osseous changes similar to those seen in vitamin D deficient non-adults. However, rickets is generally identified by the presence of the bowing of the long bones and/or the presence of widened, cupped and porous/frayed ('brush end') epiphyses, sternal rib-end flaring ('rachitic rosary') and cranial vault thinning (Waldron, 2009, p. 129). Additional manifestations of the disease in non-adults can also include orbital roof porosity, deformation of the mandibular ramus, porosity of the sternal rib-ends and deformation of the ribs (Brickley et al., 2005; Brickley and Ives, 2010; Mays et al., 2006; Ortner, 2003; Ortner and Mays, 1998). Using the published diagnostic criteria outlined in Table 6.7, macroscopic lesions were recorded as either present or absent. A diagnosis of rickets was only recorded if three or more probable rachitic features were present, or if there were bending deformities of the long bones plus one other feature. Individuals exhibiting no probable features but three or more possible features alongside any non-diagnostic features were considered as possibly rachitic. Using Ortner and Mays (1998) definition, a distinction was also made between healed and active rickets.

| Diagnostic category | Probable            | Possible                         | Non diagnostic                         |
|---------------------|---------------------|----------------------------------|--|
| Cranial             | Deformed mandibular | Cranial vault porosity           | Delayed Closure of                     |
|                     | ramus               |                                  | frontanelles                           |
|                     |                     | Orbital roof porosity            | Cranial bone thinning                  |
|                     |                     | Layers of spiculated,            | Frontal and parietal                   |
|                     |                     | irregular porous bone            | bossing                                |
|                     |                     | can occur during                 |  |
|                     |                     | healing when osteiod is          |  |
|                     |                     | millerunsed                      | Craniotabes (softening                 |
|                     |                     |                                  | over occipital region &                |
|                     |                     |                                  | adjacent to lambdoid<br>suture)        |
|                     |                     |                                  | Formation of large, square-shaped head |
| Post-cranial        | Deformed arm bones  | Flaring of sternal rib-<br>ends  | Superior flattening of the femora      |
|                     | Deformed leg bones  | Porosity of sternal rib-<br>ends |  |
|                     | Ilium concavity     | Long-bone                        |  |
|                     | A 1. 1 11 1         | metaphyseal flaring              |  |
|                     | Altered rib angle   | Long-bone thickening             |  |
|                     |                     | Porous roughening of             |  |
|                     |                     | long-bone metaphyses             |  |
|                     |                     | Long-bone concave                |  |
|                     |                     | curvature porosity               |  |

Table 6.7 – Rachitic lesions used in the identification of rickets and their diagnostic category.

(after Brickley and Ives, 2010; Hess, 1930; Mays et al., 2006; Ortner and Mays, 1998; Pettifor, 2011)

## 6.4.3.5 Scurvy

Pathological alterations indicative of scurvy primarily consist of porotic hyperostosis, cribra orbitalia and abnormal porosity (often with periosteal new bone formation) in the scapulae, long bone metaphyses, and mandible (Waldron, 2009, p. 132). These lesions tend to manifest bilaterally and are thought to be caused by chronic, low-grade haemorrhage of weakened blood vessels, predominantly at muscle attachment sites, which stimulates an inflammatory response (Ortner et al., 2001, 1999; Ortner and Ericksen, 1997). Although abnormal porosity is the primary lesion associated with scurvy, it is also common to many other disease processes such as specific and nonspecific infection, haemoglobinopathies, anaemias, and other metabolic disorders (Brown and Ortner, 2011; Lagia et al., 2007). It is therefore important to analyse the porosity in relation to its distribution across the entire skeleton. Using the published diagnostic criteria outlined in Table 6.8, macroscopic lesions were recorded as either present or absent. If three or more probable scorbutic features were present, the individual was recorded as scorbutic, while individuals exhibiting no probable features but three or more possible features alongside any non-diagnostic features were considered as possibly scorbutic.

| Diagnostic category | Probable   | Possible   | Non diagnostic   |
|---------------------|--|--|--|
| Cranial             | Porosity and/or new<br>bone formation on the<br>greater wing of the<br>sphenoid            | Porosity in the<br>mandibular coronoid<br>fossae                               | Porosity on the palate<br>of the maxilla                               |
|                     | Porosity on the<br>posterior aspect of the<br>mandible                                     | Porosity and/or new<br>bone formation on the<br>lesser wing of the<br>sphenoid | Porosity on the<br>maxillary and/or<br>mandibular alveola<br>processes |
|                     | Porosity in the temporal bone  | Porosity at the<br>infraorbital foramen on<br>the maxilla                      | Porosity and/or new<br>bone formation on the<br>endocranium            |
|                     |  | Porosity and/or new<br>bone formation on the<br>orbital roof                   |  |
|                     |  | Porosity and/or new<br>bone formation on the<br>pars basilaris                 |  |
| Post-cranial        | Porosity and/or new<br>bone formation in the<br>supraspinous and/or<br>infraspinous fossae | Metaphyseal flaring of long bones  | Porosity and/or new<br>bone formation on the<br>long bones             |
|                     |  | Flaring of sternal rib-<br>ends  | Metaphyseal porosity   |

| Table  | 6.8 -     | Scorbutic | lesions | used | in | the | identification | of | scurvy | and | their |
|--------|-----------|-----------|---------|------|----|-----|----------------|----|--------|-----|-------|
| diagno | ostic cat | tegory.   |         |      |    |     |                |    |        |     |       |

(after Brickley and Ives, 2010, 2006; Geber and Murphy, 2012; Moore and Koon, 2017; Ortner, 2003; Ortner et al., 2001, 1999; Ortner and Ericksen, 1997)

# 6.5 Data recording

Osteological and palaeopathological data were recorded on recording forms adapted from Buikstra and Ubelaker (1994) and Brickley and McKinley (2004). A Microsoft Excel database was created to collate osteological, contextual and isotope data. Dropdown options were inserted into the database where applicable to standardise and reduce errors in data entry.

## **6.6 Sampling strategy**

Due to its high resistance to diagenetic alteration and retention of *in vivo* elemental concentrations, dental enamel was targeted for trace element and isotope ratio analyses. All of the teeth selected for analysis were, where possible, free from pathological alterations (e.g. enamel hypoplasia etc.) and dental modifications, and where preservation allowed the selected tooth's antimere was present. Depending on availability, samples from 12 adults (six females and six males) and 20 non-adults (10 with skeletal evidence of metabolic disease and 10 without) were to be collected from each site.

## 6.7 Tooth selection

As different teeth form and mineralise at different times during childhood, the lead contained within different teeth reflects the lead acquired at different times of life. To ensure the tooth enamel analysed for lead isotope ratios did not contain any lead from the mother (acquired *in utero* and during breastfeeding), who may have had a different geographical origin to her child, the 2<sup>nd</sup> molar or 2<sup>nd</sup> premolar were selected from adult individuals. Both of these teeth begin forming at around three years of age and should provide isotope ratios congruent with the geographical region in which the individuals spent their childhood. Trace element and isotope ratio analyses of the adult individuals was also carried out on permanent 2<sup>nd</sup> molars or 2<sup>nd</sup> premolars, the mineralisation times of these teeth overlap with those of deciduous teeth allowing comparisons of childhood lead exposure. In non-adult individuals, deciduous incisors were preferentially selected for sampling as they are the earliest forming teeth and therefore facilitate the analysis of children as young as 30 weeks *in utero*. If these preferred teeth were unavailable, either

due to insufficient skeletal preservation or incompleteness, the earliest forming tooth available was selected in non-adults while the 3<sup>rd</sup> molar was used as a substitute in adult individuals. Tables 6.9 and 6.10 summarise the samples collected during the course of this study.

|                          | Adult | Adult  | Adult  | Tooth Type |        |    |           |    |    |  |
|--------------------------|-------|--------|--------|------------|--------|----|-----------|----|----|--|
| Site                     | male  | female | Indet. | Ma         | ndibul | ar | Maxillary |    |    |  |
|                          |       |        |        | PM2        | M2     | M3 | PM2       | M2 | M3 |  |
| Alba Iulia,<br>Romania   | 5     | 5      | 1      | 2          | 2      | 2  | 2         | 3  | -  |  |
| Barcelona, Spain         | 5     | 7      | 1      | 1          | 4      | 1  | 4         | 2  | 1  |  |
| Beirut, Lebanon          | 6     | 6      | 4      | 7          | 4      | -  | 5         | -  | -  |  |
| Caen,<br>France          | 7     | 7      | -      | 3          | 4      | -  | 3         | 2  | 2  |  |
| Ljubljana,<br>Slovenia   | -     | -      | 5      | 2          | -      | -  | 1         | 2  | -  |  |
| Musselburgh,<br>Scotland | 6     | -      | -      | 2          | 4      | -  | -         | -  | -  |  |
| Tarragona, Spain         | 7     | 5      | -      | -          | 3      | -  | 2         | 6  | 1  |  |
| Lead coffins,<br>England | 1     | 1      | -      | -          | 2      | -  | -         | -  | -  |  |
| Total                    | 37    | 31     | 11     | 17         | 23     | 3  | 17        | 15 | 4  |  |

Table 6.9 – Summary of permanent teeth from adults sampled for lead isotope and trace element analyses.

Key: Indet. = Indeterminate; PM2 = Permanent  $2^{nd}$  premolar; M2 = Permanent  $2^{nd}$  molar; M3 = Permanent  $3^{rd}$  molar.

|                        | No          | Tooth Type |      |       |       |      |           |       |       |  |
|------------------------|-------------|------------|------|-------|-------|------|-----------|-------|-------|--|
| Site                   | Individuals | Mandibular |      |       |       |      | Maxillary |       |       |  |
|                        |             | I(d)       | C(d) | M1(d) | M2(d) | I(d) | C(d)      | M1(d) | M2(d) |  |
| Alba Iulia,<br>Romania | 22          | 6          | -    | 4     | 2     | 9    | 1         | -     | -     |  |
| Barcelona,<br>Spain    | 21          | 5          | -    | 1     | 1     | 9    | 2         | -     | 3     |  |
| Beirut,<br>Lebanon     | 20          | 3          | 2    | 4     | -     | 6    | 4         | -     | 1     |  |
| Caen,<br>France        | 22          | 3          | -    | -     | 1     | 11   | 1         | 3     | 3     |  |
| Ljubljana,<br>Slovenia | 3           | -          | -    | 1     | -     | -    | -         | -     | 2     |  |
| Tarragona,<br>Spain    | 15          | -          | -    | -     | 2     | 12   | -         | -     | 1     |  |
| Total                  | 103         | 17         | 2    | 10    | 6     | 47   | 8         | 3     | 10    |  |

Table 6.10 – Summary of deciduous teeth from non-adults sampled for trace element analyses.

Key: I(d) = Deciduous incisor; C(d) = Deciduous canine; M1(d) = Deciduous 1<sup>st</sup> molar; M2(d) = Deciduous 2<sup>nd</sup> molar

## 6.8 Isotope analysis

Two of the main aims of this study are to explore how exposure to anthropogenic lead affected childhood health and investigate the variability of lead isotope ratios within the Roman Empire. In order to assess how lead burdens affected childhood health in Roman populations trace element analysis was carried out on all permanent and deciduous teeth collected during this study to determine their lead concentrations (ppm). To explore the variability of lead isotopes within the Roman Empire lead isotope analysis was carried out, and although not a main focus of this research, strontium was also analysed. Strontium isotope ratios are a well-established method for exploring mobility in bioarchaeological studies (see Chapter 3). However, as similar terrains are found across large expanses of northern Europe and Britain, strontium isotope ratios are often insufficiently unique enough to differentiate between people from different regions that have similar geology. As lead in Roman individuals is thought to derive mostly from anthropogenic ore exposure not solely local geology, it is possible that lead may show more cultural variability across Europe than strontium. To explore the variability of lead in comparison to strontium within the Roman Empire and to assess whether the combination of lead and strontium can improve not only our ability to identify migrants within skeletal populations but also narrow down possible regions of origin, both strontium and lead isotope ratios were analysed in all permanent teeth collected from adult individuals.

### **6.8.1 Sample preparation**

Initial sample preparation was carried out at Durham University Isotope Laboratory, following procedures outlined by Montgomery (2002). The enamel surface was abraded using a tungsten carbide dental bur to remove surface contamination. Following this, a chip of core enamel approximately 20–30 mg in weight was removed using a flexible diamond edged rotary saw, all exposed surfaces of the chip were abraded to remove any adhering dentine and potential sources of contamination. All dental tools were cleaned between samples via ultrasonication in Decon for 5 minutes and rinsed three times with ultra-pure deionised water. Clean core enamel chips were sealed in microcentrifuge tubes and transferred to the Class 100, HEPA filtered laboratory facilities at the Natural Environment Research Council Isotope and Geoscience Laboratory (NIGL) in Keyworth, Nottingham.

At NIGL all enamel samples were rinsed three times with high purity water (Millipore Alpha Q), and then soaked at 60 °C for one hour. The samples were then rinsed again before being leached with 0.2M HCl for five minutes. After a final rinse, the samples were dried and transferred into pre-cleaned Teflon beakers and dissolved in 8M HNO<sub>3</sub>.

Samples were converted to chloride using 6 M HCl, taken up in titrated 2.5 M HCl and pipetted onto ion-exchange chromatography columns. The strontium was separated using a Dowex<sup>®</sup> resin. The washes from the chloride samples were collected, dried down and converted to a bromide form using HBr to facilitate the separation of lead using conventional anion exchange methods. In preparation for lead concentration analysis, which was carried out separately to lead isotope ratio determination, all samples were diluted with 1% v/v HNO<sub>3</sub>. 0.5% v/v HCl before ICP-MS analysis was carried out.

#### **6.8.2 Mass spectrometry**

#### **6.8.2.1 Strontium isotope analysis**

Strontium isotope ratios were determined by thermal ionisation mass spectrometry (TIMS) using a ThermoTriton automated multi-collector mass spectrometer. Using a method adapted from Birck (1986), samples were loaded onto single Rhenium (Re) filaments with TaF activator to enhance stability and sensitivity. The reproducibility of the international standard reference material (NBS987) was  $0.710255 \pm 0.000010$  (n=54, 2 sd), and all data was normalised to the NBS987 accepted value 0.710250.

## 6.8.2.2 Lead isotope analysis

Lead isotope ratios were determined by multi-collector inductively coupled plasma mass spectrometry (MC-ICP-MS) using a Nu Plasma HR with an average reproducibility (2 sd) of  ${}^{206}Pb/{}^{204}Pb = 0.009\%$ ;  ${}^{207}Pb/{}^{204}Pb = 0.008\%$ ;  ${}^{208}Pb/{}^{204}Pb = 0.003\%$ ;  ${}^{208}Pb/{}^{206}Pb = 0.005\%$ . Before analysis all samples were spiked with a thallium (Tl) solution and normalised to NBS981. When lead concentration yields were low, MC-ICP-MS using a Thermo Fisher Scientific

NEPTUNE *Plus* with an X-Skimmer cone was used with average reproducibility (2 sd) of  ${}^{206}\text{Pb}/{}^{204}\text{Pb} = 0.010\%$ ;  ${}^{207}\text{Pb}/{}^{204}\text{Pb} = 0.014\%$ ;  ${}^{208}\text{Pb}/{}^{204}\text{Pb} = 0.019\%$ ,  ${}^{207}\text{Pb}/{}^{206}\text{Pb} = 0.006\%$ ;  ${}^{208}\text{Pb}/{}^{206}\text{Pb} = 0.012\%$ .

## 6.8.2.3 Trace element analysis

Trace element analysis to determine lead concentration results was conducted using inductively coupled plasma mass spectrometry (ICP-MS) using an Agilent 7500cx ICP-MS fitted with a CETAC ASX-520 autosampler with an average reproducibility (2 sd) of 0.01%. The transfer of samples to the ICP-MS from the autosampler was controlled by a CETAC ASXpress + vacuum pump. Multi-element quality control check standards were analysed at the start and end of each run and after no more than every 20 samples. To overcome polyatomic interferences the ICP-MS collision cell was operated in He mode at a flow rate of 5.5 ml min<sup>-1</sup> for all analytes except Se, for which H<sub>2</sub> gas was used at 4.5ml min<sup>-1</sup>. Quantitative data analysis was carried out using MassHunter Workstation software (Agilent).

# **6.9 Data comparisons**

When comparing lead concentrations between non-adults with evidence of disease and those without it was imperative to be certain that those in the category 'without disease' showed no evidence of metabolic stress. Therefore, only non-adult individuals that could be identified with certainty as not having any skeletal manifestations of metabolic disease were included in the comparison of palaeopathological data and lead concentrations. Any non-adult individual with low preservation and/or completeness levels that inhibited the assessment of pathological alterations to the entire skeleton were excluded from this phase of the analysis. Although this reduced sample sizes it did ensure that individuals were not assigned to incorrect categories.

Open access databases such as the Oxford Archaeological Lead Isotope Database (OXALID) were used to gather comparative lead isotope datasets to determine whether the data from the human samples reflected local lead isotope characteristics. Isotope ratios from contemporaneous human samples were also included in comparisons where published data was available.

## 6.10 Statistical analyses

Due to the non-parametric nature of the data, box and whisker plots were produced to graphically display the range and skewness of the data. To better visualise the differences between certain groups some y-axis ranges were reduced to enlarge the plots. Where this resulted in the exclusion of some extreme outliers from the plots, the outlying data points were described in the figure captions. For the statistical analysis of two independent groups the Mann-Whitney U test was applied, and when comparing more than two independent groups the Kruskal-Wallace test was used. Both statistical tests were run using the IBM SPSS programme version 0.2 for Microsoft Windows and a p-value of 0.05 was set as the level of significance to best correct for type one and type two errors (Madrigal, 2012, p. 94).

# CHAPTER SEVEN

# Lead Concentrations and Health

## 7.1 Introduction

By their very nature human skeletal remains offer a direct link to people in the past, providing a rich source of information pertaining to the lives and living conditions of past populations (Scott, 2013). Paired osteological evidence of disease and tooth enamel lead concentration (ppm) analyses allows exploitation of this link, facilitating a better understanding of how lead exposure affected the health and mortality of children within the Roman Empire. This chapter will discuss the results of the lead concentration analysis and how it relates to Roman childhood health and mortality from different regions of the empire. The results are separated into two sections. The first section assesses inter-dental variations and population differences in lead concentrations, while the second section focuses on how lead exposure impacted upon childhood health and mortality. The results are then discussed together (section 7.6) to present a comprehensive review of how trace element analysis of polluted populations can inform our understanding of the lives of people in the past.

The study population includes 176 individuals (66 adults and 110 non-adults) from five, 1<sup>st</sup> to 4<sup>th</sup> century AD sites located in different regions of the Roman Empire. Skeletal material was assessed to determine age-at-death, sex (of adults only) and the presence of disease (see Table 7.1). A detailed summary of the results from the lead and osteological analyses of each individual included in this study is tabulated in Appendix A1.

| Site               | Location               | Total<br>Adults | Male | Female | Total<br>Non-<br>adults | Metabolic<br>disease |
|--------------------|------------------------|-----------------|------|--------|-------------------------|----------------------|
| Dealul<br>Furcilor | Alba Iulia,<br>Romania | 10              | 5    | 5      | 27                      | 7                    |
| Michelet           | Caen,<br>France        | 14              | 7    | 7      | 23                      | 9                    |
| Multiple           | Beirut,<br>Lebanon     | 17              | 6    | 10     | 23                      | 0                    |
| PERI 2             | Tarragona,<br>Spain    | 12              | 7    | 5      | 15                      | 7                    |
| Santa<br>Caterina  | Barcelona,<br>Spain    | 13              | 5    | 7      | 23                      | 10                   |
|                    | Total                  | 66              | 30   | 34     | 110                     | 33                   |

Table 7.1 –Summary of the number of individuals in the sample populations.

## 7.2 Inter-dental variations

Within archaeological contexts teeth tend to have the highest level of preservation of any surviving skeletal material, and are highly resistant to diagenetic alteration. Thus, making them prime sampling material for trace element analysis. Understanding how heavy metals such as lead are incorporated into and distributed between different tooth types is important not only for the interpretation of such data, but also in how data between studies can be compared (Rabinowitz et al., 1991). The majority of published research on dental lead concentrations has been conducted on modern populations as a means of assessing the usefulness of teeth as an indicator of environmental lead exposure (Amr et al., 2010; Arora et al., 2006; Bayo et al., 2001; Kamberi et al., 2011; Karahalil et al., 2007; Negrea et al., 2008; Olympio et al., 2010). Due to their natural exfoliation, and therefore easy collection, deciduous teeth are often used as an indicator of childhood lead exposure and consequently, body lead burdens (Barbosa et al., 2005; Fergusson et al., 1988; Malara et al., 2006; Needleman et al., 1972). Comparison of tooth enamel lead concentrations from the permanent (n = 92) and deciduous (n = 84) teeth analysed in this study showed that deciduous teeth had higher lead concentrations (median = 5.8 ppm) than permanent teeth (median = 2.4 ppm) (see Fig. 7.1). This is consistent with previous studies that found that deciduous teeth had statistically significantly higher lead concentrations than permanent teeth (Shapiro et al., 1972). It is thought that this difference in lead concentrations is due to age related differences in lead absorption rates from the gastrointestinal tract. Although enamel lead concentrations in both deciduous and permanent teeth represent childhood lead exposure, different teeth form at different ages. As absorption rates from ingested lead decrease with increasing age, the disparity between permanent and deciduous tooth enamel lead concentrations could represent higher lead absorption rates during the development of earlier forming teeth (Paterson et al., 1988; Purchase and Fergusson, 1986; Rabinowitz et al., 1991; Selypes et al., 1997). Diet could also be linked to this. Studies have demonstrated that diet and nutrition can have significant effects on the absorption rates of lead from the gastrointestinal tract (Barltrop and Khoo, 1975). For example, diets low in iron or high in vitamin D and fats tend to increase the amount of lead absorbed, while diets high in fibre decrease lead absorption (Baernstein and Grand, 1942; Barltrop and Khoo, 1975; Sobel et al., 1940). Additionally, liquid diets such as the predominantly milk-based diet fed to infants and young children, also increase the amount of lead absorbed through the gastrointestinal tract (Kello and Kostial, 1973). Although a diet high in mineral content has proven to reduce lead absorption, the high calcium content of milk does not appear to be able to completely counteract the increased absorption rates associated with a liquid diet (Barltrop and Khoo, 1975). This may be due to the high fat and protein content of milk, both of which have been shown to increase the uptake of dietary lead. As such, the different types of diets given to

children during different stages of childhood, and therefore while different teeth are mineralising, may also influence the amount of lead incorporated into deciduous and permanent teeth.





Permanent: median = 2.4 ppm (n = 92), Deciduous: median = 5.8 ppm (n = 84). An outlier in the deciduous group with a lead concentration of 187 ppm is not shown on the plot due to reduced y-axis scale.

A number of studies have demonstrated that factors such as tooth type, tooth location (mandibular or maxillary), and dental material analysed (dentine, enamel etc.), may significantly influence the lead concentrations obtained from teeth (Kamberi et al., 2012). If, as suggested above, higher lead concentrations in deciduous teeth were a result of higher absorption rates in young children, a negative correlation between lead concentration and age of tooth formation would be expected. However, this is not the case. Some studies have shown that deciduous incisors have higher lead concentrations than deciduous molars (Mackie et al., 1977; Pinchin et al., 1978), while others have

reported contrasting patterns in lead concentrations between deciduous incisors and deciduous canines (Paterson et al., 1988; Shapiro et al., 1975).

When lead concentrations between tooth types were compared in this study, no significant differences were observed between the permanent tooth types (see Fig. 7.2). Comparison of the deciduous teeth showed that canines had higher lead concentrations (median = 12.2 ppm) than the deciduous incisors (median = 5.8 ppm), first molars (median = 5.2 ppm) and second molars (median = 3.3 ppm) (see Fig. 7.3). Although the canines had more than double the lead concentrations seen in the other deciduous tooth types, only the second molars showed a significant difference (Kruskal-Wallis  $X^2$  = 3.841, p = 0.0256). Canines begin development approximately one month after incisors (Gustafson and Koch, 1974). Therefore, if lead levels were simply a function of age, incisors (the earliest forming tooth) would be expected to have the highest lead concentrations. An important consideration here is also the length of time it takes for each tooth type to reach complete mineralisation of the crown enamel. It is reasonable to suggest that the longer tooth enamel takes to mineralise the more opportunity there is for the accumulation of lead within the mineral matrices. If this were a limiting factor to lead acquisition in tooth enamel the teeth that take the longest to mineralise, such as the deciduous canines and second molars (approximately 13 months), would have consistently higher lead concentrations than deciduous incisors and first molars which take approximately 9 - 10 months to complete mineralisation (Gustafson and Koch, 1974). While the median lead concentration obtained for the deciduous incisors, canines and first molars in this study do fit with this hypothesis, the second molars do not. As such, these results support the presupposition that physiological factors such as age and enamel mineralisation time are unlikely to be the dominant mechanism behind lead accumulation in teeth. However, other physiological facors such as periods of rapid

growth (growth spurts) and changes to diet must be considered. It is well documented that children have growth spurts during the first year of life, again between the ages of six to eight years (mid-growth spurt), and then finally during adolescence between the ages of 13 to 18 years (pubertal growth spurt) (Tanner, 1988). If these growth spurts and changes in diet such as weaning, which tends to occur around the age of two to three years (Dupras, 2001) affected natural accumulation of lead in tooth enamel then increases in lead concentrations would be expected to be seen in teeth mineralising during these times. Again, this is not the case, and studies have shown that after the removal of the very outer surface of enamel to eliminate environmental contamination, lead concentrations do not significantly vary spatially within deciduous teeth, suggesting that tooth enamel matures homogenously before eruption (Tacail et al., 2017). Therefore, although the mechanisms behind the incorporation of lead in to tooth enamel is highly complex and poorly understood, changes in levels of exposure during the formation of different tooth types is the most likely cause for these disparate concentrations.



