REGIONAL INSIGHTS INTO CHILDHOOD LEAD EXPOSURE FROM HISTORICAL NORTH AMERICAN SKELETAL REMAINS

By

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ABSTRACT

Lead (Pb) concentration and isotope analyses of human remains have been used to examine differences in Pb exposure to examine the movement of peoples and Pb-containing cultural materials. The thesis reviewed 55 bioarchaeological studies of Pb concentrations and isotope analyses of teeth and identified limitations in the archaeological, ethnographic, and historical documentation, and variability in tooth choice and analytical approach that complicate transparency and interpretation. Early fishing settlements (17th to 19th century) in Newfoundland and Nova Scotia lacked local anthropogenic Pb exposure. Pb concentrations in tooth enamel of 46 individuals from St. Paul's Anglican Church (Harbour Grace), Foxtrap-2 (Foxtrap), Wester Point (Portugal Cove-St. Philip's), Tors Cove, St. Luke's Anglican Church (Placentia), and Block-3 (Louisbourg) cemeteries ranged from 0.1 to 28.9 ppm. These were compared to 19 individuals associated with the mid-18th-century St. John's Royal Naval cemetery, Newfoundland who showed levels that ranged from 0.2 to 24.6 ppm. These levels showed that childhood exposures to Pb in these early fishing communities was as much as 20 times higher than expected, suggesting anthropogenic Pb sources were available in the absence of local mining.

Although there were many potential imported Pb-containing products available to the fishers, ceramic materials were known to have a Pb glaze and are frequently found on archaeological sites. Ferryland and Anse à Bertrand were associated with English and French fisheries, respectively, and Pb concentrations of 23 ceramic glazes analyzed from these sites ranged from 28.3% to 58.9% Pb. Pb isotope ratios in both the ceramic glazes

and human tooth enamel showed a strong association with isotope ratios of English/Welsh and Western European ore sources during the 17th to 18th centuries. However, individual outliers, including one born in Newfoundland, showed evidence of exposure to American Pb ore. The "de-focusing" of values in the 19th century as American Pb-containing products were exported highlight challenges with associating isotope ratios to cultural affiliation. This study contributes to understanding the roles of the physical versus the cultural environments in Pb exposure and emphasizes the importance of considering contributions from both environmental and cultural sources when interpreting exposure data, migration, and cultural histories.

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Manuscripts and Published Reports

Scott, A., MacInnes, S., **Munkittrick**, **T.J.A.**, Garlie, M., & Grimes, V. (in review). A "foggy, desolate death trap": Exploring the French and English experiences of the 18th century Fortress of Louisbourg, Canada. In M. Danforth (Ed.), *Bioarchaeology of New France*.

Grimes, V., Lear, M., **Munkittrick**, **J.**, & Lacy, R. (2018) Excavation and preliminary analysis of a historical burial ground at Foxtrap-2 (CjAf-10). Foxtrap, Newfoundland. In S. Hull & M. Delphina (Eds.), *Provincial Archaeology Office Annual Review for 2017 Field Season* (pp. 131-140). Provincial Archaeology Office Department of Tourism, Culture, Industry and Innovation Government of Newfoundland and Labrador.

Munkittrick, T.J.A. (2016) The Southside Cemetery (CjAe-54) and its Association with the St. John's Naval Hospital. *Archaeology Review: Provincial Archaeology Office* 14:142-147.

Rand, A., Bower, M., **Munkittrick**, **J.**, Harris, A., Burchell, M., & Grimes, V. (2015) Comparison of three bone collagen extraction procedures: the effect of preservation on δ^{13}C and δ^{15}N values. *North Atlantic Archaeology Journal 4*, 93-114.

Conference Papers

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Scott, A., MacInnes, S., Pitcher, D., Grimes, V., **Munkittrick**, T.J.A., Garlie, M., Moran, M., & Fonzo, M. (2019). *The Rochefort Point Cemetery: an interdisciplinary approach to understanding mortuary patterns and cemetery composition at the 18th century Fortress of Louisbourg*. Paper presented at the 47th Annual Meeting of the Canadian Association of Physical Anthropology.

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Munkittrick, T.J.A., Grimes, V., & Scott, A. (2017). *Investigating childhood lead exposure of early 18th century French inhabitants from the Fortress of Louisbourg, NS.* Paper presented at the 46th Annual Meeting of Canadian Association for Physical Anthropology.

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Conference Posters

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Swanston, T, Coulthard, I., Murphy, R., Cooper, D., Grimes, V., **Munkittrick, T.J.A., T.J.A.,** Jankauskas, R., & Varney, T. (2017). *Confirming bone diagenesis via high resolution synchrotron imaging of lead*. Poster presented at the 46th Annual Meeting of Canadian Association for Physical Anthropology.

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Rand, A., **Munkittrick**, **T.J.A.**, Harris, A., Bower, M., Burchell, M., & Grimes, V. (2014). *Comparing three collagen extraction procedures for stable carbon and nitrogen isotope analyses of archaeological and modern Bone*. Poster presented at the 42nd Annual Meeting of the Canadian Association of Physical Anthropology.

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1 THESIS INTRODUCTION

1.1 Overview of the Project

Lead (Pb) is a highly toxic substance to humans, even in low amounts, and its poisonous effects have been recognized since at least the 2nd century BCE in Greece (Needleman, 2004). There is a long history of examining past Pb exposure using Pb trace element and isotope ratio data in archaeological remains, which reflect the amount and sources of exposure (e.g., Jaworowski, 1968; Grandjean and Holma, 1973; Waldron et al., 1979). Researchers have used this data to examine the changes in the amount of Pb exposure over time in different populations, as well as the source of the Pb to infer the mobility of people and or cultural affiliations (e.g., Jaworowski et al., 1985; Carlson, 1996; Budd et al., 2004; Millard et al., 2014; Price et al., 2017). These topics were originally examined using bone (Jarcho, 1964; Jaworowski, 1968), which represents the last ~15–30 years before death, depending on the bone (O'Flaherty, 1998:1498). However, concerns regarding diagenetic alteration of *in vivo* bone Pb originally addressed in the 1980s (Waldron et al., 1979; Aufderheide et al., 1985; Patterson et al., 1987) remains unresolved (Swanston et al., 2012; Choudhury et al., 2016, 2017). Instead, research has shifted to tooth enamel, a denser and more highly mineralized dental tissue. Tooth enamel forms during childhood and does not alter (Budd et al., 2000a; Hillson, 2014). Being resistant to diagenetic alteration due to high crystallinity and small pore spaces, makes it a reliable in-vivo record of an individual's interactions with their environments (Budd et al., 1998). Subsequently, this tissue presents an intriguing means of examining movement and sources of childhood Pb.

There is a growing literature on Pb exposure in medieval and post-medieval populations that provide valuable information regarding childhood health and mobility (i.e., Schroeder et al., 2013; Millard et al., 2014; Price et al., 2017; Walser et al., 2019; Quinn et al., 2020; King et al., 2021). These studies suggest that childhood Pb exposure was high and originated from common geological sources during these periods, allowing researchers to identify migrants into or between cultural and geographic regions (Montgomery et al., 2005). These techniques are increasingly applied in North American populations during both pre-contact and historic periods (i.e., Fitch et al., 2012; Keller et al., 2016; Aronsen et al., 2019; Laffoon et al., 2020; Samuelson and Potra, 2020; Keenleyside et al., 2021). However, it remains mostly focused on changes in Pb exposure and sources of Pb in pre-contact Indigenous populations over time or due to migration (Patterson et al., 1991; Carlson, 1996; Farell et al., 2013; Dudás et al., 2016; Jones et al., 2017; Samuelson and Potra, 2020), with the understanding being that Pb isotope ratios would likely be associated with the underlying geology in these presumed non-anthropogenic contexts. Conversely, migration events in 19th century industrial contexts are also of interest (Fitch et al., 2012; Keller et al., 2016; Aronsen et al., 2019; Keenleyside et al., 2021), as is the forced migration and life histories of enslaved or free individuals of African descent (Bellis, 2009; Schroeder et al., 2013; Laffoon et al., 2020; Quinn et al., 2020). In these anthropogenic Pb contexts, North American born individuals would have high Pb exposure that is associated with local Pb ore or other local environmental sources. However, it remains unclear how effective such applications of Pb exposure and sourcing would be in pre-industrial Northeastern North American contexts.

Teeth are used widely in archaeology to examine such themes as mobility through strontium ($^{87}Sr/^{86}Sr$) and/or oxygen ($\delta^{18}O$) isotope analyses (e.g., Evans et al., 2012), or childhood diet using carbon isotopes ($\delta^{13}C$) in enamel carbonate or the bone collagen from dentine (e.g., France et al., 2014; Beaumont et al., 2015). There are recent applications of these techniques to 17th- to 19th-century European descent settlements in North America and Oceania, to explore questions regarding migration, diet, health of populations, and individual life histories (see: King et al., 2020). Pb trace element and isotope analyses can also provide insight into mobility in past populations, yet there are very few studies that use these techniques with tooth enamel, particularly in North American contexts.

To achieve this insight, it is necessary to have data regarding the origins of these individuals. However, many Western European and Northeastern North American regions have overlapping expected isotope values, particularly of $\delta^{18}O$ and $^{87}Sr/^{86}Sr$ (see Munkittrick et al., 2019; Scott et al., 2023). As stated, Pb isotope ratios have been used with varied success to address such concerns (e.g., Fitch et al., 2012; Keller et al., 2016; Aronsen et al., 2019). However, the applicability of such techniques given the frequent trade of Pb-containing cultural materials should be examined.

North Atlantic fisheries are an interesting context in which to explore Pb exposure. European fishers in North America were highly focused on the fishing industry, so did not participate in activities that contributed significantly to environmental Pb pollution. This minimal environmental pollution can further be inferred from the lack of bioavailable Pb within environmental archives. Lake cores from the St. John's area were dated and analyzed for heavy metals, including Pb concentration and $^{206}Pb/^{207}Pb$ values

(Christopher, 1999). Christopher (1999) found an increase of Pb pollution in the lake cores immediately around the harbour which he associated with burning imported coal for heat which began in about 1800, and the concentration increased significantly in 1845 with industrial use. Since this increase was not seen until the mid 1800s and inhalation of coal smoke is not expected to be a significant source of Pb due to the size of the particle emitted and has been shown not to be a significant contributor in industrial urban English contexts (Millard et al., 2014), this is an unlikely source of Pb exposure to these populations. In turn, while low levels of exposure from local unpolluted environment would occur, their only means of exposure would be from imported Pb and Pb-containing materials. One such example that contributed to anthropogenic Pb exposure and are ubiquitous in archaeological collections are Pb-glazed ceramics that were imported from production sites in Europe and American colonies/states.

This study will explore both Pb concentration and isotope ratios of individuals from the 18th to mid-19th centuries in Northeastern North America as part of a regional analyses of mobility and insights into the potential influence of trade on childhood Pb exposure. Developing a broader understanding of Pb in teeth will establish the potential for using it as a tool to look at Pb and other contaminants as evidence of trade and mobility in other North American archaeological populations. In adapting established protocols for such analyses, the research related to this dissertation has expanded the capabilities of the Memorial Applied Archaeological Sciences Laboratory to produce meaningful Pb concentration and isotope ratio data that could be utilized in future studies.

1.2 Structure of the Thesis and Research Objectives

This research examined the source and level of Pb exposure during childhood and its connections to biocultural determinants related to where individuals in the populations were raised. The dissertation is divided into five chapters, including a General Introduction (Chapter 1) and a Final Discussion (Chapter 5). Broadly, the thesis addressed the questions: *To what extent were children exposed to Pb in fishing populations during the late-17th to mid-19th centuries and how did that compare to other periods and regions?* And, *Are there differences in Pb sources and pathways between the fishing populations associated with England or France?*

The thesis research chapters are written in manuscript format. Chapter 2 is a literature review of Pb concentration and isotope analyses in tooth enamel to determine how these techniques have been applied, and the challenges and limitations that result from intrinsic factors, and assumptions made during interpretation (Objective 1). Chapter 3 is a comparative application that explores Pb exposure and sources within individuals from early European populations involved with fishing in Newfoundland and Nova Scotia to examine the extent of exposure to children in an otherwise non-Pb-polluted environment (Objective 2). It includes an examination of potential exposure vectors, and whether the origins of the geological Pb that resulted in exposure differed between fishing or other early settlement populations in Northeastern North America (Objective 3). Chapter 4 directly samples Pb-glazed ceramics similar to those available to fishing populations and compares them to Pb isotope values from European and American ore bodies, and English, French and New England populations. This was to determine if Pb within

ceramic glazes was sufficient to result in Pb exposure to those utilizing them, then explore where the Pb used in the glaze originated (Objective 4). Pb isotope ratios in ceramic glazes available in French and English/Irish settlements were compared as a bioavailable range to values within tooth enamel of contemporary fishery communities. The objective was to evaluate how applicable such a method could be relative to using the broader Pb isotope ratios associated with known ore bodies. It also included an examination of the timing of American sourced Pb availability and implications for the applicability of Pb isotopes for migration in an Atlantic World with significant trade and movement of Pb-containing materials (Objective 5).

The dissertation finishes with an Appendix (Section 7) that includes further historical and archaeological background, individual data, and more details on methodological and analytical procedures and results.

1.3 Chapter status

Because this dissertation was written in a manuscript style, the chapters are in various stages of publication, and were written in collaboration with coauthors. The dissertation author was the primary author of the chapters presented within this dissertation and is the primary author of all resulting publications. While the coauthors of Chapters 3 and 4 provided samples, resources, supervision, and comments on previous drafts, the dissertation author was responsible for the research design, sample acquisition and preparation, data interpretation, manuscript drafting and editing the chapters of this dissertation, submitting the resulting manuscripts for review, and incorporating reviewer and coauthor comments into subsequent drafts prior to publication. Details regarding the

coauthors and publication venues for the data presented within this dissertation are provided below.

The research topic in Chapter 2 originated as part of a comprehensive exam essay question provided by Dr. Janet Montgomery (Durham University, U.K.) regarding the use and abuse of Pb in bioarchaeology. The design of the review, sourcing and analysis of papers, and manuscript preparation were all performed by the primary author. Manuscript commentary was provided by co-authors both for initial drafting and after submission of manuscript for review. The incorporation of reviewer and co-author comments into subsequent drafts prior to publication was the responsibility of the primary author.

The research topic and design in Chapter 3 were the conception of the primary author. Samples were provided by The Rooms Corporation, and the Provincial Archaeology Office under the supervision of Dr. Grimes and by Parks Canada under the supervision of Dr. Amy Scott. All tooth sampling and chemical pre-treatment was performed by the primary author. Dr. Lam performed the initial set up and calibration of the Neptune multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS), but sample analyses were performed by the primary author as was the interpretation of the resulting Pb isotope data. Pb concentration analyses were performed by Dr. Nimal da Silva at University of Ottawa, and at Memorial University by Dr. Inês Nobre Silva or an Associate. A portion of the sample dilution and analyses were performed by the primary author to learn the technique but was often performed by the analytical technicians. Data calibration was all performed by technicians, though some additional calibration due to volumetric versus gravimetric mass calculations were necessary and were performed by the primary author. Pb concentration estimations from samples

analyzed on the Neptune MC-ICP-MS were performed using raw data by the primary author.

Drs. Grimes and Scott provided samples and laboratory space for sampling. Dr. Grimes performed initial training on Pb elution techniques and oversaw the protocol establishment for the laboratory. Drs. Grimes and Varney provided funding for analyses. The manuscript was fully drafted by the primary author, as was the integration of comments from each co-author.

The research topic and design of Chapter 4 were both the conception of the primary author. Specific ceramics were chosen from available materials with general directions from the primary author by Dr. Barry Gaulton for the Ferryland, Newfoundland materials and chosen together by the primary author, Dr. Catherine Losier, and fellow graduate students Meghann Livingston and Mallory Champagne for the materials from Anse à Bertrand from Saint-Pierre et Miquelon. Initial sampling using scanning electron microscopy (SEM) was performed by the primary author, but due to analytical malfunctions they could not be present for the laser ablation split-stream (LASS) analysis. This was performed by Drs. Rebecca Lam, Markus Wälle and Vaughan Grimes with direction on location for sampling by the primary author. Initial sample calibration and correction was performed by Dr. Wälle (ICP-MS) and Dr. Lam (MC-ICP-MS).

The additional tooth samples were provided by Provincial Archaeology Office under the supervision of Dr. Grimes and Parks Canada under the supervision of Dr. Amy Scott. Laboratory space and materials were provided by Dr. Grimes. Sampling and pre-treatment of all samples was performed by the primary author, as was the analysis of the Pb isotope data and estimated concentrations. Dr. Lam performed the initial set up and

calibration of the Neptune (MC-ICP-MS), but daily analyses were performed by the primary author. Pb concentration analyses and initial data calibration were performed by Dr. Nobre Silva. Analysis of the resulting data and the manuscript drafting were performed by the primary author. Any comments from coauthors were integrated by the primary author.

2 THE USE AND ABUSE OF PB IN BIOARCHAEOLOGICAL STUDIES: A REVIEW OF PB CONCENTRATION AND ISOTOPE ANALYSES OF TEETH [1]

2.1 Introduction

Over the past 20 years, lead (Pb) has re-emerged as an archaeological tool to improve our understanding of past human movement and the historical extent of pollution, either by the comparison of Pb concentrations (e.g., Budd et al., 2004; Schroeder et al., 2013) or by differences in the Pb isotope ratios of bioavailable Pb in the natural environment (e.g., Montgomery et al., 2000, 2005; Valentine et al., 2008; Fitch et al., 2012; Price et al., 2017). Initial studies examined Pb concentration and isotope ratios in bone (Jarcho, 1964; Jaworowski, 1968; Kowal et al., 1991), but tooth enamel allows researchers to assess childhood exposure while limiting the potential of diagenetic alteration from the burial environment and has become the preferred tissue choice. Diagenesis being the chemical alteration of bone and teeth that occurs from interactions with the burial environment (Kendall et al., 2018). Although there have been reviews on Pb use (Gulson, 2008) and a meta-analysis of modern and archaeological Pb data (Kamenov and Gulson, 2014), a critical assessment of the state of the field is warranted. Environmental and anthropogenic Pb exposure is used in archaeology to examine health, movement, and social dynamics in past populations; however, aspects of the application have also been misused, which weakens the conclusions. This article reviews published research on Pb concentrations

[1] This chapter was published in 2023 within the Journal of Archaeological Science volume 156 as article 105803 (doi:10.1016/j.jas.2023.105803) with Drs. Tamara L. Varney (Dept. Anthropology, Lakehead University) and Vaughan Grimes (Depts. Archaeology and Earth Sciences, Memorial University of Newfoundland) as co-authors.

and isotope analyses of teeth from archaeological populations to identify current trends, limitations due to misuse or insufficient information, fundamental or knowledge-based limitations, and suggests future research. Together, these considerations will improve the Pb analyses' ability to provide insight into childhood exposures, movement, and exchange of cultural materials in the past.

Here, we first give a basic background of Pb research, Pb sources and uptake, and then present a critical review of studies found through a systematic query of research search engines and journals that frequently publish Pb data. The review focuses on how techniques are applied, and the approaches taken for interpretation. The first concern is sample selection and how it affects the interpretation of *in vivo* exposure, and the consideration of the child's experiences during the period of tooth development. The second concern is data interpretation and has two components: a) how the concentration in teeth can be meaningfully related to health issues, the timing of exposure, and population experiences, and b) the relationship between Pb isotope ratios and the characterization of both bioavailable and anthropogenic Pb sources.

2.2 Background to Pb Studies

2.2.1 History of the use of Pb in bioarchaeology

The use of Pb concentration and isotope analyses in a bioarchaeological context developed in three main phases: measuring levels in bone, rising concerns about diagenetic influences, and a switch to using teeth. The first phase occurred in the 1960s to '70s when the initial implementation of Pb trace element analysis focused on bone as the analyte. Some of the main research themes from this period included examining what the

archaeological 'natural' and anthropogenic exposure levels were compared to the modern period. Since atmospheric Pb pollution from leaded-gasoline and paint was a contemporary concern, researchers wanted to understand how modern exposures compared to those in past populations. Comparisons included populations with no known anthropogenic exposure (Jaworowski, 1968; Grandjean and Holma, 1973) and those with at least some anthropogenic Pb exposure (Jarcho, 1964). There was also interest in Romano-British populations since Pb was widely used during that period (Mackie et al., 1975; Waldron et al., 1976). While studies in this first phase hypothesized that exposure was relatively low during periods with no known anthropogenic Pb sources, some studies did show that past exposures could be much higher than those in the modern period (i.e., Jaworowski, 1968; Grandjean and Holma, 1973). However, the Pb levels detected were accepted as accurate, and they did not question if the interpretation of exposures was affected by contamination in the burial environment.

The second development phase occurred in the 1980s and '90s when researchers began recognizing the effects of diagenesis on bone Pb concentrations. Waldron et al. (1979) first questioned the potential for diagenesis to impact the Pb content in bones since they found no correlation between soil Pb concentrations and bone concentrations, suggesting that the values were biogenic. In subsequent research, Waldron (1981; 1983) tested other sites and found that Pb uptake from the soil into bone occurred in some situations and potentially impacted the interpretation of *in vivo* Pb exposures. Studies continued to use bone Pb concentrations but utilized revised methods that were believed to remove the bone's diagenetic material (Aufderheide et al., 1985; Jaworowski et al., 1985; Patterson et al., 1987). However, the success of these methods could be variable,

and studies concluded that bone was unlikely to retain biogenic values, even when treated to remove diagenetic material (Jaworowski et al., 1985; Patterson et al., 1991). This revelation resulted in a drop in the number of bioarchaeological studies that examined Pb in bulk samples of bone.

This second period was also when researchers began to apply these data and methods to examine social differences between populations based on the levels of Pb exposure (Corruccini et al., 1987; Aufderheide et al., 1988). Though it was not widespread, studies incorporated methods such as bone Pb isotope analysis (Kowal et al., 1991; Carlson, 1996) and examined Pb concentrations in teeth (Ericson et al., 1979; Whittaker and Stack, 1984; Grandjean and Jørgenson, 1990; Patterson et al., 1991).

The most recent Pb research phase began in the early 2000s when bioarchaeological research increased tooth enamel use to look at past exposure levels (Figure 1). In parallel, the debate of whether biogenic values of Pb isotope or trace element concentrations are accessible through bone continued and remains a contentious issue, with many arguing that biogenic values are not accessible, as is the case for strontium (Sr) in bone (Millard, 2006; Zapata et al., 2006; Wittmers et al., 2008). However, this is not universally accepted, and bone tissue use continues with limited consideration of diagenesis in some cases (Bower et al., 2007; Rasmussen et al., 2015; Rasmussen et al., 2019; López-Costas et al., 2020). Many papers since 2011, when there was a resurgence of bone Pb studies, are exploring means of accessing biogenic Pb and the role of diagenesis in affecting these values (Swanston et al., 2012, 2018; McMillan et al., 2019, Simpson et al., 2021).

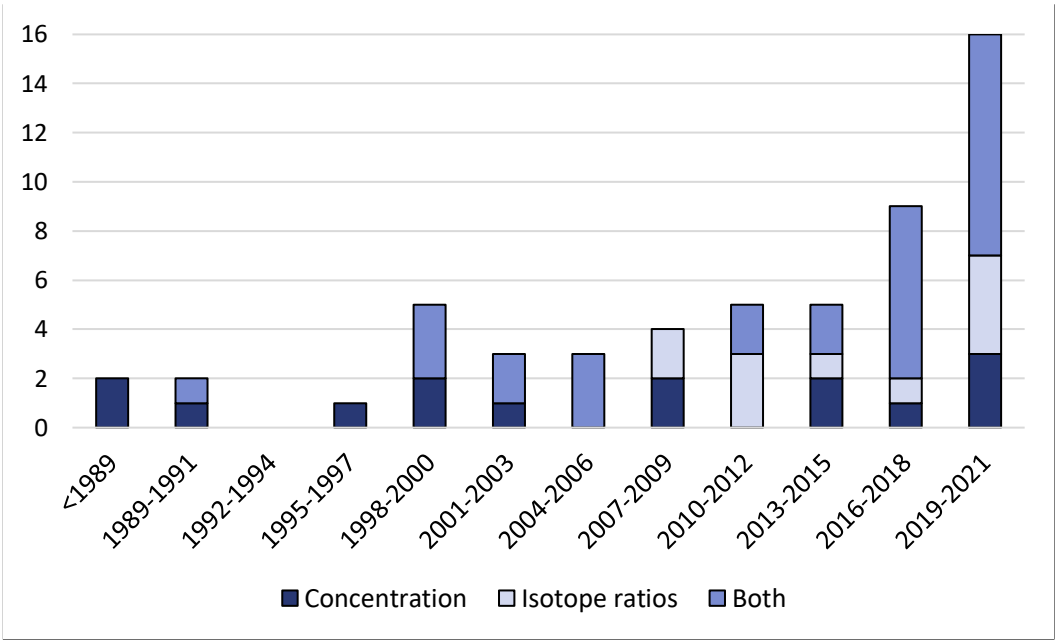

Figure 1. Number of studies using Pb concentration and/or isotope ratios of archaeological teeth per year. Note the increase in studies since 1997 and the start of publications using only Pb isotope ratios in 2008.

While there was a substantial increase in studies that exclusively used tooth enamel during this phase (e.g., Budd et al., 2000a; 2004; Montgomery et al., 2005; 2010; Valentine et al., 2008; 2015), there was also an increase in research using both bone and enamel, particularly in association with Sr isotope analysis to examine migration (Åberg et al., 1998; Budd et al., 2000a; Montgomery et al., 2000). The recent analysis themes include examining changes in Pb exposure and sources of exposure over time, similar to previous approaches involving bone but using teeth as an analyte (Budd et al., 2000a; 2004). There is also a consideration of childhood health using enamel Pb concentrations as a proxy for blood Pb concentrations (Schroeder et al., 2013; Millard et al., 2014; Jones, 2015; Quinn et al., 2020; Moore et al., 2021). Studies are also considering migration, both through differences in Pb concentrations between anthropogenic and natural exposure

regions (Schroeder et al., 2013; Jones, 2015) and by examining differences in Pb isotope ratios (Montgomery et al., 2005; Fitch et al., 2012; Millard et al., 2014; Shaw et al., 2016; Laffoon et al., 2020).

Cultural affiliation is an important area of interest, with studies examining the 'cultural focusing' of Pb isotope ratios that result from an averaging of Pb sources within a population (Montgomery et al., 2005; Millard et al., 2014; Shaw et al., 2016). Some have used this to examine life histories of individuals' health and movement (e.g., Valentine et al., 2008; Fitch et al., 2012; Price et al., 2017; Laffoon et al., 2020). More recently, the stability or inertness of enamel has been questioned, suggesting that diagenetic Pb may be more prevalent in tooth enamel than previously believed (e.g., Beherec et al., 2016; Kamenov et al., 2018; King et al., 2020; Simonetti et al., 2021). From this brief overview of the development of Pb analyses in bioarchaeology, it is clear that Pb is still utilized to address meaningful research questions. However, there are also inconsistencies in how the technique is applied that must be further explored to understand challenges affecting the interpretation of Pb concentration and isotope ratios from archaeological skeletal material.

2.2.2 Sources of Pb

To examine how human skeletal materials are used and misused to address bioarchaeological questions, it is essential first to consider the sources of Pb within humans. Even without anthropogenic contamination, natural processes result in different Pb concentrations and isotope ratios within human tissues. Additionally, physiological stages and cultural influences will also affect the deposition of Pb in skeletal materials.

Understanding these sources of variability allows for questions concerning such themes as movement and Pb toxicity to be investigated.

There are four stable isotopes of Pb typically measured in bioarchaeological analyses: ^{206}Pb, ^{207}Pb, and ^{208}Pb, which are radiogenic, and ^{204}Pb, which is primordial (Faure and Mensing, 2005). The three radiogenic isotopes are the stable daughter isotopes of uranium (^{238}U to ^{206}Pb, ^{235}U to ^{207}Pb) and thorium (^{232}Th to ^{208}Pb) decay chains with different half-lives (Doe, 1970; Faure and Mensing, 2005). The isotopes have different natural abundances, with ^{204}Pb being the lowest at 1.48%, ^{206}Pb and ^{207}Pb are similar with 23.5% and 22.6%, respectively, and ^{208}Pb has the highest abundance at 52.3% (Adriano, 1986; Faure and Mensing, 2005). The Pb isotope half-lives, the varied original isotopic abundances in rock, the age since and any mixing during its formation, all create the variability of Pb isotope ratios of geological formations (Gulson, 1986; Adriano, 1986; Teutsch et al., 2001; Faure and Mensing, 2005). From these formations, there is no measurable isotopic fractionation of Pb as it moves into and through the biosphere (Rabinowitz and Wetherill, 1972; Gulson, 1986), incorporating in the skeletal material of humans where they act as a reservoir of the exposure history.

Pb is contained within the mineral galena (PbS), which was a common Pb source for metal-using societies and was also mined for the silver within galena deposits, in addition to other minerals in lower concentrations (Adriano, 1986; Brännvall et al., 1999). In addition to the parental rock material, various external sources can add to the concentration of soil Pb, including the dry deposition of dust, wet deposition via rainwater, and the deposition of eroded soil through river run-off (Adriano, 1986). Pb has low natural concentrations in water of aquatic systems such as rivers, rain, and ocean

water (Schaule and Patterson, 1981; Bacon and Bain, 1995). The turnover time is brief for river and rainwater (Bacon and Bain, 1995). While this can be 80–100 years for deep oceans (Schaule and Patterson, 1981), it is much shorter than strontium, which has a residence time of more than one million years (Veizer et al., 1997) and is therefore too short to result in global homogeneity. While local rocks are the main source to Pb in soils, these additional sources, which could have disparate Pb isotope ratios can influence the resulting values (Samuelsen and Potra, 2020). The soil, plants, and animals that occur and live within an area constitute the biologically available Pb to humans when there is no anthropogenic exposure (Figure 2).

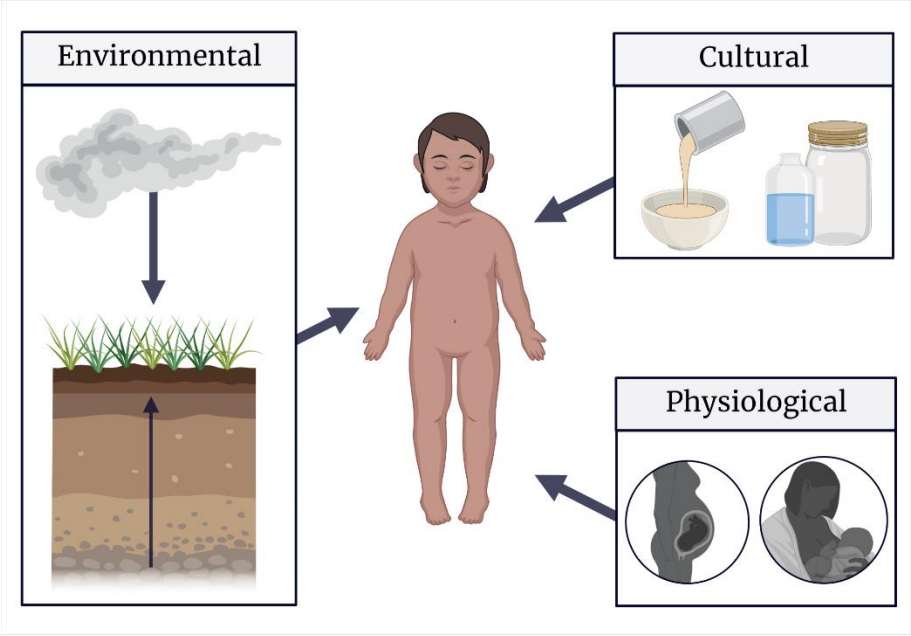

Figure 2. The sources of geogenic and anthropogenic Pb to a human include environmental, cultural material, and physiological. The arrows depict the movement of Pb from the underlying bedrock into the soil and plants, but anthropogenic sources in the atmosphere can swamp the naturally occurring Pb. Food and drink that are stored in Pb-containing materials can leach the Pb resulting in further contamination. Finally, a mother or caregiver's previous Pb can be reabsorbed into the blood and released into a growing fetus or through breastmilk.

Many cultures have processed and incorporated Pb into cultural materials. However, Montgomery et al. (2010) argued that, at least in prehistoric Britain, higher exposure was not associated with a society's ability to mine; instead, the contamination of the food and drink consumed resulted in high Pb exposure levels. Mining and smelting of Pb result in dust and particulate air contamination, which settles onto surfaces and into the soil where it can be ingested or incorporated when food is grown (Figure 2; Gulson et al., 2004). Pb contamination from mining is not as bioavailable as with smelting due in part to the much larger size of the particles (>150 µm) (Steele et al., 1990; Rieuwerts et al., 2000). In turn, within 2 km of modern smelters, bioavailable Pb tends to be much higher, particularly in comparison to mining (Mahaffey, 1978; Rieuwerts et al., 2000). It should be noted that Pb from mining dust is not necessarily the cause of Pb toxicity even within mining contexts as seen in the mining community at Broken Hill, Australia, where paint and/or gasoline were the predominant source of exposure (Gulson et al., 1994). Since it is Pb ions that substitute within biological systems, the specific molecules of the particle, the matrix the Pb is encased in, the pH of soil or water it is deposited into, and their solubility also has an impact on bioavailability (Hemphill et al., 1991; Ram et al., 2021). For example, particulates from wastewater released at these mining and smelting sites may have high particulates, but sulfates are less soluble than Pb oxides so not all of the particulates would be bioavailable (Ghotbizadeh et al., 2022). The mined Pb was often further processed and incorporated into food and drink storage vessels as an alloy or as an oxide into the glaze of ceramics, which then leeches into the food or drink within (e.g., Beagrie, 1989; Tite et al., 1998). Pb could also be consumed inadvertently if Pb shot was not

removed from a harvested animal before preparation and the shot does not pass the gastrointestinal tract (Lévesque et al., 2003; Knott et al., 2010).

2.2.3 Pb uptake by humans

These natural and anthropogenic Pb sources make their way into the biosphere, where the primary means of exposure to humans is through dust, food, or cultural materials. Initial Pb exposure can occur through ingestion, inhalation, and skin contact (Flora et al., 2006). The main pathway of human exposure to Pb is through ingestion since it is often found in natural or contaminated food and drink (Flora et al., 2006). Plants accumulate most of their Pb in the root system via the soil, while the leaves are only a substantial repository of Pb when atmospheric pollution is present (Elias et al., 1982; Adriano, 1986; Sharma and Dubey, 2005). Dust is an additional source for humans, which is consumed, particularly by children, via hand-to-mouth activities (Mushak, 1991; Gulson et al., 2004). Particle size plays a large role in the bioavailability of ingested Pb, with those between 63 μm and 250 μm accounting for most of the exposure (Rieuwerts et al., 2000; Bright et al., 2006). Since the gastrointestinal tracts of children are not fully developed, they absorb and retain a higher proportion of Pb consumed with food (40%) when compared with adults (5–15%) (Mushak, 1991; Flora et al., 2006). Additionally, dietary Pb is more effectively absorbed after fasting or with water (~100% in children and 35–60% in adults), so the medium of exposure impacts the amount of Pb absorbed (Rabinowitz et al., 1980:1788; ATSDR, 2020). Conversely, the consumption of high concentrations of calcium, iron, magnesium, and phosphate can inhibit Pb absorption (Rabinowitz et al., 1980; Gulson et al., 2003).

Exposure via inhalation occurs from the suspended particulates in the air either from natural or anthropogenic sources, such as smelting emissions (Gulson et al., 2004; Flora et al., 2006). The amount of Pb deposited into the respiratory tract varies, but for inorganic Pb it is approximately 25% (Morrow et al., 1980). Of that which is deposited into the bronchial or alveolar regions, particle size has a significant impact on bioavailability. Particles >10 μm are easily expelled from the upper respiratory tract, particle size plays a large role in the bioavailability of inhaled Pb (Hornberg et al., 1998). However, those >2.5 μm are likely expelled and moved out of the bronchia with the mucociliary escalator and, if not fully expelled, are consumed into the gastrointestinal tract (Oberdörster, 1993; James et al., 1994; ATSDR, 2020). While the absorption of Pb deposited into lung alveoli is more effective than the gut (90–95% versus 5–40%, respectively; Hursh et al., 1969; Wells et al., 1975; Flora et al., 2006:166), the concentration of atmospheric Pb is much lower than in food resulting in less exposure via air. It is estimated that 1–3% of Pb in humans comes from inhalation of atmospheric sources (Thornton et al., 1990) and that overall inhalation is a less likely exposure pathway for children than occupationally exposed adults (WHO, 2010; Zhou et al., 2014).