#### Figure 7.2 – Comparison of lead concentrations by permanent tooth type.

Incisor: median = 3.8 ppm (n = 14),  $2^{nd}$  Premolars: median = 2.2 ppm (n = 36),  $2^{nd}$  Molars: median = 2.8 ppm (n = 39) and  $3^{rd}$  Molars: median = 2.23 ppm (n = 7). An outlier in the incisor group with a lead concentration of 59.62 ppm is not shown on the plot due to reduced y-axis scale.





Incisors: median = 5.8 ppm (n = 49), Canines: median = 12.2 ppm (n = 10),  $1^{st}$  Molars: median = 5.2 ppm (n = 11) and  $2^{nd}$  Molars: median = 3.3 ppm (n = 14). An outlier in the incisor group with a lead concentration of 187 ppm is not shown on the plot due to reduced y-axis scale.

As with tooth type, comparisons of lead concentrations in mandibular and maxillary teeth have demonstrated great variability between studies. Some indicating maxillary teeth have higher lead concentrations than mandibular teeth (Pinchin et al., 1978), and others demonstrating the opposite (Smith et al., 1983). Although mandibular teeth begin developing slightly earlier (approximately 1 month) than maxillary teeth (Gustafson and Koch, 1974) a comparison of the two in this study showed that maxillary teeth had higher lead concentrations than mandibular teeth in both the permanent and deciduous dentition (see Fig. 7.4). However, these differences were not statistically significant (Kruskal-Wallis,  $X^2 = 3.841$ , p = 0.0903). When the median lead concentrations for the samples were compared by country, the same trend was seen across all four sites (see Fig. 7.5). However, the mandibular teeth from Lebanon and Romania showed greater range in lead concentrations than their maxillary counterparts. This consistency in elevated maxillary teeth lead concentrations across countries is surprising considering the divergent results seen between mandibular and maxillary teeth in other studies (Pinchin et al., 1978; Smith et al., 1983).



Figure 7.4 – Comparison of lead concentrations in maxillary and mandibular dentition. Deciduous maxillary: median = 6.8 ppm (n = 57), Deciduous mandibular: median = 5.2 ppm (n = 27), Permanent maxillary: median = 3.8 ppm (n = 53) and Permanent mandibular: median = 2.4 ppm (n = 39). An outlier in the deciduous maxillary group with a lead concentration of 187 ppm is not shown on the plot due to reduced y-axis scale.



Figure 7.5 – Comparison of lead concentrations in maxillary and mandibular dentition by country. Romania maxillary: median = 2.9 ppm (n = 22), Romania mandibular: median = 1.7 ppm (n = 15), France maxillary: median = 4.5 ppm (n = 26), France mandibular: median = 1.9 ppm (n = 11), Lebanon maxillary: median = 6.7 ppm (n = 19), Lebanon mandibular: median = 5.0 ppm (n = 21), Spain maxillary: median = 6.5 ppm (n = 44) and Spain mandibular: median = 3.9 ppm (n = 19). An outlier in the Spain maxillary group (187 ppm) and Spain mandibular group (59.63 ppm) are not shown on the plot due to reduced y-axis scale.

The results of this study, alongside previously published research (Pinchin et al., 1978; Smith et al., 1983) demonstrate the non-uniform distribution of lead within teeth. There is no consensus on which dental arcade (maxillary or mandibular) provides the highest concentrations, or whether there are any patterns to which type of tooth will yield the highest amount of lead. The significant discrepancies between studies suggest that these variations in lead concentrations are probably not due to physiological factors (blood supply, time of formation etc.), but rather a product of fluctuations in environmental exposure. It is doubtful that anyone experiences a continuous and consistent level of lead exposure for any prolonged period of time. In fact, modern studies have shown that children's levels of lead exposure have a tendency to fluctuate seasonally (Kemp et al., 2007; Laidlaw et al., 2005; Yiin et al., 2000; Zahran et al., 2013). As different tooth types develop at different times throughout an individual's childhood (Schour and Massler, 1941), it is likely that inter-dental variations in lead concentration are population specific and reflect an individual's level of lead exposure at the time of tooth formation.

Modern studies do not have the same contamination risks to consider as archaeological studies, and as a result often obtain data from the analysis of the whole tooth or dentine samples. Such variations in sampling techniques, as well as analytical methodology make comparison between studies problematic (Rabinowitz et al., 1991), especially if attempting to relate archaeological lead concentrations to those obtained from modern studies when assessing how the data reflects exposure. Irrespective of the type of sample used there are undoubtedly significant inconsistencies in inter-dental lead concentrations. Therefore, it would be optimal to compare lead concentrations from teeth of the same type and tooth position (Bercovitz and Laufer, 1990). However, this has the potential to be extremely limiting, especially in archaeological studies where

skeletal preservation and completeness dictates which teeth can be sampled. If standardisation of tooth type were to be implemented, sample sizes would be greatly reduced (Tvinnereim et al., 2000).

# 7.3 Geographical variations

From the limited number of studies that have published lead concentration data from Romano-British skeletal material, it is clear that there is great variability in the range of lead concentrations (Montgomery, 2002; Montgomery et al., 2010; Shaw et al., 2016). The results of this study were no different (see Fig 7.6), with statistically significant variations in median lead concentrations between all four countries (Kruskal-Wallis,  $X^2$  (3) = 7.815, p = 0.0002). The Romanian population had the lowest lead concentrations with values ranging from 0.03 ppm to 10.67 ppm (median = 2.4 ppm), while the Spanish individuals had the highest concentrations that ranged from 0.7 ppm to 187 ppm (median = 7.3 ppm).



Figure 7.6 – Comparison of lead concentrations by country.

Romania: median = 2.4 ppm (n = 37), France: median = 3.6 ppm (n = 37), Lebanon: median = 6.3 ppm (n = 40) and Spain: median = 7.3 ppm (n = 63). An outlier in the Spain group with a lead concentration of 187 ppm is not shown on the plot due to reduced y-axis scale.

Differences in average lead concentrations are not only seen between countries, but also between sites within the same country. Although geographically only approximately 96 kilometres apart, the Spanish sites at Tarragona and Barcelona exhibited statistically significant differences in median lead concentrations (see Fig 7.7). As geogenic environmental lead concentrations have been shown to vary geographically (Reimann et al., 2012), it stands to reason that human lead concentrations could also vary geographically. Added to this, is the variability in anthropogenic lead exposure between Roman populations both within and between countries. Therefore, it is likely that Roman human lead concentrations are population specific and reflect local levels of anthropogenic lead exposure. The geology of the local areas surrounding populations is also an important factor to consider when exploring what may influence the absorption of lead between different populations. Studies have shown that the mineral content of

drinking water can affect the absorption of lead, with hard water (high mineral content) acting as a natural buffer against the absorption of the heavy metal (Levander, 1979). Therefore if a population's drinking water was running off calcium carbonate rich terrains they might absorb lower concentrations of lead than a population whose drinking water originated from granitic terrains, for example. To this end it is interesting to note that the geologies surrounding Barcelona and Tarragona and the river that supply their respective drinking water differ quite substantially despite their close proximity to each other. The Río Francolí which supplies water to Tarragona traverses calcium carbonate rich terrains such as limestone, sandstone and marls, while Barcelona's water supply from the Río Besós flows through a terrain composed of granites and gravel (Llamas, 1969). Despite the likelihood that Tarragona's water had higher calcium content than Barcelona, the Tarragona population still accumulated higher levels of lead. Thus it may be that the significant difference seen between the two Spanish populations has a more anthropological than geological cause, such as mining pollution. Unlike Barcelona which was predominantly mined for the green minerals calaite and variscite for jewellery, Tarragona was a major source of lead rich galena and extensively mined throughout the Roman period (UNESCO, 2007). The increased pollution in the local area that would have been created during the processes involved in lead mining and metalworking may account for the variability seen between these two Spanish sites.



Figure 7.7 – Comparison of lead concentrations from the Tarragona and Barcelona individuals. Tarragona, Spain: median = 11.2 ppm (n = 27), Barcelona, Spain: median = 3.3 ppm (n = 35). An outlier in the Tarragona group (187 ppm) and Barcelona group (59.62 ppm) are not shown on the plot due to reduced y-axis scale.

## 7.4 Comparing males and females

Lead exposure can fluctuate depending on a number of variables, including occupation, status, availability of certain foodstuffs and lead containing products. Assessing the difference in lead concentrations between certain groups (males and females, rural and urban inhabitants etc.) can provide a wealth of information pertaining to a population's socioeconomic status. To further understand how exposure patterns may have varied within the Roman Empire, differences between male and female lead concentrations were assessed. Archaeological studies investigating childhood lead burdens are scarce and modern studies have reported conflicting results. Some studies stating that there is no significant difference between male and female lead concentrations (Baghurst et al.,

1992; Strömberg et al., 2008, 2003), while others show evidence for higher lead concentrations in male children (Claymaet et al., 1991; Costa de Almeida et al., 2010; Meyer et al., 1998; Paoliello et al., 2002; Roels et al., 1980; Trepka et al., 1997).

Analysis of the tooth enamel lead concentrations from the 64 sexed adult individuals in this study allowed exploration of any differences in Roman childhood lead concentrations between males and females. Collectively, males (median = 3.8 ppm) had higher lead concentrations than females (median = 2.4 ppm) (see Fig. 7.8). When compared by country, again males tended to have higher lead concentrations than females (see Fig. 7.9). The only exceptions to this were the Spanish samples, where females (median = 4.2 ppm) exhibited higher lead concentrations than males (median = 3.8 ppm). Although these differences were not statistically significant, they raise interesting questions about why Spanish individuals go against the trend seen in the majority of both archaeological and modern lead concentration studies.



**Figure 7.8** – **Comparison of male and female lead concentrations.** Male: median = 3.8 ppm (n = 30) and Female: median = 2.4 ppm (n = 34).



**Figure 7.9** – **Comparison of male and female lead concentrations by country.** Romania male: median = 2.9 ppm (n = 5), Romania female: median = 1.5 ppm (n = 5), France male: median = 1.9 ppm (n = 7), France female: median = 1.8 ppm (n = 7), Lebanon male: median = 4.7 ppm (n = 6), Lebanon female: median = 2.5 ppm (n = 10), Spain male: median = 3.8 ppm (n = 12) and Spain female: median = 4.2 ppm (n = 12).

Recent studies into sex differences in body lead burdens have mainly focused on differences in adults. These studies found that males tend to have higher lead concentrations than females (Barry, 1975; Brown and Margolis, 2012; Theppeang et al., 2008). It is thought that the increased haematocrit levels and higher exposure patterns seen in males are the main reasons for the difference between the sexes (Becker et al., 2002; Pirkle et al., 1998). However, other studies have proposed that sex dependent differences in metabolism and genetic regulation are responsible for higher lead concentrations in males (Björkman et al., 2000; Vahter et al., 2007). The majority of lead within the body is stored within the skeletal tissues (Gulson et al., 1997, 1995), and premenopausal females have been shown to release this stored lead at slower rates than males (Popovic et al., 2005; Roberts and Cox, 2003). This is in part due to oestrogen levels. The hormone reduces osteoclast activity, decreasing the rate of bone resorption

and therefore the subsequent release of lead stored within the skeleton (Goldberg et al., 2016; Kameda et al., 1997; Oursler et al., 1991; Parikka et al., 2001).

Hereditary factors are also thought to contribute to the elevated lead concentrations seen in males. Genetic studies have proposed that up to 40% of female lead burdens are influenced by genetic factors, while almost 95% of male lead burdens result from environmental exposure (Björkman et al., 2000). Hormonal regulation of the three polymorphic genes known to influence the toxicodynamics of lead (ALAD, Vitamin D Receptor and HFE protein) are thought to account for this genetic variation in sex specific absorption, retention and excretion of lead (Onalaja and Claudio, 2000; Vahter et al., 2007). Unfortunately, these explanations for the higher lead concentrations in males are not applicable to archaeological lead concentration data obtained from tooth enamel. The data obtained from tooth enamel represents the lead acquired during the time of tooth enamel mineralisation. In deciduous dentition, permanent 2<sup>nd</sup> molars and premolars, as used in this study, enamel mineralisation is complete before adolescence. During this prepubescent phase girls and boys have very similar hormone levels (Bidlingmaier et al., 1975, 1973). However, modern studies have shown that there tend to be small surges in sex hormones during the early development of infants. In boys testosterone levels have been shown to increase during four to six weeks gestation and again between one to six months after birth (Forest et al., 1976; Raivio et al., 2003; Alexander, 2014), and in girls oestrogen levels have been shown to increase during the first six months of life (Bidlingmaier et al., 1987; Kuiri-Hanninen et al., 2013). After these postnatal endocrine surges sex hormone levels fall and remains stable until puberty (Ostanikova et al., 2002). Although both boys and girls experience short periods of increased sex hormones early in life, multiple studies have demonstrated that there is no difference in prenatal or postnatal lead concentrations between males and

females (Vahter et al., 2007; Baghurst et al., 1992; Stromberg et al., 2003; Dietrich et al., 2001; Yabe et al., 2015; Taylor et al., 2017). Therefore, it is likely that hormonal regulation of gene expression or endocrine function would not be a major influential factor in early childhood lead acquisition.

It is therefore likely that the differences between Roman male and female lead concentrations seen in this study are due to differences in levels of exposure. During childhood, the predominant method of lead ingestion is through hand to mouth activity (Raymond, 2017; Sahmel et al., 2015; Sayre et al., 1974; Schnur and John, 2014; Watt et al., 1993). Boys may have been engaging in a wider range of activities that facilitated this type of exposure. The higher lead concentrations seen in the Spanish female populations also suggests that environmental exposure has a more significant effect on childhood lead burdens than genetic influences. If metabolic and genetic mechanism were influencing childhood lead concentrations, males would be expected to have the highest lead concentrations irrespective of the population they came from. However, at both the Tarragona and Barcelona sites, females had marginally higher lead concentrations than males (see Fig. 7.10).



Figure 7.10 – Comparison of the male and female lead concentrations from the Tarragona and Barcelona sites in Spain. Tarragona males: median 8.9 ppm (n = 7), Tarragona females: median = 10.2 ppm (n = 5), Barcelona males: median = 1.7 ppm (n = 5) and Barcelona females: median = 2.6 (n = 7).

Comparative archaeological studies that have explored sex differences in lead concentrations are few. Millard et al., (2014) found no significant difference in the lead concentrations of males and females from a 19th century London population. However, Japanese studies of 17th – 19th century AD samurai, merchant and farmer populations found significantly higher lead concentrations in the females from the samurai class, but no difference between the sexes in the merchant and farmer classes (Nakashima et al., 2007, 1998). Explanations for this centre on status and its accompanying exposure levels. The samurai class were considered a high-status population, while the merchants and farmers were considered low status. It is therefore posited that the higher status females would have had greater exposure to lead containing products (e.g. cosmetics) and foodstuffs (e.g. wine) than lower status females (Nakashima et al., 2007, 1998).
Therefore, a difference in status may also account for the high lead concentrations seen in the Spanish individuals analysed here.

The type and quantity of grave goods included in burials have been shown to vary by age, ethnicity and status and are often used to determine the status of an individual (Philpott, 1991). Data pertaining to grave goods and burial type were not available for the Tarragona individuals, therefore the effect that status may have had upon these lead concentrations could not be determined. However, at the Barcelona site, records show that two of the sampled females were buried in mausolea, while the remaining individuals were buried in tile-capped graves (tegula). These 'a cappuccina' burials are common in Roman Spain and thought to be representative of lower status individuals (Bruun and Edmondson, 2015). Burials in mausolea, however, are thought to represent high status individuals. Although any conclusions drawn from the comparison of these twelve Barcelona individuals is limited due to the small sample size, results show that the individuals buried in mausolea had markedly higher lead concentrations than the individuals buried in tegula graves (see Fig. 7.11). These results show that higher status in these Barcelona individuals is accompanied by elevated lead concentrations, suggesting that status may have influenced female lead exposure in Spain during the Roman period. Although this pilot study has generated promising results, it is limited by the small sample size and therefore the differences in lead concentration in relation to status cannot be generalised to encompass the entire Roman Empire.



Figure 7.11 – Comparison of the lead concentrations from Barcelona individuals buried in tegula (low status) and mausolea (high status) graves. Tegula graves: median = 2.0 ppm (n = 10) and Mausolea: median = 8.4 ppm (n = 2).

# 7.5 Health and mortality

### 7.5.1 Lead and mortality

Lead is a cumulative poison, and one to which children are particularly susceptible (Hursh and Suomela, 1968; Rabinowitz et al., 1976). The increased bioavailability of lead during the Roman period, due to high environmental lead pollution and the use of lead compounds rendered the empire's children at greater risk than ever before (Mackie et al., 1975; Montgomery et al., 2010). There is no doubt that childhood was a perilous time throughout the Roman period, with failure to thrive being an all too common occurrence, to which the high prevalence of infant and juvenile remains in Roman

skeletal populations stands testament (Carroll, 2014). Despite this, and the documentary evidence of lead poisoning being responsible for stillbirths, spontaneous abortion, and deformities in Roman infants (Gilfillan, 1965; Nriagu, 1983; Waldron et al., 1979; Woolley, 1984), little has been done to explore any link between childhood lead exposure and high infant mortality rates. With this shortage of evidence to support or indeed refute the degree to which lead impacted upon the health and mortality of Roman populations, attention must be directed to the skeletal remains from the period. To address this, lead concentrations in tooth enamel from deciduous teeth and permanent teeth were compared to explore any differences in childhood lead concentrations between individuals that died during childhood and those that survived into adulthood. However, when comparing deciduous and permanent tooth enamel samples it is important to consider total enamel thickness. As discussed above (section 4.5.3), enamel lead concentrations have been shown to be higher on the outer surface of the crown as well as next to the enamel dentine junction (Budd et al., 1998; Robbins et al., 2010). Therefore, core enamel samples are commonly used to avoid these areas of variability (Montgomery, 2002; Budd et al., 2004). Although deciduous tooth enamel is thin, making core enamel samples more difficult to obtain, recent studies have demonstrated that deciduous tooth enamel composition remains virtually unchanged throughout the tooth (Müller et al., 2019). Therefore, once surface enamel has been removed deciduous enamel samples should be comparable with core enamel samples from permanent teeth.

A comparison of the lead concentration data from the adult and non-adult individuals shows that those who survived into adulthood had lower childhood lead burdens (median = 2.6 ppm) than those that died during childhood (median = 7.2 ppm) (see Fig. 7.12). The results of a Kruskal-Wallis test showed that the median lead concentrations in these two groups were statistically significantly different ( $X^2 = 12.181$ , p = 0.0005). Children have more than double the lead concentrations observed in adults, suggesting that higher lead burdens are accompanied by lower life expectancies. Although unsurprising given the toxic nature of lead, these results suggest that lead poisoning was an issue for citizens of the Roman Empire, and offer the first bioarchaeological evidence for such a claim.



**Figure 7.12** – **Comparison of adult and non-adult lead concentrations.** Adults: median = 2.6 ppm (n = 66) and Non-adults: median = 7.2 ppm (n = 110).

It is evident from the archaeological record that there is a real failure to thrive in children throughout the Roman period. It is estimated that up to 50% of children died before the age of 10 years old, with 20–40% of these not reaching one year of age (Carroll, 2018, 2014). It is also widely accepted that children are more susceptible to lead poisoning than adults as their developing bodies are prone to absorbing higher

quantities of ingested lead (Alexander et al., 1974; Hursh and Suomela, 1968; Rabinowitz et al., 1976; Ziegler et al., 1978). To explore whether the high lead burdens characteristic of Roman individuals contributed to the high childhood mortality rates in the Roman Empire, lead concentrations were further compared to age-at-death (see Fig. 7.13). A negative correlation between lead concentration and age is clearly evident, again indicating that individuals with lower lead burdens lived longer than those with higher lead burdens.

This is particularly notable with regards to children under the age of one year. Known to be a prevalent demographic within Roman cemetery populations, explanations for their high mortality rates have ranged from malnutrition and disease to infanticide and exposure (Gowland et al., 2014; Mays, 1993; Pilkington, 2013). As infants under one year of age are the most at risk group in terms of the lethality of lead poisoning these results offer new insights into the previously overlooked role lead may have had in these high infant mortality rates. Unfortunately, little research has been done to understand how lead concentrations in tooth enamel reflect in *vivo* lead burdens, or how they correlate to manifestations of lead poisoning during life (Grobler et al., 2000; Rabinowitz et al., 1993). As such, identifying high lead concentrations in archaeological remains alone is unlikely to be sufficient to determine those who may have succumbed to lead poisoning. However, using modern clinical literature and the known biochemical pathogenesis of lead toxicity it may be possible to further elucidate the effect of lead poisoning on the health of archaeological populations.



Figure 7.13 – Comparison of lead concentrations by age-at-death. Foetal: median = 7.5 ppm (n = 10), 0-1 year: median = 7.4 ppm (n = 24), 2-6 years: median = 5.9 ppm (n = 35), 7-12 years: median = 3.3 ppm (n = 29), 13-18 years: median = 3.2 ppm (n = 12) and Adults: median = 2.6 ppm (n = 66).

### 7.5.2 Lead and disease

Lead is an insidious poison and the gradual accumulation of the metal in bodily tissues becomes increasingly toxic. Due to the systemic nature of lead poisoning, the clinical manifestations of toxicity are varied and complex. With the exception of growth plate lead lines visible on radiographs, no specific skeletal lesions have been associated with lead poisoning (Smith et al., 2015). This is most likely due to the toxicodynamics of absorbed lead culminating in clinical manifestations that are common to many other disease processes. However, with its propensity to disrupt metabolic pathways, it is unsurprising that both modern and historical clinical literature associate lead poisoning with a number of metabolic diseases, such as rickets, scurvy and anaemia (Caffey, 1938; Waldron, 1966). Therefore, it is probable that individuals who died suffering the ill effects of chronic lead poisoning would exhibit pathological skeletal alterations consistent with these metabolic diseases.

The presence of metabolic disease in the non-adult population was diagnosed according to published criteria (see Chapter 6), and examples of the pathological alterations consistent with metabolic disease observed in the sample population are presented in Figure 7.14. When compared with lead concentration data, the non-adult individuals exhibiting pathological alterations consistent with metabolic disease had significantly higher lead concentrations (median = 8.1 ppm) than those without (median = 4.9 ppm) (see Fig. 7.15). When compared using a Kruskal-Wallis test the difference in median lead concentrations between these two groups were shown to be statistically significant ( $X^2 = 4.007$ , p = 0.0453). This supports the presupposition that skeletal markers of metabolic disease will manifest in conjunction with high lead concentrations, and that lead poisoning can be tentatively identified in archaeological human remains through the combination of palaeopathological and trace element analyses.



Figure 7.14 – Examples of the pathological lesions observed in some of the non-adult individuals



Figure 7.15 – Comparison of lead concentrations from non-adults with skeletal evidence of metabolic disease (excluding individuals with cribra orbitalia) and non-adults without pathological alterations. Non-adults with lesions: median = 8.1 ppm (n = 25) and Non-adults without lesions: median = 4.9 ppm (n = 35)

To investigate any patterns regarding the types of metabolic diseases likely to be concurrent with high lead burdens, lead concentrations were further compared by type of disease (see Fig. 7.16). Both rickets (median = 10.7 ppm) and scurvy (median = 8.1 ppm) are accompanied by higher median lead concentrations than individuals with no evidence of disease (median = 4.9 ppm). However, when compared using a Kruskal-Wallis test only the rachitic group have statistically significantly different lead concentrations to individuals without disease ( $X^2 = 3.841$ , p = 0.0209). These results indicate that the elevated levels of environmental lead pollution characteristic of the Roman period did have a negative impact upon childhood health. Lending support to the hypothesis that anthropogenically induced increases in lead exposure throughout the

Roman Empire was deleterious to health, especially in those exhibiting high lead burdens coupled with palaeopathological evidence of metabolic disease. As such, it is likely that lead exposure was a contributing factor to the ill health and failure to thrive seen in many non-adult Roman skeletal populations.