The final pathway of Pb absorption is direct skin exposure. This contribution is typically minimal to an individual's overall Pb burden since the skin does not readily absorb unpowdered Pb compounds (Lilley et al., 1988; Filon et al., 2006; Flora et al., 2006; Pan et al., 2010). However, this likely had a considerable impact on those working with Pb dust from mining, powdered Pb galena or compounds in pottery, or those who practiced body painting. One example is kohl, which can be made with ground galena (lead sulfide, PbS; Tapsoba et al., 2010). While kohl recipes vary and were not always

Pb-based (Riesmeier et al., 2022), modern studies did find that many Egyptian mummies' eye makeup or makeup containers from as early as the 19th century BCE contained Pb (Tapsoba et al., 2010; Riesmeier et al., 2022). The use of kohl would have resulted in some Pb exposure as is shown in modern studies (Al-Ashban et al., 2004). There is also accounts in the 18th and 19th centuries of Pb used as a dye for cloth or to thicken silk fabric, which physicians at the time suggested were a source of acute toxicity (Jonasson & Afshari, 2017).While not a concern for archaeological populations, organic Pb, or tetraethyl Pb, was widely used as a fuel additive from the 1920s until it was phased out in many countries by the mid-1990s and is easily absorbed by the skin (Needleman, 2004).

While tactile, inhalation, and ingestion are initial means of exposure, mothers and caregivers can pass on their previous Pb exposure to fetuses and infants through maternal transfer before birth or via human milk. If mothers or caregivers have insufficient calcium (Ca) in their diet during pregnancy and lactation, bone turnover rates increase, releasing stored Ca and Pb (Gulson et al., 2003). Pb is then released into the bloodstream and can be passed onto the developing fetus or breastfeeding infant. Considering that deciduous teeth form *in utero* and during early infancy, and that some permanent teeth develop while the infant may still be breastfeeding, the source of the Pb in the first teeth formed originates from the mother and/or a caregiver, and potentially remobilized Pb from those individuals (Gulson et al., 2003).

Once incorporated into the body, Pb is absorbed and distributed into three main systems (Rabinowitz et al., 1976). While blood contains a small portion of Pb compared to total body burden, it has a relatively fast turnover rate (36 ± 5 days), making it crucial for transporting Pb throughout the body (Rabinowitz et al., 1976). When Pb exposure is

low, it is present in the red blood cells, but if high exposure occurs, plasma contains the majority of blood Pb (O'Flaherty, 1998; Hernández-Avila et al., 1998; Fleming et al., 1999). Pb is also incorporated into soft tissues with a slightly longer turnover rate than blood and has been reported to be between 30 and 55 days (Rabinowitz et al., 1976). Pb is excreted from the body through feces, urine, and sweat (Rabinowitz et al., 1976; Flora et al., 2006). While soft tissue and blood are important for the absorption of Pb into humans, they do not often survive unless in unique archaeological contexts (O'Connell and Hedges, 1999; Sharp et al., 2003; Finucane, 2007).

Unlike blood and soft tissue, Pb has a prolonged turnover rate in bone, resulting in greater accumulation. Pb is also deposited within teeth; however, Pb in dental tissues makes up a much smaller portion of the skeletal system. There remain uncertainties with how Pb is incorporated into bioapatite, the mineral portion of bone and teeth. While many researchers suggest a simple substitution of Pb at Ca sites in hydroxyapatite (i.e., Verbeeck et al., 1981), Montgomery et al. (2010:211) pointed out that this ignores the chemical dissimilarities and original research by Neuman and Neuman (1958:94–95) who argued that it was not straightforward. Pb is also not uniformly distributed within teeth, with the outer 30 µm of enamel having a significantly higher concentration of Pb than the core enamel (Budd et al., 1998). This spike in concentration is present from the tooth's formation, since unerupted and modern teeth have similar patterns, though some post-eruption addition is still possible (Budd et al., 1998). Dentine has about double the Pb concentration of the core enamel in both modern and archaeological teeth (Budd et al., 1998).

The use of archaeological teeth to examine Pb exposure has increased over the last 20 years. Multiple factors make this tissue a valuable resource to researchers. First, since teeth form at various physiological ages (which are correlated to chronological ages) , multiple periods of exposure can be examined (Gulson and Wilson, 1994; AlQahtani et al., 2010; see Table 1). Additionally, once the enamel forms and matures, the tissue does not remodel, meaning that the concentration and sources of Pb can be used to examine exposure during specific periods of life (Budd et al., 2000a; Hillson, 2014).

Table 1. Timing of the start of tooth formation, crown completion, and eruption of permanent teeth. Based on AlQahtani et al., 2010. *AJPA* 142:481-490

Tooth type	**Initial cusp formation**		**Crown completed with defined pulp roof**		**Occlusal surface in plane**	
	maxilla	*mandible*	*maxilla*	*mandible*	*maxilla*	*mandible*
I1	4.5 mo	7.5 mo	4.5y	3.5 y	7.5 y	7.5 y
I2	7.5 mo	10.5 mo	5.5 y	4.5 y	8.5 y	7.5 y
C	7.5 mo	10.5 mo	5.5 y	5.5 y	12.5 y	10.5 y
P1	2.5 y	2.5 y	6.5 y	6.5 y	11.5 y	11.5 y
P2	3.5 y	3.5 y	6.5 y	6.5 y	12.5 y	12.5 y
M1	4.5 mo	4.5 mo	3.5 y	3.5 y	6.5 y	6.5 y
M2	2.5 y	2.5 y	8.5 y	8.5 y	12.5 y	12.5 y
M3	8.5 y	8.5 y	14.5 y	14.5 y	> 15.5 y	> 15.5 y

Unlike bone, enamel is mainly bioapatite with very little organic content (Hillson, 2014). Enamel has very small pore spaces and the mineral is generally assumed to be highly stable, which would minimize the likelihood of contamination or diagenetic alteration. While this is repeated in Pb research, it should be noted that the foundational studies are not in the same context, with Budd et al. (2000b) examining Sr and

Sponheimer and Lee-Thorp (2006) using fossilized teeth. Though, some studies suggest that enamel may be more prone to diagenesis than previously determined (Beherec et al., 2016; King et al., 2020), particularly in cases where under-mineralized enamel is used (Montgomery, 2002). When properly sampled and vetted for diagenetic alteration, however, the Pb in biological tissues holds a wealth of information about past exposures.

In cases of 'natural' exposure, where minimal or no anthropogenic Pb exposure occurred, humans' Pb isotope ratios will represent an average of the sources consumed (Montgomery et al., 2005:129). This is not the case with anthropogenic Pb. It is essential to consider that there are typically a small number of anthropogenic Pb sources used by a culture at any given time, and all individuals exposed acquire Pb isotope ratios representing those sources; a process that has been named 'cultural focusing' (Montgomery et al., 2005). While Pb isotope analysis can be used to identify the movement or migration of peoples with natural Pb exposure, differences in the isotopic ratios from anthropogenic exposure define cultural affinity (Montgomery et al., 2005).

2.3 Use, Misuse, and Challenges in Pb analyses

This review specifically examines Pb enamel concentration and isotope ratio studies of archaeological human remains that were previously buried. We found journal articles and book chapters using the search engines: Google Scholar, Web of Science, and OneSearch. Search terms included variations of 'Pb isotope', 'Pb concentration', and 'trace element' with the terms 'archaeology', 'tooth', and 'enamel'. Papers were also found in citations from other articles, but dissertations were excluded from the analysis to avoid repetition of material and to allow time for publication. We examined each article

using the same criteria. The article citations and a summarized spreadsheet of the analysis results can be found in supplemental materials (see Appendix Section 7.1).

A total of 55 papers were reviewed (see Appendix Table 8 to Table 16). They were published between 1979 and 2021. These studies examined populations from the Neolithic to the late-19th century from all occupied continents. Of these, 15 examined Pb concentration, 11 Pb isotope ratios, and 29 examined both. The following discussion examines the approaches to sample choice, preparation, and the interpretation of Pb concentration and isotope ratios from the papers reviewed. Each section includes a summary of Pb's use, areas of insufficient information being given or factors influencing conclusions that are overlooked, limitations due to data acquisition or interpretation challenges, and a description of the types of studies needed to help advance this analytical approach.

2.3.1 Sample selection for Pb measurement in teeth

Researchers have used a variety of types and components of teeth to examine Pb exposure. The choice of an initial sample for analysis is key to avoiding potential contamination or interpretation problems. Most (n = 36, 65%) used only the enamel portion of the tooth for the analysis of biogenic Pb or examined the enamel and dentine separately (n = 13, 24%). Nevertheless, some studies did use whole teeth or only dentine (n = 6, 11%). Due to the potential changes in exposure during an individual's childhood and the age at which teeth form, it is crucial to describe which teeth are sampled. Chosen teeth included deciduous teeth (n = 15, 27%), permanent incisors (n = 7, 13%), canines (n = 17, 31%), premolars (n = 23, 42%), and molars (n = 39, 71%). Eleven (20%) did not

specify which type of tooth was sampled. A quarter of studies (n = 14, 25%) examined multiple types of teeth per individual, usually teeth that form at different ages.

Of those that specified which tooth was sampled, more than half (n = 30, 55%) expressed that they systematically chose a specific tooth, but of these, only half (n = 18) made a choice based on the age of the individual when the tooth developed. The teeth preferred most often were second molars (n = 17, 31%) or second premolars (n = 11, 20%), which form (depending on the cultural practices) after breastfeeding had ceased (AlQahtani et al., 2010, see Table 1). Others examined teeth from the earliest available developmental period, such as the first molars (n = 14, 25%), or, less often, the first premolars (n = 5, 9%), incisors (n = 3, 5%), and canines (n = 3, 5%). Some studies specifically chose third molars (n = 6, 11%), which form variably but in later adolescence. Note that the frequencies will not equal 100% since many papers have multiple options of preferred teeth.

There are a few fundamental challenges that arise in sample choice, most of which are common to any isotopic study using teeth. The first is that specific teeth are not always available. This may be due to ante-mortem or post-mortem tooth loss, pathology, taphonomy, previous sampling, or restrictions from the institutions or ancestral groups representing the deceased. Substantial wear or damage can also limit tooth choice and, in some cases, confidence in positive tooth identification. Since it is common to have small sample sizes and a desire to increase the representation of the population, it is sometimes unavoidable to use non-ideal teeth. However, it is essential that studies report which specific tooth was sampled to link it to the analysis and to consider this for any variations during interpretation.

The preservation of potential sample teeth needs to be considered, though only 18 (33%) specified that they were doing so, and less than half of those (n = 8, 44%) defined what that was. Common problems that justify the avoidance of a tooth or areas on a tooth include caries, staining, and cracks because of concerns about contamination after burial (Montgomery et al., 2005; Valentine et al., 2008; Fitch et al., 2012; Shaw et al., 2016). Montgomery (2002) found that poor results of preservation indicators (e.g., surficial staining) did not correlate with the likelihood of the value being diagenetically altered. However, using undermineralized teeth was problematic; this includes using unerupted teeth and enamel close to the cementoenamel junction.

Various components of chosen teeth were measured in the reviewed papers, including enamel, primary dentine, secondary dentine, and cementum. There are multiple studies establishing that dentine may be more likely affected by diagenetic alteration (Carvalho et al., 2007), although more has been published on this topic in regards to Sr studies (e.g., Montgomery et al., 2000; Montgomery, 2002) . While there are studies that suggest at least some dentine samples may be unaltered (Dudás et al., 2016; Guede et al., 2017), there remains a lack of proper examination of this issue regarding Pb. The foundational studies with Sr were using teeth from basic soils, which are more likely to affect Sr than Pb (Montgomery, 2002). How this may translate to acidic soils needs to be examined.

Since 2003, almost all studies have exclusively used enamel to interpret *in vivo* values. Not all explicitly stated that specific teeth were avoided, so there may be many researchers who did not avoid or remove areas susceptible to contamination in the burial environment. While it is possible they were taking these precautions, it is necessary to

make sure that method descriptions are clear and ideally that methodologies are employed to evaluate their samples for diagenetic alteration (i.e., Kamenov et al., 2018) to minimize possible counterarguments to interpretations and to assist researchers in setting up their own sample protocols.

As part of the initial sample preparation, contamination from cutting and cleaning instruments should be considered. Montgomery (2002) found that the diamond-tipped blades or burrs and agate mortar and pestles were a potential Pb source, instead suggesting to use a tungsten carbide bit to clean all cut surfaces and for the initial surface abrasion or removal of dentine from enamel. While this may be specific to the materials used, this should be considered as a potential source of laboratory contamination that would be difficult to identify. While many studies did not specify the specific materials used for cutting or powdering (n = 16, 29%) or cleaning teeth (n = 11, 20%), some that used diamond-tipped or unspecified burrs or saws (n = 13, 24%) without using tungsten carbide bits to clean after, and others used mortar and pestles for powdering the samples (n = 4, 7%). All of these were utilized without specifying whether they had tested their materials for Pb, nor what, if any, laboratory practices or original protocol methods were in place to ensure that this was not a source of contamination.

Part of minimizing the impact of contamination is considering the expected Pb concentration of the final sample and how this compares to sample loss and instrumentation sensitivity for both Pb concentration and Pb isotope analyses. As of now, instrumentation does not allow for Pb isotope analyses using micro-sampling or laser ablation techniques, therefore all Pb isotope data are from bulk dental samples. While most Pb concentration studies also used bulk samples, laser ablation was sometimes used,

for which some concerns are addressed later. The ideal bulk sample mass will vary greatly depending on multiple factors: expected concentration (natural is likely below 1 ppm, and anthropogenic could be much higher, but could still result in <1 ppm concentrations), contamination from pre-treatments and solutions (this will be higher in isotope pre-treatments since there is interaction with more solutions), expected loss of material during pre-treatment (either from taking aliquots for different analyses from a single bulk digest, or from the column chemistry to elute Pb for isotopic analyses), the target voltage intensity for analysis (ensuring it is at least an order of magnitude about the limit of detection and quantification), the final solution concentration, and if applicable, volume, needed to reach that target intensity. While Dudás et al. (2016) examined the low limit of sample size mass, it was unclear if heterogeneity below 10 mg was from an unknown preparation contaminant or *in vivo* variation. When specific methods were stated in the reviewed papers, it was often reported that 10 mg was used for concentration and a further 50 mg for Pb isotope analyses; however, most gave no or insufficient information regarding sample mass. While there is no *universal* ideal sample mass, future studies should be more detailed in terms of the specific steps taken to avoid influences on concentration and isotope ratios from contamination.

While consideration of avoiding diagenetic materials and post-depositional contamination is important to data quality, there is a current challenge in whether to use a bulk or micro sampling technique when acquiring Pb concentrations. Laser ablation has been utilized in method development to examine Pb concentration patterns within teeth (e.g., Budd et al., 1998; Webb et al., 2005), but more recently, studies have linked changes in concentration to the age of formation (Jones, 2015; Quinn et al., 2020).

However, there is ongoing debate as to whether the offset in the relationship between the timing of tooth formation and enamel maturation is sufficient to undermine the usefulness of serial sampling connected to physiological age (e.g., Smith and Tafforeau, 2008; Montgomery et al., 2010; Simmons et al., 2013). When enamel forms, the initial composition is rhythmic, creating cross striations and striae of Retzuis, which can be used to estimate the formation time (Lacruz et al., 2017; Berkovitz et al., 2017). The mineralization of the resulting enamel happens in two phases: secretion and mineralization. The initial secretion is more organic and is replaced with highly mineralized hydroxyapatite, which matures from the enamel-dentine junction towards the surface (Al-Mosawi et al., 2018). There is a slight offset in timing between secretion and mineralization, unlike in dentine. However, the specific pattern is still unclear and may be affected by overprinting or averaging of the elemental/isotopic history of the enamel. While there is promising evidence to suggest that the effect of offset and overprinting is minimal (Austin et al., 2016; Green et al., 2018; Smith et al., 2018; Müller et al., 2019; Dean et al., 2020), and that the Pb present in the final mineralized material was from the initial secretion (Müller et al., 2019), the amount of time represented is still unclear, particularly due to the long residence time of Pb in the body compared to other elements examined in the studies. Though there is less temporal specificity, a more certain method to examine change over time within an individual is by using bulk samples from multiple teeth (i.e., Valentine et al., 2015).

While many studies systematically choose which teeth to sample, the lack of reported details in the sample choice procedure creates two challenges for data interpretation: the potential remobilization of Pb from past exposure and the impact of social age on

interpretation. Deciduous teeth form *in-utero* and early infancy, while adult teeth form after birth (AlQahtani et al., 2010). Pb concentrations in first molars, or other teeth that form at a similar time (see Table 1), provide a record of the period closest to the time of birth when infants and children are most susceptible to Pb through hand to mouth activities and via their underdeveloped digestive system. However, remobilized maternal Pb can impact teeth that form early in development or during breastfeeding (Gulson et al., 2003). Any exposure from the physical environment to infants could be mixed with maternally transferred Pb. In situations where the mother has moved to an area with different bioavailable Pb isotope ratios, the Pb exposed to the developing fetus/infant would be mixed with, if not entirely swamped by, non-local values (Gulson et al., 2003). This challenge is compounded when the study does not identify the specific tooth sampled for each individual, thus making it difficult for readers to assess if this should have been a potential source considered by the researchers, given the age at development.

Understanding specifics about the type of tooth sampled is vital because of both physiological and social differences in the potential of Pb exposure and its pathways. There are (at least) three types of age: chronological, biological (physiological), and social (Kamp, 2001:3; Baxter, 2005:19; Halcrow and Tayles, 2011:335). Chronological age is the time elapsed since birth, either in days, months, or years. Biological age considers the biological changes that occur to an individual, such as physical growth or the development and eruption of dentition – these developments demarcate biological age groupings. Social age groups are boundaries of normative behaviour and are based on culturally constructed views of maturation. Bioarchaeologists use biological age data, which they infer as chronological data, then look for patterns to determine the

chronological boundaries of social age (Sofaer, 2011:290). However, these boundaries can be challenging to conceptualize, particularly for societies and periods with little to no written records regarding children (Sofaer, 2011; Agarwal and Glencross, 2011). As Halcrow and Tayles (2011:251–252) suggested, there is no easy answer to this problem, and more work needs to be done with theoretical views of the body.

Studies must approach skeletal remains while considering that these individuals, even as children, were active members of society who experienced numerous physical and social changes. In turn, differences in exposure between teeth may be due to social differences, not just changes in geographical environments. Without a systematic sample choice that includes the consideration of social factors during childhood, the original researchers' data could be misinterpreted and does not allow those examining childhood in a social setting to re-examine the work in the future. Although not all bioarchaeologists need to adopt the childhood social theory framework, these issues need to be considered. Until such a reconciliation can be made, what is most important is to be clear about the methods used, the terminology employed, and to integrate historical and archaeological context whenever possible.

2.3.2 Interpretation of Pb concentrations over time

Pb concentration of tooth enamel in contemporary populations is a valuable tool to track exposure during tooth formation (e.g., Gulson, 1996; Robbins et al., 2010). These data were used for several purposes, including method development or refinement, examining change in concentration over time, comparing the general pattern between populations, inferring health effects, and examining diagenesis. Some studies did not

interpret the values in the papers (n = 4, 10%), either because the purpose was method development (Budd et al., 1998) or other stable isotope systems were more effective at identifying outliers or dietary patterns within the population (Szostek and Głąb, 2001; Müller et al., 2003; Keller et al., 2016).

Studies were inconsistent in the amount of information given regarding concentration methods, and less than half gave sufficient analytical methodology information for the Pb concentration results to be replicable (n = 19, 43%). Many lacked basic information on the initial dilutions, instrumentation, standards, error (of standards and individual samples), and calibration corrections. Earlier research used atomic absorption spectroscopy (AAS) or isotope dilution thermal ionization mass spectrometry (ID-TIMS) to determine concentrations, but as of 2001 studies have forgone AAS and it is more common to use hexapole or quadrupole inductively coupled plasma mass spectrometry (Q ICP-MS) or ID-TIMS. Rather than analyze cross sections of enamel using laser ablation (n = 5, 11%), most digested the samples in acid prior to analysis (n = 35, 80%), and of these, almost all utilized the bulk digest solution (n = 30, 86%). There were four studies (10%) that determined concentration after column chemistry and some without the use of an isotopic tracer. However, column chemistry techniques are rarely 100% efficient, so this can miscalculate the Pb concentration and is therefore not advisable.

For studies that interpreted the concentration values, there were differences in what the samples were compared to and how the data were converted for interpretation. The four main approaches that were used for interpreting data were: 1) to consider the general pattern of Pb exposure, often comparing it to previous modern and/or archaeological studies; 2) to map changes in Pb exposure in teeth over time to understand high modern

exposure; 3) to convert enamel to blood Pb or comparing the data to modern values to infer health; and 4) to compare trace element concentrations to previous studies, or isotopic ratios of the soil, to suggest diagenetic alteration.

The most common approach was to compare values in archaeological samples to either modern (n = 8, 18%) or archaeological (n = 16, 36%) teeth from similar contexts, or both (n = 11, 25%) (e.g., Montgomery et al., 2005; Price et al., 2017). Unfortunately, there are limited data for directly comparable modern values of Pb in teeth. Modern studies typically use surficial enamel, cervical portions of teeth, or whole teeth to produce their data (e.g., Fosse et al., 1995; Gulson, 1996; Gomes et al., 2004), but most archaeological studies sampled core enamel. This creates a problem for cross-comparison since the concentration of Pb is not uniform throughout the tooth (Budd et al., 1998). The surficial enamel is highly enriched compared to core or dentin, and cervical portions of teeth include both dentine and enamel, and since dentine has higher concentrations of Pb, comparisons will result in erroneous interpretation of differences Therefore, comparisons of concentrations taken from different portions should not be employed.

In comparing archaeological populations, part of the consideration is whether the exposure was from natural or anthropogenic Pb sources. Since natural levels of Pb exposure result in low enamel concentrations, high enamel concentrations help to identify when anthropogenic Pb exposure occurred (Montgomery et al., 2005). Several studies established that pre-Roman Britains have enamel concentrations below 0.5 µg/g (ppm) and have no influence from anthropogenic Pb ore. While individuals with concentrations above 0.5–0.9 µg/g may have intermediary values influenced by anthropogenic Pb ore, they have found that those above 0.9 µg/g consistently have isotope values representing

anthropogenic exposure (Montgomery et al., 2010; Millard et al., 2014). However, Pb concentrations can vary between individuals within populations even when the overall population exposure is exclusively high or low (Budd et al., 2004). Schroeder et al. (2013) used this concept of very low natural Pb exposure to distinguish between enslaved labourers at the Newton Plantation who were born locally in Barbados and those born in Africa. The African environment was inferred to have limited atmospheric exposure and no Pb-containing material culture. These concentrations were further extrapolated to examine health impacts on those working at the plantation. Although there is a trend in bioarchaeology to examine life histories of individuals (Katzenberg, 2008), limitations in methods converting enamel to blood Pb concentration must be considered.

Some of the interest in using Pb concentrations in teeth is to examine the potential health effects of past exposures. A fundamental challenge with interpreting Pb concentration data is the uncertainty in converting Pb enamel concentrations to blood Pb. While there is considerable information on blood Pb levels and their association with adverse health impairments (Needleman, 2004), the extrapolation of using tooth enamel Pb concentration to determine the extent of impacts on health is poorly understood. To examine these health impacts, several studies (Montgomery et al., 2010; Schroeder et al., 2013; Quinn et al., 2020; Moore et al., 2021) converted Pb concentration in enamel to blood Pb using a 10:1 blood-to-enamel ratio proposed by Grobler et al. (2000). Grobler et al. (2000) state that the conversion they developed should not be used with high enamel concentrations or in situations where people are moving between regions. Furthermore, the 10:1 ratio proposed ignores the variability of both blood and enamel data from Grobler et al.'s (2000) initial study. The coefficient of variation was greater than 70% of

the mean (enamel: 0.33 ± 0.27 mg/kg and blood: 0.031 ± 0.022 mg/kg). Therefore, the ratio could actually vary between 1:1.3 and 67:1, which is too large a difference to be applicable without additional study. Finally, the teeth used in the study by Grobler et al. (2000) formed *in utero*, yet the blood concentrations were taken from children at 6–8 years old. Therefore, there may be discrepancies in the amount of Pb to which a fetus versus a child were exposed. Unfortunately, researchers have not acknowledged these limitations, and while natural versus anthropogenic exposure can be interpreted at the *individual* level, the absence of a simple conversion method means it is only possible to interpret potential health effects at the population level as seen by the approach of Montgomery et al. (2010).

At the population level, it is possible to compare between sites. A meta-analysis comparing modern and archaeological tooth concentrations would be beneficial to understand the change of exposure over time and to allow for better inferences regarding past health. While it may not be possible to develop an accurate equation to correlate enamel concentrations to blood values to make inferences about health, it is useful to explore this possibility. Additionally, while we now have a good estimation of what the natural Pb exposure was in archaeological populations of Britain (Montgomery et al., 2010; Millard et al., 2014), this has not been examined in other geographic areas. As such, studies are needed on the concentrations of Pb in archaeological populations from more geographically and culturally diverse settings to better delineate the cut-off between natural and anthropogenic exposure.

2.3.3 Interpretation of potential Pb sources

Pb isotope ratios are used in the archaeological examination of populations in several ways, including diachronic change over time, geographic movement, and diagenesis. While early research compared modern and archaeological exposure to see change over time and the relationship to atmospheric pollution (Patterson et al., 1991; Budd et al., 2000a; Budd et al., 2004), Whitaker and Stack (1984) were the first to apply Pb isotope ratios in teeth to examine human geographic movement, and Montgomery et al. (2000) within enamel. Fundamental to addressing these topics is the acquisition of high-quality data.

Pb isotope data are acquired using a few different approaches but starts with column chemistry to isolate the Pb. Two main forms of resins are used: anion resin (i.e., AG-1X8 or Dowex 1x8; n = 23, 58%) or a crown ether resin (i.e., Sr-Spec; n = 6, 12%). The column chemistry required to elute Pb from these resins varies, but regardless has the basic steps of cleaning, conditioning, loading sample, washing, and elution – with specific solution volumes. In the last few years many authors do give sufficient information, either in the paper or through citations, to replicate their method (n = 10, 25% total and n = 7, 18%, partially), but some still include broad descriptors such as HBr ion exchange (n = 7, 18%) or anion exchange column chemistry (n = 5, 13%). Beherec et al. (2016) suggested that Sr spec resin results in too high of blanks, but this may depend on the column chemistry involved, which was not stated. The impact of the blanks would also depend on what the expected concentration of the sample was and the initial sample mass. Blanks have not been a problem for the other studies that used this technique, and the resin

allows for Sr to be eluted before Pb (Pin et al., 2014) during the same column chemistry. It is possible to collect the washes from the anion exchange chemistry and use it to load onto a separate Sr specific resin column after, but this increases cost and time.

With Pb eluted, the solution is analyzed using an MC-ICP-MS (n = 23, 58%), or dried onto a filament and analyzed on a TIMS (n = 14, 35%). While the specific analytical parameters will depend on the analytical machinery available, certain information remains important to state. Parameters include the analytical mode (dynamic/state), cones used, faraday array, the timing of baseline and integration along with the number of blocks of data. Furthermore, there remains little explanation of calibration methods. Though many who analyzed isotope ratios on MC-ICP-MS used Thallium (Tl) corrections with a thallium spike into all samples and SRM 981 as a standard (n = 18, 78%, n = 5, 22% not specified) to account for mass bias, Kamenov et al. (2004) found that there is significant variation in the $^{205}Tl/^{203}Tl$ ratio, so the time elapsed since solution mixing needs to be controlled and accounted for, but is not always reported in these studies. Data can also be calibrated using the most probable values of standards (i.e., SRM 981, n = 6, 15%) which are bracketed after a number of samples and using an average or linear equation to account for drift during between the brackets of data. It should be noted that there are some different Pb isotope values of SRM 981 that data can be normalized to, but few authors were clear about what values or at least citations to these values they were. All these parameters and calibration techniques have impacts on data quality, but there remains a need to examine with interlaboratory studies how big of an impact such facts cause for archaeological data. It was slightly more common that Pb analytical methods included sufficient information for the data to be replicable (n = 21,

48%) or partially so (n = 14, 35%). However, when sample procedure, pre-treatment, and analytical sample methodology are all considered only three studies truly gave enough information for their studies to be reproducible: Turner et al. (2009), Millard et al. (2014), and King et al. (2021).

Related to standards, is the reporting of data quality. To assess the quality of data, both percent error to understand accuracy and variability to understand repeatability need to be reported. However, only one (3%) study gave information on both accuracy and repeatability, many provided only accuracy (n = 15, 41%) or insufficient information (n = 8, 22%). Those that gave information on variability (n = 16, 43%) in all but three instances (81%) gave sufficient information to back calculate the accuracy, even though not directly addressed. Since absolute differences in Pb isotope ratios of standards to known values and the 2σ of the analyses are often in the 2nd–3rd decimal for $^{20n}Pb/^{204}Pb$ and 3rd–4th decimal for $^{20n}Pb/^{206}Pb$, and meaningful differences are often close to this level of error, high quality data is necessary. The propagated error (2σ) of each individual sample should also be given, which will account for both the variation in the individual analysis and that added from the calibration. It should also be noted that reporting on data used for calibration as a means of addressing quality is not appropriate, so additional analyses of SRM 981 during a run, or a different check standard, is necessary. For example, SRM 1400 (Bone Ash) is a bioapatite standard with most probable values published in Hinners et al. (1998). Since it is processed through the same column chemistry procedure as the tooth samples, it is matrix matched, and in turn can account for any such problems during analysis. Either way, researchers should be clear about the errors of the samples.

Migration studies remain the primary application of Pb isotope data, but within these, there are three approaches. The first examines populations that are determined to be free of potential anthropogenic Pb exposure and identifies non-locals when their Pb isotope ratios do not fall within the expected local isotopic range (e.g., Budd et al., 2000a; Dudás et al., 2016; Samuelsen and Potra, 2020). The second approach infers a cultural focusing of Pb isotope ratios with an increase in Pb concentrations and identifies non-locals to a cultural region (e.g., Budd et al., 2004; Montgomery et al., 2005). The third combines the first two but uses the $^{238}U/^{204}Pb$ ratio, which aids in separating between natural (silicate) and anthropogenic (sulphide) Pb sources (Evans et al., 2018a, 2018b). While movement studies mainly focus on the population level, researchers are increasingly examining the life histories of individuals (e.g., Budd et al., 2000a; Valentine et al., 2008; Lamb et al., 2014; Laffoon et al., 2020). A related but separate use of Pb, is for the examination of diagenetic alteration of tooth enamel by considering Pb isotope ratios and other trace elements as markers of potential influences on data interpretation (e.g., Dudás et al., 2016; King et al., 2020; Simonetti et al., 2021).

To infer movement, the locally available Pb source must be characterized. There are a few general approaches, though many studies used more than one approach. The first is using existing or new data regarding the local environment (n = 28, 70%). An alternative is to utilize existing publications that characterized the Pb ore known to be available locally (n = 19, 49%). Finally, some researchers use ranges from previously published archaeological human comparative data (n = 20, 45%). The approach taken depends on whether it is determined that the population was exposed to anthropogenic or natural Pb. In the case of natural exposure, it is not simply the local geology, but the bioavailable Pb

that requires isotopic characterization. However, of the 28 studies that examined natural exposure, this concept was mentioned or considered by only 19 (68%) and of those, defined by 13 (46%).

There is no universal method to define the bioavailable Pb in natural exposure contexts using the local environment. Many looked at the underlying geology (n = 15, 54%), but no studies used only this method. It was also common to sample the burial soil matrix or local subsoil (n = 14, 50%), and archaeological bone or teeth from fauna (n = 8, 29%). Less often, the mean and standard deviation of bone, dentine, or enamel Pb isotope ratios from the population itself were used (n = 5, 18%).

While numerous reviews (e.g., Price et al., 2004; Bataille et al., 2020; Holt et al., 2021) and applications define bioavailable Sr ratios at the local or regional level (e.g.; Montgomery et al., 2010; Willmes et al., 2018; Janzen et al., 2020; Wang and Tang, 2020; Frank et al., 2021; Washburn et al., 2021), there is limited discussion on how to apply Pb for this purpose (Giovas et al., 2016; Sharpe et al., 2016; Tschetsch et al., 2020; Samuelsen and Potra, 2020; Price et al., 2021; Sharpe et al., 2022). While for Sr it is ideal to sample flora, fauna, and or water samples to characterize the bioavailable values (Bentley, 2006; Evans et al., 2010, 2012), contamination by the widespread use of anthropogenic Pb since industrialization complicates this process with Pb (Samuelsen and Potra, 2020). Therefore, context-specific archaeological faunal must be sampled; though this can be difficult since animals themselves can be imported.

While several studies have followed this approach (i.e., Valentine et al., 2008, 2015; Turner et al., 2009; Turner and Armelagos, 2012; Price et al., 2017, 2019; Evans et al., 2018a; Samuelsen and Potra, 2020), there is limited discussion in the discipline of what

animals (wild or domesticated) are acceptable for such studies and there is not enough attention to characterizing the variability to be able to define an adequate sample size to detect change. For example, Sharpe et al. (2022) has suggested that decisions of animals will depend on the availability and likelihood of local geology to not diagenetically alter the materials, but that thicker enamel may fair better. Many studies use soil/sediment samples (n = 14), but it is questionable whether they are sufficient to characterize local bioavailable Pb, particularly given the potential of anthropogenic soil contamination. This also assumes that the soil in which people are buried represents the land where they hunted, gathered, and or grew their food, which is certainly not always the case. While variability will depend on the complexity of the geology, foraging or agricultural practices, etc., statistical power and sample size calculations warrants further focus. It is possible to calculate the number of samples needed to identify a change from locally available values (i.e., reject the null hypothesis), or the number needed to create that local baseline (Dupont & Plummer, 1990; 1998). This, and the statistical power available to determine a difference given the sample size available can be easily calculated using a program like that created by Dupont and Plummer available through Vanderbilt (Dupont & Plummer, 2018). Sample choice is often dictated by what is available to the researchers, but the limitations of this approach need to be articulated and considered during the interpretation of results.

Studies that attributed exposure to anthropogenic sources (n = 24, 60%) often examined sources in their cultural environments because of the cultural focusing of Pb. Studies have attempted to characterize the cultural environment through the use of either Pb isotope ratios at the mines that were known or suspected to be the primary sources of

Pb to that population (e.g., Fitch et al., 2012), or used atmospheric deposits to establish expected bioavailable values (Fitch et al., 2012; Keller et al., 2016; Aronsen et al., 2019; Eshel et al., 2020). Budd et al. (2004) established that there is no direct connection between human Pb isotope ratios and those in the physical environment (i.e., soil cores and other atmospheric deposits), so the applicability of atmospherically derived baselines is unclear.

Defining bioavailable sources in populations exposed to anthropogenic Pb presents a different challenge. Part of this stems from the assumption that exposure patterns in modern contexts hold true in the past. Kamenov (2008) argued that Pb in teeth from modern Bulgarian individuals is mainly from dust associated with tetraethyl Pb in gasoline that is inhaled or ingested, which is an observation supported by studies on other modern populations (Mielke and Reagan 1998; Zhang et al., 1998; Gwiazda et al., 2005; Laidlaw et al., 2005). However, unlike many populations in the past, none of these modern populations frequently used material objects containing high amounts of Pb. Therefore, exposure to these materials likely contributes significantly to the body's Pb burden. In turn, the idea that Pb comes mostly from inhalation rather than ingestion cannot be indiscriminately employed in past populations. Yet, this concept is occasionally applied both in natural (Turner et al., 2009; Jones et al., 2017, Price et al., 2017; Scaffidi et al., 2021; Turner, 2021) and anthropogenic exposure contexts (Beherec et al., 2016; Keller et al., 2016; Price et al., 2019), sometimes leading authors to limit their consideration of other Pb-containing materials.