Figure 7.16 – Comparison of lead concentrations from individuals with metabolic disease. Rickets: median = 10.7 ppm (n = 6), Scurvy: median = 8.1 ppm (n = 11), Rickets and scurvy: median = 5.7 ppm (n = 8), Cribra orbitalia: median = 3.0 ppm (n = 19) and No disease: median = 4.9 ppm (n = 35)

### 7.5.3 Cribra orbitalia

Cribra orbitalia is a descriptive term used to describe abnormal pitting and porosity on the external surface of the orbital roofs. Presence of this rather distinctive pathological lesion has been observed in skeletal remains from multiple time periods across the world, and is typically used as a marker of general health and nutritional status (Steyn et al., 2002, 2016; Walker et al., 2009). Since the early 1960s bioarchaeologists have theorised that the pathological alteration was a result of hypertrophy of the red bone marrow, and therefore provided unequivocal proof that an individual suffered from anaemia (Carlson et al., 1974; Stuart-Macadam, 1987). Active cribra orbitalia is only seen in non-adult individuals due to a shift in the body's centres for haemopoiesis away from the cranial vault and long bone medullary cavities to vertebral bodies and sterno-costal regions after adolescence. As a result, adult individuals usually only ever exhibit an inactive form of the lesion in various stages of healing (Lewis, 2007; Stuart-Macadam, 1992; Walker, 1986).

The sensitivity of the haem system to extremely low lead concentrations (see Chapter 4) makes lead-induced anaemia a common symptom in those afflicted by lead poisoning. It is the omnipresence of anaemia in the clinical literature associated with lead poisoning that makes the skeletal manifestations of the disorder a popular target in bioarchaeological studies exploring lead exposure in past populations. Indeed, for decades studies have tried to identify lead poisoning in archaeological human remains by attempting to correlate lead concentrations with the presence of cribra orbitalia (Facchini et al., 2004; Gleń-Haduch et al., 1997; Millard et al., 2014; Zarifa et al., 2016). Despite the commonality of anaemia in those suffering from lead poisoning, the majority of bioarchaeological studies have found no correlation between the presence of cribra orbitalia and high lead concentrations. The results of this study were no different. In fact, individuals with cribra orbitalia exhibited lower median lead concentrations (median = 3.0 ppm) than individuals without the lesion (median = 4.9 ppm) (see Fig. 7.17). Statistical comparison using the Mann-Whitney U test confirmed that there was no significant difference in lead concentrations between individuals with cribra orbitalia and those without (U = 241, p = 0.09894). This is likely due to the aetiology of the

lesion itself. Therefore, it can be concluded that cribra orbitalia should not be included as an indicator of lead toxicity in bioarchaeological studies attempting to identify individuals that suffered from lead poisoning. Furthermore, these results highlight the importance of understanding not only the biochemical pathogenesis of lead poisoning, but also the often-complex aetiologies of the pathological skeletal alterations used to assess the impact of lead exposure on the health of past populations.



Figure 7.17 – Comparison of lead concentrations from non-adults with cribra orbitalia and non-adults without pathological alterations. Cribra orbitalia: median = 3.0 ppm (n = 19) and No lesions: median = 4.9 ppm (n = 35).

### 7.5.4 Poison or paucity?

Rickets and osteomalacia is caused by a deficiency in vitamin D (Brickley et al., 2018; Mays and Brickley, 2018; Pilz et al., 2019). Vitamin D is important for the gastrointestinal absorption of calcium and phosphorous, both of which are integral to the development of healthy bone mineral (Ortner, 2003; Resnick, 2002). A deficiency in this vitamin disrupts the metabolic homeostasis of these minerals, resulting poorly mineralised bones. The skeletal manifestations of which include diffuse pitting, flaring of metaphyses and softened bones prone to pathological bowing if weight bearing (Mays et al., 2006; Ortner and Mays, 1998; Resnick, 2002). Although vitamin D can be acquired through diet (mainly oily fish and some animal fats), the majority of the prohormone is synthesised during exposure of the skin to sunlight (Pearce and Cheetham, 2010). More often than not, poor nutrition or socio-cultural practices such as swaddling or restricted outdoor activities are proposed as explanations for the presence of rickets in non-adult individuals from past populations throughout Europe (Brickley et al., 2014; Giuffra et al., 2015). While the cause of rickets may well be limited to these sociocultural and nutritional aetiologies, it appears that environmental pollution may have been overlooked. It seems unlikely that a lack of UVB exposure or access to sufficiently nutritious foods could be the sole cause of high prevalence rates throughout the Roman Empire, especially in Mediterranean regions where sunlight and vitamin D-rich foodstuffs such as fish were plentiful (Marzano, 2013). Despite this, environmental factors such as pollution are rarely considered, and when they are it is usually only in reference to the smog that frequently plagued 19th-century urban populations (Hardy, 2003; O'Riordan and Bijvoet, 2014; Wallach, 2014). However, it is notable that the historical literature and bioarchaeological evidence from the Roman period and 19th century demonstrate high prevalence rates for rickets, and are also the two periods that created significant peaks in environmental lead pollution (Patterson, 1965; Roberts and Cox, 2003).

Roberts and Cox's (2003) diachronic study of disease prevalence in Britain demonstrates how the developments in industry, agriculture, trade and cultural practices that came with the Roman occupation of Britain also brought about the first instances of scurvy, rickets and osteomalacia. Like most archaeological studies of metabolic disease the authors go on to describe how these maladies were most likely a result of increased urbanisation and reduced crop quality (Roberts and Cox, 2003). However, they do briefly mention evidence for lead pollution but do not elaborate on any consequence this might have had upon health (Roberts and Cox, 2003, p. 389). That work goes on to demonstrate a decrease in the prevalence of metabolic diseases in the early medieval period, as societies moved away from the socio-economic practices characteristic of Roman populations and back towards a simpler, rural way of life (Higham, 2004; Roberts and Cox, 2003). During the 19th century, which brought about a second surge in industrial development and widespread urbanisation as the industrial revolution took a hold of Britain, prevalence rates of metabolic diseases, especially in children, significantly increase from those seen in previous periods (Newman and Gowland, 2017; Ortner, 2003, p. 393; Pettifor, 2003, p. 543; Pinhasi and Mays, 2008, p. 220). The parallels that can be drawn between the prevalence rates of rickets and levels of environmental lead pollution during both the Roman period and the 19th century are striking. Bringing about the question, could anthropogenic lead pollution have been a contributing factor to these disease prevalence rates? Whilst it must be acknowledged that disease prevalence is often multi-factorial and correlation does not mean causation, lead poisoning should be included in the differentials when the aetiology of rickets is being considered during these periods.

The clinical literature is filled with case studies detailing individuals that presented with rickets induced by lead poisoning (Caffey, 1938; Chisolm et al., 1955; Holt, 1923; Hunter, 1977), and so it is unsurprising to find that rachitic children had the highest median lead concentrations (median = 10.7 ppm) of any subgroup in this study. However, what was unexpected were the low lead concentrations observed in the adult individuals exhibiting skeletal evidence of healed rickets (median = 1.2 ppm). This group of individuals had the lowest lead concentrations observed in this study; with median concentrations half that of those observed in adults with no evidence of disease and almost six times lower than the median lead concentrations of those with active rickets (see Fig. 7.18). Comparing these medians using the Kruskal-Wallis test showed that there was a statistically significant difference in lead concentrations of those with rickets and those with healed rickets, as well as individuals showing no skeletal evidence of metabolic disease ( $X^2$  (2) = 5.991, p = 0.0052).



Figure 7.18 – Comparison of lead concentrations from rachitic individuals and individuals with no pathological alterations. Rickets: median = 10.7 ppm (n = 6), Healed rickets: median = 1.2 ppm (n = 5) and No lesions: median = 4.9 ppm (n = 35).

While there is no doubt that living conditions, status and the quality of foodstuffs play a significant role in the prevalence of these diseases; the results of this study clearly show a correlation between increased lead burdens and the presence of rickets in Roman period children. This indicates that the exploitation of lead and its products during this period contributed to the high prevalence rates of the disease throughout the Empire. These results, alongside the analogous fluctuations in rickets prevalence rates and levels of environmental lead pollution through time strengthen the argument for the inclusion of lead exposure when considering the cause of rickets in populations with known high levels of exposure. It is interesting that adult individuals with skeletal alterations consistent with healed rickets have the lowest childhood lead concentrations of any subgroup included in this study. As vitamin D promotes the absorption of lead in the gastrointestinal tract, these individuals may represent those with a nutritional deficiency, which acted as a buffer against the acquisition of lead. Conversely, the rachitic individuals with high lead concentrations may have exceeded the blood lead threshold at which the enzymatic conversion of vitamin D into is active form is disrupted, thereby initiating a lead-induced deficiency in the vitamin. If this could be verified through further research it would provide the potential to differentiate between rickets induced by lead poisoning and rickets caused by nutritional paucity. This would allow archaeologists to more precisely assess how peoples' interactions with their environment impacted upon their health, as well as gain a more accurate representation of populations' nutritional statuses. Of course, there will inevitably be overlaps in rachitic children with low lead concentrations or adults with high childhood lead concentrations as the toxicodynamics of lead poisoning and how it relates to lead concentrations in archaeological remains is complex and poorly understood. So while these results in no way suggest that all rachitic children in Roman cemetery populations are exhibiting

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lead-induced manifestations of the disease and all those with healed rickets were protected against lead poisoning, it does indicate that lead exposure is likely contributing to the high number of individuals with the disease during this period.

# 7.6 Summary

It is clear from the results discussed above that the pairing of osteological and lead concentration analyses provide a wealth of information pertaining to the socioeconomic status and general wellbeing of archaeological populations. This study offers the first international comparisons of childhood lead concentrations from Roman period individuals. The results show that lead concentrations vary both between and within countries. The wide ranges in concentration values exhibited in these populations' highlights the unpredictability of lead burdens even in environments shown to be highly polluted with the toxic metal. The factors influencing lead exposure levels and absorption rates are clearly multifarious and complex, lead burdens are not just simply linked to how polluted an individual's environment is. This fact is highlighted by the results of this study, which demonstrates how variable lead burdens were within an empire notorious for high levels of lead pollution. The only consistency across all sites was that individuals who died during childhood had higher lead concentrations than individuals that survived into adulthood.

With regards to health and mortality, these results provide the first bioarchaeological evidence that lead poisoning may have been deleterious to childhood health. There is strong evidence to suggest that anthropogenic lead pollution contributed to the high prevalence rates of metabolic diseases, especially rickets, seen during the Roman period. Analysis of lead concentrations in rachitic individuals also revealed that nuances in the aetiology of the disease (caused by nutritional paucity or pollution) could be elucidated.

Comparisons with age-at-death also implicate elevated lead concentrations in the high infant mortality rates seen in Roman skeletal populations. The introduction of a bioarchaeological perspective to the decades-old debate surrounding how lead affected health during the Roman period offers new insights into the impact of environmental lead pollution on the fragility of childhood health throughout the empire.

The proposed increased availability of lead-containing products (e.g. wine, cosmetics, and medicines) to wealthier, higher status individuals has led to the suggestion that status is an influential factor in the bioaccumulation of lead. The results of this study support this presupposition, with higher lead concentrations seen in Spanish individuals interred in mausolea as opposed to tegula graves. Making inferences about certain aspects of identity, such as status, are often difficult in archaeological contexts. The addition of an objective method such as lead concentration analysis to the current means of assessing status, which are often biased by our expectations (wealthier individuals are likely to have more elaborate burials than those of a lower socioeconomic standing), can only serve to enhance our interpretive capabilities.

# CHAPTER EIGHT

# Lead Isotope Ratios and Migration

## 8.1 Introduction

A main focus of this study is the use of lead isotope ratios as an additional discriminatory tool to further discern places of origin in archaeological migration studies. When using any isotope system as a tracer in human mobility studies it is imperative that there is a high level of confidence that the measured isotopes represent entirely *in vivo* acquisition. Unlike bone, tooth enamel has shown to be extremely diagenetically stable due to its dense structure and low porosity, leaving limited opportunities for mineral infiltration and ion exchange between the enamel and its burial environment. Therefore, to ensure the integrity of the isotopic data collected only tooth enamel samples are analysed in this study. As isotopes are incorporated into tooth enamel as the tissue mineralises, this sampling strategy has restricted assessment to childhood movements within the Roman Empire.

Until recently lead isotopes have received little attention in bioarchaeological studies (Harris et al., 2017; Jones et al., 2017; Lamb et al., 2014; Millard et al., 2014; Montgomery, 2002; Montgomery et al., 2005, 2010, 2014, Price et al., 2017a, 2017b, 2017c; Shaw et al., 2016). The majority of these studies were not conducted on Roman populations and, with the exception of Montgomery et al., (2010), none of the studies included more than one skeletal population from the same time period. The analysis of lead isotope ratios from six Roman populations from different regions of the Roman Empire in this current study provides a means of assessing how well lead isotope ratios

can discriminate between contemporaneous individuals from different countries. This approach is, to date, unique in bioarchaeological studies.

This chapter presents and discusses the results of the lead isotope analysis, and has been separated into two sections. The first section assesses the differences in human lead isotope ratios between countries, working towards developing an understanding of what constitutes a local isotope range in different regions of the Roman Empire. The second section focuses on combining lead isotope data with other isotope systems and contextual information to determine if there is enough intercontinental variation in lead isotope ratios to facilitate the identification of migrants within the Roman Empire. The results are then discussed together (section 8.7) to present a comprehensive review of how lead isotope analysis of polluted populations can inform our understanding of migration in a population known for its movement of peoples. A detailed summary of the results from the isotope and osteological analyses of each individual included in this study is tabulated in Appendix A2.

## 8.2 Cultural focusing

The cultural focusing of lead isotope ratios in humans is a phenomenon associated with anthropogenic lead pollution (see Chapter 2). Bioarchaeological studies have demonstrated how prehistoric populations and those with limited technological advancements have low lead burdens accompanied by divergent isotope ratios (Budd et al., 2004; Montgomery, 2002; Montgomery et al., 2010). However, later populations with metallurgical technologies tend to have higher lead burdens accompanied by homogenous isotope ratios. The Romans are one such population, with studies showing that individuals from the same site tend to exhibit high lead concentrations and similar lead isotope compositions (Montgomery et al., 2010; Shaw et al., 2016).

Figures 8.1a and 8.1b present <sup>208</sup>Pb/<sup>204</sup>Pb verses <sup>206</sup>Pb/<sup>204</sup>Pb isotope ratios grouped by lead concentration for each country included in this study. These plots allow visualisation of how patterns in thorogenic and uranogenic lead isotope ratios in human tooth enamel alter with increasing lead concentrations. The ovals highlighting trends in the data on all plots have no statistical significance and are merely to guide the reader. As expected for populations in anthropogenically-polluted regions, the majority of data points from all five countries produce linear arrays characteristic of lead ore field isotope ranges. Within these constrained fields (delineated by red ovals on each plot), lead concentration groups cluster with increasing closeness as lead concentrations increase. With the exception of Spain, all data points that plot outside the ovals are individuals with lead concentrations under 1 ppm. The individual in the Barcelona dataset (UF748) that plots below the rest of the Spain assemblage had a lead concentration of 2.37 ppm, indicating anthropogenic exposure. As individual UF748's <sup>207</sup>Pb/<sup>204</sup>Pb and <sup>208</sup>Pb/<sup>204</sup>Pb isotope ratios are lower than those seen in the rest of the Spanish assemblage it is likely that this individual was exposed to a different lead ore source during their childhood (discussed further in section 8.6).

It is clear that with increasing lead concentration there is a reduction in isotope variability. As seen in previous studies, low lead concentrations, particularly those under 1 ppm are thought to represent geogenic exposure and have been shown to exhibit a higher degree of variability in their isotopic ranges than lead acquired through anthropogenic exposure (Budd et al., 2004; Montgomery, 2002; Montgomery et al., 2010). This is best visualised in Figure 8.2, which shows <sup>207</sup>Pb/<sup>206</sup>Pb against lead concentration (ppm). The lead isotope ratios that are accompanied by low lead concentrations spread out across the bottom of the plot. Those plotting below the geogenic threshold of 1 ppm (dashed line) (Montgomery et al., 2010), show the highest

variability, and likely represent individuals who were not exposed to significant amounts of anthropogenic lead during their childhood. As lead concentrations increase the <sup>207</sup>Pb/<sup>206</sup>Pb isotope ratios shift to the left of the plot and cluster with increasing closeness. The coloured lines in Figure 8.2 have no statistical significance but are included to enable the reader to easily visualise this trend, which appear to show the transition from geogenic exposure to anthropogenic exposure within a population.

Although all individuals in this study are from 1<sup>st</sup> to 6<sup>th</sup> century AD populations within the Roman Empire, each location contains at least one individual with lead concentrations below 1 ppm. This indicates that simply living within the Roman Empire did not guarantee high levels of exposure despite the vast quantities of lead being mined, utilised and traded within its borders. Previously, lead concentrations above 1 ppm have been used to demonstrate that an individual is from a metallurgical population, with those exhibiting concentrations less than 1 ppm thought to be predominantly prehistoric (Budd et al., 2004; Montgomery et al., 2010). The majority of the individuals in this study come from multi-period cemeteries; therefore, radiocarbon dating would be useful here because the low-concentration individuals may actually be pre-Roman in date, or may reflect a temporal change in lead use throughout the Roman period. It is also possible that these low lead concentrations indicate a childhood spent in a rural location where exposure to pollution was minimal. More work on rural cemeteries is needed to establish an expected range of lead concentrations from low exposure regions of the Roman Empire. Without these clarifications the fact that 19.8% (19 of 96 samples) of the individuals in this study have lead concentrations below 1 ppm suggests that lead concentration cannot be used in isolation to separate Roman from pre-Roman populations.



Figure 8.1a – Bivariate plots showing <sup>208</sup>Pb/<sup>204</sup>Pb against <sup>206</sup>Pb/<sup>204</sup>Pb. Individuals are grouped according to the lead concentrations present in their tooth enamel. Red ovals highlight the linear groupings of individuals.





Figure 8.1b – Bivariate plots showing <sup>208</sup>Pb/<sup>204</sup>Pb against <sup>206</sup>Pb/<sup>204</sup>Pb. Individuals are grouped according to the lead concentrations present in their tooth enamel. Red ovals highlight the linear groupings of individuals.



Figure 8.2 – Bivariate plot showing <sup>207</sup>Pb/<sup>206</sup>Pb against lead concentrations (ppm) in human tooth enamel. The dashed horizontal line indicates maximum level of lead (1 ppm) that can be acquired in unpolluted individuals and the grey box delineates the British anthropogenic range identified by Montgomery et al. (2010). Analytical error is within the symbols

## 8.3 Establishing local ranges

To facilitate the identification of migrants (outliers) within a population it is necessary to establish what constitutes the local isotopic range. Usually, environmental samples (plants, soils, water etc.) are used to define local ranges for isotope systems such as strontium and oxygen (Slovak and Paytan, 2012). These can either be site specific, taken from samples in and around the excavated area, or regional/country specific topographic maps (isoscapes) created by systematic sampling of large geographic areas. However, anthropogenic pollution makes this method difficult when creating lead isoscapes intended for archaeological comparisons. Lead isoscapes must not only be spatially specific but also temporally specific. Neither the modern isoscapes of lead in European agricultural soils by Reimann et al., (2012) nor the 19<sup>th</sup> century European isoscape by Keller et al., (2016) would reflect the human lead isotope ratios measured in Roman individuals. A viable alternative to this would be to use published lead isotope datasets from contemporary human tooth enamel of geographically constrained origins. This would provide the most accurate comparative dataset, as it would most closely reflect the combination of bioavailable sources that a population was exposed to. However, there is a notable lack of comparable human lead isotope data from regions of the Empire outside of Britain. It is therefore common practice to use published datasets from lead ore and lead artefacts of known provenance (coins etc.) to define the local ranges expected for any given country (Millard et al., 2014; Montgomery et al., 2010; Shaw et al., 2016).

The human lead isotope data generated in this study has been plotted alongside lead ore isotope ratios from OXALID, an open access lead ore database, and, where available, data from Roman coins (Butcher and Ponting, 2014). These datasets give broad ranges of mineralisation (ore deposit) values for a region and have been used as proxies for the expected local anthropogenic lead isotope ranges in each country. The current study uses lead isotope ratio data for lead ores defined within specific modern national borders. While it is acknowledged that these borders differ from those of Roman provinces, and that a comprehensive review of contiguous national datasets may identity a fuller isotope baseline, the approach adopted here is in line with previous bioarchaeological studies exploring mobility using lead isotope ratios, and has proved effective in establishing the local anthropogenic lead isotope ratio ranges for the region (Montgomery 2002; Montgomery et al., 2010; Millard et al., 2014; Shaw et al., 2016).

The graphical representation of lead data is best plotted in a way that highlights the spread and separation of the data points. Therefore, all data is presented in the conventional bivariate plots using the <sup>204</sup>Pb isotope ratios (<sup>206</sup>Pb/<sup>204</sup>Pb, <sup>207</sup>Pb/<sup>204</sup>Pb and <sup>208</sup>Pb/<sup>204</sup>Pb), with the inclusion of the Stacey and Kramer (1975) growth curve for reference. To avoid the higher uncertainties surrounding <sup>204</sup>Pb measurements, <sup>208</sup>Pb/<sup>206</sup>Pb against <sup>207</sup>Pb/<sup>208</sup>Pb plots have often been used in archaeological literature. These plots benefit from higher precision, but compress the data fields. Therefore, the subtle variations that frequently differentiate one lead field from another may be lost. As these plots are commonly used in archaeological literature they have also been included here. Any outliers within the datasets have been described but will be further discussed in section 8.5.

### 8.3.1 Romania

A comparison of the Romania individuals with data from Romanian lead ore (Baron et al., 2011; Marcoux et al., 2002) shows how the human data forms a linear array that spreads to the left of the ore field (Fig. 8.3). When the data is presented using a

<sup>207</sup>Pb/<sup>206</sup>Pb vs. <sup>208</sup>Pb/<sup>206</sup>Pb bivariate plot the Romania individuals appear to split into two groups (Group A and Group B), and the shift away from the Romanian lead ore field becomes clearer (Fig 8.4). This plot inverts the lead isotope ratios; therefore the Romania individuals now plot above and to the right of the Romanian ore field.

Group A plots closely with the upper end of the Romania lead ore field indicating that the individuals in this cluster provide a good baseline for expected human lead isotope ratios in individuals with childhood origins in Romania. The individuals in Group B cluster loosely together, away from the Romania ore field. As all of Group A plot at the very edge of the Romanian ore field and Group B remains in-line with the linear array produced by Group A and the Romania ore field, the difference in Group B's isotope ratios may be due to exposure to an additional, older Romanian lead source more depleted in <sup>206</sup>Pb/<sup>204</sup>Pb than the comparative ore data plotted here. As it is currently unclear whether the four individuals in Group B represent migrants to Romania or Romanians exposed to an additional ore source, their isotope ratios should be used with caution when defining the local lead isotope range expected in ancient Romanian individuals. There is one individual that plots away from the rest of the Romanian assemblage; individual M160b sits in the upper right of the plot (Fig. 8.4), away from Group A and B. The lead isotope ratios exhibited by M160b are sufficiently different from the rest of the Romania individuals and Romanian lead ore as to suggest that they spent their childhood in a different geographic location that has older lead sources, for example, somewhere with Precambrian orogeny.



Figure 8.3 – Plots of <sup>208</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (a) and <sup>207</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (b) showing the relationship between Romania tooth enamel samples and the Romanian lead ore field. Ore data taken from Marcoux et al., (2002), and Baron et al., (2011). Analytical error shown to 2 sd.



Figure 8.4 –Plot of <sup>208</sup>Pb/<sup>206</sup>Pb versus <sup>206</sup>Pb/<sup>206</sup>Pb showing the relationship between Romania tooth enamel samples and the Romanian lead ore field. Ore data taken from Marcoux et al., (2002), and Baron et al., (2011). Analytical error is within the symbols 193

### 8.3.2 France

A comparison of the individuals from the France assemblage with data from French lead ore (OXALID, 2018) and Roman coins from Gaul (Butcher and Ponting, 2014) demonstrates how the individuals group within the isotope field created by the French lead ore and coin data (Fig. 8.5). It is interesting that the lead isotope ratios from the France individuals plot more closely with the Roman coin data than the French lead ore data, both of which cluster in the centre of the French lead ore field. As outlined in Chapter 2, human anthropogenic lead isotope ratios are culturally focussed, clustering together in a narrow range reflecting the lead ore sources used by the population (Montgomery, 2002; Montgomery et al., 2010). To some extent metals used in artefacts (in this case coins), are also culturally focussed due to the reworking of ores and metals (Montgomery et al., 2010; Shaw et al., 2016), this is particularly true of Roman coins, which were recycled every few years (Harl, 1996). Therefore, data from artefacts of known provenances are likely to provide more realistic representations of the expected human isotope composition in anthropogenically-polluted regions.

As with the Romania individuals, the France individuals also split into two groups. Group A all have lead concentrations above 1 ppm and cluster tightly together in the centre of the French lead ore and Gaul coin data. However, three individuals have lead concentrations below 1 ppm (Group B). These individuals plot close together on the upper edge of the French ore field (see Fig. 8.6). Their low lead burdens suggest natural, geogenic exposure rather than anthropogenic exposure and this probably accounts for lower <sup>206</sup>Pb/<sup>204</sup>Pb isotope ratios than those observed in Group A. The Michelet cemetery is also a multi-phase cemetery; therefore it is possible that the difference in isotope ratios between Group A and B represent a temporal shift in the lead source used by the

population. The congruency of Group A's tooth enamel isotope ratios with the French lead ore field suggests that these human samples provide a good baseline for the expected lead isotope ratios in Roman individuals with childhood origins in Northern France. The remaining two individuals that plot away from the majority of the population (S394 and S854) also have lead concentrations below 1 ppm and plot outside the French lead ore field but within the spread of isotope ratios provided by the coins from Gaul. This suggests that individuals S394 and S854 were exposed to low levels of anthropogenic lead pollution during their childhoods, and that this was likely from an ore source in the Gaul region of the Roman Empire.



Figure 8.5 –Plots of <sup>208</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (a) and <sup>207</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (b) showing the relationship between France tooth enamel samples and the France lead ore field. Ore data taken from Butcher and Ponting, (2014) and OXALID. Analytical error shown to 2 sd.



Figure 8.6 –Plot of <sup>208</sup>Pb/<sup>206</sup>Pb versus <sup>207</sup>Pb/<sup>206</sup>Pb showing the relationship between France tooth enamel samples and the France lead ore field. Ore data taken from Butcher and Ponting, (2014) and OXALID. Analytical error is within the symbols

#### 8.3.3 Lebanon

There is currently no lead ore data available from Lebanon; therefore, comparative data from Israeli and Syrian lead ores have been used (OXALID, 2018). As these two countries border Lebanon to the north, south and east they are likely to have similar anthropogenic lead isotope characteristics. When the human samples from Lebanon were compared to these datasets, the majority of individuals from Lebanon plot within the isotope field created by the Israeli lead ore (Fig. 8.7). However, on the uranium-derived plot, all of the Lebanese individuals plot above the Syrian lead ore (Fig. 8.7b), indicating that the Lebanese individuals were exposed to lead sources that had higher <sup>207</sup>Pb/<sup>204</sup>Pb isotope ratios relative to Syrian lead ore. This suggests that lead ore from Israel can be used as a proxy for the local lead isotope range for Lebanon but Syrian lead ore cannot.

The majority of the Lebanon individuals plot close together within the Israeli lead ore field. However, two individuals (SK431 and SK1004) plot below and to the left of the main cluster (Fig. 8.7). How different these two individuals are compared to the rest of the Lebanon assemblage is best visualised using the <sup>207</sup>Pb/<sup>206</sup>Pb vs. <sup>208</sup>Pb/<sup>206</sup>Pb bivariate plot (Fig. 8.8). It is clear in Figure 8.8 that SK431 and SK1004 are sufficiently different from the Lebanon assemblage and the Israeli lead ore as to suggest that they originate from a different population. With the exception of these two outliers the lead isotope ratios from the Lebanon individuals appears to provide a good baseline for the expected lead isotope characteristics of Roman individuals with childhood origins in Lebanon or Israel.



Figure 8.7 – Plots of <sup>208</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (a) and <sup>207</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (b) showing the relationship between Lebanon tooth enamel samples and the Israeli and Syrian lead ore fields. Ore data taken from OXALID. Analytical error shown to 2 sd.


Figure 8.8 –Plot of <sup>208</sup>Pb/<sup>206</sup>Pb versus <sup>207</sup>Pb/<sup>206</sup>Pb showing the relationship between Lebanon tooth enamel samples and the Israeli and Syrian lead ore fields. Ore data taken from OXALID. Analytical error is within

## 8.3.4 Spain

A comparison of the Tarragona and Barcelona individuals with data from Spanish lead ore (OXALID, 2018) and Roman coins (Butcher and Ponting, 2014) of known provenance shows how the Spain individuals group within the isotope field created by the Spanish lead ore and coin data (Fig. 8.9). The Tarragona individuals show tighter clustering than the Barcelona individuals, which might be due to the significantly higher lead concentrations exhibited by the assemblage (see Chapter 7). As seen with the individuals from France, the Spain individuals also plot more closely with the Roman coin data than the Spanish lead ore data, again indicating that Roman artefacts of known provenance provide the most accurate source of proxy data for Roman population origin studies.

When the Spain data is presented using a <sup>207</sup>Pb/<sup>206</sup>Pb vs. <sup>208</sup>Pb/<sup>206</sup>Pb bivariate plot it becomes apparent that there are five outliers within the assemblage (Fig 8.10). Individuals UF217 and UF748 from the Barcelona site plot below and to the left of the main Spain cluster, while Barcelona individual T3 and Tarragona individuals UF2 and UF14 plot above and to the right of the main group. The close correlation of the Spain individuals with the Spanish coin data, both of which plot in the centre of the Spanish lead ore field, suggests that the range of lead isotope ratios exhibited by the Barcelona and Tarragona individuals provides a good baseline for the expected lead isotope characteristics of Roman individuals with childhood origins in Spain.



Figure 8.9 –Plots of <sup>208</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (a) and <sup>207</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (b) showing the relationship between Spain tooth enamel samples and the Spain lead ore field. Ore data taken from Butcher and Ponting (2014) and OXALID. Analytical error shown to 2 sd.



Figure 8.10 –Plot of <sup>208</sup>Pb/<sup>206</sup>Pb versus <sup>207</sup>Pb/<sup>206</sup>Pb showing the relationship between Spain tooth enamel samples and the Spanish lead ore field. Ore data taken from Butcher and Ponting, (2014) and OXALID. Analytical error is within the symbols

### 8.3.5 Slovenia

The Slovenia individuals do not have lead isotope ratios consistent with Slovenian lead ore (Henjes-Kunst et al., 2017). Instead the Slovenia individuals have higher <sup>206</sup>Pb/<sup>204</sup>Pb and <sup>208</sup>Pb/<sup>204</sup>Pb isotope ratios relative to Slovenian ore and therefore plot above and to the right of the Slovenian ore field (see Fig. 8.11 and 8.12). Although small in number these Slovenia individuals are an interesting assemblage. From the five individuals analysed, three have very low lead concentrations ranging from 0.22 ppm to 0.40 ppm, indicating predominantly geogenic exposure. It is possible that living in rural locations could account for the low lead concentrations due to the reduced exposure to pollutants. However, if this did account for their low lead burdens it would suggest that these three individuals were not local to the area as Emona (now Ljubljana) was a prosperous Roman city with strong links to Italy (Zupanek and Mlekuz, 2001) and therefore lead burdens reflecting urban living would be expected.

With regards to the two remaining individuals labelled in Figure 8.12, JM03 had a high lead concentration of 509 ppm. This exceedingly high concentration is unlikely to represent *in vivo* acquisition as it would equate to a blood lead level of 509  $\mu$ g/dL (Grobler et al., 2000), and as death occurs with blood lead levels of  $\geq$ 150  $\mu$ g/dL this value is incompatible with life (Bellinger and Bellinger, 2006). It was noted during sample processing that the enamel from JM03 was soft and slightly discoloured, therefore, individual JM03's lead composition is most likely contaminated from the burial environment and must be excluded from further analysis. The final Slovenia individual (JM04) had an anthropogenic lead burden of 6.43 ppm and plots between contaminated individual JM03 and the other three individuals (see Fig. 8.12). The lead isotope ratios observed in this individual indicate that they were exposed to a younger

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lead source than the comparative Slovenian lead ore data plotted in figures 8.11 and 8.12. However, as JM04 plots closely with JM03, who has diagenetic lead isotope ratios that reflect the local lead in the soils, it is also possible that JM04 is local.



Figure 8.11 –Plots of <sup>208</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (a) and <sup>207</sup>Pb/<sup>204</sup>Pb versus <sup>206</sup>Pb/<sup>204</sup>Pb (b) showing the relationship between Slovenia tooth enamel samples and the Slovenian lead ore field. Ore data taken from Henjes-Kunst, (2017). Analytical error shown to 2 sd.



Figure 8.12 –Plot of <sup>208</sup>Pb/<sup>206</sup>Pb versus <sup>207</sup>Pb/<sup>206</sup>Pb showing the relationship between Slovenia tooth enamel samples and the Slovenian lead ore field. Ore data taken from Henjes-Kunst, (2017). Analytical error is within the symbols

# 8.4 Geographic variation

This study has already demonstrated that Roman tooth enamel contains in vivo lead isotope ratios consistent with the lead ore field of the country in which the individuals were recovered (see section 8.3). This overlap between the isotopic composition of a country's lead ore and the corresponding human data suggests that these enamel isotope ratios provide accurate baselines for the lead isotope ratios expected in Roman populations from these regions. However, it is the variations in lead isotope ratios between countries that will determine how useful they are in identifying the origins of Roman migrants. To assess the usefulness of lead isotopes in this regard, the relationship between tooth enamel compositions from each country is illustrated in Figure 8.13a and 8.13b. In both plots the data spread creates a linear array, with the individuals from countries with younger geology (e.g. Lebanon and Romania) plotting to the top right of the plot and those from countries with older geology plotting further down towards the left (e.g. France). Although there is some degree of overlap between the countries, there are also subtle separations between populations. The plot displaying only uranium-derived lead (Fig. 8.13b) provides the clearest visualisation of these differences.



Figure 8.13 – Plots of  ${}^{208}$ Pb/ ${}^{204}$ Pb versus  ${}^{206}$ Pb/ ${}^{204}$ Pb (a) and  ${}^{207}$ Pb/ ${}^{204}$ Pb versus  ${}^{206}$ Pb/ ${}^{204}$ Pb (b) showing the relationship between all tooth enamel samples. Analytical error shown to 2 sd.

Statistical methods to identify outliers have not been applied to the lead isotope ratio datasets generated in this study. Sayre et al., (1992) state that any data plot that shows significant separation between data points or groups of data points provides sufficient evidence that there is a difference between them. This is supported by other authors who propose that if lead isotope ratio data point separations are graphically obvious there is no need to resort to statistical methods (Baxter, 1999, p.123; Scaife et al., 1996, p.306). Furthermore, Pernicka (1993, p.259) goes on to suggest that applying any statistical methods would likely lead to an overinterpretation of the data. As such, statistical methods to identify outliers within bioarchaeological lead isotope ratio datasets are not used. Instead individuals that fall outside known country sources (e.g. ore data, artefacts of known provenance), or plot visually far away from the majority of the group have been identified as outliers (Montgomery 2002; Montgomery et al., 2010; Shaw et al., 2016). The same method has been applied in this study, and once the visually different outliers (see section 8.3) from each population have been removed it becomes clear that the data separates into three distinct groups (see Fig. 8.14). The lead isotope ratios from the Slovenia, Romania and Lebanon individuals make up the first group, plotting closely together in the top right of the plot (red oval, Fig. 8.14). These individuals exhibit higher <sup>206</sup>Pb/<sup>204</sup>Pb and <sup>207</sup>Pb/<sup>204</sup>Pb isotope ratios than the Spain and France individuals. The Tarragona and Barcelona individuals exhibit lower <sup>206</sup>Pb/<sup>204</sup>Pb isotope ratios than the Slovenia, Romania and Lebanon individuals, plotting closely together to the left of group one (blue oval, Fig. 8.14). The third group contains the France individuals (orange oval, Fig. 8.14); these individuals have similar <sup>206</sup>Pb/<sup>204</sup>Pb isotope ratios to the Spain individuals but lower <sup>207</sup>Pb/<sup>204</sup>Pb isotope ratios than the other populations. The four France and three Romania individuals that do not conform to this

trend are the Group B, low lead burden individuals described above in section 8.3 and are not expected to plot closely with lead ore isotope ratios.

By grouping the data in this way it becomes apparent that there are two major trends in the dataset, and these trends also extend to previously published data from Roman populations (see Fig. 8.15). Two lines have been added to Figure 8.15 to visually help the reader see these trends. These lines have been placed where the majority of the data from Atlantic European and Mediterranean regions (red line) or Eastern and Western regions seperate (blue line). Firstly, the data produces two parallel linear arrays, separated by the red line (y = 1.6296x + 0.7053) on the plot. All of the data points below this red line are individuals buried in Atlantic European regions such as Britain, Northern France and Germany, while all of the data points above the red line are individuals from central European and circum-Mediterranean regions. This trend appears to relate to major European orogenic events (see Fig. 8.16), during which lead ores are often formed (Blichert-Toft et al., 2016; Evans et al., 2018). The human data from European regions formed during the Hercynian (c. 280 - 380 Ma) and Caledonian (c. 390 – 490 Ma) events plot below the red line, while the data from regions formed during the Alpine event (c. 60 - 2.5 Ma) plot above the red line. This demonstrates that Roman tooth enamel lead isotope composition relates to the geological age of the ore bodies being exploited in their cultural sphere, and can be useful in determining the provenance of outliers within a skeletal population.

The blue line (y = -7.25x + 8.216) in Figure 8.15 highlights the second trend in the data. Here the data separates into two groups, with the individuals from eastern European countries (Lebanon, Romania and Slovenia) plotting to the left of the blue line and the western European countries (Britain, France, Spain and Italy) plotting to the right. Both of these trends are also reflected in the lead ore data from the corresponding countries. The tendency of lead ore fields to spread over a wide range of values means that there is often overlap between lead ore fields from different countries. Due to this overlap, which is evident in both tooth enamel and lead ore data, it is clear that lead isotope ratios are not country specific. Nevertheless, this data does demonstrate that lead isotope ratios can be useful in distinguishing between broad regions of Europe, such as Eastern vs. Western Europe or Atlantic vs. circum-Mediterranean Europe. However, it is important to note that neither of the lines annotating Figure 8.15 represents an absolute separation between regions, but rather provide an indication of an individual's likely origin. These four suggested 'lead provinces' illustrated in Figure 8.15 provide a preliminary classification system for Roman lead isotope data and will need to be further tested and refined as more data becomes available in the future.



Figure 8.14 – Red oval (Group 1) comprising the central and eastern-European countries (Slovenia, Romania, Lebanon). Blue oval (Group 2) containing the circum-Mediterranean countries (Spain). Orange oval (Group 3) containing the Atlantic countries (France). Analytical error shown to 2 sd.



Figure 8.15 – Plot of <sup>208</sup>Pb/<sup>206</sup>Pb verses <sup>207</sup>Pb/<sup>206</sup>Pb showing how the tooth enamel samples and lead ore data group according to orogenic age and region of Europe (east vs. west). Analytical error is within the symbols



8.16 – Simplified map showing the major orogenic events of Europe. Purple represents Hercynian regions, green represents Alpine regions and yellow represents Precambrian and Caledonian regions. (Adapted from Muchez *et al.,* (2005).

# 8.5 Comparing lead and strontium

Strontium isotope analysis is a well-established method for investigating mobility and has been extensively used to identify non-locals in archaeological studies (Budd et al., 2001; Chenery et al., 2010; Evans et al., 2006; Montgomery et al., 2011, 2010; Valentine et al., 2015). However, similar terrains are found throughout Europe, which often renders strontium isotope ratios insufficiently unique enough to differentiate between countries (Evans et al., 2012). In Roman populations, however, lead exposure is dominated by anthropogenic not geogenic sources, therefore lead isotope ratios in

tooth enamel may show different patterns of variability across Europe than strontium isotope ratios. A difference is likely to be seen as strontium isotope ratios have been proven to be a useful discriminant in short-distance migration studies, especially in regions with heterogeneous geology, while lead isotope ratios are much more adept at identifying outliers in long-distance migration studies. Although no strontium isotope ratio data for the Slovenia individuals was obtained, strontium isotope ratios from the remaining four sites has been analysed to assess its variability across the Roman Empire and compare its effectiveness at identifying migrants with that of lead isotope ratios.

To explore the variability of lead isotope ratios between countries and in comparison to strontium isotope ratios, box and whisker plots have been used to illustrate the differences between sites. These plots provide an easy way to visualise the variation in ranges and central tendencies of the lead and strontium isotope measurements taken from each site. As can be seen in Figure 8.17, each country exhibits a smaller range in strontium isotope ratios than lead isotope ratios. The central tendencies of strontium (Fig. 17a) also exhibit less variability between countries (range = 0.00114) than the lead isotope ratios. The smaller variability in strontium isotope ratios between countries is likely to be due to the similar geology found across large expanses of Europe, and influenced by the bias in the dataset as a large proportion of the individuals analysed were from regions with carbonate geology (limestone, chalk etc.). Of the lead isotope ratios measured, the central tendencies of  $^{208}Pb/^{204}Pb$  (Fig. 17b) show the most variability between countries with a range of 0.3703, while  $^{207}Pb/^{204}Pb$  (Fig. 17c) show the least variability (range = 0.04102). This pattern of variability suggests that lead isotopes may provide a better means of differentiating between countries than strontium.



Figure 8.17 – Box and whisker plots showing the variances and central tendencies of the strontium and lead isotope ratios from each site.

Analysis of the strontium isotope ratios revealed that the Romania, France and Barcelona individuals have  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios that cover a broad range (see Fig. 8.18). The relatively wide spread of strontium isotope ratios in all three of these sites reflects the complex geology in these regions. With the exception of Barcelona, which also has areas of Cretaceous limestone, each of these regions have predominantly Palaeozoic (Cambrian and Devonian) and pre-Cambrian sediments with granitic intrusions (see Figs. 8.19 - 21), these types of terrains typically produce high  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios between 0.71101 – 0.78000 (Voerkelius et al., 2010). Conversely, the Tarragona and Lebanon individuals exhibit a narrower range of lower  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios (see Figs. 8.18) consistent with the Quaternary and Mesozoic limestone terrains that dominate these regions (see Figs. 21 and 8.22). This split in the types of terrain at each site is clearly evident in Figure 8.18 with the inclusion of the seawater value (dashed black line), here there is a clear separation of the individuals from regions with a predominantly limestone geology and those from predominately silicate regions.

The majority of individuals have strontium concentrations (ppm) below 160 ppm (median value = 111 ppm), however, 16 individuals have higher concentrations of up to 261 ppm (see Fig. 8.18). A notable feature of those with strontium concentrations above 160 ppm is that they are all from coastal cities. The high strontium concentrations found in seawater may have contributed to these high enamel concentrations as the close proximity of Tarragona, Barcelona and Beirut to the coast will have resulted in the regions being subject to sea-spray, marine-derived rainwater and even the use of marine-derived fertilisers (kelp, seaweed etc.) to grow crops, all of which would increase the concentration of bioavailable strontium (Montgomery et al., 2003).



Figure 8.18 – Bivariate plot showing the <sup>87</sup>Sr/<sup>86</sup>Sr ratios against Sr concentration (ppm) from the tooth enamel. Analytical error is within the symbols.



Figure 8.19 – Simplified geology map of Romania (Derry, 1980)

# IMAGE REMOVED BY AUTHOR FOR COPYRIGHT REASONS

Figure 8.20 – Simplified geology map of France (Derry, 1980)

IMAGE REMOVED BY AUTHOR FOR COPYRIGHT REASONS

Figure 8.21 – Simplified geology map of Spain (Derry, 1980)

# IMAGE REMOVED BY AUTHOR FOR COPYRIGHT REASONS

Figure 8.22 – Simplified geology map of Lebanon (Derry, 1980)

To explore mobility within this sample group the data from each site has been plotted separately, on conventional  ${}^{87}$ Sr/ ${}^{86}$ Sr vs. Sr ppm plots (see Figs. 8.23 – 8.26). In the absence of regionally specific bioavailable strontium ranges for each site, the estimated local ranges have been defined by ±2 sd from the mean of the combined enamel  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios (Bentley et al., 2004; Price et al., 2002). The majority of individuals plot within the estimated local ranges for their respective assemblage, however there are three outliers, one from each of the Barcelona, France and Lebanon populations.

Both the Barcelona outlier (individual T8, see Fig. 8.26) and the France outlier (individual S745, see Fig. 8.24) have higher  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratios (0.71157 and

0.71294 respectively) than the rest of their respective populations. Although T8's strontium isotope ratio remains within the bioavailable range suggested by Voekelius et al., (2010) for north-eastern Spain, T8 is sufficiently different from the rest of the assemblage as to suggest that they had childhood origins in a different region with upper Palaeozoic sediments and Mesozoic metamorphic rocks. Areas such as this could include Andorra or the border between France and Spain. The high strontium isotope ratio exhibited by the France outlier is within the bioavailable range for France identified by Goude et al., (2012), and is indicative of older Palaeozoic terrains and can be produced in very few locations in France (Voerkelius et al., 2010), most of which are located along the borders with Spain and Italy in the Pyrenees and Alpine mountain ranges. The outlier in the Lebanon assemblage (individual SK1004, see Fig. 8.25) has a lower strontium isotope ratio (0.70764) than the rest of the group. Although this strontium isotope ratio is still consistent with the bioavailable range for Israel identified by Hartman and Richards, (2016), as well as the Mesozoic limestone of the region, this geology is also common across large expanses of Europe. Therefore, although SK1004 has a strontium isotope ratio consistent with the geology found in Beirut it is sufficiently different from the rest of the Lebanon population to suggest that they originate from a different population.

It is notable to note that the strontium isotope ratios from this dataset identified fewer outliers than the lead isotope ratios, and that two of the three strontium outliers were not identified as outliers within the lead isotope dataset (see Table 8.1). This suggests that applying the combination of lead and strontium analyses can improve our ability to assess mobility within Roman populations.

| Country          | Pb outliers | Sr outliers |
|------------------|-------------|-------------|
| Romania          | M160b       | -           |
| France           | S394        | S745        |
|                  | S854        |             |
| Lebanon          | SK431       |             |
|                  | SK1004      | SK1004      |
| Barcelona, Spain | T3          | T8          |
|                  | UF217       |             |
|                  | UF748       |             |
| Tarragona, Spain | UF3         | -           |
|                  | UF14        |             |

Table 8.1 – Summary of outliers identified in the lead (Pb) and strontium (Sr) datasets.



Figure 8.23 – Bivariate plot showing the <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios against Sr concentration (ppm) from the tooth enamel of the Romania individuals. The shaded area represents a comparative bioavailable strontium isotope range from Romania and Serbia (Borić and Price, 2012). Analytical error is within the symbols.



Figure 8.24 – Bivariate plot showing the <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios against Sr concentration (ppm) from the tooth enamel of the France individuals. The shaded area represents a comparative bioavailable strontium isotope range from France (Goude et al., 2012). Analytical error is within the symbols.



Figure 8.25 – Bivariate plot showing the <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios against Sr concentration (ppm) from the tooth enamel of the Lebanon individuals. The shaded area represents a comparative bioavailable strontium isotope range from Israel (Hartman and Richards, 2014). Analytical error is within the symbols.



Figure 8.26 – Bivariate plot showing the <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios against Sr concentration (ppm) from the tooth enamel of the Spain individuals. The shaded area represents a comparative bioavailable strontium isotope range from Spain (Valenzuela-Lamas et al., 2016). Analytical error is within the symbols.

# 8.6 Identifying migrants across a continent

Of the 96 individuals in this study, 11 exhibit strontium or lead isotope ratios that suggest that they did not originate from the same geographical region as the other individuals in their respective populations. These 11 individuals are discussed below.

## 8.6.1 Romania

There was only one outlier within the Romania population, a middle adult male (M160b) with a low lead burden of 0.23 ppm. This low lead concentration suggests

exposure to geogenic sources of lead and therefore should not be expected to conform to the isotopic signature of the Romanian ore field. Geogenic lead isotope ratios do not relate to anthropogenic lead isotope ratios as they derive from the local underlying geology, whereas anthropogenic lead isotope ratios reflect the geological age and composition of the lead ore body that has been exploited (Faure, 1986). Therefore, interpreting these geogenic lead isotope ratios in archaeological populations is difficult due to widespread environmental lead pollution (Evans et al., 2015). The extensive lead mining and metalworking throughout history has caused contamination of soils, swamping any local geogenic lead isotope ratios and replacing them with lead isotope ratios consistent with exploited lead ore bodies (Montgomery 2002; Montgomery et al., 2010; Gulson, 1986). Although some studies have shown that burial soil leaches can provide a guide to the expected geogenic lead isotope ratios for the local area (Budd et al., 2000; Montgomery 2002; Montgomery et al., 2010), soil samples from each of the sites within this study were not available, therefore soil leaches could not be carried out to assess the local geogenic lead isotope composition for the local region.