Rather than assume that sources from the physical environment are the likely contributor, the material cultural environment must be explicitly considered. Although,

even in modern contexts, a local mine is not often the exclusive Pb source to a population (Gulson, 2008), it is common for studies to consider only the locally mined Pb (i.e., Fitch et al., 2012). Presumably, this is assuming that the mined Pb would pollute the environment they are living in and cause exposure. Based on Mahaffey (1978), Montgomery et al. (2010:219) suggested that there were three aspects to exposure from Pb: "1) Drinking soft water or acidic fluid from Pb pipes, cisterns or vessels; 2) Ingestion of bioavailable Pb; or; 3) accidental ingestion of Pb from glazed pottery." Ingestion or inhalation of dust would fall into the second category, yet many studies ignore the others. While this highlights accidental ingestion of food or drink from Pb glazed pottery or Pb vessels, there are countless other means for accidental ingestion including other Pb-containing alloys like pewter, or even the consumption of Pb paint which is common in young children (Åberg et al., 1998; Gulson et al., 1996a). Arguably, we can add a fourth possibility of dermal Pb exposure through body paint or makeup. These potential sources could easily contribute to the overall exposure of a population or even mask the role of the ingested or inhaled bioavailable Pb from the physical environment. Since Pb used in those materials may not match any pollution to the local environment, all sources of Pb to a population must be considered.

Attributing Pb to anthropogenic or natural exposure involves considering a population's sources of Pb, both from the environment and the potential material culture that was locally sourced and produced or imported. While it is impossible to map all potential sources, there is information in archaeological materials, ethnographic accounts, and historical records that help suggest the specific materials. Following this, consideration of the Pb source within the materials should be examined. Part of the

anthropogenic exposure can be through material goods, even imported goods. Although some studies do mention the potential Pb exposure to such materials, it is largely discounted or ignored entirely (e.g., Fitch et al., 2012; Keller et al., 2016; Aronsen et al., 2019; Quinn et al., 2020), or the characterization of all Pb sources to a population is ignored (e.g., King et al., 2020).

An additional limitation in the approach to Pb is in regard to what populations should be considered to have 'natural' rather than 'anthropogenic' Pb exposure. This can affect both when exposure is attributed to only natural or only anthropogenic sources. Referencing Turner et al. (2009), Kamenov and Gulson (2014) present a version of this critique by asserting that the Inca did have access to Pb-containing materials. However, this has gone unheeded by some recent studies and remains a potential alternative interpretation rather than an indication of diagenetic alteration (i.e., Turner et al. 2009; Scaffidi et al., 2021; Simonetti et al., 2021). Considering possible Pb sources outside of populations typically thought to have metallurgy is important since researchers may be overlooking some anthropogenic Pb sources. For example, Jones et al. (2017) found that some individuals at a site in the Lower Illinois River Valley during the Middle Woodland Period (2100–1700 BP) had Pb isotope ratios similar to locally available galena and proposed that it may be associated with later ethnographic accounts of its use as a ceremonial pigment. This possibility is compelling and highlights the potential of ethnographic and archaeological records to support alternative conclusions.

In cases where it is unclear whether exposure occurred exclusively from anthropogenic sources, it is advisable to not only examine the Pb concentration of the enamel, but also characterize the local Pb environment, ideally using faunal samples (the

number of which determined through power analyses). This is in addition to considering the Pb isotope values of anthropogenic sources. If the values of the anthropogenic sources are too similar to the local environment, attribution to anthropogenic exposure may not be possible, however incorporating $^{238}U/^{204}Pb$ ratio (Evans et al., 2018a, 2018b) may help differentiate between natural (silicate) and anthropogenic (sulphide) Pb sources. Focusing on the concept of 'natural' exposure was effective in the 1980s to early 2000s in establishing high levels of contemporary Pb. However, it is not always clear what populations had solely 'natural' exposure levels. Although there are cases where interaction with anthropogenic Pb occurred, that is not always the case. Studies should consider the cultural environment and materials people may have interacted with when determining potential Pb exposure sources.

Rather than assume that populations were exposed to solely 'natural' Pb from the underlying geology, archaeological and historical evidence along with Pb concentrations should be considered to critically assess the point. Whether natural exposure is concluded, or it is determined that anthropogenic exposure occurred, this should remain contextualized in the study using the archaeological and historical data by examining what Pb geological sources were common, and what objects could they have been using (both locally produced and imported).

The final concern about interpreting potential movement and sources of Pb relates to how diagenesis might alter Pb concentrations and Pb isotope ratios. Of the studies reviewed, 46 (84%) considered diagenesis. Of those that concluded diagenesis possibly or did occur (n = 18, 33%), three methods were used to come to this conclusion. First, was when elemental concentrations or ratios were considered to be high or were high in

comparison to specific cut-offs established by Patterson et al. (1991) or Kamenov et al. (2018) (n = 8, 44%). Next, if the Pb concentration was higher than expected in cases of natural exposure or they were too high to be biologically possible (n = 5, 28%). Finally, when Pb isotope ratios were outside of the local or expected range or suggest an additional source through a linear array/mixing model (n = 3, 17%). Of those that concluded diagenesis occurred, only five (28%) specified that they were considering the preservation of the individual teeth, and one defined what characteristics were considered in establishing the level of preservation. However, half of the examined studies did consider but did not identify that diagenesis occurred (n = 28, 51%). While many of these did not examine any criteria (n = 17, 37%), instead saying enamel was stable (n = 12), the same techniques used by those who determined diagenesis occurred were applied. Therefore, not finding diagenesis was not exclusively an issue of papers not having examined the issue.

Considering the methods applied to identify diagenetic alteration, studies may be misattributing the population's use of Pb or the results from poor sample choice and preparation to the effects of diagenesis. Additionally, establishing arbitrary cut-offs for what is expected for 'normal/natural' Pb concentrations in teeth (i.e., Dúdas et al., 2016) is not advisable without solid consideration of data error and reliability, and possible anthropogenic sources available during life. While identification of diagenetic alteration is possible by comparing Pb isotope ratios in soil and teeth as is commonly done with bone (Carlson, 1996; Bower et al., 2005; Montgomery, 2002; DeMuynck et al., 2008), this requires properly identifying all end members, and not ignoring potential anthropogenic sources that may be imported. Finally, there are no direct quality checks to

determine if diagenesis has occurred beyond examining if other trace elements are above what is expected. Kamenov et al. (2018) established parameters of what should be considered normal using specific trace element concentrations; however, what is considered within the 'normal' range is based on modern enamel studies and a small data size from Patterson et al. (1991). Moving forward, more consideration of how sample choice, analytical methodology, and potential locally available and imported Pb-containing materials may influence the concentrations of Pb in teeth should be considered when interpreting diagenesis.

2.4 Conclusion

In bioarchaeology, Pb analyses are currently used to address differences in Pb exposure between populations, identify instances of natural vs. anthropogenic exposure, and reference overarching health concerns. Pb isotope analysis is applied to examine the movement of people in cases of natural exposure and movement out of a broader region if anthropogenic exposure occurred. Pb is a powerful analytical tool, particularly when used with other isotopic systems ($\delta^{18}O$, $^{87}Sr/^{86}Sr$). However, a few underlying themes of misuse need to be considered, that are summarized in Table 2. First, the assumption of anthropogenic versus natural exposure must be critically evaluated using archaeological, ethnographic, and historical documentation. Only with this information can we more holistically consider the sources of Pb, including the materials that result in Pb exposure. Additionally, studies give insufficient information on sample choice, preparation, and data acquisition, which means that the methods are not reproducible, thus negatively affecting the interpretation of the data. Moreover, the readers cannot assess if

Table 2. Summary of topics regarding use or application, misuse or insufficient information given, challenges from current limitations, and studies needed within each theme explored as part of the review.

Issue	**Use**	**Insufficient information or consideration**	**Current limitation**	**Study Needed**
Sample choice	- Mostly enamel - Most frequently premolars or molars - Some systematically for age -Bulk or micro sampling	- Preservation - Contamination by cleaning/cutting - Unidentified teeth - Remobilization of Pb during pregnancy and breastfeeding	- Tooth availability -Tooth enamel maturation timing - Biological versus cultural age	- Dentine diagenetic alteration
Interpretation: Concentration	- Method development - Comparing general population patterns - Inferring health effects - Examining diagenesis	- Insufficient method information - Converting enamel Pb to blood Pb - Concentration not always examined in "natural" exposure contexts	- No directly comparable modern values	- Meta-analysis of modern and archaeological tooth values - 'Natural' Pb exposure in other regions
Interpretation: Isotopic ratios	- Movement by identifying individuals outside local bioavailable range or outside range for locally used ore - Examine diagenetic alteration	- Insufficient method information - Not considering both atmospheric and cultural material exposure - Many Indigenous populations assumed to have no anthropogenic exposure	- No standard for defining bioavailable Pb - Identifying all Pb sources to a population - Identification of diagenetically altered samples	- Best practices for establishing bioavailable values

methodological, physiological, or cultural limitations have been considered, which weakens the studies' conclusions. Finally, we do not have a means to directly identify if Pb diagenesis has occurred. While some have compared the Pb isotope ratios in burial soil and teeth, this can only suggest that there has been no change if there is a difference between the two, and even then, there can be significant ambiguity that requires interpretation and requires the researcher to properly define all end members. The most common method is via proxies of other elements that do not necessarily have the same reactions in the burial environment. This is compounded by insufficient reporting of sample preparation and analytical methodology. Without properly defining potential Pb sources to a population and potential sampling or analytical methods defined, the arguments questioning the stability of enamel bioapatite are greatly weakened.

Based on this review of 55 papers dealing with Pb concentration and isotope analyses in bioarchaeological contexts, we make the following recommendations. First, studies must specify the specific tooth sampled for each individual. This is important because of remobilized Pb during breastfeeding and the potential for cultural influences that can affect how an individual may be exposed to Pb over the course of their childhood. While laser ablation is used, it is still uncertain if changes in Pb exposure relate to a specific age of development due to the unclear zonation in teeth. Therefore, more Pb studies may be necessary before we can assess the specific health experiences of an individual at an annual or sub-annual level. Though it cannot specify health impacts to an individual, determining Pb concentration is important to assess differences in Pb exposure between populations and for identifying an individual's potential for anthropogenic exposure. However, many studies examining 'natural' Pb are not acquiring concentration data,

which would be an important additional means of supporting the assumption that there was no anthropogenic exposure to a study population.

Finally, there need to be more studies on characterizing bioavailable Pb isotope ratios in anthropogenic and natural contexts. Pb from material goods is often underemphasized or ignored in anthropogenic Pb source interpretation. These can be a substantial source of Pb to a population. Yet, there remains a focus in North American studies on just the atmospheric Pb or ore and a disregard for the Pb-containing objects with which populations would have been interacting. While Samuelsen and Potra (2020) asserted that animal teeth are ideal for characterizing bioavailable Pb isotope values in natural contexts, the number of samples necessary will depend on data variability and should be explicitly examined using power analyses.

It is hoped that the guidance and recommendations provided in this review will increase confidence in results and improve the ability to interpret specifics about Pb data reported, to compare studies, and to transfer protocols. Appropriate use of the techniques requires careful consideration of sample choice, determination of possible contamination, and data interpretation would benefit from improving the contextual information available from archaeological, ethnographic, and historical documentation.

3 EVIDENCE OF CHILDHOOD Pb EXPOSURE IN TOOTH ENAMEL FROM 18TH–19TH CENTURY NORTH ATLANTIC FISHING COMMUNITIES[2]

3.1 Introduction

Lead (Pb) isotope ratios in archaeological tooth enamel can help to identify the movement of cultural materials and the migration of people. While Pb isotope analysis is frequently used on archaeological populations in Europe (e.g., Budd et al., 2004; Blackström et al., 2018; Walser et al., 2019; Moore et al., 2020), a limited number of studies have examined Pb in Indigenous (Carlson, 1996; Dudas et al., 2016) or European-descent settlements of North America with most focused specifically on the industrial period in areas with local Pb mining (Fitch et al., 2012; Keller et al., 2016). More recent studies have examined Pb in 19th century USA and New Zealand (Aronsen et al., 2019; King et al., 2020) that are away from or predate local mining. However, these nor the Industrial period USA studies have considered the importation of European goods as a possible *in vivo* source of exposure. Instead, the North American studies emphasized local environmental pollution as the source of exposure. In contrast, Laffoon et al. (2020) found that Barbados-born enslaved labourers had Pb isotope ratios consistent with England, likely due to local rum production using distillation pipes made with imported English Pb. This raises an important question: could the pattern of Pb isotope ratios consistent with European sources be more widespread in local North American

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populations? To help address this question, here we examine Pb exposure and sources in North American contexts with no local anthropogenic environmental Pb exposure but with strong trade ties throughout the Atlantic World.

Enamel represents an ideal sampling tissue for chemical and isotopic analyses since it contains small pore spaces and is highly mineralized, thus minimizing the likelihood of contamination or diagenetic alteration (Budd et al., 2000b:692-693; Sponheimer and Lee-Thorp, 2006:1650-51). Once enamel forms and matures the tissue does not remodel, meaning that it's Pb concentration and isotopic ratios reflect an individual's exposure during specific developmental periods and ages (Budd et al., 2000a:3; Hillson, 2014). This allows researchers to access information regarding childhood, a life stage often under-researched in archaeology (Alfonso-Durruty et al., 2014; Lillehammer, 2015). Tooth enamel is used in modern and archaeological contexts to examine the source and extent of Pb exposure using concentrations and isotopic ratios (Kamenov & Gulson, 2014).

Four stable isotopes of Pb are typically measured as part of bioarchaeological migration studies: ^{204}Pb, which is primordial, and ^{206}Pb, ^{207}Pb, and ^{208}Pb, which are radiogenic (Faure & Mensing, 2005). The different original concentrations of rocks (parent isotopes), the age since formation (half-lives), and mixing during formation create the geological variability of Pb isotope ratios (Gulson, 1986; Adriano, 1986; Teutsch et al., 2001; Faure & Mensing, 2005). These Pb isotope ratios can help identify a 'non-local' when an individual's values are outside the expected local ranges. Local ranges are commonly determined using faunal remains in non-anthropogenic (natural) Pb contexts (Samuelson & Potra, 2020) or through characterizing isotope ratios of local ore sources in

anthropogenic contexts (Montgomery et al., 2005; see Chapter 3). When populations are exposed to anthropogenic Pb, there are typically only a few geological sources and, at least in Britain, Pb was frequently melted down and reused, meaning that it is progressively averaged with time (Montgomery et al., 2005; Montgomery et al., 2010). Therefore, there is a narrow range of expected Pb isotope ratios within a population, termed 'cultural focusing' (Montgomery et al., 2005).

This study examined Pb concentrations in six North Atlantic fishing communities to explore the extent of exposure and whether there were differences between communities based on cultural influences (Table 3; Figure 3). These include burial grounds from four English or Irish fishing villages in Newfoundland (NL), the largest of which was in the large fishing and mercantile center of Harbour Grace (St. Paul's Anglican Church cemetery; 1764 to 1820). It also included two smaller fishing villages of individuals most likely born in NL, at Foxtrap (Foxtrap-2 burial ground; 18th century to circa 1896) and Tors Cove (Tors Cove burial ground; 1812 to late-19th c.), and a cemetery with both locally born and immigrant individuals at Portugal-Cove (Wester Point cemetery; 18th to 19th c.). A cemetery associated with what is now known as St. Luke's Anglican Church (16th to 18th c.) in Placentia, NL, has individuals of uncertain cultural affiliations (Basque, French, or English). A French cemetery on what is now the island of Cape Breton in Nova Scotia (NS) at the Fortress of Louisbourg (Block 3 cemetery; 1713 to 1723) was associated with the early establishment of the Fort and could only have included immigrants.

Data from these fishing settlements are compared to individuals buried at the contemporary St. John's Royal Naval hospital cemetery in NL (Southside Cemetery;

1750 to ca. 1825) for a snapshot of non-urban and industrial-specific childhood origins that were mainly within Britain (Munkittrick, 2015; Munkittrick et al., 2019). Further historical and archaeological contexts for these sites are included in Appendix 7.2.1. The fisheries were also compared to previously published data from England and European-descent populations in North America and New Zealand across time (Table 4). The null-hypothesis is that there are no differences between Pb exposure in Britain and North American European settlements during the 18th and early-19th centuries. The questions addressed in this study include,

1) Are there any differences in enamel Pb concentrations across the burial sites and regions in 18th and 19th century North Atlantic communities?
2) How did the concentrations in early North Atlantic settlements compare to other previously examined populations?
3) Given the archaeological and historical context, can we say anything about the origins of the anthropogenic Pb?

Table 3. Location and date of all burial sites in Newfoundland (NL) and Nova Scotia (NS) sampled in this study. It also includes the number of individuals sampled and the contextual information. This includes whether they were likely locally born or recent immigrants based on census data and previous isotopic studies (see supplementary materials), the country most closely tied to the settlement, and the type of village or institution associated with the site. Bolded portions of site names are how they are referred to in text.

Site name (Borden or Parks Canada number)	**Location**	**Dates**	**No. of samples**	**Context**
St. Paul's Anglican Church cemetery (CkAh-06)	**Harbour Grace**, NL	1764–1820	8	Locally born English and Irish, large fishing center
Foxtrap-2 burial ground (CjAf-10)	Foxtrap, NL	18th c.–ca. 1896	9	Locally born English, small fishing village
Wester Point cemetery (CjAf-08)	Portugal-Cove, NL	18th–19th c.	3	Immigrants and locally born English, small fishing village
Tors Cove burial ground (ChAf-01)	Tors Cove, NL	1812–late-19th c.	3	Locally born Irish and English, small fishing village
St. Luke's Anglican Church cemetery (ChAl-17)	**Placentia** (Plaisance), NL	16th–18th c.	4	Immigrants and locally born likely French or Basque, possibly English, large fishing center
Block 3 Cemetery (3L)	**Louisbourg**, NS	1713–1723	19	Immigrants from French territories and France, large fishing center
Southside Cemetery or St. John's **Royal Navy** Hospital cemetery (CjAe-54)	St. John's, NL	1750s–ca. 1825	19	Mostly English, St. John's British Royal Naval Hospital cemetery

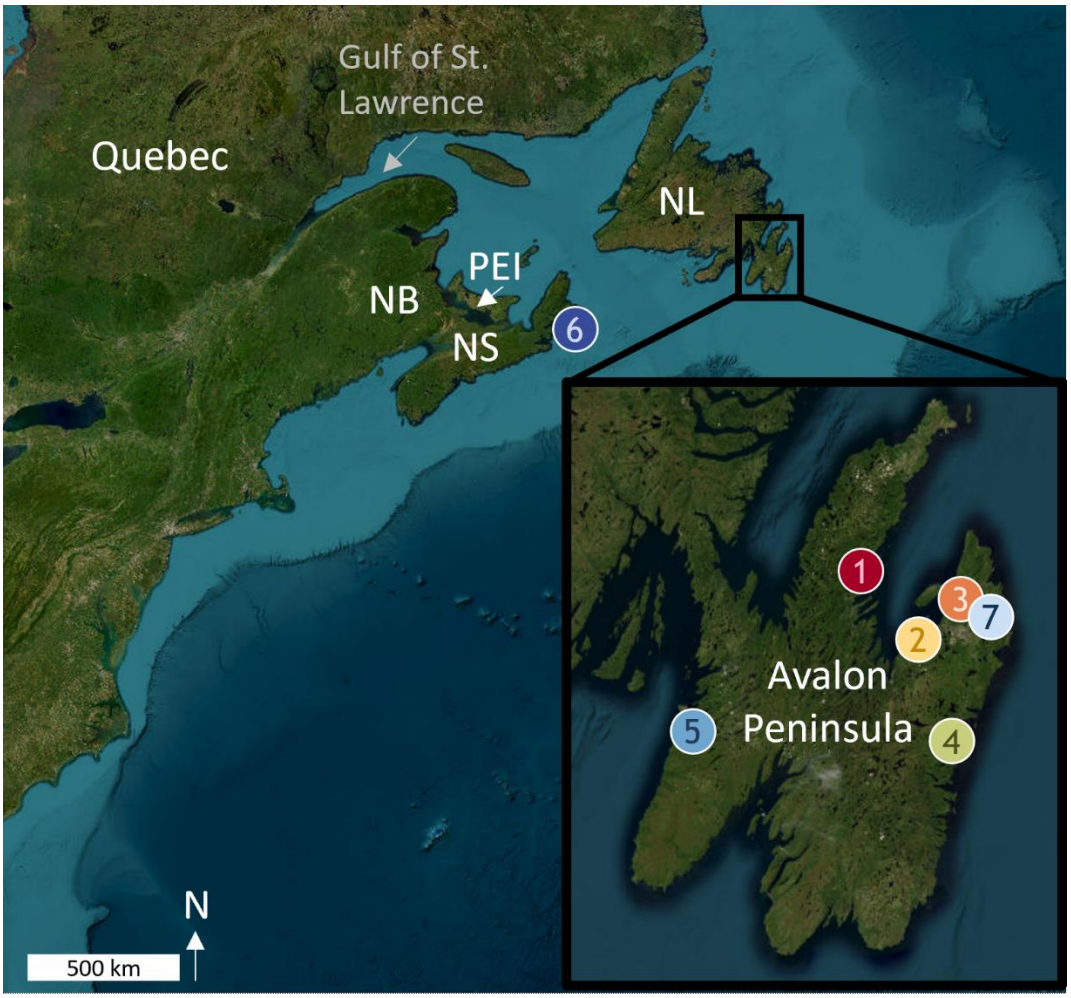

Figure 3. Map of sample site locations. English/Irish fishery sites in Newfoundland include 1) St. Paul's Anglican Church Cemetery, Harbour Grace, 2) Foxtrap-2 burial ground, Foxtrap, 3) Wester Point Cemetery, Portugal-Cove, and 4) Tors Cove burial ground, Tors Cove. Likely French or Basque cemetery is 5) St. Luke's Anglican Church Cemetery, Placentia (Plaisance). The French fishery site in Nova Scotia is 6) Block 3 Cemetery at the Fortress of Louisbourg. The comparative Royal Naval Hospital site in Newfoundland is 7) Southside Cemetery, St. John's. Map made by author using ArcGIS.

Table 4. The summary names of the comparative data used in this paper, including the number of samples and the citations. The Newfoundland (NL) and Nova Scotia (NS) archaeological sites are grouped for Pb concentration statistical comparisons, but the Louisbourg (Block 3 cemetery) is separated in parentheses for Pb isotope comparisons due to statistical differences. Bolded names are how these groups are referred to in text.

Region and period	Pb ppm count	$^{20n}Pb/^{206}Pb$ count	$^{20n}Pb/^{204}Pb$ count	citation
NL and NS Fisheries (Louisbourg is separate for isotopic analyses)	46	26(19)	26(19)	This publication
Southside Cemetery (Royal Navy, 1750s–1825)	19	19	19	This publication
Prehistoric England, Ireland, and Scotland (Neolithic, Bronze Age, Iron Age)	61	-	-	Budd et al., 2000a; Montgomery, 2002; Budd et al., 2004; Montgomery et al., 2010, Moore et al., 2020
Roman Britain (1st–4th c. CE)	38	-	-	Montgomery, 2002; Budd et al., 2004; Montgomery et al., 2010; Shaw et al., 2016; Moore et al., 2020
Early Medieval England and Scotland (5th–12th c. CE)	107	-	-	Montgomery, 2002; Budd et al., 2004; Montgomery et al., 2010; Evans et al., 2018a
Viking Scotland, Ireland, and England (8th–11th c. CE)	43	-	-	Budd et al., 2004; Montgomery et al., 2010; Harris et al., 2017; Evans et al., 2018b
Late Medieval Atlantic Archipelago (13th–15th c. CE)	27	8	5	Montgomery, 2002; Budd et al., 2004; Knüsel et al., 2010; Montgomery et al., 2010
Newton Plantation, **Barbados** (1680–1830 CE)	24	17	17	Laffoon et al., 2020
Otago, **New Zealand** (1860–1890 CE)	19	19	19	King et al., 2020
New Haven, **Connecticut**, USA (1833–1851 CE)	3	4	4	Aronsen et al., 2019
Industrial England (late-18th– mid-19th c. CE)	83	34	34	Millard et al., 2014
Industrial USA (mid-19th – late-19th c. CE)	27	16	31	Fitch et al., 2012; Keller et al., 2016

3.2 Background

Understanding cultural influences on Pb sources requires some context of the patterns of settlement in the region. Within the Beothuk and Mi'kmaq's ancestral homelands, the islands of Newfoundland and Cape Breton have rich marine life, which was also a staple within the Indigenous populations' diets (Pastore, 1989). These marine resources also became the foundation of the European-based fishing industry, where, beginning in the late-15th century, crews from ports along the western coast of Europe were fishing off the coast of Newfoundland to supply codfish for European, Mediterranean, and Caribbean markets (Candow, 2001; Pope, 2008; Holm et al., 2019). The fishery quickly grew with Basque (French and Spanish), Breton, Norman, Iberian, and West Country English based sailing vessels seasonally fishing the rich shores of Newfoundland (Cell, 1969; Pope, 2008). The earliest fishery tended to be inshore or coastal, known as the "dry" fishery, which required safe harbour and shore access to dry and lightly salt the fish. (Candow, 2001). The French began fishing the Grand Banks by the late 16th century, participating in the "wet" fishery, where they salted the fish onboard vessels, but the English did not frequently participate in this type of fishery until the early 18th century (Pope, 2004:15; Janzen, 2008; Pope, 2009:141). These types of fisheries cannot be divided by nationality, since the notion of the dry fishery being English and the wet fishery being French is a misrepresentation of history (Pope, 2008:38). Shore locations were utilized on a first-come-first-served basis, and European-based ports geographically closer to Newfoundland could access more desirable areas. By the end of the 16th-century, fishing stations established by European fishers from the same regions

tended to group in certain areas on the island (Candow, 2001; Pope, 2009). English, mostly from West Country and London, fished along the Eastern Avalon Peninsula (Cape Race to Cape Bonavista), and the French were in the Petit Nord (Bonavista to Quirpon) and Côte du Chapeau Rouge (South coast of Placentia Bay to Plaisance). The French and Basque were also present in the Gulf of St. Lawrence and Cape Breton (Hiller, 1996; Mannion, 2001; 2004; Pope, 2009).

The fishery was a lucrative industry and many nations commonly at war with one another fished in the same waters. French and English military, navy, and privateers helped protect the nations' interests (Hiller, 1996; Pritchard, 1999; Matthews, 2001; Landry, 2004). While the fishery remained largely migratory until the early 19th century, some settlements developed in major harbours. For the English, this included St. John's and Harbour Grace (Cadigan, 1995; Candow, 2010), and for the French it included the fortified settlement of Plaisance established in 1655 (later renamed Placentia by the English) in the location that the Basque used during the previous century (Loewen and Delmas, 2012; Landry, 2018).

This settlement pattern changed drastically in the 18th century. After the French lost most of their eastern North American territory following the signing of the Treaty of Utrecht in 1713, they moved their fishing headquarters from Plaisance to Île Royale (modern-day Cape Breton, Nova Scotia) and established the Fortress of Louisbourg (Head, 1976; Johnston, 2004). France retained migratory fishing rights in Newfoundland, focusing their fishery along the "French Shore" (Bonavista to Point Riche), though they were not permitted to settle unless they swore allegiance to the British crown (Hiller, 1996; Candow, 2001). Newfoundland's English settlements and permanent population

also increased during this period (Head, 1976; Handcock, 2003:98-104). No longer was the fishery exclusively a first-come-first-serve basis, as a sense of property ownership developed with fishers staying year-round (Janzen, 2008). Settling permanently in the early 17th century was partly to assert sovereignty but more directly because of the need to harvest lumber during the winter for boat and stage production and to maintain and protect fishing rooms (Matthews, 1973; Pope, 1993). The cause of the surge in population during the mid to late 18th century is multi-facetted. It was influenced by merchants transporting not only fish but extending credit to locals in exchange for goods (Janzen, 2008). The fishery was also diversified to include seals and salmon caught during spring, which effectively extended the profitable season (Candow, 2001; Janzen, 2008). Finally, the opening and closing of various markets with the American Revolution and wars between France and Great Britain around the turn of the 19th century created a greater demand for salt fish in European markets. Yet, these wars also created more danger and uncertainty in the Atlantic crossings, thereby promoting permanent, or at least temporary/seasonal, settlement (Janzen, 2008).

While there was a growing "permanent" population, it is best to recognize that it was still temporary for many, as some would eventually leave the island for New England, Canada, or even back to Europe. While the fishery, many of which were based out of West Country, did establish crews from their local English port, this was not exclusively so (Mannion, 2001). English vessels frequently stopped in Ireland for provisioning (mainly in Munster ports or Dublin) starting in the late 17th century. Along with these provisions came cheap untrained Irish labourers who, by the 18th century, became a prominent source of labour for the fisheries (Pope, 2004:161-93; Pope, 2009:146). The

early 19th century saw an almost complete collapse in the migratory fishery, in part due to embargos and war, which resulted in almost all fishing vessels being locally owned and operated (Ryan, 1983; Handcock, 2003:96). The population of Newfoundland increased from about 11,400 in 1797 to 19,000 in 1803, by which point half of the permanent population were Irish (Ryan, 1983; Mannion, 2001). Newfoundland's population continued to increase throughout this period, both through local births and additional migration, increasing between 1815 and 1891 from about 40,500 to 202,000 individuals (Ryan, 1971:209).

Tensions between the Roman Catholic Irish and Protestant English meant that many smaller communities were segregated according to religion. However, the larger towns of Harbour Grace and St. John's were more heterogeneous. Due to the shift in origins of the labour force, the composition of some communities changed over time. For example, Tors Cove (or Toads Cove) was initially English, but many Irish came in the later-18th century (Mannion, 2001). English, Irish, and French communities were never wholly homogenous. This mixed population was also true for Plaisance and early Louisbourg, with French from culturally distinct areas such as the Basque country, Normandy, Brittany, New France (Quebec and Acadia), and regions around the ports of La Rochelle, Bordeaux, St. Malo, and Nantes. Louisbourg also included enslaved African labourers and individuals from Ireland and England (Johnson, 1984; Turgeon, 2000; Nadon, 2004; McLeod-Leslie, 2014; Landry, 2018).

While these fishing communities developed permanent settlements, many residents did not occupy them year-round; exceptions included major ports like Harbour Grace, St. John's, and Plaisance/Placentia (Mannion, 2004). Many permanent residents would move

into more sheltered locations during the fall and winter, either further inland or to different areas along the coast (Smith, 1995; Venovcevs and Gaulton, 2018; Gaulton and Losier, 2020). This would protect them from the harsh coastal climate and facilitated lumber harvesting needed for the spring fishery since areas close to the coast would be exhausted of trees quickly (Smith, 1987; Nemec, 2006 cited in Gaulton and Losier, 2020). During this time, settlers would also hunt and trap to supplement their provisions (Smith, 1987).

To understand how individuals were exposed to Pb in these settlements, it is essential to consider the three general origins of exposures: environmental, cultural and physiological (see Chapter 2). The environmental Pb pathway includes exposure from dust or erosional materials due to natural or anthropogenic processes that increase concentrations in soil or air. The mining and smelting of Pb results in dust and particulate air contamination, which settles onto surfaces and soil where it can be ingested or incorporated into food crops (Gulson and Wilson, 1994). The inhaled or ingested particulates and food constitute the environmental pathway.

The second contamination pathway is via cultural materials. Mined Pb is often further processed and incorporated into food and drink storage and consumption vessels as an alloy, including pewter (a Pb-alloy), or oxide into the ceramic glaze (e.g., Beagrie, 1989; Tite et al., 1998). Pb pipes or Pb-containing solder can also be a source, particularly with soft water (Crawford & Clayton, 1973). The Pb from those vessels can then leach into the food or drink that is prepared, stored, or served within them. Pb can also be directly consumed with harvested/hunted food from animals if all the Pb shot was not removed before preparation, becomes stuck within the gastrointestinal tract of the

consumer, and is not passed immediately (Lévesque et al., 2003:694; Knott et al., 2010:98).

The final pathway of exposure is physiological. Mothers can pass on their previous Pb exposure to developing fetuses through maternal transfer or mothers and caregivers to infants via breast milk (Gulson et al., 2003:99-100). In summary, the environmental pathway in anthropogenic environments produces local Pb isotope ratios similar to local ore sources. It should be noted that local ore values are not necessarily the same as surface bedrock or soil, since galena is often from epithermal deposits or later emplacements (Milot et al., 2021). Pb from the cultural pathway (i.e., Pb-containing materials) can be transported to distant locations, making it challenging to separate the movement of people and the movement of materials as the cause for differences in Pb concentrations and isotopic values. This complex pattern of movement during early North American settlement by Europeans, changing ore sources during industrial development, and the movement of cultural materials is vital for providing context for interpreting the Pb concentration and isotope ratios in archaeological samples from this period.

3.3 Methods

Samples were collected from 46 individuals buried at five fishing settlements with strong ties to England/Ireland and Basque country/France, and another site with an uncertain date associated with Basque, French, and later the English fisheries. An additional 19 individuals buried at a Royal Naval hospital cemetery in Newfoundland were sampled as a comparative population from a non-urban specific contemporary

context, many of whom were likely born in Britain, though not exclusively (Table 3; Figure 3). See supplementary materials 7.2.1 for full archaeological site backgrounds.

Enamel of one tooth, preferably the 2nd molar, was sampled from each individual at the Memorial Applied Archaeological Science (MAAS) laboratory. Enamel samples were chosen from areas on the teeth without pathologies or obvious cracks. The summarized sampling procedure was as follows: the sample location's surface was cleaned to at least 100 μm using a tungsten carbide or diamond burr, approximately 50 mg of an enamel chunk was extracted using a diamond-coated blade, and all cut/altered surfaces were cleaned and dentine was removed using a tungsten carbide burr. Finally, the sample was ultrasonicated with deionized water and then rinsed with high purity acetone (Montgomery, 2002). The samples were then moved to the Radiogenic Isotope Laboratory at The Earth Resources Research and Analysis Facility (TERRA), Department of Earth Sciences, Memorial University to dry and for further sample digestion and Pb extraction in a HEPA filtered laminar-flow clean hood.

Samples were weighed in cleaned Savillex™ (PFA) 3 mL or 7 mL vials then digested with 2 mL of 8 M HNO_3 at 100°C for 1–2 hours. One aliquot of 0.5 mL was used for concentration analysis, the other 1.5 mL for Pb isotope analysis. Pb concentration was analyzed using the 0.5 mL aliquot at one of two laboratories, the University of Ottawa (using an Agilent ICP-QQQ) or the TERRA facilities at Memorial University (using a Perkin-Elmer Elan DRCII ICP-Q-MS or a Neptune MC-ICP-MS). The 1.5 mL aliquot was added to pre-cleaned 1 mL pipet tip columns fitted with a porous polyethylene frit for Pb extraction chromatography using Sr-spec resin (Charlier et al.,

2006; Pin et al., 2014) and Pb isotope ratios were determined on a Thermo Neptune MC-ICP-MS. See Appendix 7.2 for full method procedures.

Pb concentration data at the Memorial University TERRA facility and uOttawa was evaluated by comparing values obtained to known Pb concentrations of reference standards USGS T-143 (83.40 ppb), SRM 1400 (Bone Ash; 9.07 ppb), and uOttawa's internal standard (12.5 ppb). The standard values obtained in this study were 105.6% for USGS T-143 (88.08 ± 2.94, $n = 8$), 90.5% for SRM 1400 (8.21 ± 0.12, $n = 5$), and 100.3% for uOttawa's internal standard (12.54 ± 0.68, $n = 4$). Blank contributions were negligible on Pb concentration analyses (<2% of final concentrations), particularly for those analyzed at the TERRA facility. The one exception was for Burial 5 from Wester Point cemetery where the blank contributed ~23% of the final concentration, but all samples were blank-corrected for the uOttawa samples (full details in Appendix 7.2.4.2).