As can be seen in Figure 8.28, M160b has lead isotope ratios much lower than the rest of the Romania population, and plots away from almost all of the comparative human lead isotope data from Roman populations across the Empire. The only other individual to plot close to M160b is France outlier S394, who also has a lead burden less than 1 ppm. There is currently no method for comparing the isotopic composition of archaeological human remains with a country's geogenic lead signature due to anthropogenic environmental pollution not only during the Roman period, but also the 19<sup>th</sup> century industrial revolution and again in the 20<sup>th</sup> century with the introduction of leaded petrol (Brännvall et al., 1997; Lee and Tallis, 1973; Renberg et al., 2002, 2001). As this individual has a strontium isotope ratio consistent with the rest of the Romania

population (see Fig. 8.27), it is difficult to ascertain whether the lead composition of this individual represents a migrant to Romania or the expected isotope ratios for Romanian 'locals' with predominantly natural lead exposure.

## 8.6.2 France

There are three isotopic outliers within the France population. Two of these individuals (S394 and S854) stood out due to their low lead isotope ratios, which were not obviously consistent with the rest of the France population and the French ore field. Individual S394 was an old adult female with a lead burden of 0.78 ppm and individual S854 was a middle adult probable female with a lead burden of 0.70 ppm. As with the Romania outlier, these low lead concentrations suggest exposure to predominantly geogenic lead sources and are unlikely to conform to the isotopic signature of the French ore field. Therefore, it is difficult to ascertain whether these individuals are migrants to France or French individuals from rural areas of France with minimal exposure to anthropogenic lead sources. The strontium isotope ratio for individual S394 could not be obtained, however S854's strontium isotope ratio (0.70935) is consistent with the majority of the France population and the expected values for the region. Although the lead isotope ratios of these two individuals appeared to suggest migrants to France, their low lead burdens indicate minimal exposure to anthropogenic lead sources and therefore may in fact be locals from a rural location where exposure to lead would be limited

The third outlier, individual S745 was an old adult probable male with an elevated strontium isotope ratio relative to the rest of the France population. The high strontium isotope ratio of 0.71294 constrains possible origins to older Cambrian and Precambrian terrains found in predominantly mountainous regions of Europe. Lead analysis

demonstrated that S745 had an anthropogenic lead burden of 1.78 ppm and plotted with Group A of the France population, within the centre of the French ore field. Although S745 has lead isotope ratios consistent with French ore, when compared to contemporaneous individuals from other regions of the Roman Empire, S745 also plots with Romano-British individuals at the upper end of the Mendips ore field (see Fig. 8.28). There is overlap between the upper end of the British ore field and the lower end of the French ore field, making it difficult to determine a region of origin solely on S745's lead isotope ratios. However, the isotopic composition of this individual does suggest childhood origins in areas of Western Europe with older Hercynian or Cambrian ores, such as Britain, France or northwest Spain. Adding S745's strontium isotope ratio to this interpretation narrows down possible childhood origins to the Pyrenees and Alpine regions in France or areas of Palaeozoic sediments in north and western Britain (Derry, 1980).

### 8.6.3 Lebanon

Individual SK1004 was a young adult male, dated to the early 1<sup>st</sup> century AD (pers. comms. V. Kalendrian). This individual has a low lead burden of 0.50 ppm, which suggests geogenic lead exposure rather than anthropogenic exposure. Lebanon did not become part of the Roman Empire until Pompey the Great's conquest in BC 64 (Doyle, 2012, p. 12). It is plausible that the early date of SK1004 could account for his low lead burden as Lebanon was still a relatively new region of the Empire and may not yet have developed the high environmental lead pollution seen in other countries under Roman rule. Previous research examining an Iron Age population from Lebanon has demonstrated that pre-Roman mining and metallurgy did not result in people from this region acquiring high anthropogenic lead burdens (Beherec et al., 2015). Therefore, the

early data could explain the low lead concentrations seen in SK1004, which as seen in British populations, are more consistent with pre-Roman populations exposed to predominantly natural lead as well as the lower lead isotope ratios exhibited by this individual in comparison to the rest of the Lebanon population.

The second outlier within the Lebanon population is SK431, an old adult female dating from the 2<sup>nd</sup> – 3<sup>rd</sup> century AD (pers. comms. V. Kalendrian). SK431 has a higher lead burden of 2.62 ppm, which is indicative of anthropogenic exposure. This individual's lead isotope ratios are lower than the rest of the Lebanon population and plot outside the Israeli lead ore field used as a proxy for the expected Lebanese lead isotope range. When compared with contemporaneous human isotope ratios from other regions of the Roman Empire, SK431 plots closer to the lead isotope ratios obtained from Mediterranean individuals from Spain and Italy (see Fig. 8.28). Although SK431's strontium isotope ratio (0.70839) is consistent with the Lebanon population, it is also similar to the strontium isotope ratios obtained from regions such as Spain, Greece and Italy.

#### **8.6.4 Spain**

Lead isotope analysis of the Spain population highlights five possible migrants (Barcelona individuals UF217, UF748, T3 and Tarragona individuals UF2, UF14). These five individuals have anthropogenic lead burdens ranging between 2.37 ppm and 20.3 ppm, and plot away from the main group of Spanish individuals (see Fig. 8.28). Barcelona individual UF748, a middle adult of indeterminate sex, plots below the other Spain individuals, with lead isotope ratios more consistent with regions of older Hercynian, Cambrian or Precambrian geology rather than the Alpine orogeny found in eastern Spain. Comparisons with human data from other regions of the Roman Empire

show how individual UF748 plots with contemporaneous humans from England and France. The second possible migrant, Barcelona individual UF217, was a middle adult female interred within a mausoleum. Like the majority of the Spain individuals, UF217 had lead isotope ratios consistent with contemporaneous individuals from regions of the Empire with Alpine orogeny. However, UF217 had higher lead isotope ratios than the Spain assemblage and therefore plots to the left of the Spanish field, and with the individuals from Eastern Europe (see Fig. 8.28). Although UF217 has a strontium isotope ratio (0.70806) consistent with the Spain assemblage it is also characteristic of large areas of northern Europe. The isotopic composition of this individual suggests that they are a migrant to Spain and likely spent their childhood in Eastern Europe.

The three other possible migrants, Tarragona UF2, UF14 and Barcelona T3, all exhibit low lead isotope ratios that plot close together, above and to the right of the Spain assemblage, close to France outlier S854 (see Fig 8.28). As these individuals plot a considerable way from the rest of the Spain assemblage they appear to be migrants to Spain. However, their lead compositions are consistent with Alpine orogeny, which is found in eastern Spain, and have lead isotope ratios that plot within the upper end of the Spanish ore field. The strontium isotope ratios for these three individuals are also consistent with other Spanish individuals and Spain's geology. The dates through which the Barcelona and Tarragona necropolises were in use ( $3^{rd} - 6^{th}$  centuries AD) affords the possibility that these individuals may highlight a temporal shift in the predominant source of lead used by these Spanish populations. Therefore, it is possible that these three individuals although different to the majority of the Spain population may not be migrants to Spain from another region of the Roman Empire but rather migrants to the Catalonia region of Spain from another area within the country. Radiocarbon dating these three individuals could potentially provide additional information about how Roman lead use changed within a country and inform our understanding of how temporally specific local baselines of lead isotopes may be. Finally, individual T8, a middle adult female from the Barcelona assemblage had lead isotope ratios consistent with the anthropogenic Spanish range; however she has a higher strontium isotope ratio than the rest of the Spanish assemblage (see Fig 8.27). Although this individual still plots within the suggested bioavailable strontium range for Barcelona by Voerkelius et al., (2010), T8's isotope ratio is sufficiently different from the rest of the Spanish population as to suggest that they come from a different population. Taking into account this individual's lead isotope ratios, it is likely that individual T8 had childhood origins in another region of Spain with Middle Upper Palaeozoic sediments and Mesozoic metamorphic rocks. This could include mountainous regions such as northern Spain near the border with France or possibly Andorra.



Figure 8.27 – Bivariate plot showing the <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios against Sr concentration (ppm) from all individuals. Analytical error is within the symbols.



Figure 8.28 – Bivariate pot showing <sup>208</sup>Pb/<sup>206</sup>Pb verses <sup>207</sup>Pb/<sup>206</sup>Pb isotope ratios of all individuals from this study alongside comparative lead ore data and contemporary individuals from previously published studies

# 8.7 Isotopic investigations of intrusive burial rites

Variations in burial practices have informed archaeological interpretations of past societies for centuries, and have been used to reconstruct almost every aspect of life from ethnicity and status to religion and gender (Ekengren, 2013). While any direct relationship between burial practices and geographic origins have long been disputed (Pearce, 2000), the unusual nature of such Roman burials raises questions about their identities and whether they share any commonalities with each other. This section presents the results of the multi-isotopic analysis of six Roman skeletons recovered in Musselburgh, Scotland, and two lead coffin burials excavated in Ilchester and York, England. It aims to ascertain the geographical origins of these individuals, which have been afforded unusual burial rites within an Empire known for the widespread movements of people, and to test the four suggested 'lead provinces' identified in this study (see Fig 8.15).

## 8.7.1 Lead coffin burials

Social status has often been interpreted using archaeological evidence such as funerary and epigraphic data (Buzon and Judd, 2008), and it has been shown that burial type can often be a good indicator of social status (Hope, 2009). It is thought that cheaper burials, those that simply place the body in the ground or use plain wooden coffins, represent people with limited economic resources and are therefore, by extension, low status, while more elaborate burials using coffins made of lead or stone represent higher status individuals with the means to pay for more elaborate burials (Hope, 2009; Redfern and DeWitte, 2011; Toynbee, 1996). Lead coffin burials from Romano-British contexts have predominantly been found in urban locations, which follows the distribution of wealth within the province at the time and therefore lends support to the assumption that individuals afforded a lead coffin burial are of higher status (Toller, 1977). However, the rarity of lead coffin burials from Romano-British contexts raises questions about the identity of the individuals afforded such an elaborate and presumably expensive burial rite.

Here the isotopic compositions of four Roman coffin burials from different locations in England (see Fig. 8.29) have been compared. Two of the individuals, a 4<sup>th</sup> century female from Ilchester and a 4<sup>th</sup> century male from York were analysed during this study, while previously published (Montgomery 2002; Montgomery et al. 2010) strontium and lead isotope ratio data from a 4<sup>th</sup> century female from Spitalfields, London, and a 4<sup>th</sup> century male from Eagle Hotel, Winchester, have been included for comparison. In the case of the York individual, oxygen isotope data was also available (provided by N. Wilson, pers. comm.) and has been included to help constrain possible regions of origin. Although published work using the same techniques employed in this study have demonstrated that disparate lead isotope ratios can be obtained from a lead coffin and the individual interred within it (Montgomery, 2002; Montgomery et al., 2010), contamination from the burial environment is always a concern. Therefore, lead samples from the Ilchester and York lead coffins were also analysed to assuage any doubt about the origins of the lead measured in the tooth enamel. The isotope ratios obtained from the two lead coffin individuals and their corresponding coffin samples are presented in Table 8.2. No isotope data is available for the lead coffin from Winchester, therefore the lead isotope ratios obtained from a dentine sample from this individual has been used as a proxy for the coffin composition. Previous analysis of this sample demonstrated a lead concentration of 1540 ppm, which has been interpreted as contamination from the burial environment and likely representative of the isotopic
composition of the lead coffin within which the individual was interred (Montgomery, 2002).



Figure 8.29 – Map of Britain showing the location of the lead coffin burials

Table 8.2 – Tooth enamel and lead coffin isotope ratios from the Ilchester and York lead coffin burials (Spitalfields and Winchester data from Montgomery, 2002. Oxygen isotope data from N.Wilson, pers. comm.).

| Sample       | Element   | Sex | Age | δ <sup>18</sup> O<br>(VSMOW) | <sup>87</sup> Sr/ <sup>86</sup> Sr | Pb<br>ppm | <sup>206</sup> Pb/ <sup>204</sup> Pb | <sup>207</sup> Pb/ <sup>204</sup> Pb | <sup>208</sup> Pb/ <sup>204</sup> Pb |
|--------------|-----------|-----|-----|------------------------------|------------------------------------|-----------|--------------------------------------|--------------------------------------|--------------------------------------|
| Ilchester    | M2        | F   | YA  | -                            | 0.7089                             | 63.3      | 18.4174                              | 15.6370                              | 38.4137                              |
| Ilchester    | Pb coffin | -   | -   | -                            | -                                  | -         | 18.4131                              | 15.6351                              | 38.4167                              |
| York         | M2        | М   | MA  | 20.0                         | 0.7109                             | 93.8      | 18.5862                              | 15.6633                              | 38.6931                              |
| York         | Pb coffin | -   | -   | -                            | -                                  | -         | 18.4141                              | 15.6414                              | 38.4184                              |
| Spitalfields | PM2       | F   | YA  | -                            | 0.7099                             | 30.1      | 18.4600                              | 15.7000                              | 38.6200                              |
| Spitalfields | Pb coffin | -   | -   | -                            | -                                  | -         | 18.4001                              | 15.6291                              | 38.3686                              |
| Winchester   | M3        | М   | YA  | -                            | 0.7092                             | 41.8      | 18.3700                              | 15.5700                              | 38.2200                              |
| Winchester   | Dentine   | -   | -   | -                            | -                                  | 1540.0    | 18.4300                              | 15.6000                              | 38.3600                              |

#### 8.7.1.1 The lead coffins

With the exception of the Winchester dentine sample, all of the lead coffin samples plot within a tight cluster of points located within the expected ranges for British lead ore, at the upper end of the Mendips ore field data. The Winchester dentine sample plots further down, but still within the Mendips ore field (see Fig. 8.30). All four coffin samples plot within the bottom right 'lead province' (see Fig. 8.30) indicating lead isotope ratios consistent with region of Atlantic Europe with а Hercynian/Cambrian/Precambrain orogeny. This is congruent with British lead ore. Thus, indicating that the lead used to construct all four coffins was sourced from southern Britain. Previous studies that have analysed Romano-British lead coffin burials at Poundbury Camp, Dorset (Molleson et al., 1986) and Spitalfields, London (Montgomery 2002) have also demonstrated that the lead used to construct the coffins originated from English ore sources. Importing lead would have been an expensive and unnecessary endeavour considering the vast quantities of lead mined in Britain during this period (Toller, 1977). Therefore, it is unsurprising that people were utilising the readily available resources in southern Britain.

Comparisons of the tooth enamel lead isotope ratios with those from the lead coffin samples confirms that the lead measured in the tooth enamel represents *in vivo* lead acquisition (see Fig. 8.30). Although the Ilchester female has lead isotope ratios similar to those of her lead coffin, the fact that the other three individuals exhibit *in vivo* isotope ratios markedly different from their respective coffins leaves little reason to suspect that the Ilchester individuals does not do the same. All four individuals underwent identical sampling and processing procedures therefore it is likely that the isotope ratios obtained from the Ilchester female represent a childhood spend in Britain rather than post-mortem contamination from the burial environment.



Figure 8.30 – Bivariate plot showing <sup>208</sup>Pb/<sup>206</sup>Pb against <sup>207</sup>Pb/<sup>206</sup>Pb ratios for the coffin burials in relation to comparative datasets. All coffin samples cluster tightly within the British ore field, while the tooth enamel samples exhibit diverse characteristics. Mendips ore field data taken from Haggerty et al. (1996) and circum-Mediterranean data from Butcher and Ponting (2014)

#### 8.7.1.2 Investigating origins

The individual from York had a high  $\delta^{18}O_{VSMOW(p)}$  value (20.0‰), which falls outside the expected range for Britain (see Fig. 8.31) and thus indicates origins in a region with a warmer climate or lower altitude than Britain. There have been very few Romano-British individuals reported with values as high as this, with only three individuals from London exhibiting higher values (Redfern et al., 2016). Three individuals from York, Driffield Terrace (6Drif- 21: 19.8‰ and 3Drif-26: 22.9‰) and Trentholme Drive (TDC710: 19.7‰), have similarly high  $\delta^{18}O_{VSMOW(p)}$  values and have been interpreted as migrants to Britain from either a southern Mediterranean region of Europe or North Africa (Leach et al., 2009; Müldner et al., 2011). The isotopic and aDNA results from individual 3Drif-26 offer compelling evidence for origins in the Middle East (Martiniano et al., 2016). The corresponding drinking water value ( $\delta^{18}O_{(dw)}$ ) for this York lead coffin individual is calculated as -3.0 +/- 1.0‰, which also suggests a childhood spent in a southern Mediterranean or North African region of the Empire (Longinelli and Selmo, 2003; Lykoudis and Argiriou, 2007). While the high value obtained for this York individual does suggest origins in a warm, arid environment, previous migration studies in the Nile Valley and Nubia have demonstrated  $\delta^{18}O_{SMOW(p)}$ values significantly higher than that seen in this Roman individual (Dupras and Schwarcz, 2001; Iacumin et al., 1996; White et al., 2004). This does appear to rule out areas of North Africa such as Egypt and Sudan, however with large expanses of the continent without available comparative data, other provinces of the Roman Empire in Northern Africa may still be viable locations.



Figure 8.31 – Bivariate plot showing <sup>87</sup>Sr/<sup>86</sup>Sr against  $\delta^{18}O_{VSMOW(p)}$ . The black box represents the UK range for  $\delta^{18}O_{VSMOW(p)}$  (± 2 sd) and the shaded area represents the UK range for <sup>87</sup>Sr/<sup>86</sup>Sr (Evans et al., 2012). Comparative Roman York data from: Driffield Terrace (Müldner et al., 2011); Trentholme Drive and The Railway (Leach et al., 2009); Catterick (Chenery et al., 2011).

The strontium isotope ratio (0.71094) obtained from this York individual is common in many areas of the Roman Empire but does exclude limestone and basalt terrains found in large regions of North Africa and the Middle East. Although the strontium isotope ratio obtained from the York male sits within the expected range for Britain (see Fig. 8.31) it is slightly more radiogenic than the smaller strontium isotope range suggested for the Vale of York (between 0.7084 and 0.7102) (Chenery et al., 2011; Leach et al., 2009). However, this value is also associated with a range of Cenozoic terrains found extensively throughout Europe (Chenery et al., 2010; Evans et al., 2010). Therefore,

despite being consistent with the British range, in combination with the high  $\delta^{18}$ O value obtained for this individual, a childhood spent outside of Britain is most likely.

The use of lead isotopes to investigate the origins of this individual does help constrain possible areas of origin further. In Figure 8.32 the York individual has been plotted alongside lead ore data from broad regions of the Roman Empire, previously published Romano-British human tooth enamel results and the human data generated in this study. As with the oxygen isotope values, the lead isotope ratios appear to rule out a childhood spent in Britain, with isotope ratios plotting away from the main cluster of contemporaneous British individuals in the lower end of the circum-Mediterranean lead ore range established by Butcher and Ponting (2014) and within the Israeli and Romanian lead ore fields. This York individual has higher <sup>207</sup>Pb/<sup>206</sup>Pb lead isotope ratios than those seen in North African lead ores, which plot lower than the York individual (see Fig. 8.32), indicating exposure lead sources older than those observed in North African lead ores. The lead isotope ratios obtained from this York individual plot within the upper left 'lead province' (see Fig. 8.30), indicating exposure to anthropogenic lead ore in a region of central or circum-Mediterranean Europe with Alpine orogeny.

The York individuals' isotope characteristics are inconsistent with origins in North Africa and the Levant as archaeological individuals from these areas are reported to have much higher oxygen and lead isotope values than those seen here (Dupras and Schwarcz, 2001; Iacumin et al., 1996; Martiniano et al., 2016; White et al., 2004). The female lead coffin burial recovered from Spitalfields, London, has a similar strontium isotope ratio to this York individual, and, after comparison with isotope data from three individuals from Rome, has been interpreted as having possible origins in Italy

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(Montgomery, 2002; Montgomery et al., 2010). However, the isotope characteristics of the York individual seem inconsistent with origins in Italy. The oxygen value obtained from this individual is higher than those expected for mainland Italy ("IAEA/WMO," 2006 in Evans et al., 2012; Longinelli and Selmo, 2003), and although both the Spitalfields female and the York male have lead isotope ratios indicative of origins in a region with Alpine orogeny, the lead isotope ratios exhibited by the York individual are higher than those seen in individuals from Rome. The isotope characteristics obtained from the York lead coffin burial indicate that this male is a migrant to Roman Britain, likely from a region of the Empire with a warmer, drier climate. Although the strontium isotope ratio obtain from the tooth enamel is relatively common across Europe, the addition of lead and oxygen isotope results suggest that a childhood spent in areas of Eastern Europe (Crowder et al., (in press).

The isotopic composition of the Ilchester individual is much less varied. This individual has a similar strontium isotope ratio (0.7089) to the Winchester lead coffin individual (0.7092), both of which are consistent with the Mesozoic terrains found in England and large expanses of Europe. The lead isotope ratios for both of these individuals are also consistent with childhood origins in Britain, as both individuals plot within the expected ranges for British lead ore (Fig. 8.32).

### 8.7.1.3 Conclusions

It is evident from the disparate isotopic characteristics obtained from the tooth enamel of these intriguing burials that they spent their childhoods in very different locations. The Ilchester female and the Winchester male appear to have British origins, however, the possibility that they are second-generation migrants cannot be ruled out. Conversely, the Spitalfields female and York male are likely migrants to Britain with isotopic compositions indicative of a childhood spent in warmer regions of the Roman Empire. These results suggest that lead coffin burials in Britain were not a rite reserved exclusively for migrants. A few individuals from dispersed regions of the Empire as well as locals appear to have been interred in this way. What these results do reinforce are the multicultural nature of societies within the Roman Empire and the tenuous link between burial rites and birthplace origin.



Figure 8.32 – Bivariate plot showing Pb isotope results for the lead coffin individuals in relation to comparative datasets. Mendips ore field data taken from Haggerty et al. (1996) and circum-Mediterranean data from Butcher and Ponting (2014).

#### 8.7.2 Decapitation burials

Musselburgh is home to one of the largest known Roman settlements outside of a fort in Scotland (Jones, 2012), consisting of both military and civilian settlement types (Thomas, 1988). Given its coastal location and prominent position on Dere Street (Hanson and Breeze, 1991; Shotter, 1996), Musselburgh was likely a key part of the supply network providing provisions to the military sites built along the Forth-Clyde isthmus (Breeze, 2006; Jones, 2012; Whittaker, 2002). In an area thought to be a prime location for the convergence of people from diverse locations, intrusive burial rites here raise questions about the geographic origins of the unusual individuals. This is especially true, considering the site's close proximity to the Antonine Wall, a large military presence in a location with no history of previous occupation on such a scale (Tipping and Tisdale, 2005).

Several of the Roman skeletons excavated from Musselburgh display evidence of trauma, with at least four of the skeletons exhibiting trauma patterns consistent with decapitation (Anderson, 2011). Although not a common burial rite in Scotland, decapitation burials have been identified in other Roman period contexts (Philpott, 1991; Tucker et al., 2014). Decapitation burial is a common minority rite in Roman-Britain (Tucker et al., 2014, p.213), and at sites where it has been identified, the crude prevalence rate is approximately 5–10% (Müldner et al., 2011; Philpott, 1991; Roberts and Cox, 2003) with the majority of examples dating to the 4<sup>th</sup> century AD (Philpott, 1991). However, at the late Roman ( $2^{nd} - 4^{th}$  century AD) cemetery at Driffield Terrace, York, close to 80% of the predominantly male skeletons exhibited evidence for decapitation. However, there is currently no consensus as to why this is the case (Martiniano et al., 2016; Montgomery et al., 2011; Müldner et al., 2011). It has been

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theorised that decapitation was a form of post-mortem burial ritual (Mattingly 2006, p.478; Timberlake, 2007, p.57; McKinley and Egging-Dinwiddy, 2009, p.58; Taylor 2008; Jones, 2003). However, a review of the osteological evidence has demonstrated that a large number of individuals was decapitated as a mechanism of death either as a live sacrifice or judicial execution (Tucker et al., 2014, p230).