Pb isotope ratios for SRM 1400 were compared to most probable values published by Hinners et al. (1998) and values obtained in this study were 100.002% for $^{206}Pb/^{204}Pb$ (mean ± 2σ, $n = 25$; 18.3662 ± 0.0210), 100.034% for $^{207}Pb/^{204}Pb$ (15.6773 ± 0.0162), and 100.072% for $^{208}Pb/^{204}Pb$ (38.6378 ± 0.0560). Since Tl correction was not performed for the Pb isotope data, the precision and accuracy are important for interpretation. Full error details for all isotopic ratios are provided in the Appendix 7.2.4.3.

Data were compared to previously published Pb concentration and isotope data, as seen in Table 4 (full details in Appendix Table 17 and Table 18). If multiple teeth per individual were sampled, second molars or teeth that develop at a similar age were considered. For the King et al. (2020) paper, the samples from deciduous teeth, as assumed by skeletal age, were removed for statistical analysis. King et al. (2020)

identified that all but one had high Pb concentrations (>500 ppm), were very cracked, friable, and were excavated from a high-water table area with low pH and poor drainage. This would create a higher potential for diagenetic alteration in the deciduous teeth that are smaller, and therefore have a larger probability of cracking to contribute to surface area compared to core enamel material and may account for the high concentrations seen. All statistical analyses were performed with R (R Core Team, 2022) and the Kruskal-Wallis test used the core program (R Core Team, 2022). When differences were found, a Dunn (1964) Kruskal-Wallis multiple comparison test was performed, and *p*-values were adjusted with the Benjamin-Hochberg method (Ogle et al., 2020). While statistical tests were performed, this was done with consideration of the American Statistical Associations statement on *p*-values (Wasserstein & Lazar, 2016; Wasserstein et al., 2019). As such, visual interpretation using box and scatter plots was also performed and figures are presented in both the results and appendices.

3.4 Results

3.4.1 Diagenesis

When sampled correctly, enamel, with its low porosity and highly mineralized nature, is minimally susceptible to soil-derived contamination from the burial environment (Budd et al., 2000; Montgomery, 2002 see Chapter 2). While a comparison of the Pb isotope ratios in burial soil, dentine, and enamel can indicate if diagenesis has occurred (e.g., Montgomery et al., 2000; Montgomery, 2002; Simonetti et al., 2021), this is only possible to identify if enamel values fall outside of what is expected. In this case, data from burial soil were not available. Studies have found that enamel retains isotope

ratios representing *in vivo* exposure when sampled from well preserved teeth, including from entirely lead coffins (Montgomery et al., 2010). There is a recent trend in research to identify whether diagenetic alteration occurred by analyzing enamel concentrations of proxy elements (Beherec et al., 2016; Kamenov et al., 2018; Simonetti et al., 2021; Kamenov & Krigbaum, 2023). Kamenov et al. (2018) suggested that diagenetic alteration of the enamel is identifiable using concentrations of elements compared to a maximum threshold concentration created using data from modern individuals. This specific method suggests that if the concentration to maximum threshold concentration (MTC) ratio is greater than one, some alteration had occurred; if many element's MTC were high, there was a concern for major alteration (Kamenov et al., 2018).

Memorial University's TERRA facility's methodology analyses seven of the ten suggested elements (Ca, V, Mn, Fe, La, Ce, and U; missing Nd, Dy, Yb, and Th). Data from St. Luke's, Block 3, and Southside cemeteries were available for this comparison, and full results are available in the Appendix as Table 28). Most of the samples had MTC values below 1.0 for all elements. For some individuals analysed, elements had concentrations below the limit of detection. However, the MTC ratio was calculated assuming the smallest sample size and worst methodological limit of detection over the four days of analyses.

A few individuals with single elements had concentrations above the MTC. This included the elements V, Mn, and Ce. It should be noted that while several individuals from Block 3 had slightly high Mn concentrations, the analysis day for most of these samples had a higher Mn error (10.8%) than other analysis runs. This may, in part, account for the borderline Mn values (MTC values of 1.0 to 3.5) within the samples.

Three individuals from the Royal Navy (B1) and Louisbourg (F7 and F19) cemeteries had multiple elements above the MTC. However, all were borderline, and one of these three individuals had a Pb concentration well above the expected natural Pb concentrations, and Pb isotope ratios were consistent with other individuals. Another individual from Louisbourg (F10) had two elements above the MTC and low Pb so their concentration and isotope ratios may possibly be affected. In turn, efforts were made not to overinterpret the isotope ratios. The Kamenov et al. (2018) methodology assumes that if one or multiple elements are affected, Pb is likely as well. However, based on previous research, we know that elements are not necessarily co-affected by diagenesis (Radosevich, 1993). Given this and the long-term research showing the resistance of enamel to diagenetic change, we decided to include the Pb concentration and isotope ratios for these four individuals in further analyses of the data, despite their borderline MTC values.

3.4.2 Pb concentrations

Individuals from the six fishing populations have mean concentrations in the range of 2.7 to 10.1 ppm and medians from 2.6 to 10.8 ppm (Table 5, additional information available in Appendix Table 20). Within these fisheries, the lowest concentration was 0.1 ppm (Wester Point B5) and the highest was 28.9 (Harbour Grace, F7). There was no difference in concentrations between the fishery populations ($p = 0.201$, Kruskal-Wallis chi-squared = 7.2874, $df = 5$). Therefore, data from the fishing communities were pooled for future comparisons to other burial sites and regions.

The Pb concentrations from the collective fisheries were compared to samples from the Royal Navy in St. John's (Southside cemetery). The mean concentration of the British Royal Naval population was 6.0 ppm, with a median of 2.9 ppm, and ranged from 0.2 to 24.6 ppm (B10 and A6, respectively). The fisheries were also compared to archaeological sites in the Atlantic Archipelago (the archipelago includes: Great Britain, Ireland, Isle of Mann, Inner and Outer Hebrides, Orkney, and Shetland) dating from the Neolithic to 19th century CE, or to sites from other regions that date between the 16th and 19th centuries CE to see if they lived under similar Pb exposure situations (Table 5). There were differences between these populations ($p < 0.001$, Kruskal-Wallis chi-squared = 320.43, $df = 11$), but since these were populations from different periods, we could reject that these were coming from the same exposure situations. In turn, a Dunn Test was performed to identify what pairs of sites had different exposures (Table 5). The fishing populations differed from Industrial England and USA, which had higher concentrations, and from Prehistoric, Early Medieval, and Viking populations in the Atlantic Archipelago, with much lower concentrations. Therefore, the fisheries were from similar exposure conditions to the other sites, including Roman-Britain, Late Medieval Atlantic Archipelago, Newton Plantation cemetery Barbados, New Haven Connecticut, and the Southside cemetery ($p > 0.256$) (Figure 4; Table 5).

3.4.3 Pb isotope comparisons

While the Pb isotope ratios of the fishing populations are quite similar, there were differences between the archaeological sites in $^{208}Pb/^{206}Pb$ ($p < 0.001$), $^{207}Pb/^{206}Pb$ ($p = 0.003$), $^{208}Pb/^{204}Pb$ ($p = 0.091$), $^{207}Pb/^{204}Pb$ ($p = 0.625$), and $^{206}Pb/^{204}Pb$ ($p = 0.008$) (Pb

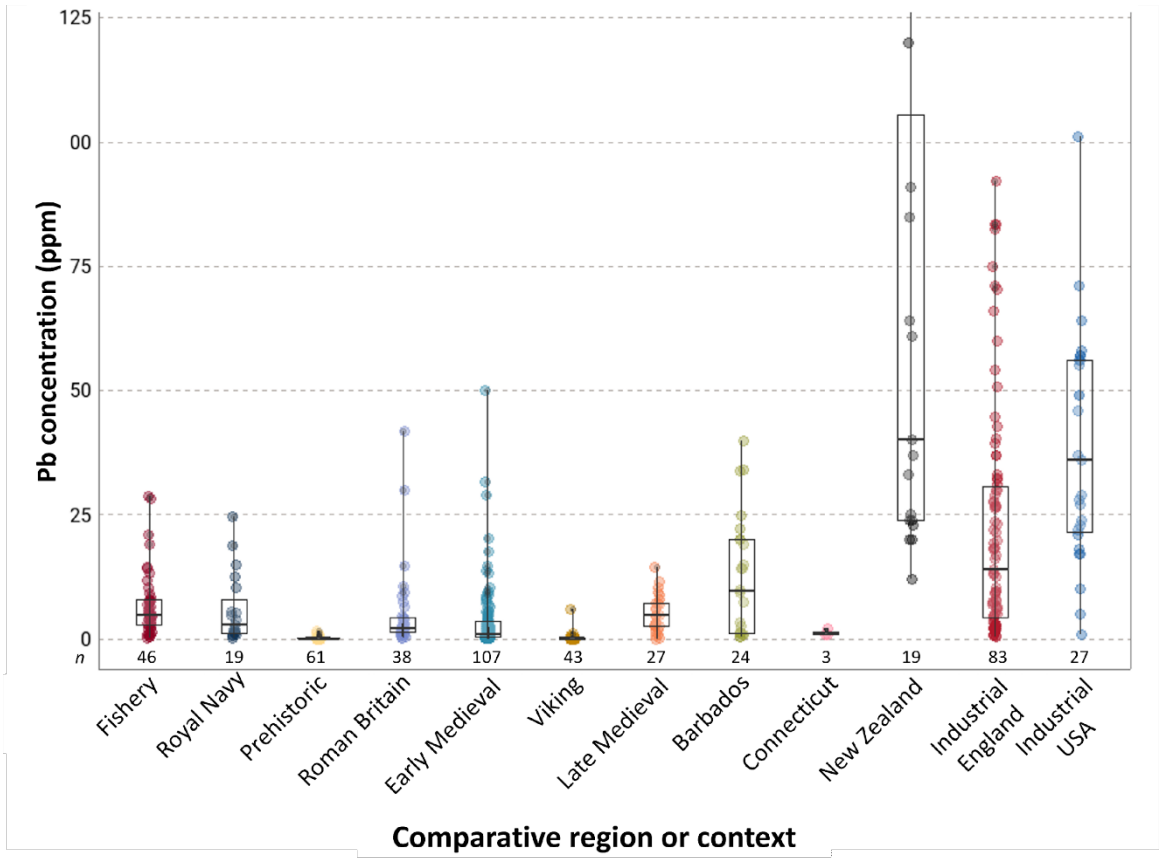

Figure 4. Box-plot of Pb concentration of comparative archaeological sites to summarized fisheries sites. Summarized **Fishery** and **Royal Naval** data are from this study. Other groupings are: **Prehistoric** England, Ireland, and Scotland; **Roman Britain**; **Early Medieval** England and Scotland; **Viking** Scotland, Ireland, and England; and **Late Medieval** Atlantic Archipelago; 16–19th century Newton Plantation, **Barbados**; 19th-century New Haven, **Connecticut**; 19th century **New Zealand**; **Industrial England**; and **Industrial USA**. Bolded names are those used in the figure. The y-axis is stopped at 125 to allow for lower concentrations to be visible, but this cut off some of the NZ samples. All citations are available in Table 4.

Table 5. Summary statistics of Pb concentration (ppm) from all analyzed burial sites. English/Irish or French fishing cemeteries in NL and NS are indicated in parentheses. Basque, French, and English used St. Luke's Anglican Church cemetery, Placentia. The summarized fisheries concentrations are followed by the comparative sites or groups. The Southside Cemetery, St. John's, is associated with the British Royal Navy. Also included are previously published materials. *P*-values from a Dunn Test result for Pb concentrations of the summarized fishery populations compared to other sites are indicated. Citations for comparative sites are available in Table 4.*Only samples that, based on skeletal age, were from permanent teeth were included for statistical comparison.

Site/Comparative Group	*n* =	mean	1σ	median	min	max	IQR	*p*-value
St. Paul's Anglican Church Cemetery, **Harbour Grace** (Eng./Irish)	8	8.89	8.41	6.15	2.8	28.9	2.78	-
Foxtrap-2 burial ground (Eng.)	9	5.6	1.68	4.9	3.1	7.9	2.8	-
Wester Point cemetery (Eng./Irish)	3	8.07	9.51	5.5	0.1	18.6	9.25	-
Tors Cove burial ground (Irish/Eng.)	3	2.67	1.80	2.6	0.9	4.5	1.8	-
St. Luke's Anglican Church cemetery, **Placentia** (Basque/Fr./Eng.)	4	10.1	4.64	10.8	4.3	14.4	6.12	-
Block 3 cemetery, **Louisbourg** (Fr.)	19	6.42	7.63	3.1	0.2	28.3	6.9	-
Summarized fisheries	46	6.90	6.60	4.8	0.2	28.7	5.03	-
Southside Cemetery (St. John's Royal Naval Hospital)	19	5.97	6.99	2.9	0.2	24.6	6.75	0.586
Prehistoric England, Ireland, and Scotland	61	0.188	0.288	0.09	0.003	1.7	0.1	<0.001
Roman Britain	38	4.99	8.15	2.18	0.21	41.8	3.0	0.256
Early Medieval England and Scotland	107	3.69	7.0	0.99	0.09	50.0	3.34	0.001
Viking Scotland, Ireland, and England	43	0.322	0.894	0.11	0.02	5.88	0.205	<0.001
Late Medieval Atlantic Archipelago	27	5.05	3.7	4.68	0.02	14.5	3.62	0.605
Newton Plantation, Barbados	24	12.4	12.1	9.6	0.4	39.9	18.9	0.595
Otago, New Zealand*	19	89.7	133.0	40	12	608	81.5	<0.001
New Haven, Connecticut, USA	3	1.33	0.577	1	1	2	0.5	0.338
Industrial England	83	22.2	22.3	14	0.47	92.2	26.2	0.016
Industrial USA	27	38.3	23.1	36	1	101	34.5	<0.001

isotope results for each individual are found in the Appendix as Table 26 and Table 27, and Kruskal-Wallis test results in Table 29). A Dunn-test showed there were no differences between Pb sources at the sites ($p > 0.068$, all results are found in the Appendix as Table 30) except for $^{208}Pb/^{206}Pb$, $^{207}Pb/^{206}Pb$, and $^{206}Pb/^{204}Pb$ values for Louisbourg and Placentia cemeteries ($p = 0.005$, 0.009, and 0.018, respectively) and Louisbourg and Foxtrap only for $^{208}Pb/^{206}Pb$ ($p < 0.001$). These differences do not fully portray the overlap in some of the values of individuals (Figure 5). Despite the population at Louisbourg differing from Placentia and Foxtrap, there is overlap in the Pb isotope ratios of some individuals within these site clusters as well as the other fisheries.

Since all the fishery sites could not be pooled, the Louisbourg cemetery was left separate for broader comparisons. The Pb isotope ratios from both groups were compared to the St. John's Royal Naval Hospital cemetery in NL, as well as other contemporaneous sites with human enamel data, including New Zealand, urban industrial England, and three industrial USA cemeteries: two in areas associated with mining (Colorado – Keller et al., 2016; Illinois – Fitch et al., 2012) and the other from an urban immigrant community (New Haven, Connecticut – Aronsen et al., 2019). Despite being contemporaneous, the Newton plantation burials from Barbados included many individuals suspected to have spent their childhoods in Africa, which is not a relevant region of origin for these fishery populations. Therefore, only those identified by Laffoon et al. (2020) as not having spent their childhood in Africa were included for isotopic comparative analyses. There were differences between groups in all isotope ratios (p-value < 0.001, available in Appendix Table 31), suggesting that Pb was not sourced from similar mines or mines with a similar composition and formation age. Pb isotope ratios

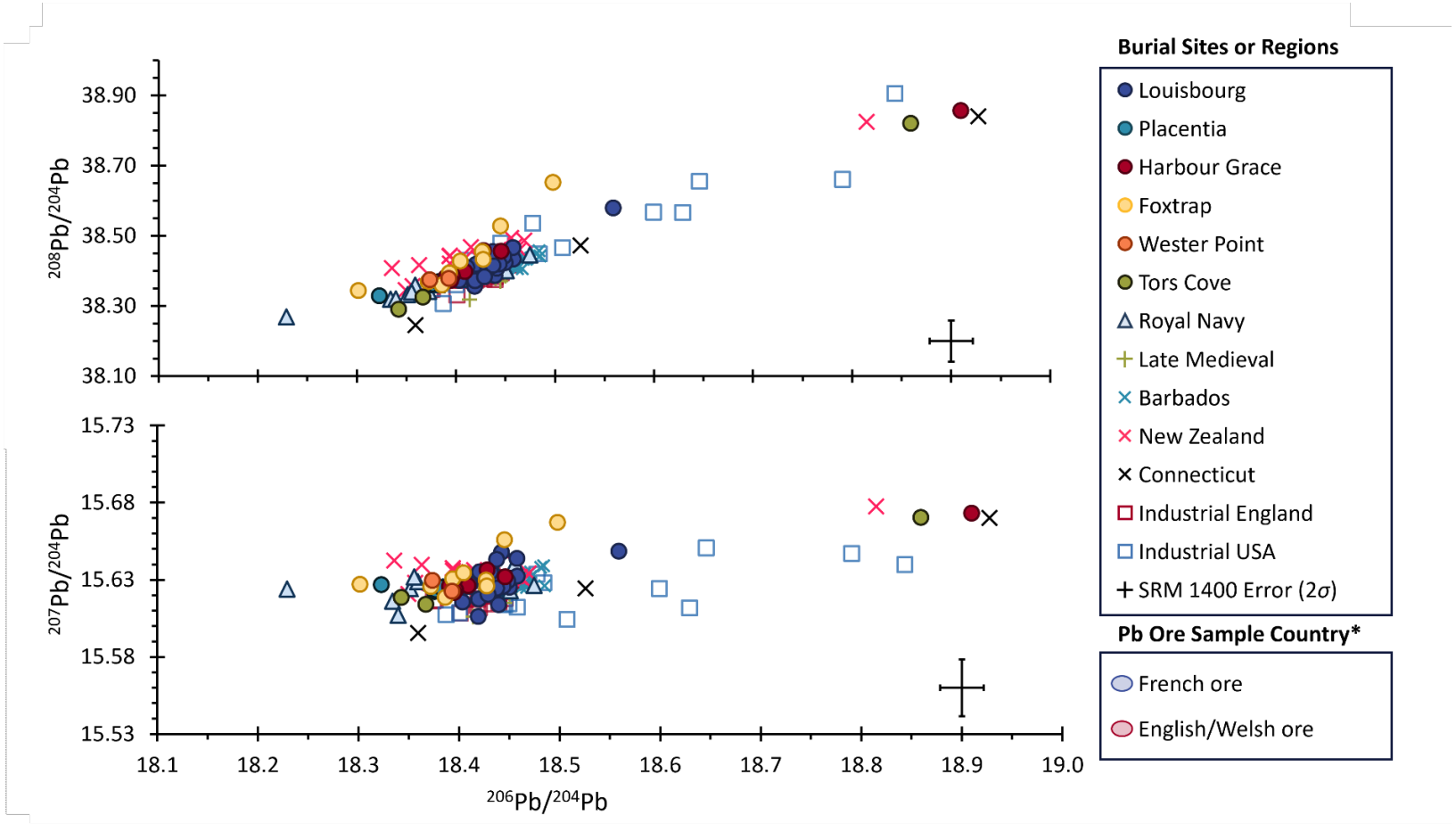

Figure 5. Comparison of $^{206}Pb/^{204}Pb$ to $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ between Newfoundland fisheries, Royal Navy, and other previously published contemporaneous sites. Pb ore field isotope value ranges for France, English, and Wales are outlined. All comparative citations are available in Table 4 and Pb ore citations are in Table 45. Note that the axes are restricted, cutting off some individuals from Industrial USA with higher isotope values. Between assay variability is presented as 2σ as determined by analysis of SRM 1400.

are visually presented comparing and $^{208}Pb/^{204}Pb$ and $^{207}Pb/^{204}Pb$ to $^{206}Pb/^{204}Pb$ (Figure 5) (the comparison of $^{208}Pb/^{206}Pb$ to $^{207}Pb/^{206}Pb$ values are presented in Appendix Figure 24).

Since there is not as much power in these statistical tests to show differences, we are more likely to make a type II error and wrongfully conclude that there was no difference between sites when one may exist. Detecting differences with low sample sizes suggest that these differences are real. Not all sites differed in each Pb isotope ratio (all comparisons available in Appendix Table 32). There were no differences between Louisbourg and the Royal Navy, New Haven Connecticut, or Industrial England ($p > 0.075$), nor between the broader fishery and the Royal Navy or Connecticut ($p > 0.170$). However, Louisbourg was different in all isotope ratios compared to Industrial North America ($p < 0.024$).

There are two methods of comparison employed between the fisheries, Louisbourg, and the other comparative sites/regions: statistical and visual. Interpretation of p-values must be done within context (Wasserstein & Lazar, 2016) and this is particularly important in Pb analyses since the ratios do not necessarily co-vary, errors can be at the point of interpretation, and bioarchaeologists commonly seek to gain information on the individuals rather than exclusively at the population level. The range of values within clusters of the Fisheries are visually most similar to 18th and early 19th century Royal Navy in NL (this study), the mid-19th century New Haven, Connecticut cemetery (Aronsen et al., 2019), or mid to late 19th century Otago, New Zealand (King et al., 2020). The outliers from New Zealand and Connecticut also overlap with the outliers seen in the fisheries at Harbour Grace and Tors Cove. While some of the values overlap, even

considering error, there are also differences to the enslaved laborer cemetery in Barbados (Laffoon et al., 2020), Late Medieval Atlantic Archipelago (Montgomery, 2002; Budd et al., 2004; Knüsel et al., 2010), and Industrial England (Millard et al., 2014), which have more constrained values (Figure 6). The two cemeteries in 19th century USA within mining communities had some individuals with similar values to Britain (Fitch et al., 2012; Keller et al., 2016), but most of the $^{20n}Pb/^{204}Pb$ values are much higher than those from Britain.

Fisheries had a much wider range of Pb isotope ratios when compared with Louisbourg, especially when considering the Block 3 outlier was not from anthropogenic Pb exposure due to the low Pb concentration. The tight cluster is very similar, particularly when considering analytical error, to Late Medieval Atlantic Archipelago (Montgomery, 2002; Budd et al., 2004; Knüsel et al., 2010; Millard et al., 2014), Industrial England (Millard et al., 2014), and the enslaved laborer cemetery in Barbados (Laffoon et al., 2020). The only outliers for Late Medieval Atlantic Archipelago were due to non-anthropogenic Pb exposure. The Royal Navy (this study), Connecticut (Aronsen et al. 2019), and New Zealand (King et al., 2020) have much larger ranges than the main cluster of Louisbourg. The Industrial USA cemeteries had some similar values (Fitch et al., 2012; Keller et al., 2016), but like the fisheries samples, most values are much higher.

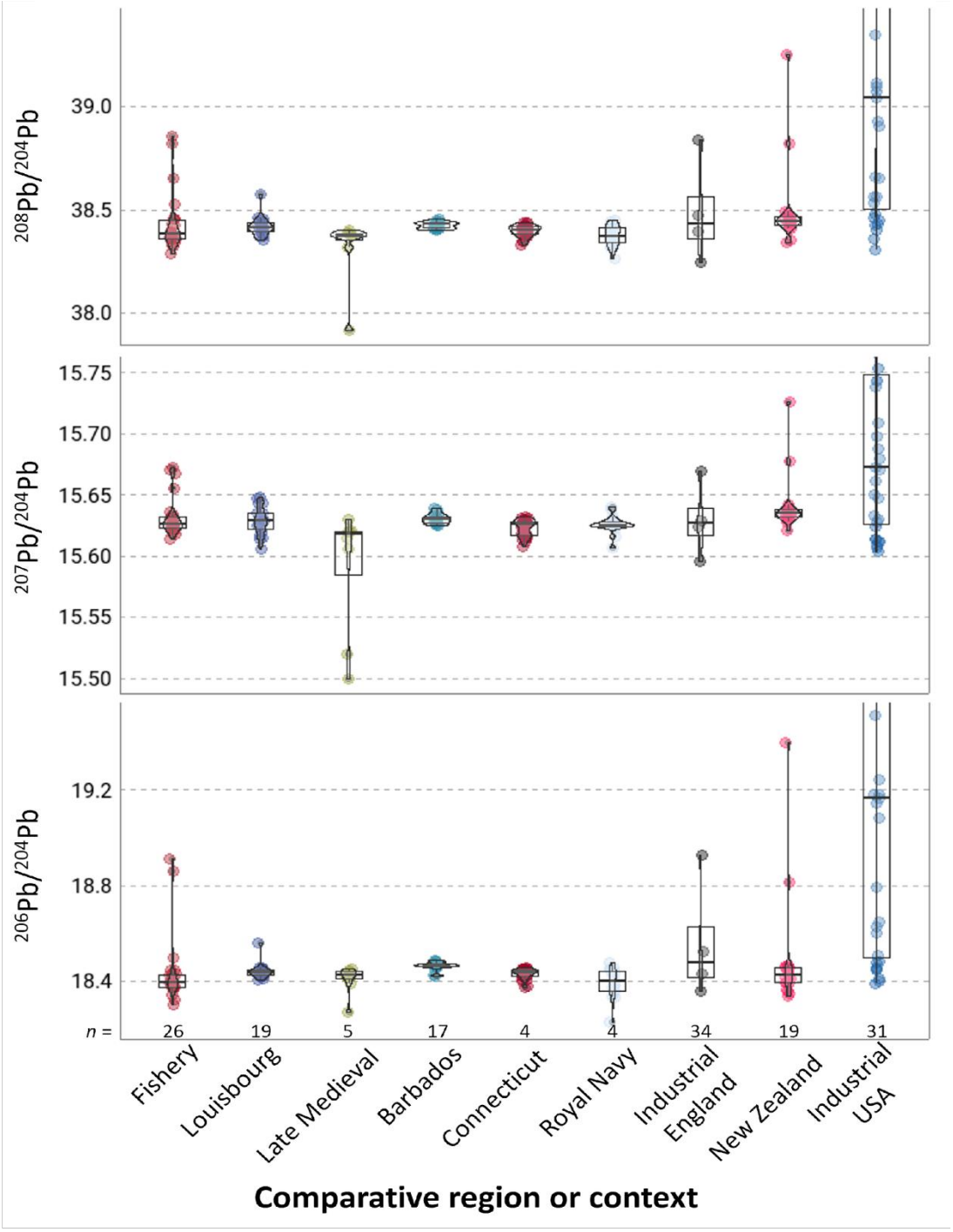

Figure 6. Violin box plots of Pb isotope distributions from the NL fisheries, Louisbourg, and Atlantic World comparative sites (see Table 4 for citations). Note that the axes are restricted, cutting off the full distribution of Industrial USA values.

3.5 Discussion

3.5.1 Anthropogenic Pb exposure

The Pb concentrations of individuals show only slight variation between the fishing sites sampled. While established for Britain, Montgomery et al. (2010) and Millard et al. (2014) found that concentrations above 0.8 ppm could not be explained by exposure to natural environmental Pb alone. When the fisheries sites are combined, 91% (n = 42) of individuals have concentrations above 0.8 ppm. The only individuals below this threshold are from Wester Point (B5) and Louisbourg (F2, F10, and F20). It should be noted that having concentrations below 0.8 ppm does not preclude the possibility of exposure to some anthropogenic Pb sources (Montgomery et al., 2010). Therefore, the low concentrations do not inherently suggest that they were living in different environments.

The fishery populations' exposure levels were dissimilar to the late-18th- to mid-19th-century English (Millard et al., 2014) or mid- to late-19th-century American (Keller et al., 2016) populations, likely due to the higher Pb exposure from living in industrial contexts. They were also dissimilar to Neolithic, Bronze Age, and Iron Age sites (Budd et al., 2004; Montgomery et al., 2010; Evans et al., 2018a) since the latter exposure was solely from natural Pb exposure. Concentrations are similar to individuals from the Royal Navy (Southside cemetery; this study), Roman Britain (Montgomery et al., 2010; Shaw et al., 2016), Late Medieval Atlantic Archipelago (Budd et al., 2004), and the Newton Plantation (Laffoon et al., 2020). This is not to suggest that these populations were all living in similar Pb environments or using similar Pb products, rather their environments resulted in similar exposure distributions.

The comparison of mean concentrations done through statistical comparisons cannot solely characterize the experiences of individuals within the population. There are large ranges, particularly for industrial period England and USA. Therefore, there is overlap in the ranges between those with industrial exposure and the fisheries. It can be stated that, on the whole, the fisheries populations were not interacting with sufficient Pb to result in overall high concentrations. Similarly, the Prehistoric or Viking sites in the Atlantic Archipelago have a range that extends into the low concentrations seen in the fisheries. Yet, as a whole, the fishers have much higher values suggesting that they were living in an anthropogenically derived Pb environment.

While temporally the fisheries sites overlap with those in Industrial England (Millard et al., 2014), the cultural environments in London and Coventry resulted in many individuals with more Pb exposure. The other sites that were contemporary with the fisheries did, however, have similar concentrations and isotope ranges. This includes the Royal Naval sailors, many of whom likely originated in Britain (see Appendix 7.2.1.7 for discussion on Royal Navy origins), but did not exclusively come from the urban industrial setting as examined by Millard et al. (2014). Since the fishers had similar amounts of Pb exposure during childhood to those who ended up in the Royal Navy, the enslaved labourers at the Newton Plantation (particularly those identified as not originating from Africa; Laffoon et al., 2020), and early 19th century Connecticut (Aronsen et al, 2019), it suggests that the Pb exposure in Newfoundland was broadly similar to the concentrations found in non-industrial environments the Atlantic World during the 17th to mid-19th centuries.

The three pathways of exposure (physiological, environmental, and cultural) can be considered to determine where the Pb exposure of the fisher peoples originated. Physiological pathways were avoided by sampling teeth that form after weaning is complete. Environmental exposure to Pb occurs regardless of cultural contexts, though unless it has been anthropogenically enriched from mining, smelting, or burning processes, it would be in low concentrations. However, as shown by the comparison to non-anthropogenic Pb context populations and the 0.8 ppm cut of suggested at least for Britain, almost all individuals from these fisheries sites had Pb concentrations that were too high to be from natural sources alone. This leaves polluted environmental exposure; however, there was no mining in NL or NS until the mid-19th century (Martin, 1983), with the exception of small-scale NS coal mining in the early 1700s (McIntosh, 2000; Falcon-Lang et al., 2009). Therefore, it could not have been a local environmental contaminant that contributed to fisher peoples' exposure. The NL exception to this was a small Pb outcrop at La Manche (between Ferryland and Tors Cove), where there are records of fishermen using this source to make fishing weights (Martin, 1983). A study of lake cores from around the St. John's area suggest that there was minimal environmental Pb pollution until the early- to mid-19th century, and when it did occur, it was likely caused by burning imported coal (Christopher, 1999). While this suggests that there was some Pb pollution in the environment, it is expected that the size of Pb contained within coal particles are too large to be bioavailable (WHO, 2010; Millard et al., 2014). Therefore, environmental exposure can be excluded for consideration. This leaves Pb-containing cultural materials to explain most of the exposure in locally born individuals.

Many of the cultural materials responsible for local Pb exposures would have been related to food or drink-based contacts. Permanent and migratory fisheries in Newfoundland (including Plaisance, where many individuals in the Block 3 cemetery of Louisbourg may have originated; see Appendix 7.2.1 for site histories) relied on imported goods. While some food was supplemented through kitchen gardens and farming (MacKinnon, 1991), all cooking, storage, and serving dishes were imported for use in the English and French fisheries (Head, 1976; Crompton, 2012; Losier et al., 2018). The imported wares included Pb-glazed ceramics and pewter, but of which would have contributed to Pb exposure. Ceramics are ubiquitous on archaeological fishery sites, but pewter is found less frequently found since it was easily melted down and reused (Pope, 2008).

Due to the transhumant nature of many fishers, there is the potential for changes in Pb availability throughout the year. Many permanent settlers outside of more urban contexts, including their children, were transhumant, moving to more sheltered areas during the late fall to early spring (Smith, 1995). Yet, storage vessels are not commonly found at overwintering sites, with the interpretation being that they were reliant on hunted resources or goods not stored in ceramic vessels (Venovcevs and Gaulton, 2018). However, only four winter houses have been excavated, one from the 17th century and three from the mid-19th century (Venovcevs and Gaulton, 2018), so it is unclear how representative this is of all experiences. Since these were more ephemeral dwellings, Pb pipes and cisterns would not have been used for transport and storage of water. Granted, it is unlikely that these were present even in their summer houses during the relevant time periods (Thomas, 1989; Ingram, 2014).

Regardless of the potential lower prevalence of glazed earthenware in winter housing, the general use of Pb-glazed ceramics and even pewter remains a very likely source of exposure. Practices such as cooking and storage in an acidic liquid like vinegar, result in significant leaching of Pb from the glaze (González de Mejía and Craigmill, 1996; Tunstall and Amarasiriwardena, 2002; Rasmussen et al., 2022). While there were also serving dishes that were either Pb-glazed or Pewter (a Pb alloy) and are a potential vector, the relatively shorter time for exposure means that they are less likely to contribute than cooking or storage vessels. It must be stipulated that Pb-glazed cooking and storage vessel use decreased in the 19th century, as did the amount of Pb within many glaze recipes. Therefore, these materials may have contributed less to overall exposure. Another Pb source from drink was the imported rum, which was often distilled using pipes containing Pb. Rum was imported to Newfoundland from American colonies and the Caribbean (Head, 1976); however, it is a less likely vector of exposure to these individuals at the young age of tooth formation.

Another means of exposure, particularly in the 19th century, were the myriad of medical treatments that contained Pb, particularly Pb acetate. Many medical treatments used Pb acetate, including as a panacea or 'cure all', but also for specific ailments (Jonasson & Afshari, 2017). These examples are not specific to medical practice in NL, but it is helpful to consider how broadly it was used. It was particularly common in treatment for gastrointestinal related problems including dysentery, cholera, and diarrhea, but also many other complains like typhoid pneumonia, tuberculosis, or even nose bleeds (Jonasson & Afshari, 2017). While not a source for the samples in question since they were children at the time of exposure and maternal/caregiver exposure is avoided through

sampling later mineralizing teeth, Pb acetate was also a common treatment for heavy menstrual flow and uterine hemorrhage (Jonasson & Afshari, 2017).

There are specific vectors of exposure relating to childhood that should be considered, although given the time period and the age of exposure represented by the individuals sampled, they are less likely. Nipple guards used during breastfeeding were commonly made from lead and would have resulted in exposure (Jonassen & Afshari, 2017). Though, this would be avoided in all but two individuals (Harbour Grace NP264B/2 & Wester Point B4) since the teeth sampled formed after they were likely weaned. Hand to mouth practices in infants and young children are very common, making small pieces of Pb, which are sweet in taste, a significant problem. While timing of Pb paint for domestic use is unclear, particularly in NL, it was certainly used by the late 19th century and chipped paint is a common source of acute Pb toxicity in children. In addition to the Pb sources that contaminated the food and drink, or were directly consumed by young children, there were other potential sources of Pb to the fishers.

Occupational Pb exposure itself was not a likely vector, either from occupations associated with Pb exposure or the fishery itself. There were no active Pb mines or other Pb processing factories, nor large scale practices such as potteries that used white or red Pb (Pb acetate in powdered form), on the island until the late 19th century. Additionally, since the samples reflect exposure during childhood, such occupations would only affect the children if they were working themselves, or if dust containing the Pb was blown into their residence or brought home on clothing, then exposed through hand to mouth activities (see Gulson et al., 1996b).

While there are many materials associated with the fishery that contained Pb, on a whole these likely minimally contributed to the exposure for fishers, including their children. Although the Pb weights used in jiggers and nets were an essential part of the fishery (Pope, 2004), touch these solid items is a less likely vector for contamination since non-organic Pb is removed through sweat (Rabinowitz et al., 1976). Hunting was crucial to life, particularly during the winter. Pb shot pellets have been found to result in acute toxicity (Lévesque et al., 2003; Knott et al., 2010), it would be rare for an individual to swallow a Pb ball that was not removed from the harvested meat and the Pb ball not pass through the gastrointestinal tract in a timely manner. Therefore, it is not a likely source of widespread community exposure.