The isotope ratios obtained from these six Roman Musselburgh individuals are presented in Table 8.3; one Iron Age individual from the same site is also included to establish the local human isotope range. Figure 8.33 shows the Musselburgh individuals plotted against datasets from Roman coins of known provenance (Butcher and Ponting, 2014) and the Mendips ore field data (Haggerty et al., 1996), which provide comparative ranges for Mediterranean and English lead isotope ratios respectively. Additional Roman migration studies (Montgomery et al., 2010; Shaw et al., 2016) and data collected in this study offer further comparative human data and are also included here.

Table 8.3 – Multi-isotope results from the Musselburgh Roman and Iron Age individuals tooth enamel samples (Strontium and oxygen data from Moore et al., in prep)

| Sample  | Element | Period      | Sex | Age       | Decap | δ <sup>18</sup><br>Οc | <sup>87</sup> Sr/ <sup>86</sup> Sr | Pb<br>ppm | <sup>206</sup> Pb/ <sup>204</sup> Pb | <sup>207</sup> Pb/ <sup>204</sup> Pb | <sup>208</sup> Pb/ <sup>204</sup> Pb |
|---------|---------|-------------|-----|-----------|-------|-----------------------|------------------------------------|-----------|--------------------------------------|--------------------------------------|--------------------------------------|
| PPCM235 | PM2     | Roman       | М   | MA-<br>OA | Yes   | 25.3                  | 0.71411                            | 1.82      | 18.3372                              | 15.6376                              | 38.4341                              |
| PPCM316 | PM2     | Roman       | М   | MA-<br>OA | Yes   | 25.5                  | 0.71403                            | 0.61      | 18.4663                              | 15.6367                              | 38.5100                              |
| PPCM323 | PM2     | Roman       | М   | YA        | -     | 25.3                  | 0.70896                            | 0.35      | 18.5377                              | 15.6528                              | 38.4793                              |
| PPCM420 | M2      | Roman       | М   | А         | Yes   | 25.3                  | 0.71245                            | 2.17      | 18.3821                              | 15.6296                              | 38.3994                              |
| PPCM451 | M2      | Roman       | М   | MA-<br>OA | -     | 26.5                  | 0.71397                            | 2.20      | 18.3783                              | 15.6313                              | 38.3900                              |
| PPCM630 | PM2     | Roman       | М   | MA-<br>OA | Yes   | 27.3                  | 0.70980                            | 6.57      | 18.4395                              | 15.6321                              | 38.4515                              |
| PPCM864 | PM2     | Iron<br>Age | ?   | YA        | -     | 26.3                  | 0.70973                            | 0.13      | 18.3396                              | 15.5947                              | 38.3280                              |

The Iron Age individual (PPCM864) exhibited a  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratio and  $\delta^{18}$ O<sub>dw</sub> value that was consistent with local origins in the East Lothian area (see Fig. 8.34) (Moore et

al., in prep). This individual also had a very low lead burden which is inconsistent with exposure to the anthropogenic lead pollution of the Roman Empire and in line with prehistoric lead levels (Montgomery, 2002; Montgomery et al., 2010). The lead isotope ratios from this Iron Age individual are also comparable with other prehistoric individuals from Scotland, indicating exposure to low level geogenic lead sources (Montgomery et al., 2005, 2010). Therefore, this individual provides a good baseline against which to compare the intrusive Roman burials at Musselburgh.



Figure 8.33 – Bivariate plot showing the <sup>208</sup>Pb/<sup>206</sup> against <sup>207</sup>Pb/<sup>206</sup>Pb ratios from the Musselburgh individuals tooth enamel in relation to comparative datasets. Data for comparative contemporary human tooth enamel samples were taken from: England Shaw et al., (2016) and Montgomery et al., (2010); Scotland and Italy samples from Montgomery et al., (2010). Mendips ore field data from Haggerty et al., (1996) and circum-Mediterranean data from Butcher and Ponting (2014).



Figure 8.34 – <sup>87</sup>Sr/<sup>86</sup>Sr and  $\delta^{18}O_{dw}$  values for enamel samples from the Musselburgh Roman (n = 6) and Iron Age (n = 1) individuals. Also showing the estimated range of UK  $\delta^{18}O_{dw}$  (mean -7.4 ± 1.7‰, 2 $\sigma$ ) and <sup>87</sup>Sr/<sup>86</sup>Sr (0.7078 - 0.7165) values (dashed box) obtained from previous archaeological studies (Evans et al., 2012b) as well as the estimated <sup>87</sup>Sr/<sup>86</sup>Sr and  $\delta^{18}O_{dw}$  range for the Lothian area (dotted box) according to (Darling and Talbot, 2003). <sup>87</sup>Sr/<sup>86</sup>Sr 2 $\sigma$  errors are within the symbol

Considering the local isotope ratios provided by the Iron Age individual, it is clear that the Roman individuals at Musselburgh exhibit extremely variable tooth enamel isotope characteristics and do not appear to have originated from the Lothian area. When compared to other Romano-British sites, few Roman Period individuals have strontium isotope ratios as high as those found at Musselburgh. In fact, British values above 0.714 are rare in all periods (Evans et al., 2012). The only individual that displays higher strontium isotope ratios was recorded at Driffield Terrace, York, and when considered in conjunction with a high  $\delta^{18}$ O value, it was concluded that this individual was likely to have originated from an area with more radiogenic geology, such as Sardinia, Corsica, northern Italy or northern Africa (Montgomery et al., 2011). Four of the Musselburgh individuals have low  $\delta^{18}O_{dw}$  values (c. –9.1‰) and high <sup>87</sup>Sr/<sup>86</sup>Sr values above 0.7120, which are not common within British burials. Decapitated individual PCM316 had a low  $\delta^{18}O_{dw}$  value consistent with origins in eastern Britain or northern Europe together with the high <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratio indicative of a childhood spent in a region with a granitic terrain. However, PPCM316's lead burden was low and accompanied by lead isotope ratios inconsistent with Scottish ore sources (Rohl, 1996). This data suggests exposure to natural lead sources from younger rocks such as those found in England (Rohl, 1996). Very similar lead isotope ratios have been obtained from a male burial at Driffield Terrace whose  $\delta^{18}O_{dw}$  value and aDNA indicate that origins in the Levant are extremely likely (Martiniano et al., 2016). However the high <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratio and low  $\delta^{18}O_{dw}$  value make such origins for PPCM316 unlikely.

The remaining three individuals (PPCM235, PPCM420 and PPCM451) with high <sup>87</sup>Sr/<sup>86</sup>Sr isotope ratios are also linked through their lead isotope characteristics. Their low lead burdens indicate low level childhood exposure to anthropogenic lead pollution from ore sources inconsistent with those found in England. The lead isotope ratios that accompany these low lead burdens are more indicative of older lead ores such as those found in Scotland (Rohl, 1996). High <sup>207</sup>Pb/<sup>206</sup>Pb isotope ratios, similar to those of PPCM235, have been observed in a small number of individuals from the Scottish Isles (Montgomery et al., 2010). However, PPCM235 has a higher <sup>208</sup>Pb/<sup>206</sup>Pb isotope ratio than these Scottish individuals, indicating childhood origins in a geographical region with Alpine orogeny rather than the Cambrian orogeny found in Scotland.

Skeletal preservation is often poor in northern Scotland and there are very few comparative sites with which to compare the Musselburgh individuals. Nonetheless, a small Beaker Period assemblage from northern Scotland exhibits the same high strontium and low oxygen isotope characteristics seen in PPCM235, PPCM316 and PPCM420 (Pearson et al., 2016). Similar lead and strontium isotope characteristics have been obtained from a child excavated in Roman London. This individual is thought to originate from Germany (Shaw et al., 2016), and while origins in western Europe are certainly possible for these individuals origins within the Scottish Highlands cannot be ruled out.

In contrast to the rest of the Musselburgh assemblage, individuals PPCM323 and PPCM630 exhibit low strontium isotope ratios more typical of Mesozoic and Cenozoic sediments (Evans et al., 2010). This limits the efficacy of their strontium isotope ratios in constraining their area of geographical origin, as these sediment types are prevalent throughout Europe and most of Britain. It is clear that decapitated male PPCM630 does not originate from the local area. Despite the rather common and therefore undiagnostic  ${}^{87}$ Sr/ ${}^{86}$ Sr isotope ratio, PPCM630's  $\delta^{18}$ O<sub>dw</sub> value is too high for origins in the Lothian area and most of eastern Britain, with a value more consistent with areas of western or southern Britain. PPCM630 also has the highest lead burden amongst the Musselburgh group, with an enamel concentration of 6.57 ppm. When interpreted in conjunction with the lead isotope ratios, PPCM630 plots tightly with contemporary burials from England (see Fig. 8.33) and within the English ore field (Haggerty et al., 1996). These results indicate anthropogenic exposure to English lead ore (Montgomery, 2002; Montgomery et al., 2010) and suggests that this individual spent his childhood within the Roman Empire, with likely origins in southern Britain. Conversely the low  $\delta^{18}O_{dw}$  value seen in PPCM323 is not common in Romano-British individuals and when considered in

conjunction with the individuals' low lead isotope ratios, it is likely that PPCM323 originated from overseas. Indeed, PPCM323 has low  $\delta^{18}O_{dw}$  values common in central and Eastern Europe (Crowder et al., in press; Hakenbeck et al., 2017; "IAEA/WMO," 2006 in Evans et al., 2012), and while the low lead isotope ratios for this individual appear to exclude the Mediterranean as a region of origin, it is possible that PPCM323 originates from another region of the Roman Empire.

Using a multi-isotope approach, this study has explored the link between intrusive burial rites and geographic origins at the Antonine Wall frontier, and like the majority of human provenance studies, the results have affirmed the current understanding that a multifarious relationship exists between the two. The isotopic data does suggest that the decapitation burials at Musselburgh do not represent the continuation of a native Iron Age population. Furthermore, while their commonality as possible migrants to southern Scotland links the burials, the individuals do not appear to share a common geographical origin, suggesting that something more complex than a shared ethnicity unites the individuals. However, it is apparent that a more comprehensive understanding of the isotopic variation amongst archaeological populations with childhoods spent in Scotland is needed before non-British origins for these Musselburgh individuals can be confidently established. Finally it should be noted that the Musselburgh decapitation burials represent the earliest known examples of Roman inhumation and decapitation in Britain (Kirby, 2016), and their apparent ethnic diversity coupled with a shared burial rite reflects the cosmopolitan nature of the Roman army.

# 8.8 Summary

One of the main aims of this study was to establish what constitutes the local lead isotope range for Roman populations from five regions within the Roman Empire. This was achieved for four of the five regions included in the study, with the results demonstrating that Roman tooth enamel exhibits lead isotope ratios consistent with lead ore from their corresponding country. This has resulted in successfully establishing expected 'local' lead isotope ranges for Roman individuals with childhood origins in Spain, France, Romania and Lebanon. These ranges provide a good baseline against which other isotope studies can compare their data, and should aid in the identification of possible geographic origins of any outliers in future Roman mobility studies.

In each of these four countries, individuals with lead burdens above 1 ppm exhibit the characteristic cultural focusing and linear spread associated with *in vivo* isotope ratios dominated by lead ore signatures. This indicates that the cultural shift from predominantly geogenic lead to predominantly anthropogenic lead is not limited to the British populations in which this trend has initially been reported. Although there is an overlap in these anthropogenic isotope ratios between countries, the results of this study have shown that lead isotope ratios can distinguish between individuals from either Eastern or Western European countries as individuals and lead ores provide higher <sup>206</sup>Pb/<sup>204</sup>Pb isotope ratios relative to samples from western European countries. The second feature is differentiation based on the orogenic age of the region in which an individual spent their childhood. Individuals from older Hercynian or Cambrian regions such as Britain, France and Germany have lower <sup>208</sup>Pb/<sup>206</sup>Pb ratios than younger Alpine regions such as Romania, Spain or Italy.

With regards to the Slovenian human samples which were not consistent with Slovenian ore isotope ratios, the small sample size of both human samples and lead ore make it difficult to determine whether the disparity in isotopic composition between the two is

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due to an as yet unknown end member in the lead ore field or whether the one individual with an anthropogenic lead burden is a migrant to Slovenia. However, as more enamel lead isotope data is obtained from Roman burials throughout Europe, the geographic origins of individuals with different lead isotope ratios will become identifiable with increasing confidence.

A second aim of this study was to assess whether combining lead and strontium analyses could improve not only our ability to identify migrants within skeletal populations but also narrow down possible regions of origin. Lead and strontium isotopes provide different information regarding an individual's residential history. Strontium represents the geology of the land on which an individuals food and water sources originated, while lead represents the dominant ore sources exploited by a particular population. Therefore, both isotopic systems have the potential to identify different outliers within the same assemblage and when combined, increase the resolution at which possible origins can be suggested. The results of this study support this as, although the majority of outliers were identified due to their different lead isotopes ratios, two individuals were highlighted as outliers due to their disparate strontium isotope ratios. This study has found that lead isotope analysis is an asset to migration studies on Roman populations and that not only are more outliers found if more than one isotope system is analysed together, but a better resolution on possible childhood origins can be obtained. Therefore where possible, it is best to apply a multi isotopic approach when assessing migration within skeletal populations.

# CHAPTER NINE

# **Conclusion and Future Directions**

# 9.1 Introduction

The overarching aim of this study was to explore how exposure to anthropogenic lead pollution impacted upon childhood health and mortality during the Roman period, and what this type of exposure could tell us about geographic mobility within the Empire. It focused upon the use of lead isotopes as a discriminant in migration studies in an attempt to determine the extent of variation in lead isotope ratios between countries in a highly polluted archaeological population, as well as the paired analysis of lead concentration and palaeopathological data to explore the impact of lead exposure on childhood health and mortality. In this final chapter the findings of this study are summarised and suggestions for future work, which could be undertaken to further our understanding of lead concentrations and lead isotope ratios in relation to Roman health and mobility are given.

# 9.2 Lead concentration analysis

### 9.2.1 Sample variation

An understanding of how lead is incorporated into and distributed between different tooth types is important not only for the interpretation of trace elemental data from tooth enamel, but also in how data between studies can be compared. To address this, this study explored the differences in lead concentrations between tooth types and tooth positions. The results produced here mirror those of previously published work (Kamberi et al., 2012; Mackie et al., 1977; Pinchin et al., 1978), demonstrating that the distribution of lead within teeth is non-uniform. There are undoubtedly significant inconsistencies in inter-dental lead concentrations with no consensus on which dental arcade provides the highest concentrations, or if there are any patterns to which type of tooth will yield the highest amount of lead. It is likely that these inter-dental variations in lead concentrations are population specific and reflect not only the innate physiological factors related to lead absorption but also an individual's level of lead exposure at the time of tooth formation. As an individual's exposure cannot be controlled for, standardising the type and position of the tooth sampled would be the first step towards reducing any variability driven by physiological factors and would therefore improve comparability between studies. However, this has the potential to be extremely limiting, especially in archaeological studies where skeletal preservation and completeness dictates which teeth can be sampled. If standardisation of tooth type were to be implemented, sample sizes would likely be greatly reduced.

## 9.2.2 Comparing males and females

To further understand how exposure patterns may have varied within the Roman Empire, differences between male and female lead concentrations were assessed. In this study the majority of males had higher lead concentrations than females, which was consistent with modern studies examining sex differences in lead burdens (Claymaet et al., 1991; Costa de Almeida et al., 2010; Paoliello et al., 2002). Although intrinsic factors such as gene expression and hormone levels have been suggested as the cause of these differences (Björkman et al., 2000; Vahter et al., 2007), studies have shown that the short periods of postnatal endocrine surges do not result in any differences in lead concentrations between the sexes. As the values obtained in this study represent

prepubescent childhood, a time of life when the biochemistry of boys and girls is very similar (Bidlingmaier et al., 1975, 1973), any sex differences observed in archaeological lead burdens are most likely the result of differences in levels of environmental exposure. Thus, these results suggest that during the Roman period boys were engaging in a wider range of activities that facilitated higher levels of lead exposure than girls.

A notable finding in this study was that higher lead concentrations were observed in the female individuals from Barcelona and Tarragona than the male individuals from the same sites. Although these differences were not statistically significant, they raise interesting questions about why Spanish individuals go against the trend seen in the majority of research into human lead concentrations. Archaeological studies that have identified this trend have attributed it to the status of the individuals (Nakashima et al., 2007, 1998), with wealthier, higher status females thought to have increased access to lead containing products than lower status females. A comparison of burial rites in the Barcelona population (mausolea verses tegula graves) supports the presupposition that wealthier females have higher lead burdens than lower status females. Inferences about certain aspects of identity, such as status, are often difficult in archaeological contexts as assessments are often biased by our own expectations (i.e. grave goods). The use of lead concentration analysis to address questions surrounding status may provide an additional, objective method to the current means of assessment.

## 9.2.3 Health and mortality

This study has provided the first bioarchaeological evidence that lead poisoning may have been an influential factor in the poor childhood health observed throughout the Roman Empire. The elevated lead concentrations seen in non-adults with metabolic disease in comparison to non-adults without skeletal markers for disease offers strong evidence to suggest that anthropogenic lead pollution contributed to the high prevalence rates of metabolic diseases, especially rickets, in Roman populations. Analysis of lead concentrations in rachitic individuals also revealed that nuances in the aetiology of the disease (caused by nutritional paucity or pollution) could also be tentatively established. The negative correlation observed between lead concentrations and age-at-death also implicates elevated lead burdens in the preponderance of Roman infant remains present in skeletal assemblages, suggesting that lead exposure contributed to the high infant mortality rates seen in Roman populations. The introduction of a bioarchaeological perspective to the decades-old debate surrounding how lead affected health during the Roman period has provided new insights into the impact of environmental lead pollution on the fragility of childhood health throughout the Empire.

# 9.3 Lead isotope analysis

#### 9.3.1 Cultural focusing

As demonstrated in previous studies, lead concentrations under 1 ppm are thought to represent geogenic exposure and have been shown to exhibit a higher degree of variability in their isotopic ranges than lead acquired through anthropogenic exposure (Montgomery, 2002; Montgomery et al., 2010; Shaw et al., 2016). While individuals with lead burdens above 1 ppm exhibit the characteristic cultural focusing and linear spread associated with *in vivo* isotope ratios dominated by lead ore signatures. As expected for populations in anthropogenically-polluted regions, the majority of data points from all five countries in this study produced linear arrays characteristic of lead ore field isotope ranges. Within these isotope fields there was a reduction in isotope variability with increasing lead concentration. This indicates that the cultural shift from

predominantly geogenic lead to predominantly anthropogenic lead is not limited to the British populations in which this trend has initially been reported, and that cultural focusing is universal phenomenon associated with anthropogenic lead use.

#### 9.3.2 Establishing local ranges

One of the main aims of this study was to establish what constitutes the local lead isotope range for Roman populations from five countries within the Roman Empire. This was achieved for four of the five countries included in the study, with the results demonstrating that Roman tooth enamel exhibits lead isotope ratios consistent with lead ore from their corresponding country. This has resulted in successfully establishing expected 'local' lead isotope ranges for Roman individuals with childhood origins in Spain, France, Romania and Lebanon. However, it must be noted that the tendency of lead ore fields to spread over a wide range of values means that there is often overlap between lead ore fields from different countries. Due to this overlap, which is evident in both tooth enamel and lead ore data, it is clear that lead isotope ratios can be useful in distinguishing between broad regions of Europe, such as Eastern vs. Western Europe. Thus, the data produced in this study provides a good baseline to which other isotope studies can compare their data, and should aid in the identification of possible geographic origins of any outliers in future Roman mobility studies.

## 9.3.3 Geographic variation

Although there is an overlap in the anthropogenic lead isotope ratios between countries, the results of this study have demonstrated that lead isotope ratios can differentiate between populations on the basis of two broad features. Firstly, lead isotope ratios can distinguish between individuals from either Eastern or Western European countries. The results of this study have demonstrated that individuals and lead ores from eastern European countries exhibit lead isotope ratios enriched in <sup>206</sup>Pb/<sup>204</sup>Pb relative to samples from western European countries. The second feature is differentiation based on the orogenic age of the region in which an individual spent their childhood. Individuals from older Hercynian or Cambrian regions such as Britain, France and Germany have lower <sup>208</sup>Pb/<sup>206</sup>Pb isotope ratios than younger Alpine regions such as Romania, Spain or Italy. Although lead isotope ratios are not country specific this study has demonstrated that they are capable of discriminating between geographical regions when other isotope system are not.

#### 9.3.4 Identifying migrants

Different isotope systems provide information pertaining to specific environmental features be that geological (Sr and Pb), climate (O) or food source (C and N). With the overlapping of environmental parameters across significant expanses of land the analysis of a singular isotope system is unlikely to ever be sufficient to identify all migrants within a population. In this study the combined use of strontium and lead isotope analysis has demonstrated that not only are more outliers found if more than one isotope system is analysed together, but a better resolution on possible childhood origins can also be obtained. Therefore where possible, applying a multi-isotopic approach is the most effective means of assessing migration within skeletal populations.

# 9.4 Limitations

Although this research has greatly contributed to our understanding of lead isotope analysis in Roman populations this section discusses some of the limitations identified during the study, the majority of which can be addressed with future research. The necessarily small samples sizes due to cost, are the major limiting factor for this study. Firstly, the patterns observed in the lead isotope ratio data from previously studies regions of the Roman Empire has highlighted four 'lead provinces' that can be used to constrain possible regions of origin in mobility studies. However, the sample sizes analysed from each region were small (n = 12), and therefore the data should be used as a guide for the expected lead isotope ratios for a particular region. As more data becomes available in the future the expected lead isotope ratio ranges for regions within the Roman Empire will become increasingly accurate and the four 'lead provinces' suggested in this study will need to be refined. Secondly, the sample sizes available for comparisons between lead concetrations and status are also extremely small (n = 12). Althought the correlation seen in this study between lead concetrations and status are interesting, the small sample size is limiting. Therefore, the results cannot be used to infer that there are general differences in lead concetrations according to status throughout the Roman Empire.

Finally, the physiological factors that underpin the acquisition of lead in deciduous and permanent tooth enamel need to be better understood. This study has demonstrated that the inconsistencies in tooth enamel lead concentrations suggest that physiological factors are only partially responsible for the acquisition of lead in tooth enamel during life, and that cultural factors can significantly influence lead concentrations. However, the extent to which physiological factors influence lead concentrations in people of different ages and between males and females is important for the interpretation of any lead concentration data. Currently these processes are poorly understood and therefore current interpretations are limited.

# 9.5 Future research

This study has greatly enhanced our understanding of how anthropogenic lead isotope ratios vary geographically and how lead exposure impacted upon health. However, it is evident that there is much more that can be done to further our understanding of geographic variation in lead isotope ratios and concentrations in Roman populations. In the following section future directions are suggested.

Firstly, comparison of lead concentration from rural and urban sites would be useful. In this study only urban sites were utilised and within these there was a wide range of lead concentrations. This demonstrates that not all inhabitants of the Roman Empire acquired lead burdens above the currently accepted anthropogenic threshold (1 ppm). By comparing individuals from rural environments with those from urban locations it may be possible to establish what the normal range of lead burdens is to be expected in individuals living in locations with low exposure risks. This would improve our interpretation of lead concentrations in Roman populations and potentially allow the identification of individuals who have moved from highly polluted, urban environments to low pollution areas such as rural settlements. From the small sample size, this study has provided promising results that suggest higher status is accompanied by higher lead concentrations. Targeted sampling of individuals of perceived high status (burials rich in grave goods) alongside contemporaneous individuals from supposedly lower status burials would help to further our understanding of how lead exposure varied with socioeconomic status, which would in turn provide information pertaining to any differences in health and wellbeing between social classes.

Secondly, although this study has successfully established baseline human lead isotope ranges for four countries within the Roman Empire, more could be done to further our understanding of geographic variations in human lead isotope ratios. Expanding this work to include more countries within the Roman Empire as well as multiple sites within the same country would help refine the expected lead isotope range for individual countries and improve our understanding of regional variation across Europe. As more enamel lead isotope data is obtained from Roman burials throughout Europe, the geographic origins of individuals with non-local lead isotope ratios will become identifiable with increasing confidence.

Finally, the methodology employed in this study limited the investigations into mobility to childhood movements. By applying lead isotope analysis to cremated remains investigations could be expanded to include movements during adulthood. This is currently untested using lead isotopes but has been successfully applied using strontium isotope analysis (Snoeck et al., 2016, 2015). Due to the high atomic weight of strontium, it does not fractionate during the cremation process and become resistant to postmortem alteration when the bone becomes fully calcined (Snoeck et al., 2016, 2015). As lead isotopes are heavier than strontium is it plausible that *in vivo* lead isotope ratios would also be preserved in fully calcined bone.