Children did participate in various activities related to the fishery, both boys and girls helping the women with drying the cod flakes in permanent settlements (Porter, 1985). Boys of unspecified ages had various responsibilities even within the migratory fishery, working as apprentices helping to move fish to be split and salted, then rinse and lay out the flakes to dry (Porter, 1985; Handcock, 1989: 191; Pope, 2003). Less is known about their activities during winter months and, therefore, their specific vectors of Pb exposure.

Given these assumptions, in spite of the absence of local anthropogenic sources, it is clear that the fishery populations' Pb exposures were elevated as seen in the tooth enamel concentrations. In turn, imported Pb-containing cultural materials must be considered. Pb isotope ratios can be used to explore the origins of the ore used in producing the cultural materials, which resulted in the heavy metal exposure.

3.5.2 Cultural environments of Pb

3.5.2.1 Fisheries and the Atlantic World

While there are some differences in Pb sources between fishery sites, except for three outliers (Harbour Grace F7, Tors Cove NP54, and Louisbourg F20), the values fall within the expected Pb isotope ratios of Pb mines in England, Wales, and France (compiled in Blichert-Toft et al., 2016; Available in Appendix 7.2.3 and Table 45). The French fishery cemetery at Louisbourg (Block 3) was different from two of the fishery sites (Foxtrap and Placentia). Though there is overlap in the values from some individuals, it suggests that most individuals associated with Louisbourg were interacting with at least some Pb-materials sourced from different mines. While French and English/Welsh mines have some deposits with indistinguishable Pb isotope ratios, there is less known about what specific French mines were in use during this period and what ore potters used for the glaze.

In addition to the two clear outliers from Harbour Grace (F7) and Tors Cove (NP54) cemeteries (Figure 5), there is one other individual with outlying Pb isotope values (>2σ of the mean). F20 from Louisbourg has a Pb concentration of only 0.2 ppm and higher $^{20n}Pb/^{204}Pb$ values than the other individuals. Given the low concentration, their outlying Pb isotope ratios are partly or entirely because of natural Pb from the soil and inhalation rather than anthropogenic sources.

While the NL and NS sites in this study clearly had English/Welsh or Western European Pb from imports, the question arises of how to consider outlying values. In the NL sites, two individuals had evidence of anthropogenic Pb exposure, but their Pb isotope

ratios are outside the values expected for English/Welsh Pb ore. The outliers have similar values to individuals from Grafton, Illinois (Fitch et al., 2012), New Haven, Connecticut (Aronsen et al., 2019), Lawrence, New Zealand (King et al., 2020), and Pueblo, Colorado (Keller et al., 2016) (Appendix Table 18). While Fitch et al. (2012) suggest that their outlier was possibly from the Austinville-Ivanhoe Pb-Zn district in Virginia, the other papers did not fully explore the origins of their exposure. In all these studies, outlying values not assumed to be from diagenesis were considered to represent 'non-local' individuals.

While it is not possible to determine the specific source of exposure for 'non-locals', it is helpful to explore possible origins. The two outlying values in this study's fishing communities are similar to the Appalachian Valley Pb ore (Figure 7; Heyl et al., 1966), but this area seems to have been actively mined since in the 1850s (Bayley, 1910:72; Mosier 1948), which makes this an unlikely source. A more appropriate interpretation of these data would suggest they were exposed to multiple sources, creating a mixed value between the English Pb ore and those from other American ores such as the Austinville-Ivanhoe and Eastern Tennessee Pb ore districts which were active since the mid-18th century (Figure 7; Foley et al., 1981; Kesler et al., 1994a). Alternatively, they were exposed to small amounts of Pb ore with much higher values such as the Upper Mississippi Valley district (Figure 7; Heyl et al., 1966; Millen et al., 1995); however, these were not active on a large scale until the 1840s (Heyl et al., 1955:232). Either way, American Pb is likely contributing to the individuals in Harbour Grace (St. Paul's cemetery) and Tors Cove, Newfoundland, but could also account for outlying values in other post-colonial contexts.

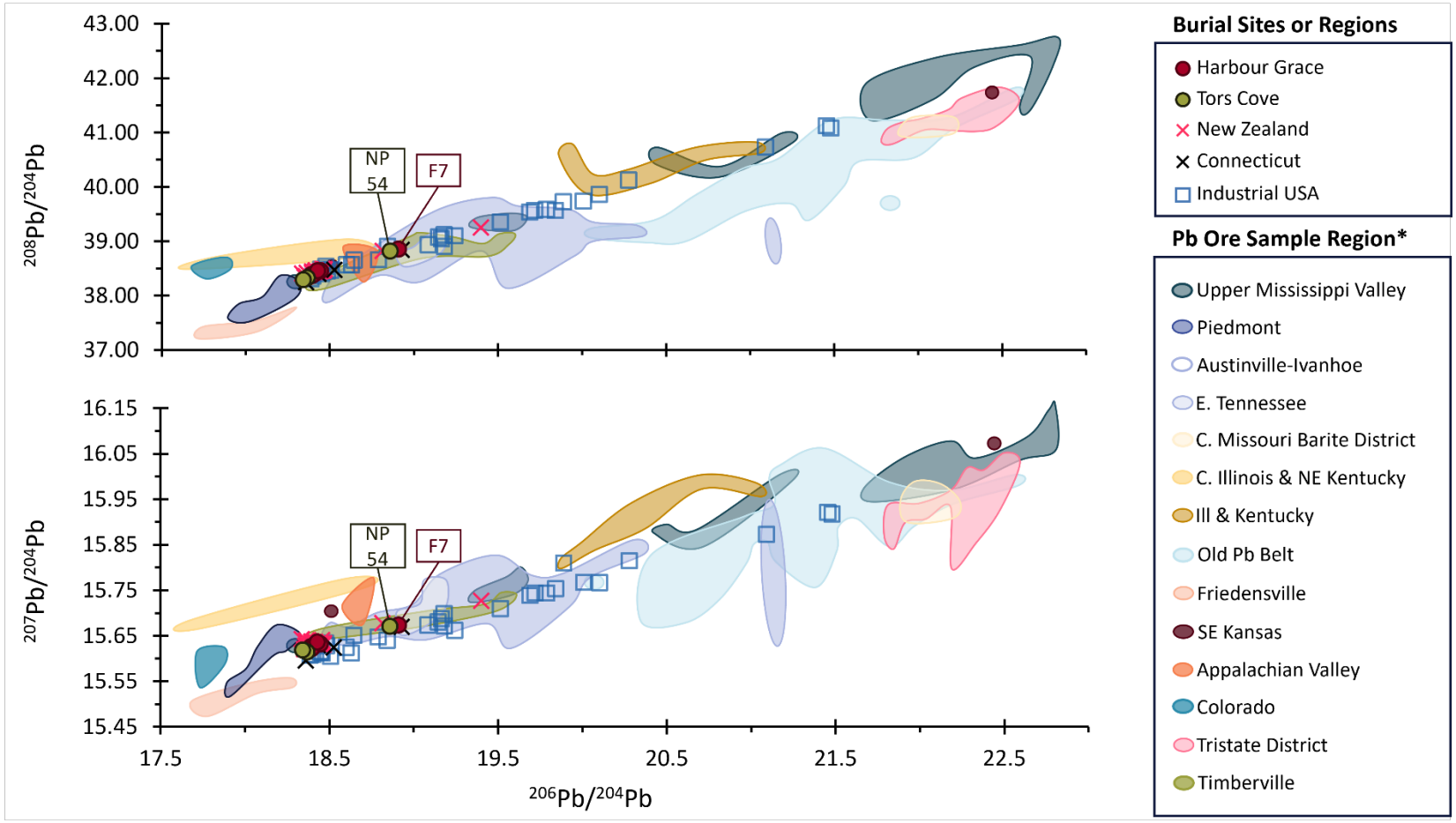

Figure 7. Comparison of $^{206}Pb/^{204}Pb$ to $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ between sites with values consistent with American Pb ore, both from the Newfoundland fisheries (Harbour Grace and Tors Cove), and previously published contemporaneous sites (Industrial USA, New Zealand, and Connecticut). Citations for all sites are available in Table 4. Pb ore field isotope value ranges for all American ore are outlined. All comparative citations are available in Table 4 and Pb ore citations are in Appendix Table 45.

Based on previous bone collagen carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotope analyses of the individuals from Harbour Grace, the significant contribution of fish to their diets as children and adults were consistent with local origins, as were their low oxygen ($\delta^{18}O$) isotope values of enamel carbonate from childhood (Munkittrick et al., 2019; Garlie, 2022). Further analyses of $\delta^{13}C$, $\delta^{15}N$, and $\delta^{18}O$ of bone collagen and enamel carbonate from the Tors Cove individuals are ongoing. So, while F7 from Harbour Grace, was likely living locally but had 'non-local' Pb isotope values consistent with at least some Pb from USA, additional data is necessary before such a claim can be made for Tors Cove. This brings further into question how to interpret 'non-local' values at other colonial or post-colonial contexts, since not only English/Welsh but American-sourced Pb materials were exported widely. Therefore, examination into the diversity of Pb use in ceramics and other Pb-containing cultural materials is required before immigrants to these regions can be identified with Pb isotope data.

Previous studies seldom considered the contribution of Pb in cultural materials as its impact on Pb isotope ratios in human teeth. Most of the individuals from the two New Zealand cemeteries (King et al., 2020), the Connecticut cemetery (Aronsen et al., 2019) and the Newton Plantation cemetery in Barbados (Laffoon et al., 2020) have similar values to these fishing settlements. Laffoon et al. (2020) concluded that, with the exception of a few individuals with low concentrations that may have been affected by diagenesis, the individuals' Pb must have come from imported Pb-containing materials. However, King et al. (2020) and Aronsen et al. (2019) suggested that the Pb isotope ratios

did not fit with local exposure. Importantly, these were based on geological or soil-core data and did not consider the import of Pb-containing cultural materials used during life as a potential source.

The implication for not fully considering imported Pb-containing materials for *in vivo* exposure is that individuals must be interpreted as immigrants, or the tissue has been diagenetically altered. Aronsen et al. (2019) concluded that these would have to be first-generation immigrants. While this is a possible interpretation, the Pb values could also have originated via exposure from their imported goods, so, at least based on Pb isotope ratios, they may or may not have been immigrants. King et al. (2020) suggested that the values from burials at St. John's, Milton were all diagenetic, based on the high concentrations and Pb isotope values matching those of British imported coffin hardware. There was not an explanation of the burials from Lawrence New Zealand that did not have pewter burial coffin hardware until a later publication (King et al., 2021), where it was hypothesized that they were all first-generation immigrants, likely from Britain. While diagenesis is likely the case for the values from the deciduous teeth from St. John's Milton, NZ (that were not considered in this comparison), the 65 ppm cut-off they used, suggested by Kamenov et al. (2018), is based on modern enamel data and uses a different methodology of Pb concentration analysis methodology so may not be applicable. Therefore, given the lack of consideration of Pb exposure during life from British imports (and no diagenetic alteration from those imports), there is an alternate interpretation that the values represent exposure to British Pb sources while living in New Zealand. In both Aronsen et al. (2019) and King et al. (2020), without better assessment of diagenesis and possible imported sources, the Pb values cannot distinguish between a conclusion

involving diagenesis, non-local origins, and local origins. Considering that both this study and Laffoon et al. (2020) found that imported materials had a significant impact on local Pb isotope values, future studies will benefit from considering these as possible sources during interpretation.

3.5.2.2 A cultural de-focusing

Montgomery et al. (2005) introduced the concept of 'cultural focusing,' a phenomenon noticed in Britain where there is a decreased variability of Pb isotope ratios and, therefore, sources of Pb as concentrations increase. This focusing occurs due to the restricted number of Pb sources available within a society and the fact that Pb containing materials were frequently melted, mixed, and re-used, thereby averaging the Pb isotope values. How this activity of material re-use translates into the Atlantic World more broadly needs to be clarified. By examining both Figure 6 and Figure 8, we can see that cultural focusing is occurring, unsurprisingly, within Late Medieval Atlantic Archipelago and Industrial England, but this also translates into Barbados and Louisbourg. Since the interpretation with the Newton plantation in Barbados was that much of the exposure was likely from the distillation processes that the enslaved labourers were working with (Laffoon et al., 2020), it is unsurprising that there is minimal variability in Pb isotope ratios as that is a restricted source of anthropogenic Pb. However, Louisbourg is a mixed-origin population with minimal variability within the Pb isotope values. While many likely came from various ports in France's North and Western coast, others came from Placentia and possibly Quebec and Acadia (Nova Scotia and New Brunswick). The Louisbourg burials date between 1713 and 1723, which is an earlier settlement than the

other fishery sites, except for possibly the St. Luke's cemetery in Placentia, which has an uncertain date. These constrained values are consistent with what would be expected with cultural focusing of Pb sources, but the other comparative sites and fisheries did not.

The comparative assessment of the NL fisheries to the Royal Navy and New Zealand populations showed larger ranges of Pb isotope ratios within the clusters, than Louisbourg (Figure 6; Figure 8). This is particularly true when not including low Pb exposure individuals (<0.8 ppm), which excludes the possibility that the outlying values were due to natural environmental sources. The large variability within these groups and the further outliers particularly within American contexts suggests that, outside of urban Britain, there is a 'cultural de-focusing' beginning at least by the 18th century. This is further supported when examining Pb concentration compared to $^{204}Pb/^{206}Pb$ (Figure 8), since the high concentrations do not preclude the variable isotope values. The reasoning for this defocusing is likely multi-faceted.

This defocusing compared to Late Medieval Atlantic Archipelago and contemporary French and urban English contexts could be due to the data currently available as well as mining and export practices. However, it could be because urban England and the broader Atlantic Archipelago that was exposed to culturally focused Pb, but more rural areas were more variable. This, at least during the Late Medieval period, seems less likely, as individuals with anthropogenic exposure from Essex, Cumbria, and Guernsey all had similar ratios to those from Blackfriars in London (Montgomery, 2002; Figure 8). However, if many of the individuals buried in the Royal Naval Hospital cemetery in St. John's were from Britain, as is supported by historical documentation and isotope

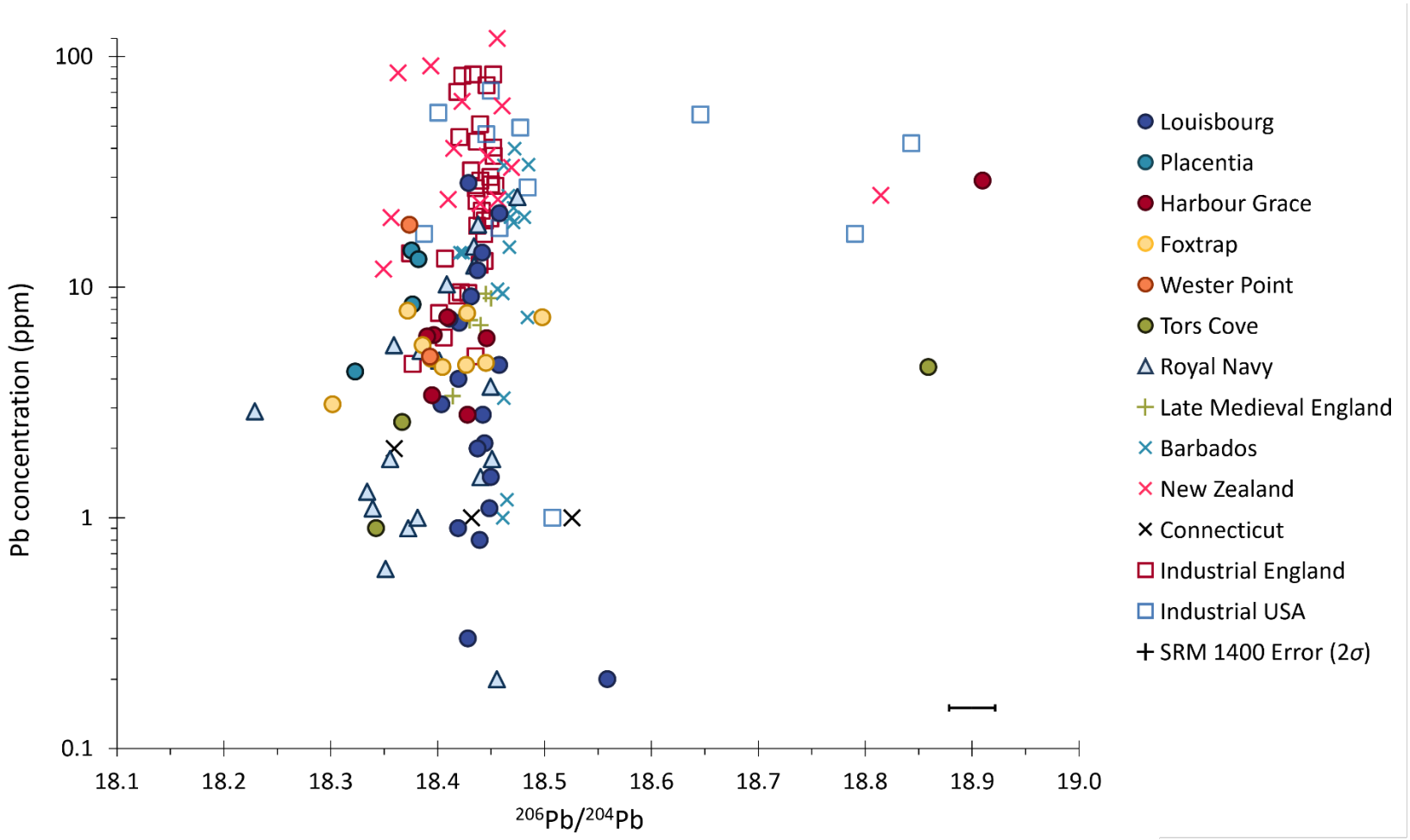

Figure 8. Comparison of $^{206}Pb/^{204}Pb$ to Pb concentration (ppm) between the Fisheries, Royal Navy, and other previously published contemporaneous sites. All comparative citations are available in Table 4. Note that the axes are restricted, cutting off some individuals from Industrial USA with higher isotope values and New Zealand individuals with high Pb concentrations. Between assay variability is presented as 2σ as determined by analysis of SRM 1400.

analyses (see 7.2.1.7; Munkittrick 2016; Munkittrick et al., 2019), then it would suggest that by the mid-18th to early-19th centuries there was an increase in the variability of sources available. This is not to say that those producing Pb-containing products were sourced from outside of Britain, but that there was no longer the very averaged values seen in those with anthropogenic Pb burdens in Roman Britain and Late Medieval Atlantic Archipelago. What could account for this increase in variability is the significant increase in Pb mining in England and America during the 18th and particularly 19th centuries (Burt, 1984). The resulting influx of more material into the market may have undone some of the focusing accomplished by the long-term re-use of Pb since Roman Britain.

3.6 Conclusion

Children had anthropogenic Pb burdens in these historic (17th–19th century) North Atlantic fishing communities, that were as much as 20 times higher than natural exposures (i.e., Prehistoric Britain, Viking Atlantic Archipelago) but not as high as industrial period individuals in England or those associated with local mining activities in the USA. Since Pb mining was nonexistent in Newfoundland and there were limited environmental sources, exposures must have been from imported cultural materials. The high Pb content in ceramic glazes would have leached into their food and drink, particularly during cooking and storage, resulting in the anthropogenic Pb exposure. Previous studies have largely ignored the role of cultural materials as an exposure source for Pb contamination during this period.

Pb isotope values in the enamel of molars, representing childhood, suggest that the dominant source to these fishing populations was English or French Pb ore deposits. Though, there were a couple individuals with outlying values that are consistent, at least in part, with exposure from American Pb sources. It remains unclear whether outlying values indicate that individuals were immigrants to the area or if some individuals and families in Harbour Grace and Tors Cove, NL used Pb-containing materials from different areas. Further isotopic studies of childhood diet (dentine $\delta^{13}C$ and $\delta^{15}N$ and carbonate $\delta^{13}C$) and origins ($\delta^{18}O$ and $^{87}Sr/^{86}Sr$) may help to clarify this point.

The importation of cultural materials cannot explain the difference in values in the French individuals from Block 3 cemetery of Louisbourg, NS, since the cemetery was only in use for nine years during the earliest period of settlement at the fort and all residents were immigrants. There is little historical information regarding the source of Pb used in French ceramics. While there is overlap with Pb isotope values of both English/Welsh and French Pb ore data, the range of values in Louisbourg suggests that some individuals were exposed to different geological sources of Pb compared with other fishing settlements before emigrating. Pb isotope studies of imported ceramics used in these communities may help clarify whether Pb used in French and American ceramics had different Pb sources to those in English ceramics.

4 CERAMIC GLAZES AS A MAJOR SOURCE OF PB EXPOSURE IN 17TH – 19TH CENTURY NORTH AMERICAN SETTLEMENTS [3]

4.1 Introduction

Lead (Pb) isotope analyses have been used since the 1960s to examine the geographic origins of humans and cultural materials in archaeological contexts (Chapter 2). More recently, there has been an interest in exploring the life histories of individuals within the Atlantic World. However, the movement of people and materials across the Atlantic Ocean from the 16th to mid-19th centuries was multi-faceted and not solely based on the perceived 'metropole'. Implications of the complexity of migration and trade have not fully been explored regarding the ability of Pb isotope analyses to interpret migration during this period.

Key to this sourcing is having good baselines of expected values and information related to geographic variability in ratios, which differ by geological source and age of formation. Yet, determining what geological or environmental Pb sources to consider when delimiting the expected 'local' range of Pb isotope ratios is not straightforward. There is some disagreement on what approach to take (see Chapter 2 for further discussion). Bioarchaeologists interpret isotope ratios in human teeth to identify if a child was raised locally and if not, to suggest possible origins (e.g., Montgomery et al., 2005; Fitch et al., 2012; Valentine et al., 2015; Laffoon et al., 2020). Pb isotope ratios can help

[3] This Chapter has been written as a manuscript for publication, and will have Drs. Amy Scott (Dept. Anthropology, University of New Brunswick), Tamara Varney (Dept. Anthropology, Lakehead University) and Vaughan Grimes (Depts. Archaeology and Earth Sciences, Memorial University of Newfoundland), as co-authors.

identify non-local individuals in cases where their values are outside the expected local ranges and/or values from underlying geology, which are determined when exposure occurred in non-anthropogenic or 'natural' environments using faunal remains. In anthropogenic contexts – as is relevant in this case – the ranges are determined by characterizing isotope ratios of local ore sources (Montgomery et al., 2005; Evans et al., 2018a; Samuelsen & Potra, 2020) and/or environmental pollution (Keller et al., 2016; Aronsen et al., 2019).

Since there are typically a limited number of geological sources of Pb locally available to a population, the Pb isotope ratios in tissues will reflect an average of available sources. In Britain, the limited number of sources and recycling of Pb-containing materials results a "cultural focusing" of Pb isotope values at a population level of individuals when their Pb enamel concentrations are increased (>0.8 ppm) above naturally occurring exposure levels (Montgomery et al., 2005). Most of the research regarding anthropogenic Pb sourcing within bioarchaeology is from England. While many of these are Romano-British studies (Montgomery et al., 2010; Shaw et al., 2016), there are also several studies from the medieval or industrial periods (e.g., Montgomery et al., 2005; Millard et al., 2014). Since England was a prominent producer of Pb and only began importing substantial amounts of Pb in the mid- to late-1800s (Burt, 1984), it is logical to use Pb isotopes from ores within England to create their baselines. However, how that applies to other historical regions during the 17th to 19th centuries is unclear.

Several North American bioarchaeological studies have focused on local environmental exposure to determine the source of Pb to populations in 19th century United States (Fitch et al., 2012; Keller et al., 2016; Aronsen et al., 2019), including

interpreting origins using a map by Keller et al. (2016) which used soil cores to estimate local sources of bioavailable Pb. However, even in mining areas, environmental pollution is not necessarily a significant source of exposure, nor are soil pollutants necessarily bioavailable. For migration studies, more broadly this brings into question what geological sources of Pb should be considered when identifying the values expected for "local" individuals.

Additionally, in the examination of tooth enamel from the late-17th to mid-19th century in Newfoundland (NL) and Nova Scotia (NS), Canada, it was shown that fishing communities experienced significant Pb exposure, even though there was no local environmental pollution (Chapter 3). The geological source(s) of this Pb within English/Irish and French communities were consistent with English/Welsh and Western European ore (Chapter 3). However, there was variability between the English/Irish and French populations and outliers consistent with contributions from American Pb ore (Chapter 3).

North American studies often underemphasize the potential of exposures from everyday cultural materials, but given the absence of local sources during this time period, it is necessary to consider the Pb isotope values of materials that they were interacting with. Since ceramics are ubiquitous on archaeological sites, they present an ideal means of exploring the variability of Pb available to English and French communities from the 17th to mid-19th centuries. This variability must be understood within a framework of transnational networks of influence and trade across the Atlantic Ocean during the 16th to mid-19th centuries, termed the Atlantic World (Appendix 7.3.2).

This paper directly explores Pb isotope values within the glaze of ceramics to better contextualize Pb exposure during early settlement in the Atlantic World, using French and English/Irish fisheries as case studies. Given the spheres of influences, this study can also speak more broadly to uncertainties involved in inferring the geographic origins of people and ceramics using Pb isotope ratios. Pb exposure information further contextualizes sources within new and previously published materials representing childhood exposure. Finally, we explore the potential contribution of Pb exposure from ceramics for identifying migration using Pb isotope ratios in the Atlantic World. This is accomplished using the following research questions:

1) Was Pb high enough in available in ceramic glazes of French and English/Irish fishing settlements to contribute to anthropogenic levels of human exposure?
2) How variable are the Pb isotope ratios within Pb glaze of ceramics available to fishing populations?
3) Do the Pb isotope values of ceramic glazes correlate with the manufacturing country or region of origin?
4) How does variability of Pb isotope ratios in ceramic glazes compare as a bioavailable range to those in human tooth enamel of NL and NS's French and English/Irish populations?
5) Using the isotope ratios of enamel from fishing populations and contemporary published materials, how applicable is the cultural focusing model in the Atlantic World?
6) Given outliers in NL populations whose isotope ratios are consistent with American Pb and the potential of cultural material contribution, comparing values

within individuals likely born in New England and other published materials, when is the potential of Pb isotope ratios for identifying non-locals possible?

4.2 Background

Modern manufacturers have strict regulations regarding the levels of heavy metals allowed in household goods, but this was not always the case. Lead (Pb) has been a common component of ceramic glazes since antiquity due to its ease of application and aesthetic, as it produces an even layer and is less likely to have crawling cracks or dark colouring (Tite et al., 2008). While Pb exposure from ceramics may only occur in small concentrations, it can accumulate in the body over time, with skeletal tissues being the predominant reservoir in humans (Rabinowitz et al., 1976; Flora et al., 2006). Storage sites include tooth enamel remain unchanged throughout life, resulting in a short-term history of exposure during the period of tooth formation (Budd et al., 2000b; AlQahtani et al., 2010).

Potters continued to use Pb in glazes until the mid-20th century. The United States Food and Drug Association only began controlling the Pb content in ceramics starting in 1971, Canada since 1976, and the European Union since 1984 (Franson et al., 1977; Directive, 84/500/EEC; Lecos, 1987). While American correspondence in the late-18th to early-19th centuries implied that the toxic effects of Pb glazes were well known (Baldwin, 1993; Janowitz, 2013), this did not stop potters from using the heavy metal in other earthenwares, nor consumers from their purchase. The focus on the effects of Pb even into the mid-20th century, particularly by physicians, was on acute accidental poisonings (Warren, 2000). This focus on acute toxicity from a single source downplayed or ignored the low cumulative exposure from interacting with everyday materials, including Pb

glazes. The trend of downplaying the potential of ceramics contributing to Pb poisoning has continued into recent archaeological studies examining human Pb exposure and sourcing the geological origins of that exposure.

The Pb within ceramic glazes or other Pb-containing cultural materials, would have originated from recently mined ore from a single or multiple geological sources, or result from the repurposing of older materials. Pb was processed in different manners depending on the final material. In the case of ceramics, crushed Pb ore, red-Pb (PbO), or white-Pb ($2PbCO_3 \cdot Pb(OH)_2$) was commonly used in glaze production (Burt, 1969; Burt, 1984). The original and inexpensive method used into the early 19th century was to place a layer of crushed ore on a dried piece of pottery then fire it. By the early 18th century crushed red-Pb, and later white-Pb in powder or fluid form, was used to create the glaze (Burt, 1969).

Pb-glazed ceramics are ubiquitous on 17th to 19th century archaeological sites in the Atlantic World; however, they were not the only vector of Pb exposure to populations. Many other cooking, serving, and storage vessels contained Pb, either as an alloy such as pewter, or as the main fabric of pipes and cisterns. Pb acetate was a common component of medicinal treatments in the 18th and 19th centuries and was even used in fabric dyeing and processing. Pb acetate used as additives would not be found on archaeological sites, and Pb itself, or its alloys, were often melted down and re-used, meaning that they are infrequent finds. These materials, be they locally produced or imported, were used by many populations on both sides of the Atlantic during this period, and it is possible to examine the geological origin of that Pb using isotope analyses.

Four isotopes of Pb are typically measured as part of bioarchaeological applications of this technique: ^{206}Pb, ^{207}Pb, and ^{208}Pb, which are radiogenic, and ^{204}Pb, which is primordial (Faure and Mensing, 2005). The different original concentrations of parent isotopes in rocks (^{238}U, ^{235}U, ^{232}Th), the age since formation (half-lives), and mixing during formation create the variability of Pb isotope ratios within geological formations (Adriano, 1986; Gulson, 1986; Teutsch et al., 2001; Faure and Mensing, 2005). There is minute measurable but not meaningful isotopic fractionation of Pb isotopes during smelting and processing into cultural materials (Budd et al., 1995; Stos-Gale and Gale, 2009; Cui and Wu, 2010). There is also no meaningful fractionation when the Pb humans are exposed to is stored in their bones and teeth (Rabinowitz and Wetherill, 1972; Gulson, 1986).

Pb is incorporated into human tissue mainly by consumption or inhalation (Adriano, 1986; Brännvall et al., 1999). While humans are exposed to small amounts of Pb that naturally occurs in the environment, depending on the environmental and cultural context, they are also exposed to this anthropogenic Pb resulting in much higher concentrations with isotope values that reflect a weighted average of those sources. Pb within cultural materials, such as ceramic glazes, can leach into the food or drink consumed from those Pb-containing materials. The stable isotope ratios of Pb differ between geological sources of ore, and the minute alterations to the isotope values (fractionation) during processing of the ore are not interpretably relevant for provenance studies (Budd et al., 1995; Stos-Gale and Gale, 2009; Cui and Wu, 2010). This means that Pb isotope ratios can be used to differentiate the origins of Pb within an analyzed material, including ceramic glazes

(Schurr et al., 2018; Métreau et al., 2021; Faure and Mensing, 2005) and human enamel (Adriano, 1986; Gulson, 1986; Teutsch et al. 2001).

Given the uncertainty over whether "cultural focusing" is applicable in the broader Atlantic World during the 17th to 19th centuries and the problems with estimating "local" bioavailable Pb, it is necessary to consider materials themselves that were available to these populations. Except for the Franklin Expedition (Kowal et al., 1991), studies have not examined the Pb isotope ratios of materials available to a cultural group. Some Roman studies have used Pb isotope ratios in coins to suggest regions of origin for individuals (Montgomery et al., 2010; Shaw et al., 2016), with the argument being that the geographic origin of a coin is within the same region as the geological source of Pb used in minting that coin. They additionally argue that the same Pb ore source was used in other cultural materials that contributed directly to human Pb exposure. While this may be reasonable, it is a way that researchers have approached typifying Pb values for different occupied regions in areas where Pb isotopes of ore bodies were not readily available. A handful of studies have suggested the specific materials that may have resulted in exposure. Laffoon et al. (2020) even linked values consistent with England to locally born Barbadian enslaved labourers, but this approach appears in the minority of studies.

While the composition of paste and glaze of ceramics has been broadly studied (Duffy et al., 2002; Tite et al., 2008; Greer et al., 2021; Greer and MacDonald, 2022), few studies have specifically addressed the Pb isotope ratios within the glaze, particularly within a post-medieval European context (Schurr et al., 2018; Métreau et al., 2021). One study of Pb in British ceramics excavated from two early-mid 19th century Indigenous

contexts in USA (Pokagon Village, Michigan and Collier Lodge, Indiana) solely involved white ware and pearlware (Schurr et al., 2018). Additionally, the Pb source from 13th- to 14th-century faïence tiles from medieval Northwest France (Chateau Suscinio, Brittany and Brain-sur-Allonnes, Anjou) attributed the source to Derbyshire England (Métreau et al., 2021). These studies were exploring Pb in ceramics to examine manufacturing techniques and chaîne opératoire. But this approach can be further extrapolated to explore the implied exposure within the populations interacting with such materials.

4.3 Materials and Methods

Ceramic materials were available from two sites with an extended occupation for French and English/Irish individuals that were involved with the Atlantic fisheries: Anse à Bertrand, Saint-Pierre and Miquelon (SPM-2) and Ferryland, NL (CgAf-2) (Figure 9). These sites have comparable contexts to the French and English/Irish fishing towns with burial grounds mentioned in Chapter 3 that were examined using Pb concentration and isotope analyses of teeth during late-17th to mid-19th centuries. These were supplemented with additional human dental enamel was sampled from two burial sites: St. Paul's Anglican Church Cemetery from Harbour Grace, NL (CkAh-06, 1764–1820 CE), and Rochefort Point burial ground, Louisbourg, NS (55L34; 1730s–1758 CE).

4.3.1 Pb-glazed ceramic samples

Ceramics were chosen from the collections associated with Anse à Bertrand, Saint-Pierre (late 17th–19th centuries), and Ferryland, NL including the Colony of Avalon (1621–1696; 18th– mid-19th centuries). Materials were chosen to represent the sites'

Figure 9. Location of archaeological sites associated with ceramic samples are in black. (1) Anse à Bertrand, and (2) Ferryland. Those in blue are associated with burial grounds: (3) St. Paul's Anglican Church Cemetery, Harbour Grace, NL, and (4) Rochefort Point burial ground, Louisbourg, NS. Comparative human remains from Munkittrick et al. (Chapter 3) were also sampled from the (3) cemetery in Harbour Grace and the (5) Block 3 Cemetery at the Fortress of Louisbourg, NS. Additional comparative sites from Newfoundland are in yellow: (6) Wester Point Cemetery, Portugal Cove, (7), Foxtrap-2 burial ground, Foxtrap, (8) Tors Cove burial ground, Tors Cove. Part of the territory the French lost before establishing the Fortress of Louisbourg is (9) Plaisance, renamed Placentia by the English. Map made by author using ArcGIS.

occupation periods and from materials found frequently in English and French archaeological contexts in the North Atlantic fishery settlements (see Appendix 7.3.2 for further details). These ceramic sherds were taken for analysis by Scanning Electron Microscopy for initial imaging and semi-quantitative concentrations, and then Pb isotope ratios and concentrations were determined by laser ablation split-stream (LASS) analysis using simultaneous single detector and multicollector inductively coupled plasma mass spectrometry (ICP-MS and MC-ICP-MS).

4.3.1.1 Ceramic Site Backgrounds

Basque fishers were active in Saint-Pierre with the migratory fishery prior to the 1570s (see citations in Losier et al., 2021). However, the year-round occupation of the French *habitants-pêcheurs* began in the 17th century, with the first evidence of occupation at the site of Anse à Bertrand likely dating to the late 17th century. Despite transitioning to British control of the island between 1713 and 1764, there is evidence to suggest continued French use of the site for at least a short period into this change of rule (Livingston & Losier, 2021). The earliest names associated specifically with the site are from 1763 are the families of Pierre Bertrand, Charles Philibert, and Pierre Dalair; the first two of whom returned in the 1780s following a short period of British rule (Livingston & Losier, 2021). During the 19th century, *petits pêcheurs*, or small-scale inshore family fishers occupied the site seasonally, spending their winters in the town of Saint-Pierre (Gaulton and Losier, 2020; Livingston & Losier, 2021; Champagne & Losier, 2021). Despite Saint-Pierre changing political control multiple times during these

centuries, there is no specific evidence of English occupation of the site (Losier et al., 2021).

Ferryland was occupied by both the migratory fishers and the Indigenous Beothuk from the early-16th century (Gaulton, 2001; Pope, 2009), but became a permanent settlement when the English established the colony of Avalon in 1621, later named the Pool Plantation after 1638 (Pope, 2004). A French raid in 1696 destroyed much of the settlement and surrounding area, but inhabitants quickly returned to Ferryland, with a significant increase of Irish occupants beginning in early 1700s and increasing drastically by the end of the century (Ryan, 1983; Mannion, 2001; Pope, 2004). While it was not the lucrative colony of the 17th century, the port was a mercantile center for the Avalon Peninsula south of St. John's (Head, 1976).

4.3.1.2 Ceramic materials and analyses

For this study, ceramic material samples were limited primarily due to the size of the laser ablation cell. Only ceramic sherds less than 55 mm in diameter and 8 mm in height were selected. Another limitation was that materials were accessed during Covid-19 restrictions in the fall of 2020 and early winter 2021. The Anse à Bertrand materials taken for analysis were from the 2019 field season housed at Memorial University and the Ferryland materials were taken from the Department of Archaeology's on-site teaching collection.

A total of nine ceramic sherds were successfully analyzed that dated between the 17th to 19th centuries from Anse à Bertrand (Table 6; Figure 10). These included seven ceramics from France and two Northern Italian sherds. There were 14 ceramic sherds

Table 6. Ceramic sample production summary information, sample number, and archaeological site name (Ferryland, Newfoundland, CgAf-2; Anse à Bertrand, Saint-Pierre, SPM2). Ceramic sherds without specific ware type identification are identified as REW (refined earthenware)

Country of origin	**Region**	**Ware type**	**Date (century CE)**	**Sample no.**	**Site**
French	Vallauris	Vallauris	17th–19th	807-4148	SPM2
French	Brittany	Pabu-Guingamp	17th–19th	807-4152	SPM2
French	Unknown	Faïance blanche	17th–18th	708-3700	SPM2
French	Unknown	Faïance blanche	17th–18th	710-3809	SPM2
French	Unknown	Faïance brune	17th–18th	508-2430	SPM2
French	Unknown	Faïence – tinglaze upper (sample), brown Pb under	17th–18th	5552	CgAf-2
French	Saintonge	Saintonge	17th–18th	708-3704	SPM2
French	Saintonge	Saintonge	17th–18th	5547	CgAf-2
French	Unknown	REW	17th–18th	5549	CgAf-2
French	Unknown	REW	17th–18th	5551	CgAf-2
English	Devon	N. Devon gravel	16th–mid 18th	5545	CgAf-2
English	Devon	N. Devon smooth	16th–mid 18th	5546	CgAf-2
English	Staffordshire	Staffordshire	18th	5560	CgAf-2
English	Unknown	CEW (possibly Midlands Purple or Manganese Mottled	17th–18th	5557	CgAf-2
English	Unknown	REW	18th	5558	CgAf-2
English	Unknown	Creamware	18th	5556	CgAf-2
English	Unknown	Whieldonware	18th	5559	CgAf-2
Dutch	Unknown	slipware	17th	5540	CgAf-2
Dutch	Unknown	slipware	17th	5541	CgAf-2
Italian	Liguria	Albisola	18th–19th	804-4054	SPM2
Italian	Liguria	Unknown	18th–19th	803-3968	SPM2
Spanish	Unknown	Spanish Heavy	16th–18th	5539	CgAf-2
Spanish	Unknown	Spanish Heavy	16th–18th	707-3648	SPM2

Figure 10. Ceramic samples separated by country of origin and date of production. The sample numbers are as follows, with additional information available in Table 6. **English**: a) 5545; b) 5546; c) 5556; d) 5557; e) 5560; f) 5558; g) 5559. **French**: a) 807-4148; b) 807-4152; c) 708-3700; d) 710-3809; e) 508-2430; f) 5552; g) 708-3704; h) 5547;ij) 5549; j) 5551. **Italian**: a) 804-4054; b) 803-3968. **Dutch**: a) 5540; b) 5541. **Spanish**: a) 5539; b) 707-3648. Ceramic samples not to scale.

successfully analyzed for glaze composition and Pb isotope ratios from Ferryland (Table 6; Figure 10). These included four 17th-century English ceramics, five 17th- to 18th-century French ceramics, six 18th-century English ceramics, two 17th-century Dutch sherds from the same vessel, and one Iberian ceramic. An additional six ceramic sherds from Ferryland and six from Anse à Bertrand were chosen for analysis. However, they were not analyzed or included here due to final size restrictions, lack of Pb glaze available for analysis, or time constraints.

Data were acquired using laser-ablation split-stream (MC-)ICP-MS over three days. Ceramic sherds were ablated using an ArF Eximer laser (GeoLas Laser UV-193nm) with concurrent trace element and Pb isotope ratio analyses using a high resolution inductively coupled plasma mass spectrometer (Thermo-Finnigan Element XR HR-ICP-MS) and a multi-collector ICP-MS (Thermo Scientific Neptune MC-ICP-MS). Samples were calibrated using the NIST 610 standard bracketed every four to ten analyses. Further methodological and analytical error details are available in the supplemental materials (Appendix 7.3.3).

4.3.2 Human dental enamel samples

4.3.2.1 Comparative collections and site contexts

Ceramics from Anse à Bertrand are used as a comparison for the fishery context at the French cemetery of Block 3, Louisbourg (n = 18; Chapter 3). This cemetery represents the first eight years of the Fortress' occupation (1713–1723; Harris, 1974; Johnston, 2004), so none of these people were born in Louisbourg. Many of the initial settlers were the original inhabitants of Plaisance, NL who relocated after France lost the

territory with the treaty of Utrecht (1713). Those in Plaisance were themselves from a mixed population, with some having been born locally, but others were from Basque country, Brittany, and Normandy, or ports such as La Rochelle, Bordeau, St. Malo, and Nantes (Johnson, 1984; Turgeon, 2000; Nadon, 2004; McLeod-Leslie, 2014; Landry, 2018). There were small numbers of other origins such as Ireland and England. Once in Louisbourg, many military officers found wives from Acadia (what is now part of Nova Scotia, New Brunswick, and Prince Edward Island). There was also at least one African enslaved labourer during the period of the cemeteries use, though the number of African labourers increased along with the resident population to be consistently about 3% of the population after 1724 (Donovan, 2004). The varied origins of those buried at Block 3 are supported by the stable isotope analyses of bone collagen and enamel representing diet ($\delta^{13}C$ and $\delta^{15}N$) and geographic origins ($^{87}Sr/^{86}Sr$) (Garlie, 2022). One individual from Block 3 (F20) was removed for this paper's reanalysis since their Pb concentration was 0.2 ppm and they had very different isotope ratios, which together suggest that the isotope values represent at least in part natural exposure sources (Montgomery et al., 2010) and that their isotope ratios would not represent anthropogenic Pb exclusively which is what is necessary for comparison to the cultural materials.

Ceramics from Ferryland are used as a comparative site for the English and Irish fishery context at cemetery sites that date roughly between the mid-18th and mid-19th centuries (Chapter 3). These cemetery sites include a significant merchant town of Harbour Grace (St. Paul's Anglican Church Cemetery, 1764–1820, n = 8), a small burial ground in Portugal-Cove (Wester Point Cemetery, 18th–19th c., n = 2), the farming and fishery area of Foxtrap (Foxtrap-2 burial ground, 18th–late-19th c., n = 9), and a small

burial ground at Tors Cove (~1812–late-19th c., $n = 3$). Based on historical documents and available isotopic values of skeletal remains ($\delta^{13}C$, $\delta^{15}N$, and $^{87}Sr/^{86}Sr$; Harris, 2015; Munkittrick et al., 2019; Garlie, 2022), it is expected that most of the individuals were born in Newfoundland. It should be noted that many of the ceramics do date towards the earlier period of comparison in the 17th to mid-18th centuries, while many of the burials likely date from mid-18th to mid-19th centuries. Including additional later ceramics would have been ideal for comparison, but it was not possible with the materials available.

Additional dental enamel samples were analyzed from two sites to further contextualize Pb exposure in these fishing settlements and the broader Atlantic World. The first was one individual (Feature 7/NP254) buried at St. Paul's Anglican Church Cemetery from Harbour Grace, NL, who had comparatively high Pb concentration and outlying isotope ratios in their second molar, which represented exposure during their childhood (2.5–8.5 y.o.; AlQhatani et al., 2010; Chapter 3). Samples were taken from their first and third molars, which represent infancy (4.5 mo.–3.5 y.o.) and adolescence (8.5–14.5 y.o.), respectively (AlQahtani et al., 2010).

To further explore the implications of imported Pb-containing goods in the Northwestern Atlantic, individuals from Rochefort Point, a mid-18th-century burial ground from at the Fortress of Louisbourg, in what is now Cape Breton, NS, were sampled. These eight adolescent individuals were previously identified based on historical contexts and mortuary contexts, and dietary isotopic values to have originated from the American colonies, likely the region of New England (Massachusetts, including the area that is now Maine, Rhode Island, Connecticut, and New Hampshire) (Scott et al., 2023). Therefore, these individuals were most likely buried between 1745 and 1748

during the New England occupation of Louisbourg. Second molars, representing childhood exposure (2.5–8.5 y.o.), were sampled from each individual.

4.3.2.2 Dental enamel Pb concentration and isotope analyses

Core enamel (50 mg) of the first and third molar of F7 from St. Paul's Anglican Church cemetery and one second molar from each individual from Rochefort Point was sampled at the Memorial Applied Archaeological Science (MAAS) laboratory. Samples were further pretreated at the Radiogenic Isotope Laboratory at Memorial University in laminar air flow clean boxes. Each sample was weighed then digested in 2 mL 8 M HNO_3 in PFA vials (Savillex). One aliquot (0.5 mL) was separated for concentration analysis, the remaining for Pb isotope analysis. The 1.5 mL aliquot for Pb isotope analyses was used for extraction chromatography using Sr-spec resin (Charlier et al., 2006; Pin et al., 2014) and was analyzed on a Neptune MC-ICP-MS. Solutions were analyzed for Pb concentration at the TERRA facilities at Memorial University (using a Perkin-Elmer Elan DRCII ICP-Q-MS). The more accurate method of Pb concentration analyses using a Q-ICPMS was only available for the samples from Rochefort Point. The concentrations of two teeth from St. Paul's were analyzed using the same eluted Pb solution using the MC-ICP-MS. Complete methodological procedures and levels of analytical errors are available in Appendix 7.3.4.

4.3.3 Comparative Pb ore data

Both the ceramic glaze and dental enamel Pb isotope ratios were compared to existing Pb ore isotope data to contextualize their origins. Blichert-Toft et al. (2016) amalgamated all European Pb ore isotope ratios along with coordinates; American ore Pb

isotope ratios were previously compiled by (Bird et al., 2019), but the individual values were re-examined in this study. See Supplementary Materials (Appendix 7.3.5) for a detailed explanation of the method used for determining the baseline comparative materials. While some studies present ore data as kernel density clusters, there are too many ore bodies used in this comparison for it to be a useful visual representation, Therefore, for the purpose of analyses, all data points were used, but are presented in text as outlines of the data.

4.4 Results

4.4.1 Ceramic values

The concentrations of the two analyses of each ceramic glaze were averaged unless there was interpretable variability that corresponded with observations during analysis (i.e., unstable and lower Pb voltage seen in the second analysis of 804-4054). The two colours of glaze from the Dutch ceramic were examined separately (5540), but the Pb concentrations and isotope ratios are averaged for this study.

The Pb concentration of the glaze from 23 ceramics ranged from 283,800 ppm (28.3%) to 588,900 ppm (58.9%), with a mean of 47.5 ± 9.7% (Available in Appendix Table 37). Of these, the French ceramics with lower Pb concentrations between 28.4% and 36.1% (n = 3: 5552, 708-3700, and 710-3809) were faïence-blanche, and all had tin (Sn) concentrations between 7.0% and 17.5%. The next highest Pb concentration was 48.5% (508-2430), which was identified as faïence-brune but did not have corresponding high Sn content (0.0001%). The lack of Sn in the faïence-brune is unsurprising since the brown portion of the glaze is not the part of the ceramic that was tin-glazed (Brongniart,

1854 cited in Blanchette, 1981). The Dutch, Italian, and Spanish glazed vessels had high Pb concentrations (41.1–58.4%) and were not tin-glazed. The English ceramics had more varied Pb concentrations, with some as high as 57.6% (5546) and as low as 31.7% (5557). None of the English ceramics had high Sn concentrations, but a few on the lower end of the Pb concentrations (5560, 557, & 5558) did have much higher Si (~20% versus 10%) in the glaze. This suggests that it was less Pb used in general, and there is comparatively more Si, from silicate, in the glaze recipe.

Most Pb isotope ratios of the French ceramic glazes clustered (Figure 11), but the Vallauris glaze (807-4148) is an outlier due to its higher ^{208} Pb/^{204}Pb and ^{207}Pb/^{204}Pb values and lower ^{206}Pb/^{204}Pb. It should be noted that differences are interpretably significant in the second decimal for ^{20n}Pb/^{204}Pb values and the third for ^{20n}Pb/^{206}Pb (see Chapter 2.3.3 for further details). The English samples clustered similarly to the French but with one visible outlier. The coarse earthenware (5557) had lower $^{208\ \&\ 206}$Pb/^{204}Pb values. The Spanish samples (5539 &707-3648) had very similar values to the French and English clusters, though they fall within Spanish ore (Figure 12). The Ligurian ceramic glaze (803-3968) was similar to the English and French clusters, but the Albisola ceramic glaze (804-4054) fell outside, with similar values to the French Vallauris sample.

The two Dutch samples (5540 and 5541) had very similar values to each other, but not the other ceramics. There was little interpretable difference in the isotope ratios between the green and brown parts of the glaze. This is likely because they are slip-glazed, so the difference in colour is coming from the additional oxides on the initial slip rather than the overlaying Pb-glaze. The similarity between the sherds is unsurprising because it is suspected that they are from the same vessel. The isotope ratios within the

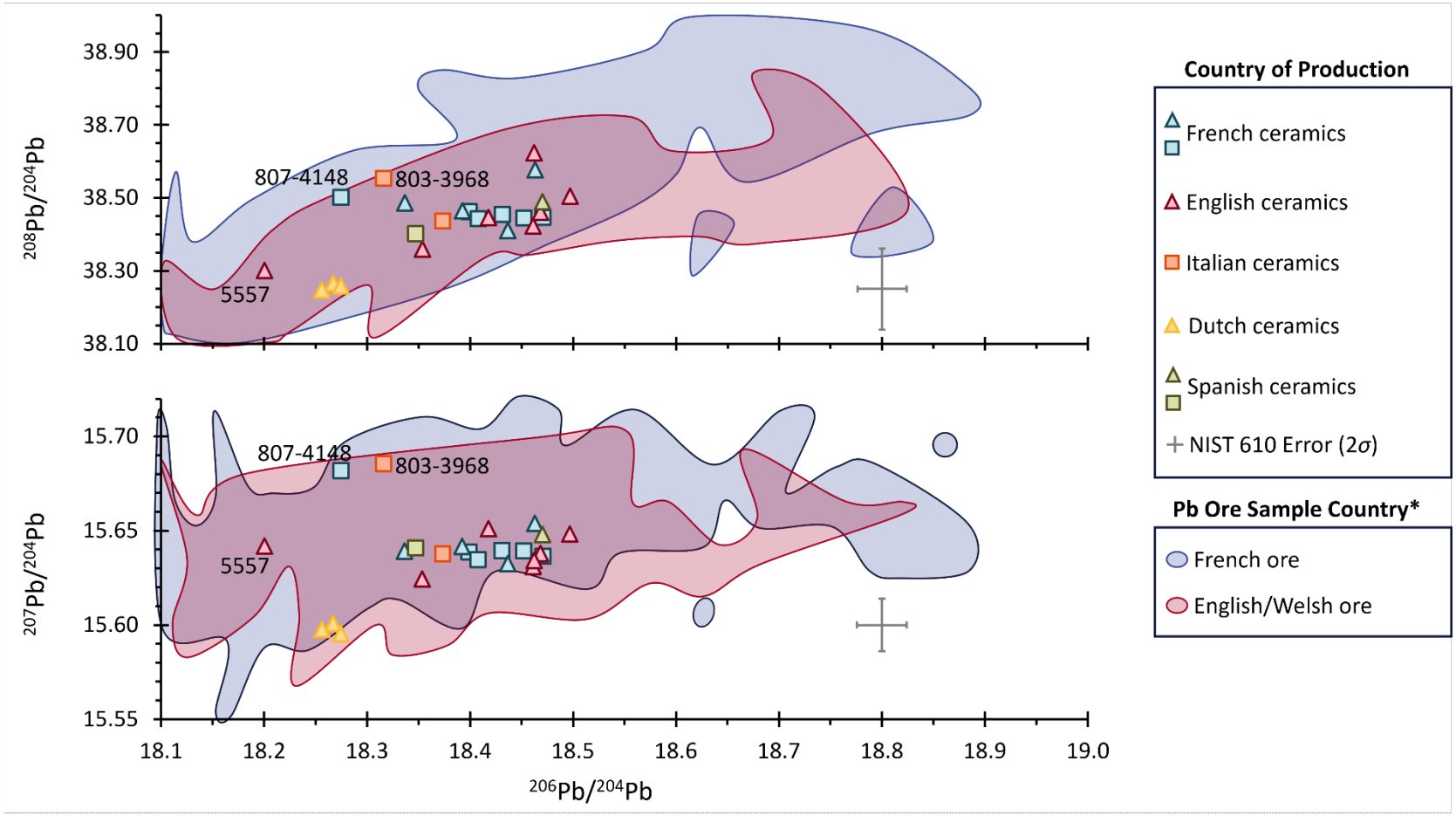

Figure 11. Comparison of $^{206}Pb/^{204}Pb$ to $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ values of ceramic glazes compared to Pb ore of France, England, and Wales are from data compiled in *Blichert-Toft et al., 2016; full ore data citations are in Table 45. Ceramic samples from Anse à Bertrand are depicted as squares and Ferryland as triangles.

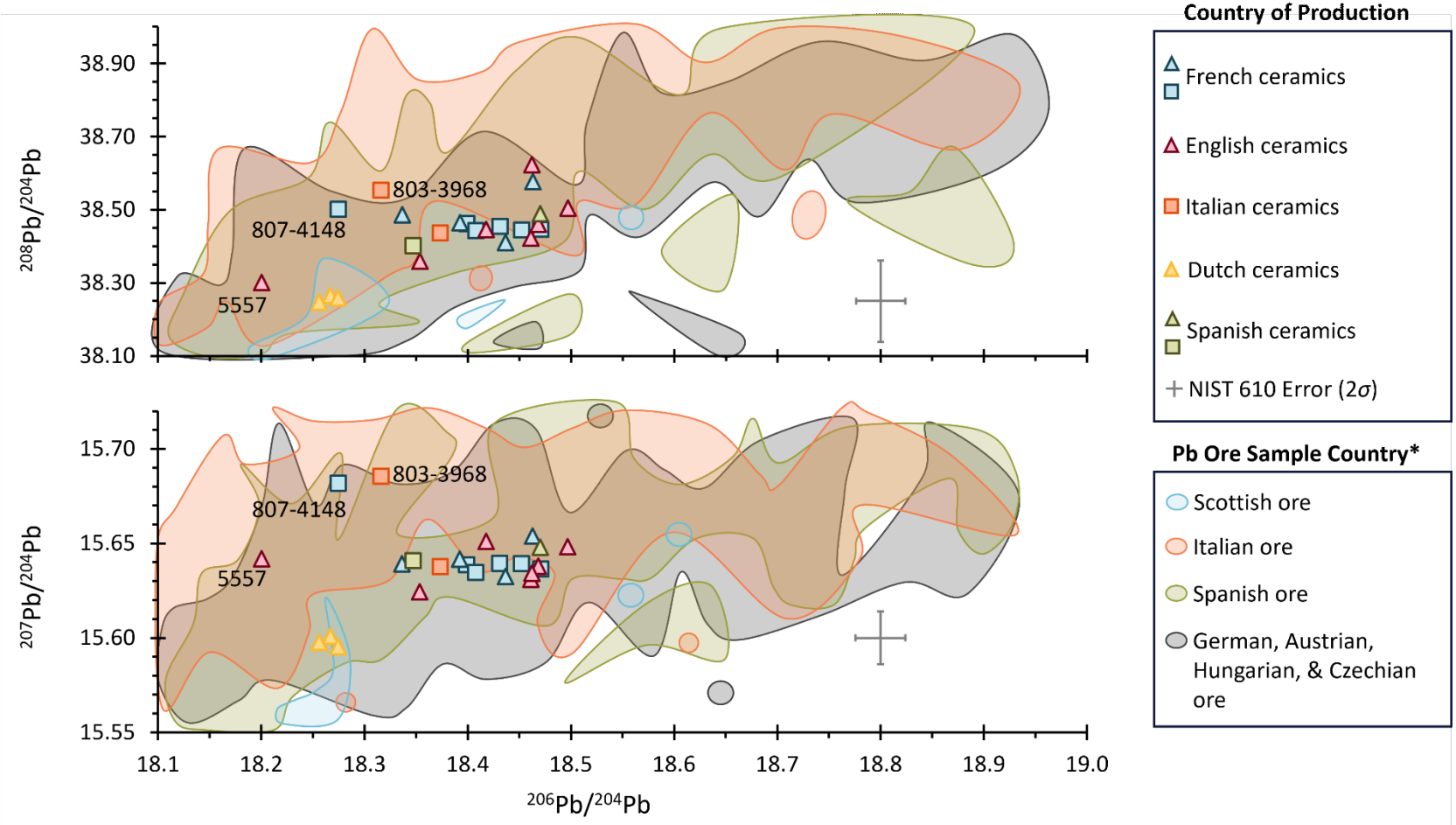

Figure 12. Comparison of $^{206}Pb/^{204}Pb$ to $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ values of ceramic glazes compared to Pb ore of Western Europe from data compiled in *Blichert-Toft et al., 2016; full ore data citations are in Table 45. Ceramic samples from Anse à Bertrand are depicted as squares and Ferryland as triangles.

glaze used for the Dutch earthenware fell well outside the English and French clusters (Figure 11; Figure 12).

4.4.2 Human values

Enamel from the first, second, and third molars (M1, M2 and M3) of the young adult female from St. Paul's Anglican Church cemetery (NP654/ F7) had variable but high concentrations of Pb (Table 7; Figure 13). While their concentration in infancy (M1) and adolescence (M3) was 18.6 ppm and 18.1 ppm, respectively, their exposure was much higher during childhood at 28.6 ppm. The Pb isotope ratios of these three teeth varied as well. For example, their $^{206}Pb/^{204}Pb$ values shifted from 18.451 to 18.910 between infancy(M1) and childhood (M2), but during adolescence (M3) they had an intermediary value of 18.625.

Enamel Pb concentrations of the eight likely New England born adolescent individuals buried at Rochefort Point, Louisbourg, NS (Scott et al., 2023), were taken from the second molars representing childhood. They had much lower concentrations than Harbour Grace F7, between 0.6 and 5.0 ppm and a mean of 2.4 ppm (Table 7). The Pb isotope ratios all clustered tightly with $^{206}Pb/^{204}Pb$ values between 18.398 and 18.438 (Table 7; Figure 13).

4.5 Discussion

4.5.1 Ceramic glaze Pb concentration

The concentrations of Pb within the ceramic glaze were variable but high (28–59%). These concentrations are consistent with those seen in other studies from 18th to 19th-

Table 7. Pb concentration and isotope ratios of the core enamel samples from the multiple teeth of F7 from St. Paul's Anglican Church Cemetery, Harbour Grace, NL, and the eight adolescent individuals identified as born in New England from Rochefort Point, Louisbourg, NS.

Individual	**Tooth**	**Pb (ppm)**	**^{208}Pb/ ^{206}Pb**	**^{206}Pb/ ^{207}Pb**	**^{208}Pb/ ^{204}Pb**	**^{207}Pb/ ^{204}Pb**	**^{206}Pb/ ^{204}Pb**	**^{208}Pb/ ^{207}Pb**	**^{207}Pb/ ^{206}Pb**
NP254 (F7)	LLM1	18.6	2.08537	1.17983	38.4761	15.6381	18.4505	2.46087	0.84758
NP254 (F7)	LLM2	28.6	2.05484	1.20652	38.8565	15.6731	18.9099	2.47965	0.82883
NP254 (F7)	LLM3	18.1	2.07476	1.18938	38.6428	15.6595	18.6251	2.46816	0.84078
1/2017	URM2	3.5	2.08665	1.17772	38.4345	15.6393	18.4187	2.45797	0.84910
4/2017	ULM2	2.8	2.08550	1.17844	38.4163	15.6316	18.4210	2.45813	0.84858
6/2017	LRM2	5.0	2.08724	1.17720	38.4174	15.6356	18.4063	2.45755	0.84947
7/2017	ULM2	0.6	2.08842	1.17635	38.4234	15.6402	18.3983	2.45717	0.85009
20/2017	ULM2	1.1	2.08612	1.17746	38.4078	15.6358	18.4103	2.45682	0.84929
22/2017	LRM2	1.5	2.08617	1.17806	38.4515	15.6458	18.4318	2.45808	0.84885
28/2018	ULM2	2.0	2.08630	1.17844	38.4669	15.6463	18.4384	2.45906	0.84858
29/2018	URM2	2.7	2.08646	1.17879	38.4684	15.6406	18.4371	2.45998	0.84833

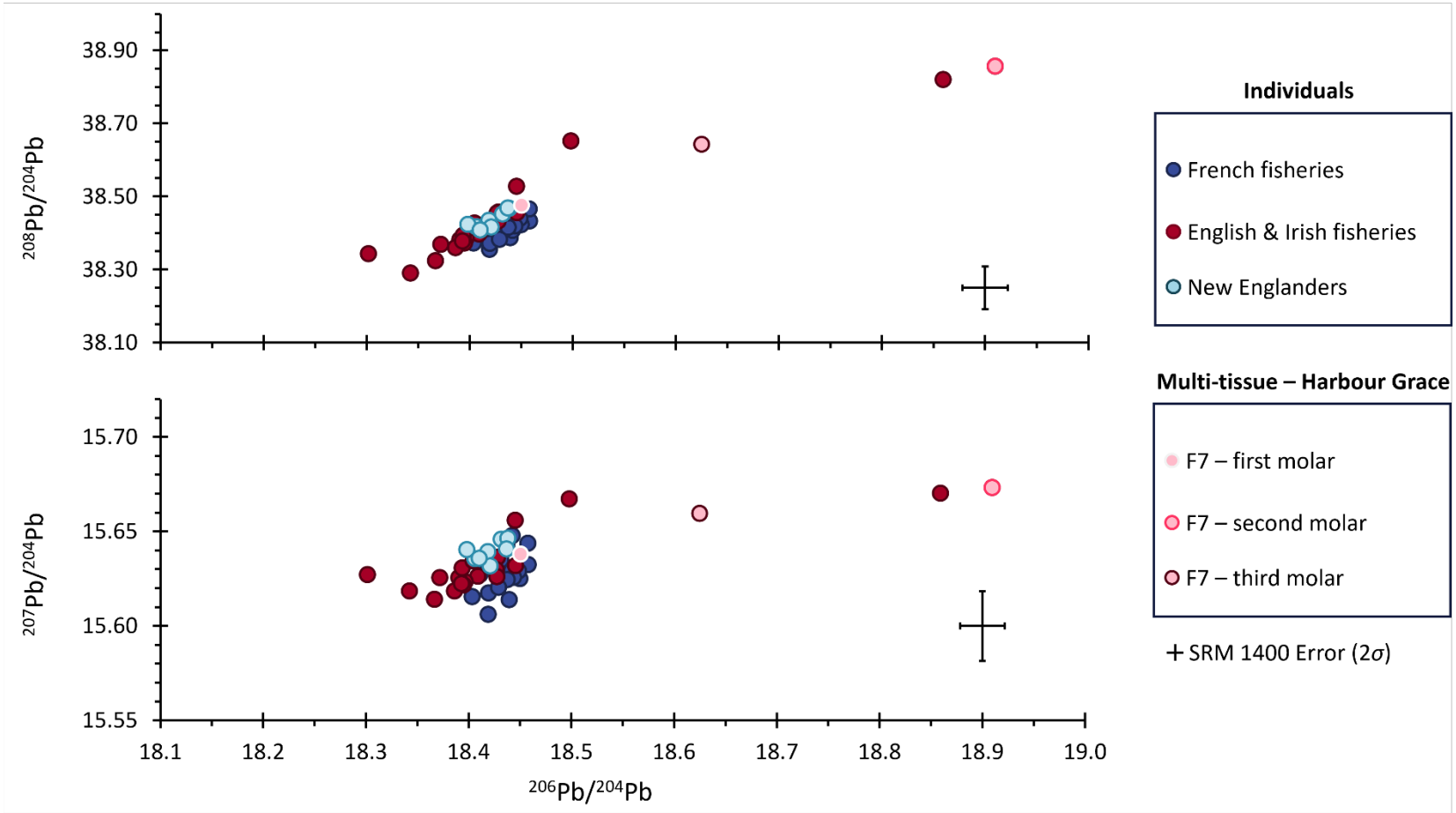

Figure 13. Dental enamel Pb isotope values representing childhood exposure of individuals from the French (Block 3) and English/Irish fisheries (Harbour Grace, Foxtrap, Wester Point, and Tors Cove). Adolescent individuals born in New England were buried at Rochefort Point, Louisbourg. First, second, and third molars from individual F-7 from Harbour Grace are depicted with white, pink, and red, respectively.

century British ceramic glazes. For example, Greer et al. (2021) examined mid-18th- to early-19th-century British ceramics and found that the glazes of creamware and pearlware were approximately 40% PbO, as seen in the 18th century creamware sherd (5556, Pb 41%) in this study. However, Greer et al. (2021) found that the later produced yellowware and whiteware were lower at 30% and 20% - though whiteware was quite variable (Greer et al., 2021). While the concentrations were similar to green and blue-edge glazes as seen in Schurr et al. (2018), they were not similar to the ~10% PbO found within red transfer print and green sponge. This difference was attributed to the decrease in Pb within glazes suggested by both historical documents and confirmed by analysis of ceramic glazes by Douglas (2000). Pre-1833 PbO within glazes was >20% and ceramic Pb contents from the 1890s could be as low as 5% (Douglas, 2000). Though in an American rather than British context, ceramics produced by various potteries in Virginia during the late-18th to late-19th centuries also had similarly high PbO in the glaze, where most were between 30% and 65% (Greer and MacDonald, 2022).

In this study, the 17th-century ceramic glaze concentrations were above 50% Pb, except the French tin-glazed ceramics. However, the lower concentration Pb in the 18th century was between 32% and 41%. While based on small sample sizes per ceramic type, most of these glazes' high concentrations are from earlier materials than examined by previous studies (Greer et al., 2021); and many of those that examine glaze focused on creamware, pearlware, and whitewares (Schurr et al., 2018; Greer et al., 2021). Part of the explanation of the higher concentrations could be the differences in glaze recipes for these other types of ceramics, or it may be due to a further decrease in the amount of Pb within glazes from the 17th to 18th centuries. Regardless of the differences between glazes,

the concentrations were high enough that leeching from normal use would have resulted in Pb exposure to the fisheries populations interacting with the food or drinks prepared, stored, or served in the vessels.

4.5.2 Ceramics compared to Pb ore

There are no distinct differences between the Pb isotope values of ceramics when separated by country of production, except for the Dutch (Figure 14). The Pb isotope values from all the ceramics unsurprisingly fall broadly within the Western European and English and Welsh Pb ore fields. However, differences in the geographic origins of Pb used in the glaze exist, including between ceramics from the same country. The following section compared Pb isotope values within ceramic glazes from each country to values from relevant ore bodies.

The main group of French ceramics fits within the Pb isotope values of ore from Cévennes, Montange Noire, and the Armorican Massif (Figure 15). While outside the range, when considering analytical error, the Central Massif and Alps also encompass the main French ceramic glazes. However, they also broadly fit within the values of Pb ore from several other areas, including English/Welsh Pb ore (Figure 16). While there are records on the export of English Pb to France (Burt, 1984), it is not clear what specific potteries used for their glazes.

Most of the Pb isotope values of glaze from English ceramics cluster together. Shropshire is the only English ore field that Pb from this main cluster could not originate from (Figure 16). Central Welsh Pb may also be a possibility for some ceramics when accounting for analytical error. Since England was a major Pb producer (Burt, 1984), it

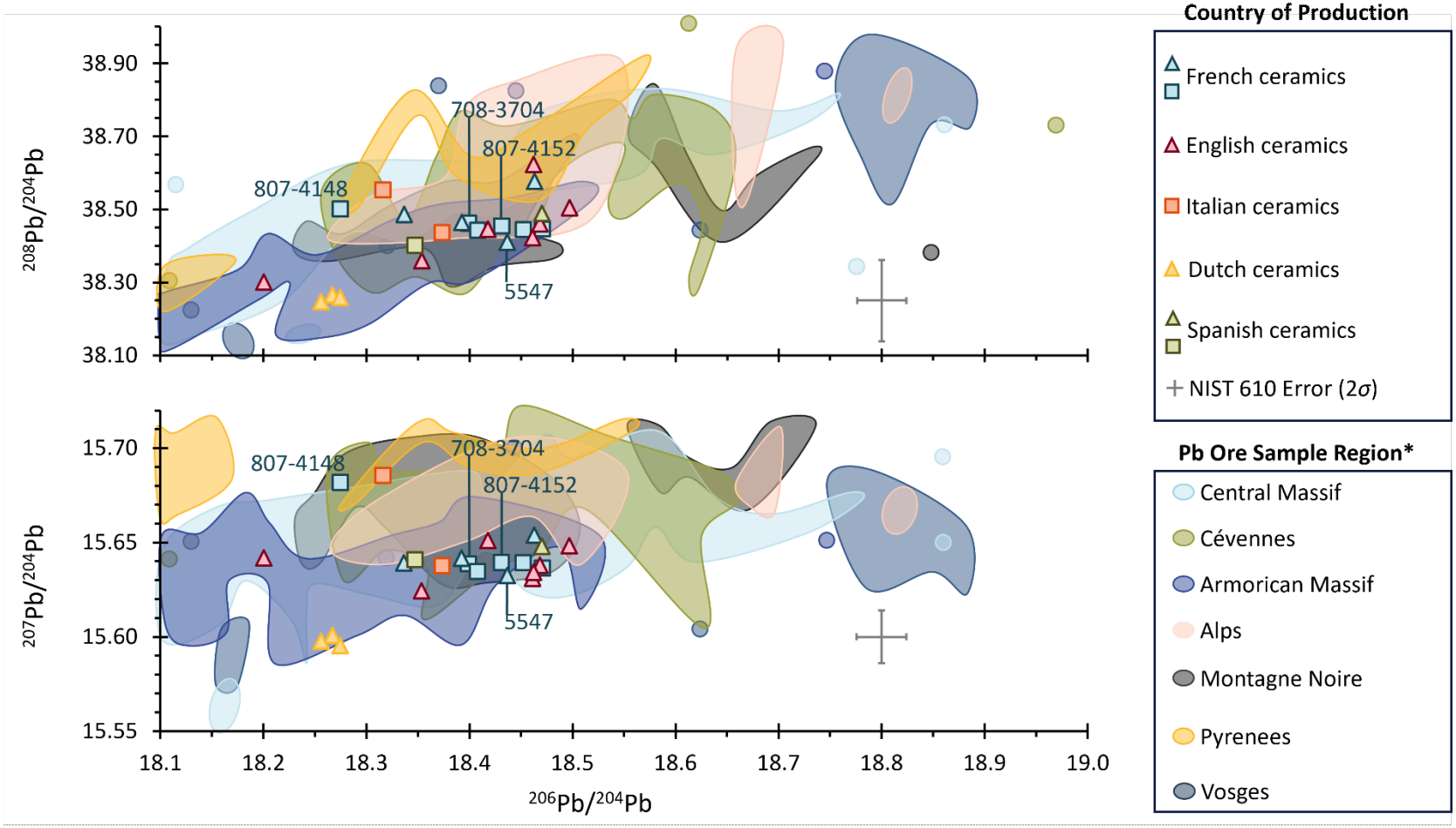

Figure 14. Pb isotope values of ceramics compared to values from ore within France (Blichert-Toft et al., 2016). Ore is separated based on geologic formations and citations are available in Appendix Table 45. Ceramic samples from Anse à Bertrand are depicted as squares and Ferryland as triangles.