# **9.6 Final conclusions**

This study has shown the effectiveness of lead isotope analysis as a tool in archaeological migration studies. The successful establishment of baseline ranges in previously unstudied regions of the Roman Empire has greatly enhanced our ability to identify the potential origins of isotopic outliers and has improved our understanding of how anthropogenic lead isotope ratios in Roman individuals varies across a continent. For the first time this study has also introduced a bioarchaeological perspective to how lead exposure affected health during the Roman period. In addition to providing strong evidence that Roman lead pollution contributed to the high prevalence of metabolic diseases during childhood, especially rickets. These results also provide the first bioarchaeological evidence implicating elevated lead burdens in the high prevalence of infant remains in Roman skeletal assemblages. Thus, offering a new narrative to existing debates over the cause of high infant mortality rates seen in Roman populations.

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| A1 | Osteological and lead concentration data (obtained from tooth enamel using ICP-MS)                          | 311 |
|----|---|-----|
| A2 | Lead and strontium isotope ratios and elemental concentrations from human tooth enamel samples using ICP-MS | 315 |

| Country | Site            | Skeleton | Age      | Pathology | Pb    |
|---------|-----------------|----------|----------|-----------|-------|
| Romania | Dealul Furcilor | M171     | Adult    |           | 0.03  |
| Romania | Dealul Furcilor | M72      | Adult    |           | 4.06  |
| Romania | Dealul Furcilor | M160b    | Adult    |           | 0.23  |
| Romania | Dealul Furcilor | M172     | Adult    |           | 0.80  |
| Romania | Dealul Furcilor | M141     | Adult    |           | 2.41  |
| Romania | Dealul Furcilor | M160     | Adult    |           | 4.69  |
| Romania | Dealul Furcilor | M42      | Adult    |           | 2.87  |
| Romania | Dealul Furcilor | M59      | Adult    |           | 0.25  |
| Romania | Dealul Furcilor | M86      | Adult    |           | 1.15  |
| Romania | Dealul Furcilor | M178     | Adult    |           | 8.29  |
| Romania | Dealul Furcilor | M111     | Adult    |           | 0.28  |
| Romania | Dealul Furcilor | M103     | Foetal   | Scurvy    | 0.92  |
| Romania | Dealul Furcilor | M115     | Foetal   |           | 9.33  |
| Romania | Dealul Furcilor | M116     | Foetal   |           | 5.44  |
| Romania | Dealul Furcilor | M124     | Foetal   |           | 4.62  |
| Romania | Dealul Furcilor | M163     | Foetal   | Scurvy    | 10.67 |
| Romania | Dealul Furcilor | M8       | Foetal   |           | 3.83  |
| Romania | Dealul Furcilor | M19      | Foetal   |           | 5.46  |
| Romania | Dealul Furcilor | M71      | 0-1yr    |           | 7.87  |
| Romania | Dealul Furcilor | M75      | 0-1yr    |           | 2.39  |
| Romania | Dealul Furcilor | M109     | 0-1yr    |           | 6.98  |
| Romania | Dealul Furcilor | M137     | 0-1yr    | Scurvy    | 5.12  |
| Romania | Dealul Furcilor | M158     | 0-1yr    |           | 4.57  |
| Romania | Dealul Furcilor | M52?     | 2-6yrs   |           | 5.81  |
| Romania | Dealul Furcilor | M161     | 2-6yrs   |           | 6.70  |
| Romania | Dealul Furcilor | M2       | 2-6yrs   |           | 3.88  |
| Romania | Dealul Furcilor | M12      | 7-12yrs  | Rickets   | 0.37  |
| Romania | Dealul Furcilor | M170     | 7-12yrs  |           | 2.37  |
| Romania | Dealul Furcilor | M26      | 7-12yrs  |           | 1.19  |
| Romania | Dealul Furcilor | M26b     | 7-12yrs  |           | 0.85  |
| Romania | Dealul Furcilor | M10      | 7-12yrs  |           | 0.31  |
| Romania | Dealul Furcilor | M10b     | 7-12yrs  |           | 0.15  |
| Romania | Dealul Furcilor | M125     | 7-12yrs  |           | 1.90  |
| Romania | Dealul Furcilor | M125b    | 7-12yrs  |           | 1.61  |
| Romania | Dealul Furcilor | M165     | 13-18yrs |           | 1.94  |
| Romania | Dealul Furcilor | M7       | 13-18yrs |           | 0.66  |
| Romania | Dealul Furcilor | M7b      | 13-18yrs |           | 3.34  |
| Spain   | Santa Caterina  | Т3       | Adult    |           | 3.90  |
| Spain   | Santa Caterina  | Т9       | Adult    |           | 0.69  |
| Spain   | Santa Caterina  | T12      | Adult    |           | 3.75  |
| Spain   | Santa Caterina  | T15 (T4) | Adult    |           | 1.73  |

Table A1 - Osteological and lead concentration data (obtained from tooth enamel using ICP-MS)

| Country | Site           | Skeleton | Age      | Pathology        | Pb    |
|---------|----------------|----------|----------|------------------|-------|
| Spain   | Santa Caterina | <br>T3   | Adult    |                  | 1 37  |
| Spain   | Santa Caterina | LIF371   | Adult    |                  | 4 79  |
| Spain   | Santa Caterina | UF755    | Adult    |                  | 1.98  |
| Spain   | Santa Caterina | UF758    | Adult    |                  | 2.60  |
| Spain   | Santa Caterina | UF217    | Adult    |                  | 12.00 |
| Spain   | Santa Caterina | UF708    | Adult    |                  | 1.95  |
| Spain   | Santa Caterina | UF729    | Adult    |                  | 7.28  |
| Spain   | Santa Caterina | T8       | Adult    |                  | 1.57  |
| Spain   | Santa Caterina | UF748    | Adult    |                  | 2.37  |
| Spain   | Santa Caterina | UF1      | Foetal   | Rickets & Scurvy | 59.62 |
| Spain   | Santa Caterina | UF382    | 0-1vr    | Idenets & Searry | 7.09  |
| Spain   | Santa Caterina | 03.40    | 0-1yr    |                  | 1.96  |
| Spain   | Santa Caterina | UF5      | 0-1yr    | Rickets & Scurvy | 4.92  |
| Spain   | Santa Caterina | UF712    | 2-6yrs   | Rickets & Scurvy | 4.03  |
| Spain   | Santa Caterina | UF722    | 2-6yrs   | Scurvy           | 8.07  |
| Spain   | Santa Caterina | T10      | 2-6yrs   |                  | 3.75  |
| Spain   | Santa Caterina | 04.81    | 2-6yrs   |                  | 6.17  |
| Spain   | Santa Caterina | 04.103   | 2-6yrs   |                  | 10.00 |
| Spain   | Santa Caterina | B1.110   | 2-6yrs   |                  | 1.80  |
| Spain   | Santa Caterina | B1.126   | 2-6yrs   | Rickets          | 2.65  |
| Spain   | Santa Caterina | B1.141   | 2-6yrs   |                  | 5.43  |
| Spain   | Santa Caterina | UF730    | 7-12vrs  |                  | 1.26  |
| Spain   | Santa Caterina | UF726    | 7-12vrs  | Rickets & Scurvy | 5.84  |
| Spain   | Santa Caterina | UF730    | 7-12yrs  | ,                | 3.25  |
| Spain   | Santa Caterina | A1.189   | 7-12yrs  | Scurvy           | 8.81  |
| Spain   | Santa Caterina | A2.106   | 7-12yrs  | 5                | 4.94  |
| Spain   | Santa Caterina | B3.099   | 7-12yrs  |                  | 1.77  |
| Spain   | Santa Caterina | UF720    | 13-18yrs |                  | 1.91  |
| Spain   | Santa Caterina | UF720    | 13-18yrs |                  | 3.01  |
| Spain   | Santa Caterina | UF718    | 13-18yrs |                  | 1.97  |
| Spain   | Santa Caterina | UF747    | 13-18yrs |                  | 2.48  |
| Lebanon | ASH 002        | SK431    | Adult    |                  | 2.62  |
| Lebanon | ASH 002        | SK456    | Adult    |                  | 9.71  |
| Lebanon | ASH 002        | SK335    | 0-1yr    |                  | 41.73 |
| Lebanon | ASH 002        | SK110    | 2-6yrs   |                  | 44.13 |
| Lebanon | ASH 002        | SK341    | 13-18yrs |                  | 11.58 |
| Lebanon | ASH 163        | SK506    | Adult    |                  | 2.41  |
| Lebanon | ASH 163        | SK1004   | Adult    |                  | 0.50  |
| Lebanon | ASH 163        | SK428    | 2-6yrs   |                  | 2.88  |
| Lebanon | ASH 163        | SK476    | 2-6yrs   |                  | 38.26 |
| Lebanon | ASH 163        | SK100    | 7-12yrs  |                  | 28.44 |
| Lebanon | ASH 163        | SK489    | 13-18yrs |                  | 4.98  |
| Lebanon | ASH 163        | SK83     | 13-18yrs |                  | 3.51  |
| Lebanon | BCH 740        | SK350    | Adult    |                  | 7.21  |

| Country | Site     | Skeleton<br>ID | Age<br>category | Pathology | Pb<br>ppm |
|---------|----------|----------------|-----------------|-----------|-----------|
| Lebanon | BCH 740  | SK611          | Adult           |           | 1.29      |
| Lebanon | BCH 740  | SK489          | Adult           |           | 9.83      |
| Lebanon | BCH 740  | SK188          | Non-adult       |           | 10.46     |
| Lebanon | BCH 740  | SK445          | Non-adult       |           | 6.66      |
| Lebanon | MDWR 02  | SK2769         | Adult           |           | 1.90      |
| Lebanon | MDWR 02  | SK2615         | Adult           |           | 3.75      |
| Lebanon | MDWR 02  | SK2840         | Adult           |           | 1.47      |
| Lebanon | MDWR 02  | SK2195         | 0-1yr           |           | 5.81      |
| Lebanon | MDWR 02  | SK1846         | 0-1yr           |           | 13.93     |
| Lebanon | MDWR 02  | SK2280         | 0-1yr           |           | 3.24      |
| Lebanon | MDWR 02  | SK1062         | 2-6yrs          |           | 33.37     |
| Lebanon | MDWR 02  | SK2143         | 2-6yrs          |           | 3.50      |
| Lebanon | MDWR 02  | SK2226         | 7-12yrs         |           | 27.19     |
| Lebanon | MDWR 468 | SK193          | Adult           |           | 1.85      |
| Lebanon | MDWR 468 | SK145          | Adult           |           | 0.56      |
| Lebanon | MDWR 468 | SK144          | 2-6yrs          |           | 14.10     |
| Lebanon | MDWR 468 | SK31           | 2-6yrs          |           | 1.64      |
| Lebanon | MDWR 468 | SK79           | 7-12yrs         |           | 4.68      |
| Lebanon | RML 2385 | SK1818         | Adult           |           | 6.68      |
| Lebanon | RML 2385 | SK2318         | Adult           |           | 5.72      |
| Lebanon | RML 2385 | SK1606         | Adult           |           | 2.96      |
| Lebanon | RML 2385 | SK2442         | Adult           |           | 22.60     |
| Lebanon | RML 2385 | SK1607         | 2-6yrs          |           | 22.49     |
| Lebanon | RML 2385 | SK2408         | 2-6yrs          |           | 21.80     |
| Lebanon | RML 2385 | SK2186         | 2-6yrs          |           | 5.89      |
| Lebanon | RML 2385 | SK1635         | 7-12yrs         |           | 24.40     |
| France  | Michelet | S359           | Adult           |           | 0.39      |
| France  | Michelet | S690           | Adult           |           | 24.30     |
| France  | Michelet | S48            | Adult           |           | 16.90     |
| France  | Michelet | S365           | Adult           |           | 2.16      |
| France  | Michelet | S132           | Adult           |           | 8.02      |
| France  | Michelet | S376           | Adult           |           | 1.88      |
| France  | Michelet | S854           | Adult           |           | 0.70      |
| France  | Michelet | S405           | Adult           |           | 0.93      |
| France  | Michelet | S762           | Adult           |           | 2.55      |
| France  | Michelet | S394           | Adult           |           | 0.78      |
| France  | Michelet | S142           | Adult           |           | 0.28      |
| France  | Michelet | S831           | Adult           |           | 7.86      |
| France  | Michelet | S335           | Adult           |           | 1.06      |
| France  | Michelet | S745           | Adult           |           | 1.78      |
| France  | Michelet | S7             | Foetal          | Scurvy    | 25.51     |
| France  | Michelet | S283           | 0-1yr           |           | 9.53      |
| France  | Michelet | S291           | 0-1yr           | Scurvy    | 5.34      |
| France  | Michelet | S134           | 0-1yr           |           | 1.06      |

| Country | Site     | Skeleton<br>ID | Age<br>category | Pathology        | Pb<br>ppm |
|---------|----------|----------------|-----------------|------------------|-----------|
| France  | Michelet | S788           | 0-1yr           |                  | 6.39      |
| France  | Michelet | S542           | 0-1yr           | Rickets & Scurvy | 8.07      |
| France  | Michelet | S123           | 2-6yrs          | Rickets & Scurvy | 5.48      |
| France  | Michelet | S318           | 2-6yrs          |                  | 1.88      |
| France  | Michelet | S347           | 2-6yrs          | Rickets & Scurvy | 9.24      |
| France  | Michelet | S423           | 2-6yrs          | Rickets & Scurvy | 3.61      |
| France  | Michelet | S485a          | 2-6yrs          |                  | 3.23      |
| France  | Michelet | S613           | 2-6yrs          | Scurvy           | 4.78      |
| France  | Michelet | S800           | 2-6yrs          | Scurvy           | 9.66      |
| France  | Michelet | S853           | 2-6yrs          |                  | 1.28      |
| France  | Michelet | S143           | 7-12yrs         |                  | 1.11      |
| France  | Michelet | S150           | 7-12yrs         |                  | 3.67      |
| France  | Michelet | S433           | 7-12yrs         |                  | 4.22      |
| France  | Michelet | S540           | 7-12yrs         |                  | 0.89      |
| France  | Michelet | S830           | 7-12yrs         |                  | 3.28      |
| France  | Michelet | S830           | 7-12yrs         |                  | 1.27      |
| France  | Michelet | S511           | 13-18yrs        |                  | 5.69      |
| France  | Michelet | S164           | 13-18yrs        |                  | 12.60     |
| Spain   | PERI 2   | 1104(UF11)     | Adult           |                  | 18.9      |
| Spain   | PERI 2   | 2129           | Adult           |                  | 2.77      |
| Spain   | PERI 2   | 2413(UF29)     | Adult           |                  | 6.48      |
| Spain   | PERI 2   | 1442(UF11)     | Adult           |                  | 11.20     |
| Spain   | PERI 2   | 1069(UF7)      | Adult           |                  | 3.55      |
| Spain   | PERI 2   | 2209(UF8)      | Adult           |                  | 13.90     |
| Spain   | PERI 2   | 5552(UF19)     | Adult           |                  | 8.86      |
| Spain   | PERI 2   | 1215(UF2)      | Adult           |                  | 1.85      |
| Spain   | PERI 2   | 1035(UF2)      | Adult           |                  | 10.20     |
| Spain   | PERI 2   | 1125/2(UF14)   | Adult           |                  | 20.30     |
| Spain   | PERI 2   | 5400(UF8)      | Adult           |                  | 15.30     |
| Spain   | PERI 2   | 2226(UF17)     | Adult           |                  | 2.46      |
| Spain   | PERI 2   | 1035           | Foetal          | Scurvy           | 187.00    |
| Spain   | PERI 2   | 1092           | 0-1yr           |                  | 12.00     |
| Spain   | PERI 2   | 1098           | 0-1yr           | Rickets          | 13.20     |
| Spain   | PERI 2   | 2024           | 0-1yr           |                  | 19.80     |
| Spain   | PERI 2   | UF1            | 0-1yr           |                  | 15.50     |
| Spain   | PERI 2   | 545(UF37)      | 0-1yr           | Rickets          | 90.40     |
| Spain   | PERI 2   | 5230           | 2-6yrs          | Rickets          | 76.10     |
| Spain   | PERI 2   | 2012           | 2-6yrs          |                  | 99.90     |
| Spain   | PERI 2   | 2430           | 2-6yrs          | Rickets          | 10.70     |
| Spain   | PERI 2   | 2490a          | 2-6vrs          |                  | 7.18      |
| Spain   | PERI 2   | 5319           | 7-12vrs         |                  | 5.55      |
| Spain   | PERI 2   | 1233(UF3)      | 7-12yrs         |                  | 8.85      |
| Spain   | PERI 2   | 2490b          | 7-12vrs         |                  | 6.78      |
| Spain   | PERI 2   | 2604           | 13-18yrs        |                  | 5.14      |

| Country | Region    | Site     | Skeleton<br>ID  | Tooth      | Age<br>Categor<br>y | Sex    | Pb<br>ppm | <sup>206</sup> Pb/<br><sup>204</sup> Pb | 2σ<br>% | <sup>207</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>207</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | Sr<br>ppm | <sup>87</sup> Sr/ <sup>86</sup> Sr |
|---------|-----------|----------|-----------------|------------|---------------------|--------|-----------|---|---------|---|-------|---|-------|---|-------|---|-------|-----------|------------------------------------|
| England | Ilchester |          | Lady in<br>lead | M2         | YA                  | Female |           | 18.4100                                 | 0.004   | 15.6365                                 | 0.005 | 38.4008                                 | 0.006 | 0.8494                                  | 0.002 | 2.0859                                  | 0.003 | -         | 0.7089                             |
| England | York      |          | -               | M2         | MA                  | Male   |           | 18.5862                                 | 0.005   | 15.6633                                 | 0.006 | 38.6931                                 | 0.007 | 0.8427                                  | 0.002 | 2.0818                                  | 0.003 | -         | 0.7109                             |
| France  | Caen      | Michelet | S359            | U.L.M2     | YA                  | Male   | 0.39      | 18.2769                                 | 0.011   | 15.6245                                 | 0.012 | 38.3335                                 | 0.013 | 0.8549                                  | 0.003 | 2.0974                                  | 0.006 | 46        | 0.7095                             |
| France  | Caen      | Michelet | S690            | L.R.M2     | YA                  | Male   | 24.30     | 18.4254                                 | 0.005   | 15.6369                                 | 0.005 | 38.4180                                 | 0.006 | 0.8487                                  | 0.001 | 2.0851                                  | 0.006 | 57        | 0.7092                             |
| France  | Caen      | Michelet | S132            | U.L.M3     | MA                  | Male   | 8.02      | 18.4091                                 | 0.004   | 15.6350                                 | 0.006 | 38.3987                                 | 0.006 | 0.8493                                  | 0.002 | 2.0859                                  | 0.006 | 157       | 0.7108                             |
| France  | Caen      | Michelet | S142            | L.R.M2     | OA                  | Male   | 0.28      | 18.2705                                 | 0.017   | 15.6445                                 | 0.020 | 38.3524                                 | 0.024 | 0.8563                                  | 0.007 | 2.0992                                  | 0.011 | 246       | 0.7086                             |
| France  | Caen      | Michelet | S831            | U.L.PM2    | OA                  | Male   | 7.86      | 18.4315                                 | 0.004   | 15.6394                                 | 0.006 | 38.4375                                 | 0.006 | 0.8485                                  | 0.002 | 2.0854                                  | 0.006 | 104       | 0.7091                             |
| France  | Caen      | Michelet | S335            | L.L.M2     | OA                  | Male   | 1.06      | 18.3655                                 | 0.005   | 15.6322                                 | 0.006 | 38.3711                                 | 0.007 | 0.8512                                  | 0.002 | 2.0893                                  | 0.006 | 98        | 0.7096                             |
| France  | Caen      | Michelet | S745            | L.R.PM2    | OA                  | Male   | 1.78      | 18.4516                                 | 0.013   | 15.6592                                 | 0.012 | 38.5122                                 | 0.014 | 0.8487                                  | 0.005 | 2.0873                                  | 0.007 | 110       | 0.7129                             |
| France  | Caen      | Michelet | S48             | U.L.M2     | YA                  | Female | 16.90     | 18.4115                                 | 0.004   | 15.6302                                 | 0.006 | 38.4014                                 | 0.007 | 0.8489                                  | 0.002 | 2.0858                                  | 0.006 | 109       | 0.7086                             |
| France  | Caen      | Michelet | S365            | U.R.M3     | YA                  | Female | 2.16      | 18.3872                                 | 0.007   | 15.6319                                 | 0.008 | 38.3816                                 | 0.008 | 0.8502                                  | 0.002 | 2.0875                                  | 0.006 | 69        | -                                  |
| France  | Caen      | Michelet | S376            | L.R.PM2    | MA                  | Female | 1.88      | 18.3854                                 | 0.008   | 15.6311                                 | 0.009 | 38.3779                                 | 0.009 | 0.8502                                  | 0.002 | 2.0874                                  | 0.006 | 74        | -                                  |
| France  | Caen      | Michelet | S854            | U.L.PM2    | MA                  | Female | 0.70      | 18.2373                                 | 0.007   | 15.6300                                 | 0.007 | 38.3946                                 | 0.008 | 0.8570                                  | 0.002 | 2.1053                                  | 0.006 | 49        | 0.7094                             |
| France  | Caen      | Michelet | S405            | U.L.PM2    | MA                  | Female | 0.93      | 18.2933                                 | 0.019   | 15.6422                                 | 0.016 | 38.3564                                 | 0.021 | 0.8551                                  | 0.015 | 2.0968                                  | 0.020 | 130       | 0.7116                             |
| France  | Caen      | Michelet | S762            | L.L.PM2    | OA                  | Female | 2.55      | 18.4220                                 | 0.005   | 15.6375                                 | 0.006 | 38.4198                                 | 0.006 | 0.8488                                  | 0.002 | 2.0856                                  | 0.006 | 97        | 0.7094                             |
| France  | Caen      | Michelet | S394            | L.LM2      | OA                  | Female | 0.78      | 18.1295                                 | 0.008   | 15.6151                                 | 0.008 | 38.4067                                 | 0.009 | 0.8613                                  | 0.002 | 2.1186                                  | 0.005 | 65        | -                                  |
| France  | Caen      | Michelet | S830            | U.L.M2 (d) | 7-12yrs             | Ι      | 3.28      | 18.4301                                 | 0.005   | 15.6408                                 | 0.006 | 38.4500                                 | 0.007 | 0.8487                                  | 0.002 | 2.0863                                  | 0.006 | 76        | -                                  |
| France  | Caen      | Michelet | S830            | U.L.PM2    | 7-12yrs             | Ι      | 1.27      | 18.4205                                 | 0.012   | 15.6395                                 | 0.012 | 38.4353                                 | 0.013 | 0.8490                                  | 0.003 | 2.0866                                  | 0.006 | 76        | 0.7092                             |
| Lebanon | Beirut    | MDWR 02  | SK2615          | L.L.PM2    | MA                  | Male   | 3.75      | 18.6080                                 | 0.006   | 15.6793                                 | 0.011 | 38.7796                                 | 0.017 | 0.8426                                  | 0.005 | 2.0841                                  | 0.011 | 62        | 0.7084                             |
| Lebanon | Beirut    | SFI 645  | SK1004          | L.R.PM2    | YA                  | Male   | 0.50      | 18.3056                                 | 0.014   | 15.6484                                 | 0.018 | 38.4231                                 | 0.022 | 0.8548                                  | 0.007 | 2.0990                                  | 0.011 | 139       | 0.7076                             |
| Lebanon | Beirut    | RML 2385 | SK1818          | U.RPM2     | OA                  | Male   | 6.68      | 18.7487                                 | 0.005   | 15.6845                                 | 0.007 | 38.8463                                 | 0.007 | 0.8366                                  | 0.002 | 2.0720                                  | 0.006 | 210       | 0.7086                             |
| Lebanon | Beirut    | RML 2385 | SK2318          | L.R.PM2    | OA                  | Male   | 5.72      | 18.6640                                 | 0.004   | 15.6777                                 | 0.005 | 38.8121                                 | 0.006 | 0.8400                                  | 0.002 | 2.0795                                  | 0.006 | 177       | 0.7088                             |
| Lebanon | Beirut    | MDWR 02  | SK2840          | L.L.M2     | OA                  | Male   | 1.47      | 18.5963                                 | 0.010   | 15.6690                                 | 0.011 | 38.7243                                 | 0.012 | 0.8426                                  | 0.002 | 2.0824                                  | 0.005 | 110       | 0.7084                             |