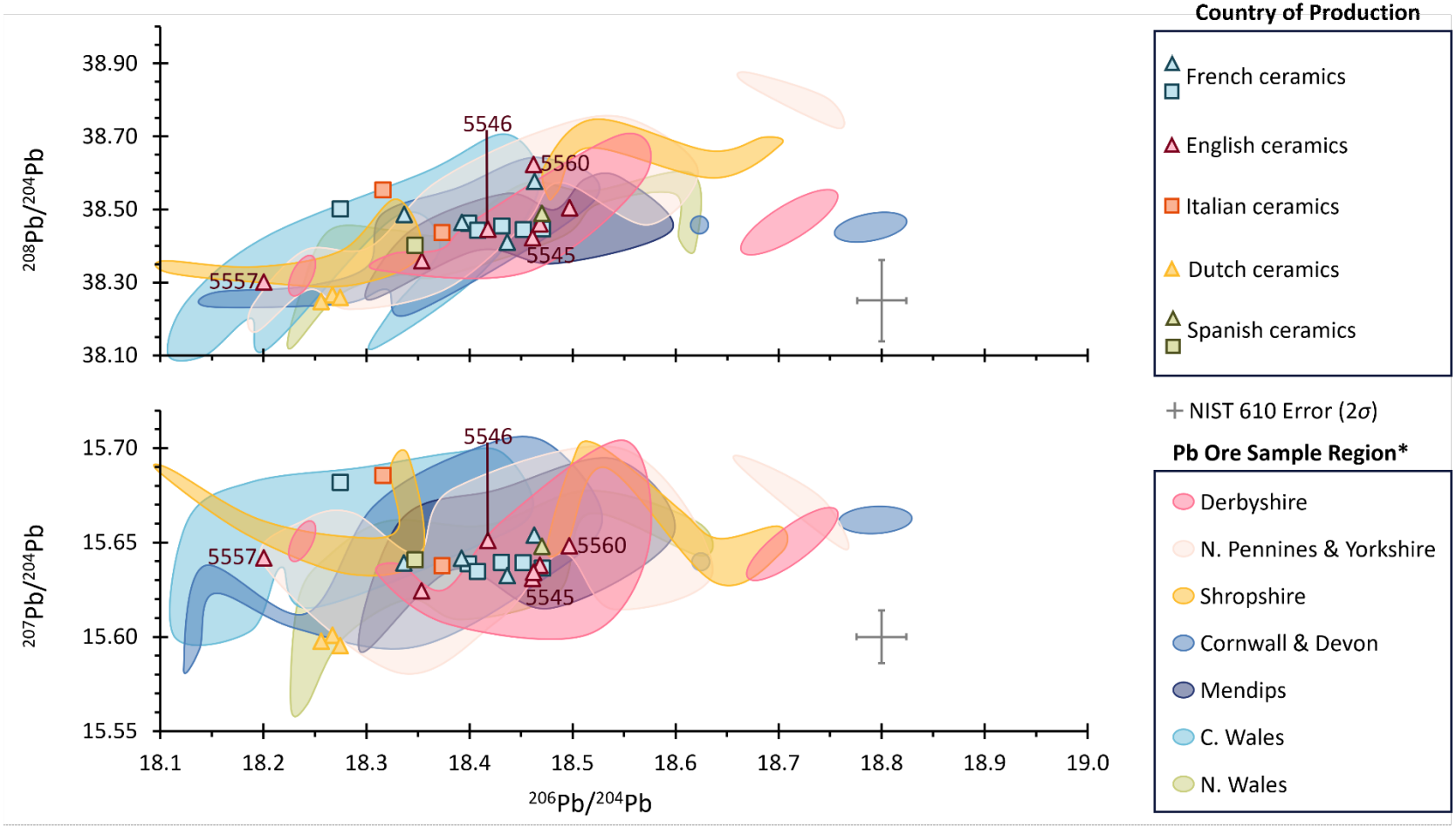

Figure 15. Pb isotope values of ceramics compared to values from ore within England and Wales (Blichert-Toft et al., 2016). Ore is separated based on historical Pb mining regions (Burt, 1969) and geographic formations (Evans et al., 2022) and citations are available in Appendix Table 45. Ceramic samples from Anse à Bertrand are depicted as squares and Ferryland as triangles.

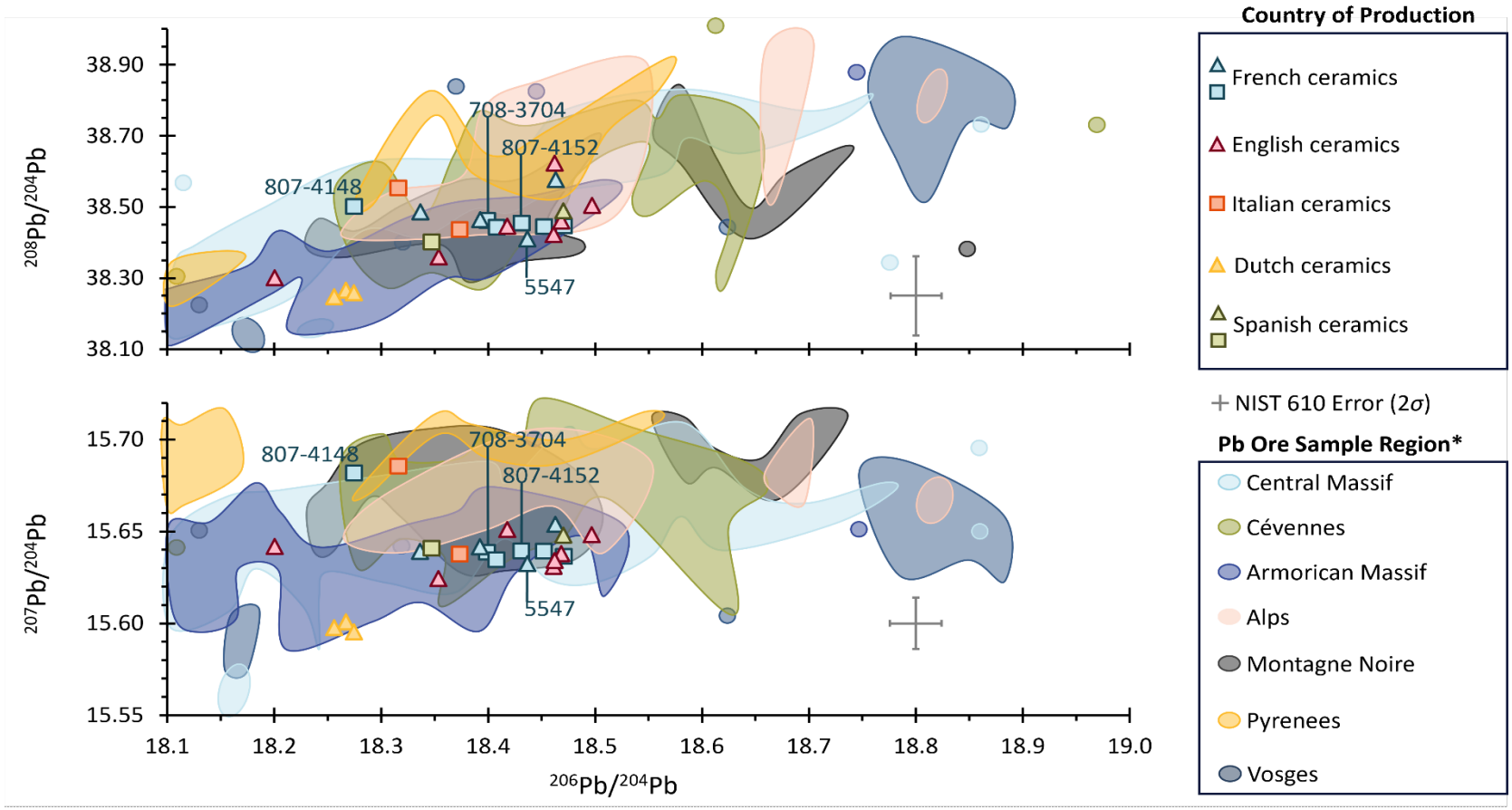

Figure 16. Pb isotope values of ceramics compared to relevant Pb ore from geological formations within France. Pb values were compiled by Blichert-Toft et al., 2016, and are separated based on geologic formations with citations available in Appendix Table 45. Ceramic samples from Anse à Bertrand are depicted as squares and Ferryland as triangles.

was unlikely that English potteries needed to import Pb for ceramic glazes, though it may have been transported throughout the country itself.

The other foreign-made ceramics had more variable origins (Figure 11). The Italian ceramics both fit within Italian Pb ore isotope values, but the Ligurian ceramic is on the outskirts of the ore data. The Spanish ceramics are consistent broadly with Spanish ore, but do not have distinct values, therefore, other areas cannot be discounted. There is no local Pb ore in the Netherlands, so the values within the glaze must be compared to ore bodies elsewhere in Europe.

The Pb isotope values of the Dutch ceramic sherds' glazes are consistent with Pb ore from a few different regions, namely, within the Armorican Massif (France; Figure 16), Cornwall and Devon (England; Figure 16), Irish deposits (Appendix Figure 29) and German Pb broadly (Figure 12; Figure 28). They are also consistent with outlying values from Scottish deposits (Figure 12). This is significant because there are later accounts of potters importing Scottish Pb ore in the 18th century. The Czech Republic and kingdoms within what is now Germany, especially Bohemia, Bavaria, and Saxony, mined significant Pb ore in the 16th century (Nef, 1987; Renberg et al., 2000; Shotyk et al., 2002). In turn, it is possible that these regions were still exporting Pb during this period, or that previously mined material was reworked into the glaze of these Dutch ceramics. More research is needed to describe potteries and where they were sourcing their materials specifically.

Some of the ceramics are from known or suspected origins within countries and, therefore, can be compared to locally available ore where available. The English Staffordshire-type ceramic (5560) while named after the region, is a misnomer since other

potteries in the North Midlands and Bristol produced this style of ware (Barker & Crompton, 2007). Though not within Staffordshire, there are a number of large mines in the area which may have produced the Pb available to the potteries. However, there are some accounts of at least one Staffordshire potter during the 1630s using local (within 6 miles) crushed galena in the glaze. Most of these galena deposits in the region are not distinguishable isotopically, but the large mining areas within or close to those potteries that produced the Staffordshire-type ceramic include Derbyshire, Bristol, and Shropshire (Burt, 1969). Isotopically, the ceramic glaze fits within the Derbyshire and Bristol ore, but not that of Shropshire (though it is on the outskirts of the data when considering analytical error; Figure 17). So, while it cannot be discounted that they used local small galena mines, as they would be isotopically indistinguishable from the Derbyshire and Bristol ores (Evans et al., 2022), the use of at least exclusively Shropshire Pb can be.

There are two other English ceramic sherds with known origins from North Devon: North Devon gravel (5545) and North Devon smooth (5546). While the glaze is isotopically consistent with multiple areas, they do fall within the expected values for local Devon deposits (Figure 17).

While not with a specific known origin of the ceramic, the English dark brown/purple coarse earthenware (5557) is possibly Midland Purple or Manganese Mottled. The Pb isotope ratios of the glaze are higher than the values expected in most English mines, but it is consistent with a central Wales ore field. In terms of the glaze composition, the dark colour is likely from the comparatively high amounts of Mn (2.8%) and Fe (1.5%) within the glaze. There are no comparative data for elemental concentrations of glazes of

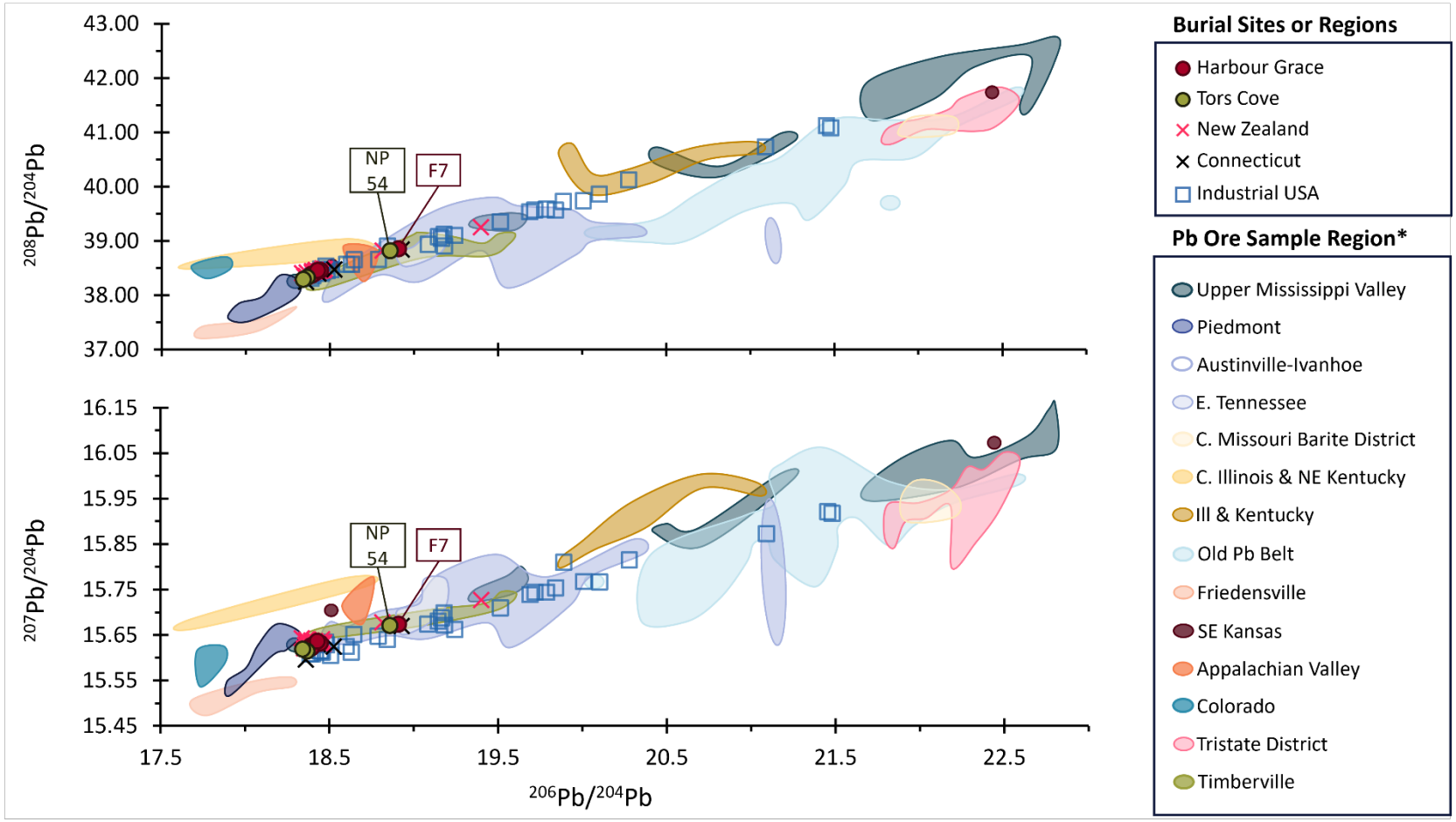

Figure 17. Pb isotope values of ceramics compared to relevant Pb ore from geological formations within England and Wales. Pb values were compiled by Blichert-Toft et al., 2016, and are separated based on historical Pb mining regions (Burt, 1969) geologic formations (Evans et al., 2022) with citations available in Appendix Table 45. Ceramic samples from Anse à Bertrand are depicted as squares and Ferryland as triangles.

Midlands Purple, but there are some data using x-ray spectrometry for glazes of Manganese Mottled. Philipot (1985) found that Manganese mottled may be a misnomer since there was very little Mn within the glaze, as was found in a previous study as well. However, this is based on two ceramics, and historic glaze recipes mentions Mn specifically as a major ingredient (Philipot, 1985). If the sherd sampled in this study is Manganese Mottled, it would suggest that there were likely regional or temporal differences in the glaze recipes. Interestingly, Manganese Mottled was produced in both Staffordshire, England, and Buckley, Wales. The Pb isotope values do not fall within those in the Midlands, but Buckley is located within the area with galena consistent to that found in the glaze. Clearly there is not enough information to use this for identification, but it would be interesting for future studies to compare the glazes of Mottled produced in the various potteries to determine if there is such a distinction.

The Breton (French) Pabu-Guingamp ceramic (807-4152) has very similar values to the bulk of the local data from the Côte d'armour within the Armorican Massif (Figure 16). Pb glazed tiles from the medieval period in Brittany did use imported Pb from Derbyshire England based on their isotope values (Métreau et al., 2021). While the Pabu-Guingamp ceramic is consistent with some English ore deposits, including Derbyshire, they do not fall outside of those possible locally.

There were two Saintonge ceramic sherds (708-3704 and 5547). There are no local Pb sources that have been sampled within the area of production (Blichert-Toft et al., 2016), nor has the author been able to find evidence of local outcrops that could have been used in the pottery industry. Isotopically, the sherds are consistent with values from Cévennes and Montagne Noire, but those deposits are on the Eastern side of France and

would necessitate long distance transport around the Iberian Peninsula, over notoriously poor roads, or through canal systems and down the coast. They are however consistent with both the Armorican Massif and on the edge of the Central Massif. The city of Saintes is the main production center of Saintonge ceramics and is along the Charente River (Barton, 1963). There are deposits along the edge of the Charente River watershed (Descy et al., 2022), which may have facilitated the galena or Pb oxide's transport up the river to Saintes. However, given the strong sea-transport ties of the city (Barton, 1963), it more likely was brought in by the coast from the Armorican Massif.

No known Pb ore source is located near the French town of Vallauris (historically or within available isotope data) that could have been used in producing the glaze of the Vallauris sherd (807-4148), and the values fall outside the available Pb ore data for the local Maures Massif just south of the region, nor the Alps more broadly (Figure 16; Blichert-Toft et al., 2016). It should be noted that there are only four data points for the nearby Maures Massif, which is smaller than the 30–50 recommended to properly characterize the Pb isotope values of the formation (Baxter et al., 2000; Stos-Gale and Gale, 2009). The Alps as a whole only have 29 data points (Blichert-Toft et al., 2016), despite their complex geology. The Vallauris sherd's glaze is consistent with materials that must be imported from further in France or abroad. Based on isotopic values, this includes Montagne Noire, Cévennes, and Italy broadly. While on the outskirts of the data, the glaze's values are also consistent with the Central Massif or Pyrenees. Considering Vallauris is located in eastern France, English Pb logistically is less likely, but the levels are only consistent with Central Welsh ore (Figure 17). While the location of the galena remains unclear, the process of making the glaze for Vallauris is recorded in primary

documents, which explain that Pb was ground into fine powder at a local granite mill, and by the 19th century, a roller mill was used (Petrucci, 1999).

The Italian Albisola sherd (804-4054) has very similar values to Vallauris. These Pb values are consistent with Pb from the area of what is now Italy (Figure 11). The island of Sardinia was an active Pb mining region since the Roman period, with production increasing during the 18th century (Pillola, 2002). Given Albisola's proximity to Sardinia, this makes it a strong possibility. An intriguing connection to Vallauris in addition to the similar Pb isotope values, is that it was recorded that clay harvested near Vallauris was exported to Albisola for their ceramic production (Petrucci, 1999).

In contrast to Albisola, the other Italian ceramic analyzed from the Ligurian region (803-3968) is on the outskirts of known values of Italian Pb ore and is not distinct from specific regions of Europe. Therefore, even though the Albisola and Ligurian sherds were both produced in the same region of Italy, distinctly different sources of Pb were used in their manufacture.

Based on the comparison of the ceramic glaze and ore Pb isotope values, it is clear that some regions likely imported their Pb, possibly from outside of the country, before incorporating it into ceramic glaze. After exposure to the Pb-glaze of ceramics, the Pb isotope values incorporated into skeletal remains would be an average of all sources available to the individual. The acidity of food or drink and length of cooking or storage time would impact the amount of Pb released and therefore the relevancy of outlier isotopic values to the final Pb burden and isotope ratio incorporated into tooth enamel. In other words, communities, or individuals within communities who frequently used

vessels, or other Pb-containing materials and products with outlying values would have Pb isotope ratios that integrate exposures across their various sources.

4.5.3 Limitations of modern ore isotope values

The use of Pb ore isotope values to contextualize isotopic values within material culture or humans is essential, but there are limitations to this method: (1) at least 30 (oftentimes more) data points are needed to contextualize deposits (Baxter et al., 2000; Stos-Gale and Gale, 2009), but many datasets do not reach this threshold; and (2) these ore samples mostly come from areas that in the modern period were important, commonly based on mining prospects. Therefore, those deposits may not have been actively mined during the period in question (in this case the 17th to mid-19th centuries). Since they are irrelevant for comparisons, their inclusion adds to the overlap of values seen in Western European deposits; and (3) not all deposits that were relevant in the past have been sampled, or they have been completely exhausted, so data from them is missing.

Such issues represent fundamental limitations since we cannot know what data are missing. Some of these problems are seen within the sampled ceramic examples, such as the lack of Pb ore isotope ratio values from the Maures massif which may have been relevant for the Vallauris ceramic. Despite the limitations, it is clear that more study is needed to identify sources and explore the full ramifications of ceramic exposure. Not enough known about relationship between Pb use in ceramic glazes and sourcing relevant ore bodies. The ceramic sampling was a pilot project and does not fully represent the likely range of values expected in the human populations. While the Pb isotope ranges from the ore bodies overestimate the possible values in enamel, it is possible that further

sampling of ceramics (and or other relevant sources) would better characterize the possible values expected within a population.

4.5.4 Ceramic glazes and human Pb exposure in the North Atlantic fisheries and New England

The measured Pb isotope ratios of ceramic glazes are somewhat variable but can be used along with Pb ore data to contextualize sources of exposure to individuals from fishery populations in contexts similar to where the ceramics were originally excavated. Additional enamel samples from the first and second molars of an individual buried in Harbour Grace, NL and samples from second molars of adolescents born in New England but buried at Rochefort Point, Louisbourg, NS, are used for further comparisons. The comparison between Pb isotope values in human enamel, ceramic glazes, and Pb ore data, focuses first on the tooth enamel Pb data sampled from the French fishery site in NS (n = 18), and the English and Irish fishing sites in NL (n = 21) (Chapter 3). By grouping the ceramic glazes by site and incorporating analytical error into the resulting range (Anse à Bertrand and Ferryland; Figure 18), broader cultural practices within the fisheries can be inferred. Assuming that these sampled ceramics represent the possible Pb exposure sources to the broader French and English/Irish fishery populations in NL and NS, we can address the potential of using Pb glazes to aid in establishing baseline anthropogenic isotope value ranges.

4.5.4.1 <u>French individuals from Block 3, Louisbourg</u>

The Pb isotope values of the French fishery individuals were within the proposed bioavailable range of anthropogenic Pb, as suggested by the French ceramic glazes

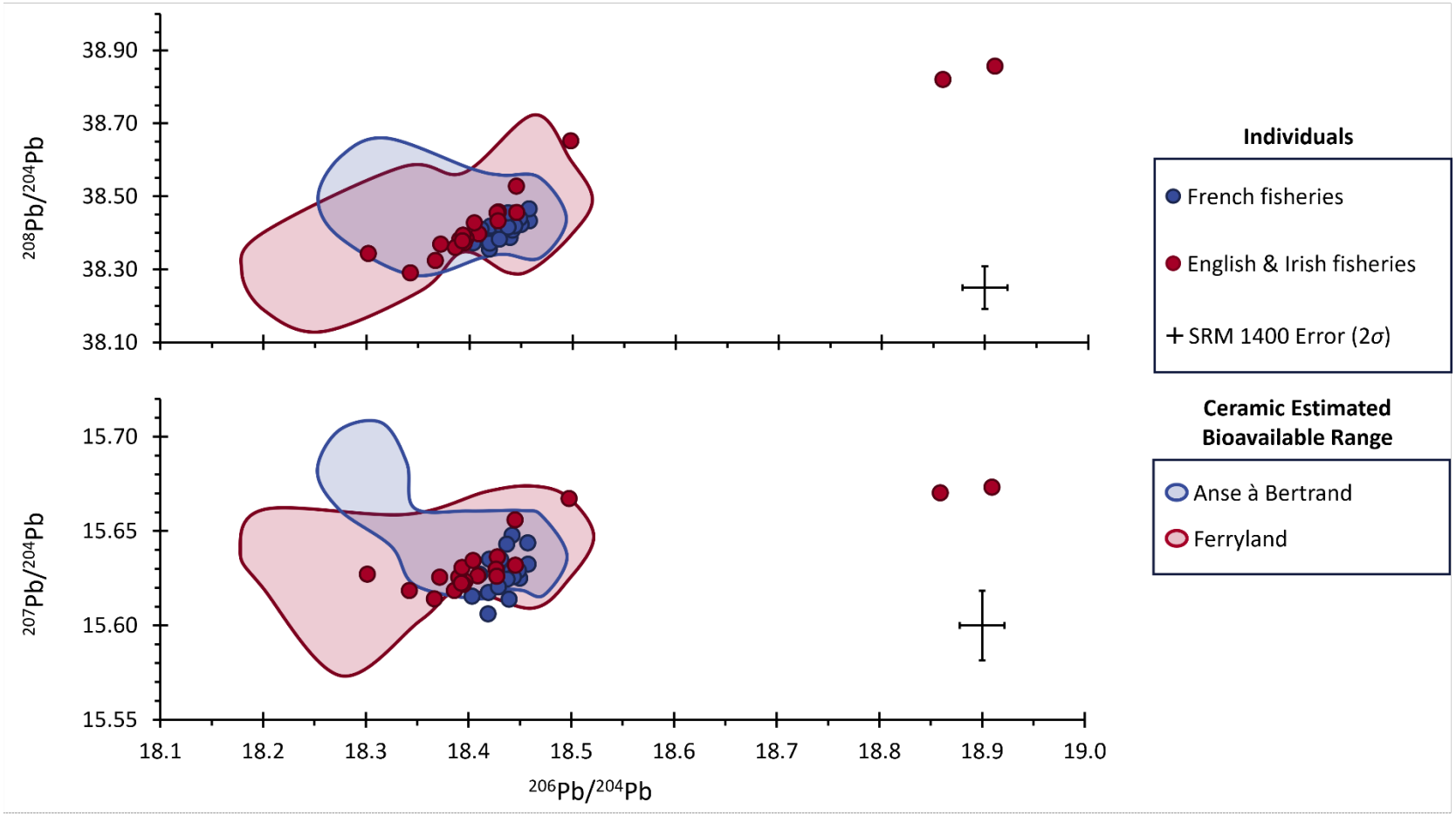

Figure 18. Pb isotope values from tooth enamel representing childhood exposure in French and English/Irish fishery populations. Values are compared to the ceramics samples from Anse à Bertrand (French), depicted as squares and Ferryland (English), as triangles. The range of values expected in French and English cultural fishery environments in Newfoundland and Saint-Pierre based on exposure to a limited number of ceramics are outlined in blue and red, respectively.

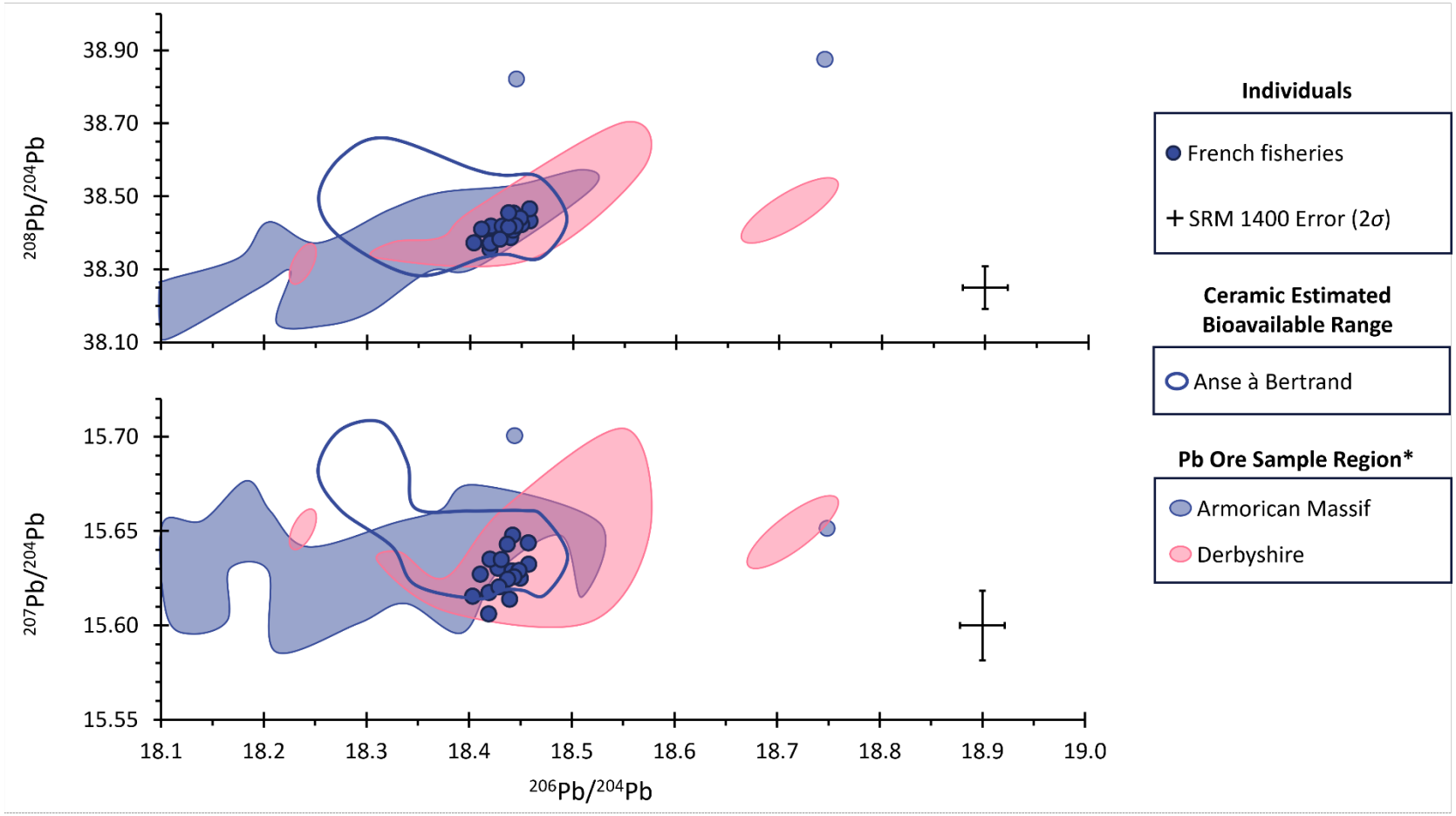

Figure 19. Pb isotope values from tooth enamel representing childhood exposure in French and English/Irish fishery populations. The range of values expected in French cultural fishery environments in Newfoundland and Saint-Pierre based on exposure to ceramics are outlined in blue. Values are also compared to relevant Pb ore from geological formations in France and Britain compiled in Blichert-Toft et al., 2016 with full citations available in Appendix Table 45.

(Figure 19). However, a couple individuals had higher $^{206}Pb/^{204}Pb$ values and lower $^{208\ \&\ 207}Pb/^{204}Pb$ values, even when considering 2σ analytical error. Therefore, there is some source(s) of Pb missing. These outliers included individuals 2 and 12 from the Block 3 cemetery, Louisbourg. It is possible that they fall outside of the range because not all of those buried at the cemetery were born into French Atlantic fishing communities. As previously stated, none of the individuals buried at Block 3 were born locally (Appendix 7.2.1.6). Since this was the earliest cemetery (1713–1723) in the newly established Fortress of Louisbourg (1714), the population is an amalgamation of people from multiple regions, mainly in France or New France (i.e., Plaisance, Quebec, and Acadia). Based on their $^{87}Sr/^{86}Sr$ values and dietary patterns, they are not from the same location, though specifically where is unclear (Garlie, 2022). Neither are inconsistent with what would be expected to be found in France. In which case, the outlying values are due to a source or sources of Pb available at some of the individuals' places of origin during childhood. The values of the individuals themselves are similar to the Armorican Massif (Figure 19), though they do not directly fall within any known values. This makes sense as a potential source, particularly considering Brittany's strong connections to Plaisance and Louisbourg. While the one Breton ceramic (807-4152) from Anse à Bertrand did not have such shifted values, the region is large, so this remains a possibility. However, if this difference is due to an averaging of isotope values, it would have to have been a source with higher $^{206}Pb/^{204}Pb$ and lower $^{208\ \&\ 207}Pb/^{204}Pb$ values to pull the average and would suggest a source outside of France.

Derbyshire ore could explain at least some of the shift in values. Adding strength to this suggestion, is that there is late medieval evidence for Derbyshire Pb use in Pb-

glazed tiles made in Brittany and Anjou (Métreau et al., 2021), so the use of this source may have continued into later periods. Therefore, it does not necessarily suggest that they were from outside of France, but that English ore was broadly exported and could affect the bioavailable anthropogenic Pb in other countries.

4.5.4.2 English and Irish fishery populations

The Pb isotope ratios of most of the English or Irish individuals are consistent with the values from the bioavailable anthropogenic Pb proposed from the ceramics; however, there are three individuals with outlying Pb isotope values (Figure 20). Unlike at Louisbourg, where the inconsistency between the range of French fishery individuals is due to a childhood engaged in a dissimilar cultural material environment, there is evidence that at least two of the English/Irish outliers grew up in NL. One of these individuals is just outside of the range when considering 2σ error (Foxtrap F12). This adult, interred with a Nova Scotia halfpenny from 1862, was a possible female whose $^{87}Sr/^{86}Sr$ values were consistent with a childhood spent in a coastal area and their $\delta^{13}C$ and $\delta^{15}N$ values of bone collagen suggested they consumed a mixed marine-C_3 diet (Grimes et al., 2018; Garlie, 2022). While it cannot be certain, the data is consistent with having grown up in Newfoundland, or more broadly in the Atlantic fisheries. Taphonomy precluded the ability for a more refined age at death, but unless she was a very old adult, there is a clear offset in the timing of most of the ceramics and what this individual would have been utilizing during childhood. The Pb isotope ratios are consistent or very similar to those found in England, including Shropshire, Northern Pennines, and Derbyshire. Therefore, it could be that the values represented in the ceramics do not fully represent

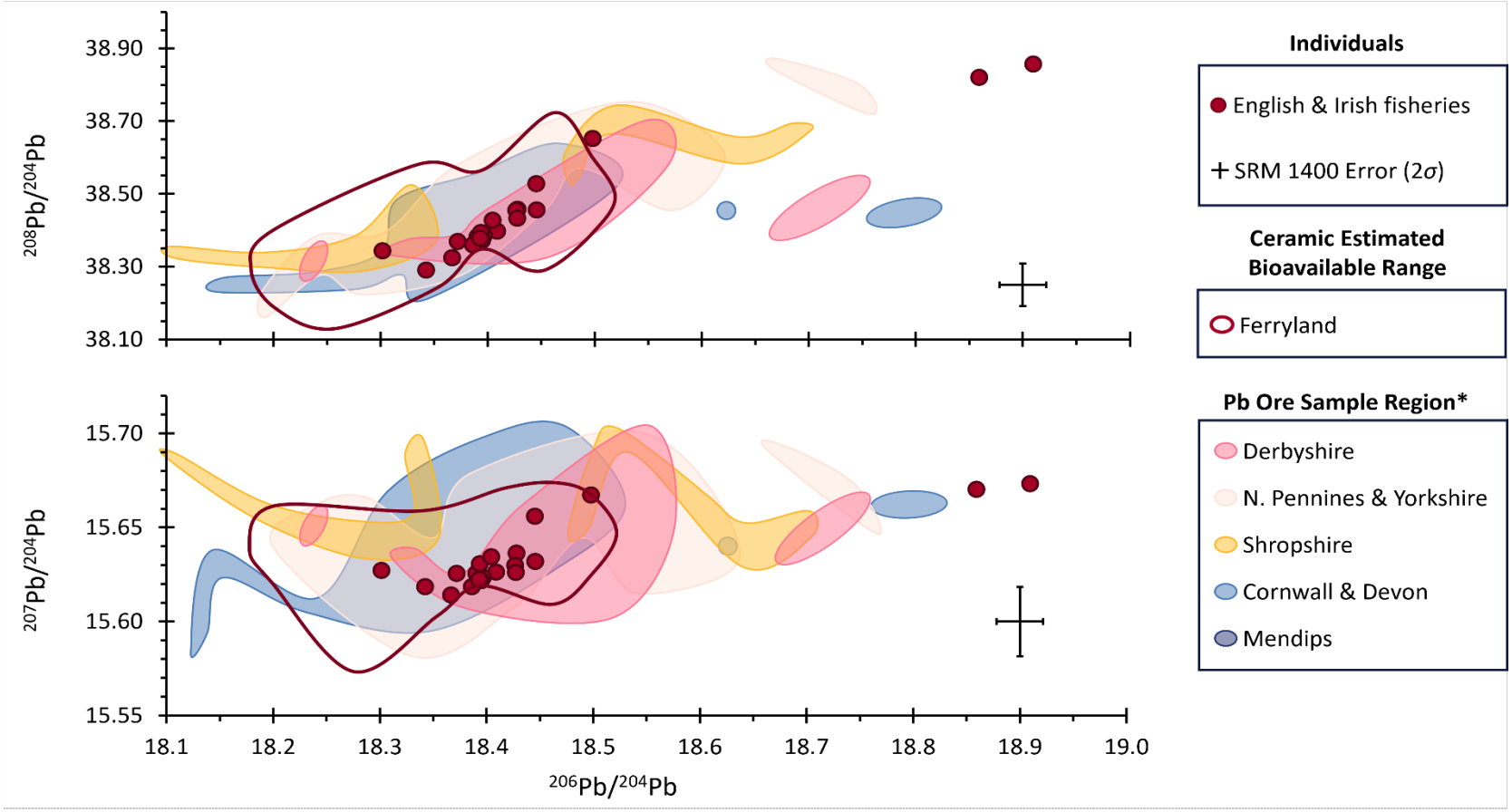

Figure 20. Pb isotope values from tooth enamel representing childhood exposure in French and English/Irish fishery populations. The range of values expected in French cultural fishery environments in Newfoundland and Saint-Pierre based on exposure to ceramics are outlined in blue. Values are also compared to relevant Pb ore from geological formations in France and Britain compiled in Blichert-Toft et al., 2016 with full citations available in Appendix Table 45.

those available in the mid 1800s even within English materials. It could also be an averaging from an American Pb ore, but based on the available data the only mines that could have a value able to average out are Appalachian Valley which were in production by the mid 1850s (Appendix Table 19), which makes the timing less likely though not impossible. The two other outliers in the English fisheries were more clearly associated with American Pb ore.