 Table A2 – Lead and strontium isotope ratios and elemental concentrations from human tooth enamel samples using ICP-MS

| Country | Region     | Site               | Skeleton ID | Tooth     | Age<br>Category | Sex    | Pb<br>ppm | <sup>206</sup> Pb/<br><sup>204</sup> Pb | 2σ<br>% | <sup>207</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>207</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | Sr<br>ppm | <sup>87</sup> Sr/ <sup>86</sup> Sr |
|---------|------------|--------------------|-------------|-----------|-----------------|--------|-----------|---|---------|---|-------|---|-------|---|-------|---|-------|-----------|------------------------------------|
| Lebanon | Beirut     | MDWR 02            | SK2769      | L.R.M2    | YA              | Female | 1.90      | 18.5087                                 | 0.010   | 15.6740                                 | 0.014 | 38.6680                                 | 0.020 | 0.8468                                  | 0.005 | 2.0892                                  | 0.011 | 76        | 0.7086                             |
| Lebanon | Beirut     | MDWR 468           | SK193       | L.R.M2    | YA              | Female | 1.85      | 18.5140                                 | 0.009   | 15.6575                                 | 0.009 | 38.6215                                 | 0.010 | 0.8457                                  | 0.003 | 2.0862                                  | 0.006 | 121       | 0.7086                             |
| Lebanon | Beirut     | SFI 655            | SK506       | U.R.PM2   | MA              | Female | 2.41      | 18.7031                                 | 0.008   | 15.6788                                 | 0.008 | 38.7486                                 | 0.008 | 0.8383                                  | 0.002 | 2.0718                                  | 0.006 | 136       | 0.7087                             |
| Lebanon | Beirut     | ASH 002            | SK431       | U.L.PM2   | OA              | Female | 2.62      | 18.3963                                 | 0.006   | 15.6494                                 | 0.007 | 38.5837                                 | 0.008 | 0.8507                                  | 0.002 | 2.0974                                  | 0.006 | 86        | 0.7084                             |
| Lebanon | Beirut     | ASH 002            | SK456       | L.L.M2    | OA              | Female | 9.71      | 18.5000                                 | 0.010   | 15.6573                                 | 0.011 | 38.6288                                 | 0.012 | 0.8463                                  | 0.002 | 2.0881                                  | 0.005 | 122       | 0.7085                             |
| Lebanon | Beirut     | RML 2385           | SK1606      | U.L.PM2   | YA              | Female | 2.96      | 18.6425                                 | 0.005   | 15.6800                                 | 0.006 | 38.7773                                 | 0.007 | 0.8411                                  | 0.002 | 2.0801                                  | 0.006 | 109       | 0.7087                             |
| Lebanon | Beirut     | RML 2385           | SK2442      | L.R.PM2   | YA              | Male   | 22.60     | 18.7129                                 | 0.004   | 15.6789                                 | 0.005 | 38.8857                                 | 0.006 | 0.8379                                  | 0.002 | 2.0780                                  | 0.005 | 156       | -                                  |
| Lebanon | Beirut     | MDWR 468           | SK145       | L.?.PM2   | MA              | Ι      | 0.56      | 18.5618                                 | 0.011   | 15.6760                                 | 0.015 | 38.6997                                 | 0.020 | 0.8445                                  | 0.006 | 2.0849                                  | 0.011 | 42        | 0.7084                             |
| Lebanon | Beirut     | BCH 740            | SK350       | U.R.PM2   | А               | Ι      | 7.21      | 18.5749                                 | 0.009   | 15.6693                                 | 0.010 | 38.7160                                 | 0.011 | 0.8436                                  | 0.002 | 2.0844                                  | 0.005 | 55        | 0.7084                             |
| Lebanon | Beirut     | BCH 740            | SK611       | L.R.PM2   | А               | Ι      | 1.29      | 18.4819                                 | 0.006   | 15.6688                                 | 0.011 | 38.6228                                 | 0.017 | 0.8478                                  | 0.005 | 2.0898                                  | 0.011 | 64        | 0.7085                             |
| Lebanon | Beirut     | BCH 740            | SK489       | L.L.PM2   | А               | Ι      | 9.83      | 18.6821                                 | 0.009   | 15.6812                                 | 0.010 | 38.8027                                 | 0.011 | 0.8394                                  | 0.002 | 2.0771                                  | 0.004 | 118       | 0.7088                             |
| Lebanon | Beirut     | MDWR 468           | SK144       | U.L.C (d) | 3-5yrs          | Ι      | 14.10     | 18.6378                                 | 0.004   | 15.6717                                 | 0.005 | 38.7744                                 | 0.006 | 0.8408                                  | 0.002 | 2.0804                                  | 0.006 | 103       | 0.7087                             |
| Lebanon | Beirut     | ASH 002            | SK341       | U.L.PM2   | 11-14yrs        | Ι      | 11.58     | 18.7110                                 | 0.009   | 15.6817                                 | 0.010 | 38.8492                                 | 0.011 | 0.8381                                  | 0.002 | 2.0763                                  | 0.004 | 192       | 0.7087                             |
| Lebanon | Beirut     | BCH 740            | SK489       | L.L.PM2   | 12yrs           | Ι      | 4.98      | 18.6782                                 | 0.005   | 15.6772                                 | 0.006 | 38.7691                                 | 0.006 | 0.8393                                  | 0.002 | 2.0756                                  | 0.005 | 128       | 0.7086                             |
| Lebanon | Beirut     | ASH 163            | SK83        | U.R.M2    | YA              | Female | 3.51      | 18.6522                                 | 0.009   | 15.6720                                 | 0.010 | 38.8070                                 | 0.011 | 0.8402                                  | 0.002 | 2.0806                                  | 0.004 | 135       | 0.7086                             |
| Lebanon | Beirut     | BCH 740            | SK445       | U.L.M2    | Adoles.         | Ι      | 6.66      | 18.6869                                 | 0.004   | 15.6781                                 | 0.005 | 38.8600                                 | 0.006 | 0.8390                                  | 0.001 | 2.0795                                  | 0.006 | 116       | 0.7087                             |
| Romania | Alba Iulia | Dealul<br>Furcilor | M72         | L.R.PM2   | MA              | Male   | 4.06      | 18.5574                                 | 0.009   | 15.6572                                 | 0.010 | 38.6648                                 | 0.011 | 0.8437                                  | 0.002 | 2.0836                                  | 0.004 | 99        | 0.7094                             |
| Romania | Alba Iulia | Dealul<br>Furcilor | M160b       | U.L.M2    | MA              | Male   | 0.23      | 18.1533                                 | 0.011   | 15.6228                                 | 0.014 | 38.4201                                 | 0.019 | 0.8606                                  | 0.006 | 2.1165                                  | 0.011 | 54        | 0.7098                             |
| Romania | Alba Iulia | Dealul<br>Furcilor | M172        | L.R.M3    | MA              | Male   | 0.80      | 18.5535                                 | 0.006   | 15.6622                                 | 0.011 | 38.6517                                 | 0.017 | 0.8442                                  | 0.005 | 2.0833                                  | 0.011 | 73        | 0.7106                             |
| Romania | Alba Iulia | Dealul<br>Furcilor | M178        | L.L.M2    | Ι               | Male   | 8.29      | 18.6531                                 | 0.007   | 15.6652                                 | 0.005 | 38.7748                                 | 0.006 | 0.8398                                  | 0.002 | 2.0788                                  | 0.004 | 103       | 0.7091                             |
| Romania | Alba Iulia | Dealul<br>Furcilor | M42         | U.L.M2    | OA              | Male   | 2.87      | 18.6003                                 | 0.006   | 15.6516                                 | 0.006 | 38.6549                                 | 0.007 | 0.8415                                  | 0.002 | 2.0782                                  | 0.004 | 84        | 0.7097                             |
| Romania | Alba Iulia | Dealul<br>Furcilor | M141        | L.R.PM2   | MA              | Female | 2.41      | 18.5758                                 | 0.006   | 15.6520                                 | 0.005 | 38.6594                                 | 0.005 | 0.8426                                  | 0.001 | 2.0812                                  | 0.004 | 95        | 0.7109                             |
| Romania | Alba Iulia | Dealul<br>Furcilor | M160        | U.L.M2    | MA              | Female | 4.69      | 18.6091                                 | 0.007   | 15.6593                                 | 0.006 | 38.7111                                 | 0.006 | 0.8415                                  | 0.002 | 2.0802                                  | 0.004 | 133       | 0.7098                             |

| Country  | Region      | Site               | Skeleton ID | Tooth         | Age<br>Category | Sex    | Pb<br>ppm | <sup>206</sup> Pb/<br><sup>204</sup> Pb | 2σ<br>% | <sup>207</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>207</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | Sr<br>ppm | <sup>87</sup> Sr/ <sup>86</sup> Sr |
|----------|-------------|--------------------|-------------|---------------|-----------------|--------|-----------|---|---------|---|-------|---|-------|---|-------|---|-------|-----------|------------------------------------|
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M86         | U.R.PM2       | OA              | Female | 1.15      | 18.3408                                 | 0.017   | 15.6520                                 | 0.018 | 38.4601                                 | 0.023 | 0.8534                                  | 0.010 | 2.0970                                  | 0.015 | 93        | 0.7098                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M111        | U.L.PM2       | Ι               | Female | 0.28      | 18.5571                                 | 0.011   | 15.6695                                 | 0.015 | 38.6615                                 | 0.020 | 0.8444                                  | 0.005 | 2.0834                                  | 0.011 | 78        | 0.7097                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M59         | L.R.M3        | OA              | Ι      | 0.25      | 18.5308                                 | 0.023   | 15.6651                                 | 0.024 | 38.6311                                 | 0.024 | 0.8454                                  | 0.004 | 2.0847                                  | 0.005 | 108       | 0.7094                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M26         | U.R.I1        | 7-12yrs         | Ι      | 1.19      | 18.4373                                 | 0.008   | 15.6534                                 | 0.012 | 38.6178                                 | 0.018 | 0.8490                                  | 0.005 | 2.0946                                  | 0.011 | 96        | 0.7092                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M26         | L.R.PM2       | 7-12yrs         | Ι      | 0.85      | 18.5427                                 | 0.007   | 15.6640                                 | 0.012 | 38.6744                                 | 0.018 | 0.8448                                  | 0.005 | 2.0857                                  | 0.011 | 114       | 0.7091                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M10         | L.R.M2<br>(d) | 7-12yrs         | Ι      | 0.31      | 18.5102                                 | 0.012   | 15.6638                                 | 0.015 | 38.6222                                 | 0.020 | 0.8462                                  | 0.005 | 2.0866                                  | 0.011 | 74        | 0.7113                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M10         | L.R.PM2       | 7-12yrs         | Ι      | 0.15      | 18.3879                                 | 0.009   | 15.6438                                 | 0.013 | 38.6045                                 | 0.018 | 0.8508                                  | 0.005 | 2.0995                                  | 0.010 | 151       | 0.7114                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M125?       | U.L.M2<br>(d) | 7-12yrs         | Ι      | 1.90      | 18.5585                                 | 0.005   | 15.6664                                 | 0.011 | 38.7037                                 | 0.017 | 0.8442                                  | 0.005 | 2.0855                                  | 0.011 | 101       | 0.7097                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M125?       | U.L.PM2       | 7-12yrs         | Ι      | 1.61      | 18.6465                                 | 0.006   | 15.6642                                 | 0.005 | 38.7497                                 | 0.005 | 0.8401                                  | 0.002 | 2.0782                                  | 0.004 | 121       | 0.7093                             |
| Romania  | Alba Iulia  | Dealul<br>Furcilor | M7          | U.L.M2        | 13-18yrs        | Ι      | 0.66      | 18.4202                                 | 0.009   | 15.6572                                 | 0.012 | 38.5038                                 | 0.018 | 0.8500                                  | 0.006 | 2.0903                                  | 0.011 | 130       | 0.7105                             |
| Scotland | Musselburgh | PHCC               | PPCM235     | PM2           | MA              | Male   | 1.82      | 18.3372                                 | 0.005   | 15.6376                                 | 0.005 | 38.4341                                 | 0.006 | 0.8528                                  | 0.002 | 2.0961                                  | 0.003 | 124       | 0.71411                            |
| Scotland | Musselburgh | PHCC               | PPCM316     | PM2           | MA              | Male   | 0.61      | 18.4663                                 | 0.011   | 15.6367                                 | 0.006 | 38.5100                                 | 0.007 | 0.8468                                  | 0.002 | 2.0855                                  | 0.003 | 125       | 0.71403                            |
| Scotland | Musselburgh | PHCC               | PPCM323     | PM2           | YA              | Male   | 0.35      | 18.5377                                 | 0.005   | 15.6528                                 | 0.012 | 38.4793                                 | 0.013 | 0.8443                                  | 0.003 | 2.0757                                  | 0.006 | 220       | 0.70896                            |
| Scotland | Musselburgh | PHCC               | PPCM630     | PM2           | MA              | Male   | 6.57      | 18.4395                                 | 0.004   | 15.6321                                 | 0.005 | 38.4515                                 | 0.006 | 0.8478                                  | 0.001 | 2.0854                                  | 0.006 | 160       | 0.70980                            |
| Scotland | Musselburgh | PHCC               | PPCM420     | M2            | YA              | Male   | 2.17      | 18.3821                                 | 0.017   | 15.6296                                 | 0.006 | 38.3994                                 | 0.006 | 0.8503                                  | 0.002 | 2.0891                                  | 0.006 | 45        | 0.71245                            |
| Scotland | Musselburgh | PHCC               | PPCM451     | M2            | MA              | Male   | 2.20      | 18.3783                                 | 0.004   | 15.6313                                 | 0.020 | 38.3900                                 | 0.024 | 0.8505                                  | 0.007 | 2.0890                                  | 0.011 | 79        | 0.71397                            |
| Slovenia | Ljubljana   | Emonske            | JM02 (8)    | L.L.M3        | -               | Ι      | 0.40      | 18.5349                                 | 0.010   | 15.6721                                 | 0.014 | 38.632                                  | 0.019 | 0.84555                                 | 0.005 | 2.0843                                  | 0.011 | -         | -                                  |
| Slovenia | Ljubljana   | Emonske            | JM03 (57)   | L.L.M3        | -               | Ι      | 509.0     | 18.5704                                 | 0.004   | 15.6800                                 | 0.005 | 38.799                                  | 0.006 | 0.84436                                 | 0.002 | 2.0893                                  | 0.003 | -         | -                                  |
| Slovenia | Ljubljana   | Emonske            | JM04        | L.L.M2        | -               | Ι      | 6.43      | 18.5465                                 | 0.008   | 15.6671                                 | 0.007 | 38.730                                  | 0.009 | 0.84477                                 | 0.002 | 2.0882                                  | 0.005 | -         | -                                  |
| Slovenia | Ljubljana   | Emonske            | JM06 (4)    | U.R.M2        | -               | Ι      | 0.23      | 18.4976                                 | 0.010   | 15.6669                                 | 0.013 | 38.627                                  | 0.019 | 0.84697                                 | 0.005 | 2.0882                                  | 0.011 | -         | -                                  |
| Slovenia | Ljubljana   | Emonske            | JM07 (9)    | L.L.M3        | -               | Ι      | 0.22      | 18.4636                                 | 0.014   | 15.6627                                 | 0.017 | 38.568                                  | 0.021 | 0.84831                                 | 0.006 | 2.0889                                  | 0.011 | -         | -                                  |
| Spain    | Barcelona   | Santa<br>Caterina  | Т3          | L.R.M2        | Ι               | Male   | 3.90      | 18.2457                                 | 0.006   | 15.6265                                 | 0.005 | 38.4548                                 | 0.006 | 0.8565                                  | 0.002 | 2.1076                                  | 0.004 | 98        | 0.7088                             |

| Country | Region    | Site              | Skeleton  | ID Tooth | Age<br>Category | Sex    | Pb<br>ppm | <sup>206</sup> Pb/<br><sup>204</sup> Pb | 2σ<br>% | <sup>207</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>207</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | Sr<br>ppm | <sup>87</sup> Sr/ <sup>86</sup> Sr |
|---------|-----------|-------------------|-----------|----------|-----------------|--------|-----------|---|---------|---|-------|---|-------|---|-------|---|-------|-----------|------------------------------------|
| Spain   | Barcelona | Santa<br>Caterina | a T9      | U.R.PM2  | Ι               | Male   | 0.69      | 18.3692                                 | 0.011   | 15.6697                                 | 0.014 | 38.5091                                 | 0.019 | 0.8530                                  | 0.005 | 2.0964                                  | 0.011 | 111       | 0.7096                             |
| Spain   | Barcelona | Santa<br>Caterina | T12       | U.L.M3   | Ι               | Male   | 3.75      | 18.4781                                 | 0.009   | 15.6648                                 | 0.010 | 38.6225                                 | 0.010 | 0.8477                                  | 0.002 | 2.0902                                  | 0.004 | 179       | -                                  |
| Spain   | Barcelona | Santa<br>Caterina | T15 (T4?) | L.R.M3   | Ι               | Male   | 1.73      | 18.4277                                 | 0.009   | 15.6547                                 | 0.010 | 38.5433                                 | 0.010 | 0.8495                                  | 0.003 | 2.0917                                  | 0.004 | 75        | 0.7099                             |
| Spain   | Barcelona | Santa<br>Caterina | T3        | U.L.PM2  | Ι               | Male   | 1.37      | 18.4421                                 | 0.008   | 15.6628                                 | 0.007 | 38.5758                                 | 0.007 | 0.8493                                  | 0.002 | 2.0918                                  | 0.005 | 206       | 0.7082                             |
| Spain   | Barcelona | Santa<br>Caterina | UF371     | U.L.M2   | Ι               | Female | 4.79      | 18.3715                                 | 0.008   | 15.6620                                 | 0.007 | 38.5321                                 | 0.007 | 0.8525                                  | 0.002 | 2.0974                                  | 0.004 | 111       | 0.7097                             |
| Spain   | Barcelona | Santa<br>Caterina | UF755     | U.L.PM2  | Ι               | Female | 1.98      | 18.3906                                 | 0.007   | 15.6534                                 | 0.006 | 38.5296                                 | 0.007 | 0.8511                                  | 0.002 | 2.0951                                  | 0.004 | 147       | 0.7106                             |
| Spain   | Barcelona | Santa<br>Caterina | UF758     | L.R.M2   | Ι               | Female | 2.60      | 18.4065                                 | 0.008   | 15.6600                                 | 0.007 | 38.5446                                 | 0.009 | 0.8508                                  | 0.002 | 2.0942                                  | 0.005 | 102       | 0.7090                             |
| Spain   | Barcelona | Santa<br>Caterina | UF217     | L.L.M2   | Ι               | Female | 12.1      | 18.5962                                 | 0.006   | 15.6658                                 | 0.005 | 38.7397                                 | 0.006 | 0.8424                                  | 0.002 | 2.0832                                  | 0.004 | 240       | 0.7081                             |
| Spain   | Barcelona | Santa<br>Caterina | UF708     | L.R.M2   | Ι               | Female | 1.95      | 18.3977                                 | 0.008   | 15.6582                                 | 0.006 | 38.5330                                 | 0.007 | 0.8511                                  | 0.002 | 2.0945                                  | 0.004 | 230       | 0.7083                             |
| Spain   | Barcelona | Santa<br>Caterina | UF729     | L.L.PM2  | Ι               | Female | 7.28      | 18.4115                                 | 0.007   | 15.6647                                 | 0.006 | 38.5862                                 | 0.008 | 0.8508                                  | 0.002 | 2.0958                                  | 0.005 | 128       | 0.7094                             |
| Spain   | Barcelona | Santa<br>Caterina | T8        | U.L.PM2  | Ι               | Female | 1.57      | 18.4138                                 | 0.013   | 15.6597                                 | 0.014 | 38.5409                                 | 0.014 | 0.8504                                  | 0.003 | 2.0932                                  | 0.005 | 96        | 0.7116                             |
| Spain   | Barcelona | Santa<br>Caterina | UF748     | U.R.M2   | Ι               | Ι      | 2.37      | 18.4222                                 | 0.007   | 15.6368                                 | 0.006 | 38.4283                                 | 0.007 | 0.8488                                  | 0.002 | 2.0860                                  | 0.005 | 123       | 0.7104                             |
| Spain   | Barcelona | Santa<br>Caterina | UF730     | U.R.M2   | 7-12yrs         | Ι      | 1.26      | 18.3449                                 | 0.011   | 15.6545                                 | 0.010 | 38.4875                                 | 0.010 | 0.8533                                  | 0.003 | 2.0980                                  | 0.005 | 79        | 0.7104                             |
| Spain   | Barcelona | Santa<br>Caterina | UF720     | U.L.PM2  | 13-18yrs        | Ι      | 1.91      | 18.4137                                 | 0.007   | 15.6753                                 | 0.012 | 38.5895                                 | 0.018 | 0.8513                                  | 0.006 | 2.0957                                  | 0.011 | 132       | 0.7084                             |
| Spain   | Tarragona | PERI 2            | 1104      | U.R.M2   | YA              | Male   | 18.9      | 18.4480                                 | 0.015   | 15.6670                                 | 0.010 | 38.6240                                 | 0.014 | 0.8493                                  | 0.008 | 2.0937                                  | 0.004 | 251       | 0.7086                             |
| Spain   | Tarragona | PERI 2            | 2129      | U.L.M2   | YA              | Male   | 2.77      | 18.4940                                 | 0.016   | 15.6750                                 | 0.011 | 38.6830                                 | 0.015 | 0.8476                                  | 0.008 | 2.0917                                  | 0.004 | 114       | 0.7096                             |
| Spain   | Tarragona | PERI 2            | 2209      | U.R.PM2  | MA              | Male   | 13.9      | 18.4920                                 | 0.014   | 15.6625                                 | 0.009 | 38.6335                                 | 0.014 | 0.8470                                  | 0.008 | 2.0892                                  | 0.004 | 179       | 0.7085                             |
| Spain   | Tarragona | PERI 2            | 2226      | U.R.PM2  | Ι               | Male   | 2.46      | 18.4610                                 | 0.015   | 15.6670                                 | 0.010 | 38.6290                                 | 0.015 | 0.8487                                  | 0.008 | 2.0925                                  | 0.004 | 166       | 0.7085                             |
| Spain   | Tarragona | PERI 2            | 2413      | U.R.M2   | YA              | Male   | 6.48      | 18.4690                                 | 0.015   | 15.6660                                 | 0.010 | 38.6320                                 | 0.015 | 0.8482                                  | 0.008 | 2.0917                                  | 0.004 | 207       | 0.7085                             |
| Spain   | Tarragona | PERI 2            | 1442      | L.R.M2   | YA              | Male   | 11.2      | 18.4690                                 | 0.015   | 15.6670                                 | 0.009 | 38.6430                                 | 0.014 | 0.8483                                  | 0.008 | 2.0923                                  | 0.004 | 261       | 0.7088                             |
| Spain   | Tarragona | PERI 2            | 5552      | U.R.M2   | MA              | Male   | 8.86      | 18.4560                                 | 0.015   | 15.6700                                 | 0.010 | 38.6410                                 | 0.015 | 0.8490                                  | 0.008 | 2.0937                                  | 0.004 | 170       | 0.7091                             |
| Spain   | Tarragona | PERI 2            | 1215      | L.L.M2   | MA              | Female | 1.85      | 18.4740                                 | 0.015   | 15.6700                                 | 0.010 | 38.6590                                 | 0.015 | 0.8482                                  | 0.008 | 2.0926                                  | 0.004 | -         | 0.7090                             |

| Country | Region    | Site   | Skeleton<br>ID | Tooth  | Age<br>Category | Sex    | Pb<br>ppm | <sup>206</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>207</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>204</sup> Pb | 2σ %  | <sup>207</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | <sup>208</sup> Pb/<br><sup>206</sup> Pb | 2σ %  | Sr<br>ppm | <sup>87</sup> Sr/ <sup>86</sup> Sr |
|---------|-----------|--------|----------------|--------|-----------------|--------|-----------|---|-------|---|-------|---|-------|---|-------|---|-------|-----------|------------------------------------|
| Spain   | Tarragona | PERI 2 | 1035           | U.L.M2 | MA              | Female | 10.2      | 18.2850                                 | 0.015 | 15.6330                                 | 0.010 | 38.4390                                 | 0.015 | 0.8550                                  | 0.008 | 2.1022                                  | 0.004 | 162       | 0.7091                             |
| Spain   | Tarragona | PERI 2 | 1069           | L.R.M2 | YA              | Female | 3.55      | 18.4700                                 | 0.015 | 15.6670                                 | 0.009 | 38.6320                                 | 0.015 | 0.8482                                  | 0.008 | 2.0916                                  | 0.004 | 121       | 0.7091                             |
| Spain   | Tarragona | PERI 2 | 1125/2         | U.R.M2 | MA              | Female | 20.3      | 18.2495                                 | 0.015 | 15.6205                                 | 0.009 | 38.3935                                 | 0.014 | 0.8559                                  | 0.008 | 2.1038                                  | 0.004 | 191       | 0.7090                             |
| Spain   | Tarragona | PERI 2 | 5400           | U.L.M3 | MA              | Female | 15.3      | 18.4500                                 | 0.015 | 15.6670                                 | 0.009 | 38.6240                                 | 0.015 | 0.8492                                  | 0.008 | 2.0934                                  | 0.004 | 222       | 0.7088                             |