The two additional individuals had outlying Pb isotope values consistent with American ore (Figure 21): a young adult woman buried in Harbour Grace (F7) and an adolescent buried in Tors Cove (NP 54). The Harbour Grace individual F7 was previously interpreted as likely living in NL, at least during childhood, while their second molar developed (Munkittrick et al., 2019). This was based on their low δ^{18}O enamel carbonate values and the high amount of fish in their diet suggested by their δ^{13}C and δ^{15}N values from bone collagen (Munkittrick et al., 2019). Similar data from the Tors Cove individual is not yet available. While an American childhood cannot be excluded for the individual from Tors Cove, available birth records from the mid-19th century suggest that residents were born in Newfoundland and the few immigrants were from Ireland (Reddigan, 2007; Losier and Lear, 2018).

This study sampled additional teeth from a woman buried in Harbour Grace (F7) to look at possible changes from infancy to adolescence. Pb isotope values during earliest infancy (first molar) were consistent with the others in the population but shifted drastically towards American Pb during childhood (second molar) along with a large increase in Pb concentration; during adolescence (third molar) their Pb concentration and isotope values was between those from earlier in life (Figure 13; Figure 21). Since the

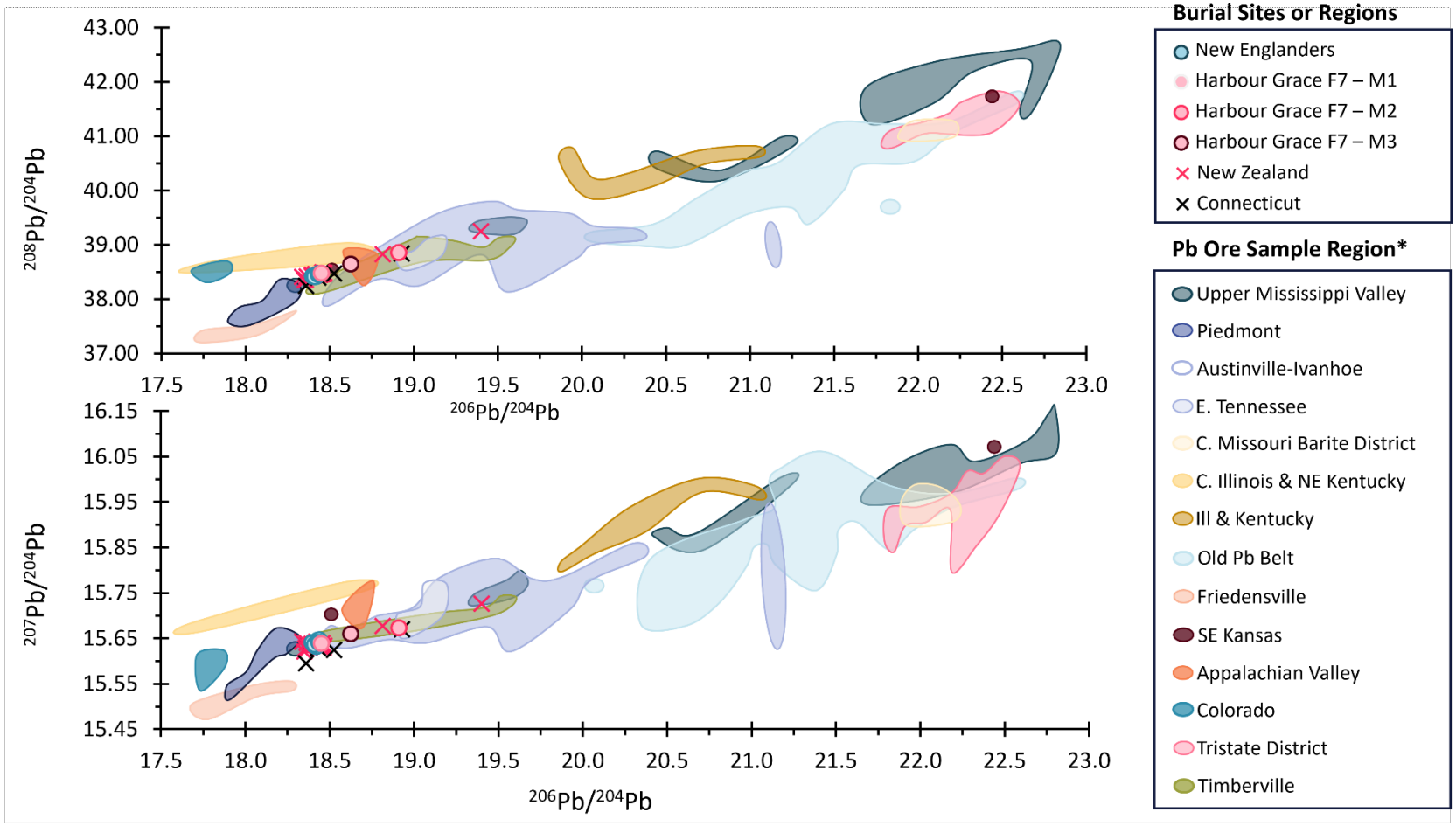

Figure 21. Pb isotope values of American ore compared to dental enamel representing childhood exposure from adolescent individuals born in New England (Rochefort Point, Louisbourg, NS). First, second, and third molars from individual F-7 (St. Paul's Anglican Church cemetery, Harbour Grace, NL) are depicted with white, pink, and red, respectively. Pb ore groupings are available in Appendix Table 45. Data is compared to human data from Otago, New Zealand (King et al., 2020), and New Haven, Connecticut, USA (Aronsen et al. 2019), with further details in Table 17.

first molar forms while breastfeeding is likely occurring, the Pb values that are consistent with English or Welsh Pb ore could originate at least in part from the mother or caregiver (Gulson et al., 2004). However, the shift back towards English/Welsh Pb during adolescence would have to be their own exposure. While the change in isotope values could suggest a change in location, when considering that it is exclusively imported Pb materials that resulted in exposure in Newfoundland, it is more likely that this was due to a change in use of Pb-containing material. Instead, they were likely introduced to a new source of Pb during childhood, but it was used less often during adolescence resulting in averaged values. It is also possible that the exposure to this source did not continue during adolescence, but their earlier exposure created a long-term averaging that was still available within the body through bone turnover during the period of tooth formation.

No American Pb-glazed ceramics were available for study, but this does not mean they were unavailable to individuals living in these contexts. Both Harbour Grace and Ferryland (close to Tors Cove), had strong trade connections with New England (Head, 1976), but very little archaeological research has been done in Harbour Grace in contexts where ceramics could be found. Such materials may have better resembled the range seen in the individuals buried in Harbour Grace and Tors Cove with outlying values. There does present the question of when materials containing American Pb were likely available to people living in North America. In turn, a site pre-dating these with outlying values but associated with New England must be examined.

The estimated bioavailable range of anthropogenic Pb does not fully encompass that of the individuals. This is likely due to a few reasons. First is the temporal offset for the English and Irish fisheries to the ceramics. The ceramics are from the 17th to 19th

centuries, with more ceramics likely representing the 18th and 19th centuries. Additionally, these individuals associated with the fishery during their later life were not all born within Plaisance or other French territory fishing communities. Many were likely born in France with different available Pb-containing materials. Therefore, it is likely that this range is fundamentally inappropriate for some of the individuals and does not encompass all of them, which is unsurprising. It is interesting, though, that despite the varied origins, there is a relatively tight cluster of values. This exercise of comparing glaze and individuals has emphasized the variability of values available within a population. The English/Irish fishers being exposed to outlying sourced while in the fishery context has implications for bioarchaeologists abilities to identify non-local individuals using Pb isotope analyses. It brings into question whether or not the British model of cultural focusing is appropriate to be applied in the broader Atlantic World.

4.5.5 Timing of American Pb access in Newfoundland

Before further exploring the applicability of cultural focusing in the Atlantic World, we must first examine the timing of American Pb ore values within currently available data. The English fisheries are not the only place that American ore values have been found. An earlier study in Colorado found many unspecified age adult individuals with American Pb ore values in a cemetery active from 1879 to 1900; they were likely exposed to this Pb in the mid to late 19th century (Keller et al., 2016). Illinois also has many individuals with similarly high Pb isotope ratios. Some of these were adults over the age of 55 and the cemetery was active from 1832 to 1873 (Fitch et al., 2012), meaning that it could have been as early as the late 1700s when these individuals were exposed. Finally,

while not within the Atlantic World specifically, there are individuals with similar values buried at a cemetery actively used between 1861 and 1863 in Oatago (King et al., 2021). This is not to say that they were necessarily American or exposed to American Pb, since one of these individuals was buried with Chinese material culture (King et al., 2023), just that there is evidence of other instances of possible American Pb ore exposure during the 19th century. Evidence of American Pb exposure in Newfoundland exposure was slightly earlier than these examples.

When the American ore data are considered with individuals from Newfoundland with similar isotope ratios, the timing of the introduction of American ore use in ceramic glazes (or other Pb-containing cultural materials) can be inferred. The date associated with the excavated section of St. Paul's Anglican Church cemetery was between 1764 and 1820 and burial F7 was a woman between 20 and 30 years of age (Pike, 2013). So, their exposure would suggest the introduction of the Pb ore sometime between 1730 and 1800. The Tors Cove individual was at least 20 years old when they died, and the burial ground is believed to be associated with the 19th century, so any exposure to American Pb would have to be post-1800.

4.5.5.1 Early-18th-century New England: Rochefort Point adolescents

The Fortress of Louisbourg was occupied by New England provincial forces between 1745 and 1748. Eight adolescents (16–25 y.o. at the time of their death; pre-1748) excavated from the Rochefort cemetery, were buried in the section identified as associated with the New England occupation. This was based on historical and burial contexts and diets consistent with consumption of significant amounts of C_4 foods both

recently and during earlier childhood (Scott et al., 2023). Since they were all adolescents and the New England occupation of Louisbourg ended in 1748, we can assume their births were between 1723 and 1729.

The Pb concentrations within tooth enamel were between 0.6 and 5.0 ppm and except for one individual (7/2017) were above the concentration of 0.8 ppm that is, at least in Britain, associated with contributions from anthropogenic exposure (Millard et al., 2014). The values fall within those expected from the French (0.2–28.3 ppm, n = 19) and English/Irish (0.4–28.7 ppm, n = 23) fishery populations, as well as other medieval and early modern sites (see Chapter 3). However, they do have a lower mean (2.4 ppm) than the French (7.7 ppm) and English/Irish (6.8 ppm), suggesting that these New Englanders buried in Rochefort Point were living within Pb environment during childhood that did not result in quite as high of exposure as populations associated with the fishery.

Of the Rochefort Point adolescents expected to have been born in the USA (Scott et al., 2023), none had shifted Pb isotope values towards known American ore (Figure 13Figure 12; Figure 21); instead, they are shifted towards English/Welsh or other Western European sources. There are some American ore mines with Pb isotope values that overlap with England, but these regions' mines were not active early enough to contribute Pb to the glazes (Appendix Table 19). This suggested that American Pb ore may not have been used in New England within ceramic glazes pre-1730. American potteries were active by this period, and American made ceramics are found on contemporaneous sites (Pendery, 1985; Deetz, 1996). A limiting point to this conclusion is that there are no Pb ore isotopic data from Massachusetts, from where most of the colonial forces originated (Wall, 1964; Rawlyk, 1999). While mining in the area was

intermittent, there was at least one active Pb-silver mine in Loudville, Massachusetts that was active during the 1680s–90s (Marshall and Dunn, 1976). So, until Pb ore isotope values for New England are available, the use of American ore cannot be completely discounted. Since many of the Pb ore fields were not yet in use or had more radiogenic isotopic values than the Rochefort Point individuals, none of the American Pb sources that have been isotopically characterized could account for the values seen in the human teeth (see Table 19).

Assuming the interpretation of the individuals' origins is correct, then materials made using American Pb were available in Newfoundland by at least the mid-18th to early-19th centuries, but not available in New England in the 1730s. Or at least, not available in a sufficient amount to shift the Pb isotope values away from English/Welsh Pb. This has important inferences for the applicability of Pb isotope ratios in estimating origins and identifying non-locals, not just in Newfoundland but more broadly in the Atlantic World.

4.5.6 Cultural defocusing

The interpretation of origins using Pb isotope values in anthropogenic contexts relies on the foundation of cultural focusing. This is Montgomery et al.'s (2005) argument that within a cultural context, individuals will have access to similar resources and, in turn, have similar Pb isotope values. This does clearly hold in many contexts, but it may not hold true on the western side of the Atlantic World.

In considering the Pb concentration and $^{206}Pb/^{204}Pb$ ratios (Figure 22), it shows that there is a continuation of cultural focusing maintained through export of British (or French) made materials into colonial contexts, followed by a defocusing. Late medieval .

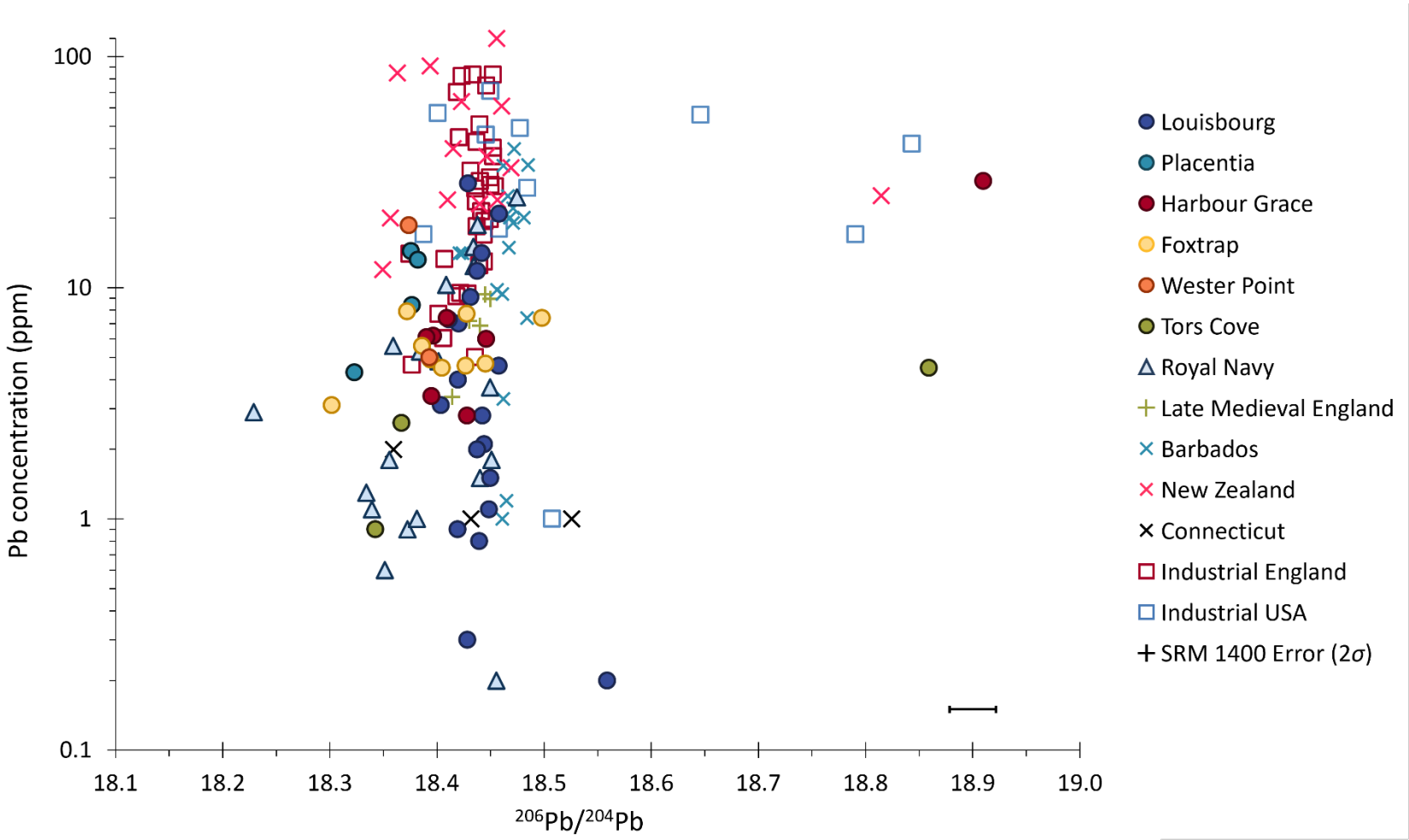

Figure 22. Cultural 'de'focusing depicted through the Pb concentration (ppm) and $^{206}Pb/^{204}Pb$ values of individuals from the French and English/Irish fisheries compared to other contemporary populations in the Atlantic World. Full comparative citations are available in Table 17.

Atlantic Archipelago contexts have a very tight cluster of Pb isotope values with higher concentrations, but very low concentrations with no or little anthropogenic Pb exposure. Within 18th and 19th century urban English contexts there are much higher concentrations with still tightly Pb isotope ratios. This is typical of the cultural focusing and is maintained when we look at 18th century New Englanders buried at Rochefort Point, Louisbourg (this study). This imported Pb also had effects on non-European descent populations including the non-African born 17th to 19th century enslaved labourers in Barbados (Laffoon et al., 2020). It is also suggested in the bone Pb isotope ratio values of individuals engaged with fur trading during the 19th century in Alberta, Canada, including possible Indigenous individuals (Carlson, 1996). Interestingly, there seems to be maintained cultural focusing in French contexts as well. Even though Block 3 were from various locations across France and French colonial territories, they had very similar Pb isotope ratios and concentrations.

However, there is also a defocusing that occurs. As seen in Chapter 3, it arguably is beginning to defocus in the English fisheries during the 18th and 19th centuries. This may be due to increased Pb mining in the 18th to 19th centuries, and or because there were more varied and rural origins of individuals compared to urban London and Coventry (Millard et al., 2014). Yet there are outliers with higher Pb isotope ratios consistent with American Pb, not only in mining contexts like Illinois (Fitch et al., 2012), but a mental hospital in Colorado (Keller et al., 2016), and possibly as far as New Zealand (King et al., 2020).

For identification of migrants there needs to be an assumption of difference, but since cultural focusing is maintained through the export of materials, then is defocused because

of the addition of more sources. The transition into the global market with the export of additional new sources, means that we cannot be certain if outlying values are from migration, or different practices of material culture use. And we hypothesize that since individual materials can contribute to acute toxicity, a single individual or family could have different values depending on the materials available to them. For example, it is possible that specific materials were imported for a family or that they acquired materials at a different time from others. It needs to be considered that families may have been taking culturally different actions; therefore, Pb isotopes alone cannot suggest outliers in origin.

4.6 Conclusion

Since ceramic glazes as a source of Pb exposure are applicable to other regions, this is a material that must be considered moving forward in interpreting mobility studies using Pb. This study has demonstrated that while Pb used for ceramic glazes could use locally acquired materials, Pb may have also been imported, sometimes from entirely different countries. Ceramics available to these Atlantic fishery settlements were not exclusive to the country or port of origin of the people themselves. There were multiple ceramics available to settlements, not just in the French and English/Irish fisheries, but in the American colonies/states. Western Atlantic colonies developed local potteries at different times, but the Pb used to make glazes has not been specifically studied. In summary, Pb-glazed ceramics were a source of Pb exposure throughout much of the Atlantic World and colonies all imported ceramics to varying extents.

Two major implications come out of these conclusions. First, the widespread imports of Pb used to produce ceramic glaze and the ceramics themselves during the 17th–19th centuries means that it remains unclear what should be used to define a baseline of the expected values within a population. Second, if the trade was so widespread, can individuals who fall outside of the majority of those within the population be identified as non-locals? Alternatively, is it that they used materials with different sources of Pb or used them more frequently than others in the settlement?

There is no simple answer on what should be used as a baseline; it will ultimately depend on the questions asked and the temporal context. However, researchers need to better understand the data limitations and the limits of our understanding of trade and production patterns. From this study it is clear that the modern ore does not fully represent the potential sources materials could be made from, but also is in part muddied by sources not contemporarily available. However, the ceramics also underrepresent the potential Pb isotope values that could result in exposure – though more comprehensive studies may aid in this. The best advice is to contextualize it within the material culture and local environmental.

The question then arises of what does cultural focusing and Pb isotope ratios look like in an Atlantic World? Cultural focusing assumes that everyone within a culture would have access to similar Pb sources and, therefore, would have similarly averaged Pb isotope values in the skeletal tissues. As a whole, it is likely that most will have similarly averaged values. But outliers in this context would be identified as non-locals. However, this study has shown that variable sources were available to populations, so such an interpretation ignores agency within the choice or differences in availability of materials.

Fundamentally, variable or outlying Pb isotope values in human teeth suggest that these individuals had access to different sources – not that they were non-local. Given what we know of the Atlantic World, ceramic production, and ceramic trade, outlying values in anthropogenic exposure contexts should be interpreted as one piece of evidence that requires further support by additional isotopic, historical, and archaeological data.

While this study is within an Atlantic World context, it has clear implications for other regions and periods. For example, looking at the origins of individuals within mining or smelting areas – where values may not indicate a local Pb source, such as seen in Sweden (Price et al., 2017) or industrial America (Fitch et al., 2012; Keller et al., 2016), or European colonies further abroad as seen in New Zealand (King et al., 2020).

Future Pb studies must integrate multiple lines of evidence from available archaeological, architectural history, and historical sources to understand the potential objects they were using that would result in Pb exposure. From there, we need to better study the materials themselves to address origins, particularly in the American contexts where we are unclear of when American Pb was used – and how widely it was exported throughout American colonies for use in the ceramics.

5 General Discussion and Conclusions

Previous Pb studies have shown the ability of Pb concentration and isotope ratios to address migration and cultural affiliation issues while adding implications for a population's health, both in anthropogenic and natural environmental contexts (e.g., Montgomery et al. 2010; Millard et al. 2014; Valentine et al. 2015; Evans et al., 2018a; Samuelson and Potra, 2020; Moore et al. 2021). A review of previously published Pb studies on archaeological teeth found that there are clear limitations to this approach, some of which are inherently part of the method's application while others involve the logic used to interpret and design Pb studies (Chapter 3). Many studies in anthropogenic contexts highlight the importance of inhalation and ingestion of environmental pollutants in terms of how humans were exposed to Pb in the past. This, in part, is based on modern studies that are impacted by the pervasiveness of Pb pollution from leaded gasoline. Additionally, many countries have regulations that limit Pb-content of everyday objects, such as paint and ceramics. Therefore, the justification of such logic applied onto past populations is not necessarily complete.

In turn, this dissertation sought to explore what happens in an environment free from anthropogenic environmental exposure. Fishery populations in the North Atlantic are an ideal context to study this because they were mainly focused on the fishing industry, so mining or other environmental contaminants were not expected. Sampling teeth representing childhood exposure in French and English fishing contexts showed that exposure levels were variable, though similar to other non-industrial anthropogenic

contexts. This is seen in the remains from the St. John's Royal Naval Hospital at Southside Cemetery, where the origins of individuals were variable, with childhood exposures likely occurring from across England. The similarity of values in individuals from French communities suggests this was also true in France.

Comparing the Pb isotope values in the tooth enamel suggests that the geological source of exposure was also similar to contemporaneous groups and consistent broadly with Western European ore. Since food and drink contaminated by Pb from glazed ceramic wares are a likely source of Pb for individuals associated with the fisheries, and exposure levels were similarly high to contemporaneous groups, the potential role of ceramic-based exposures should be considered in other 17th–19th-century settlements in addition to other locally produced or imported Pb-containing cultural materials. However, this is not often the case.

Directly sampling ceramics from Ferryland, NL and Anse à Bertrand, Saint-Pierre, allowed for the examination of the Pb values of materials available to these fishing populations. While other Pb-containing cultural materials were also available to these populations, the contribution of Pb from Pb-glazed ceramics would have been incorporated into the tooth enamel of those children interacting with such objects. The Pb isotope values of the ceramic glazes sampled in this study, do cluster within the range of isotopes for English/Welsh and Western European ore, though there is some variability. Such a comparison of ceramic and ore Pb isotope values highlights that variable sources were used within countries, and potteries did not always source Pb locally, nor were such sources always available. In turn, when considering the geological sources available to people within a community, it is insufficient to consider only the origins of the objects

themselves without also considering the materials used to create them, such as the Pb in the glaze.

Pb isotope values of the ceramics studied did not encompass the variability of materials available in the English or French fishing populations, as seen by the shifted Pb isotope values of French fishery associated humans buried at Block 3, Louisbourg, that are not accounted for by any ceramics studied. However, outliers within human Pb isotope data from Harbour Grace and Tors Cove suggest that American Pb may not be exclusively seen in American populations in the late-18th to early-19th centuries. This contrasts the Pb isotope values consistent with Western European Pb in those adolescents from Rochefort Point born in New England in the early 1700s. Such a contrast suggests that early 18th-century Americans were exposed to Pb from imported materials and that American Pb was unavailable locally until the later-18th or early-19th centuries. Together, this has major implications for the ability of researchers to address origins using Pb isotope values during this period and likely further into the 19th century globally.

Pb isotope analyses address migration in anthropogenic contexts under the premise that expected values are 'culturally focused,' since similar sources will be used within a society. However, the Atlantic and global trade networks that existed in the 18th and 19th centuries, bring into question how universal this may be. Families' agency within the market and or familial connections to areas with different geological sourced Pb-containing materials suggests that, while most individuals may hold true to the concept of 'cultural focusing,' not all do. An individual with outlying Pb isotope values, if not directly related to environmental exposure, only means that they used materials from a

different place, not that they necessarily were from there. This then undermines researchers' ability to use the single Pb isotope system to address migration questions.

5.1 Limitations and Challenges

5.1.1 Methodological details available in published studies

Data quality and reproducibility are vital aspects of the scientific method and even more ethically important when dealing with destructive analyses of human remains. The literature review (Chapter 2) highlighted the necessity for more data reproducibility since there needed to be more methodological information regarding sample tooth choice, enamel or dentine mechanical separation and cleaning, chemical pre-treatments, and the analyses themselves. This, in turn, calls into question the comparability of data, even when produced using the same analytical instruments. Studies must give sufficient detail in their methodologies to allow interpretation of the data. Recent trends in publishing to improve the availability of supplemental information creates the space for authors to include details that may have been neglected in some past studies.

5.1.2 Methodological consistency in this study

Ideally, concentrations of Pb would have been determined using Q-ICPMS in all cases. However, due to unforeseen problems with the analytical equipment available at Memorial that was prolonged by Covid-19-related shutdowns, some concentration data was estimated using the raw data from the MC-ICP-MS. This introduced more error — closer to 30% rather than less than 10% in Q-ICPMS. This error was documented by comparing the NIST SRM 1400 standard values on the analytical runs to certified values

and to tooth samples with both Q-ICPMS and MC-ICP-MS data. This increased error is unsurprising since multiple steps contribute to the determination of Pb concentration using the MC-ICP-MS data. These contributing errors include samples being analyzed using post-column solutions where collection efficiency is not 100%, high concentration solutions that exceeded the MC-ICP-MS faraday detectors (e.g., >50V on ^{208}Pb) necessitated dilution using was volumetric and not gravimetric methods since they were done as needed during analysis sessions. Finally, the Pb concentration calculation was done using bracketing with the NIST SRM 981 solution of a single concentration (100 ppb), and not through a linear equation involving three or more Pb concentrations which would better encompass the variability of the Pb isotope voltages experienced.

In one case, a SRM 1400 standard was not included with the samples and blank that were sent to the University of Ottawa for concentration analyses. While the University of Ottawa used their own standards for calibration, using a standard that could allow for inter-laboratory comparison would have been ideal.

While calculations were performed to estimate the necessary tooth enamel sample size prior to cutting a tooth (see Appendix 7.2.4.1), low Pb concentration and available enamel material resulted in a few samples having insufficient or borderline mass required for both concentration and isotope ratio determination. For Pb isotope analyses, efforts were made to bracket these particular samples with lower concentrations of SRM 981 to lower the target intensity, which did help, but was not always sufficient (see Appendix 7.2.4.3). This is particularly true for concentrations below 0.5 ppm and may mean that Pb isotope values that are shifted compared to other samples are impacted by blank contributions or error within the calibration calculation. However, a conscious effort at

the transparency of data limitations was made by clearly marking those impacted with the information given on blank contribution and the analysis voltage. Additionally, in such cases, differences in values were not over-interpreted. This will allow fellow researchers to assess the data quality themselves.

Contamination from the burial environment and laboratory instruments and procedures is always of concern. There are two points to this: avoidance of potential Pb contamination sources and identification of these sources. It was not possible to identify potential contamination during the initial sampling procedure. However, efforts were made to clean all surfaces with a tungsten carbide bit, and samples were immediately transferred to the clean lab after initial cutting and cleaning. While double-distilled acids were initially used for sampling, this became too time-restrictive, and preparation of SRM 1400 standards and blanks suggested that single-distilled acids were sufficiently clean. All instances of higher blank contribution were identified. This approach to sample preparation must be modified if applied to samples associated with non-anthropogenic or low Pb exposure contexts.

When working with previously buried materials, there is always the potential for diagenetic alteration. Efforts were made to avoid potential diagenetic material by sampling only fully mineralized teeth, avoiding cracks, caries, and dentine, and removing the cemento-enamel junction. When available, heavy metal concentrations and rare earth elements were compared to the maximum threshold concentrations (MTC) defined by Kamenov et al. (2018). Not all suggested elements were available within the concentration suite run by Q-ICPMS, which may affect the ability to identify when diagenesis occurred. Within the samples, some elements (V, Mn, Ce) were identified as

being higher than the MTC. However, it is unclear how applicable some elements are since the MTCs are based on modern contexts, which may not have naturally as high contamination depending on their living environments. While this may suggest the potential for diagenetic alteration, none of the altered values occurred in individuals with outlying Pb isotope values. Therefore, even if there was contamination, it was unlikely high enough to contaminate the high Pb concentrations. Since not all concentration samples were analyzed using Q-ICPMS, some samples were not available for this diagenetic check.

5.1.3 Archaeological and cultural contexts

Part of the analysis in this research depended on determining the cultural connections of the sampled individuals. Historical and archaeological contexts can help differentiate these in most cases, but the contexts of the individuals from St. Luke's Anglican Church cemetery in Placentia are less clear.

While it would have been ideal to incorporate additional isotope systems to address migration questions for more individuals directly, this was beyond the scope of this dissertation. The dietary ($\delta^{13}C$ and $\delta^{15}N$) and geographic origin data ($^{87}Sr/^{86}Sr$) on many of the individuals were part of an MA thesis (Garlie, 2022). Full integration of osteological and isotopic data for these populations will give a better understanding of the individuals' life histories, and particularly for the data with the Pb isotope values, particularly for Block 3, would help address variability of Pb exposure in populations buried in North American contexts.

5.1.4 Ceramic analyses

Ceramics were used as a means to better understand the Pb sources available to individuals within the French and English/Irish fisheries. Pb exposure from material culture was not only from glazed ceramics; however, other Pb-containing materials are either consumable (e.g., contaminated rum, Pb acetate for medication, etc.) or were melted and re-used (e.g., pewter) and are therefore not often found. Ceramics were chosen because they are so ubiquitous, but this also has the potential of underestimating the range of bioavailable Pb within the population.

While attempts were made to choose ceramic wares typical of those available to the French and English fishing populations, the availability was limited by a few factors. First, the laser ablation cell is 55 mm in diameter and 8 mm in height, so larger sherds would not fit. Additionally, laser ablation relies on a flat surface for ablation, so highly curved surfaces were not appropriate. Finally, this was all limited by what sherds were stored at Memorial University in January 2021. At this time, there remained limitations on the number of individuals present within a room and travel to the central repository for the archaeological collection at Ferryland was not possible. Therefore, materials were chosen from the historical archaeology teaching collection, made up of materials from Ferryland. The materials from Anse à Bertrand were only those that remained at the Historical Archaeology Laboratory for cataloguing from the 2019 excavations. This means that the available ceramics may not fully represent those materials available to the population.

Since this was a pilot study to examine the ceramic glaze, samples addressing change over time were not possible. Ideally, multiple samples from each ceramic ware representing different periods of ceramic production would have been sampled. Additionally, American-made ceramics were chosen for analysis but either did not fit within the laser cell or were not Pb-glazed.

In addressing what materials were likely available to these fishing populations, available ceramic analyses may not fully represent all of the studied fishing communities. While an extensive study by Losier et al. (2018) addressed what materials were available in the French fishing communities of Newfoundland and Saint-Pierre, a similar study was not available for English material. An informal review of theses available and papers addressed the topic of ceramic origin use by region (see Appendix 7.3.2), but there remain areas with insufficient excavations to know how representative existing studies are of English fishery settlements. These locations with limited excavations from domestic contexts include Harbour Grace, Tors Cove, and Portugal Cove.

There are two main limitations with the comparative analyses for people involved with the fisheries and their ceramics: the offset between period represented by most ceramics and people, and the applicability of available Pb ore isotope ratios. The majority of the ceramics from Ferryland used to represent potential Pb available to the English/Irish fisheries were produced in the early 17th to mid 18th centuries, but many of the individuals sampled were buried in the later 18th to 19th centuries. While most of the individuals fell within the potential bioavailable range, it must be considered that there could have been changes in the Pb available to potteries before export of their Pb-glazed ceramics. This is further supported by the improvement in transportation abilities over the

course of the 18th century through better road networks and canals (Bogard, 2007). Also, there were likely changes in how these ceramics were used, with more Pb-glazed for cooking and storing in the 17th century versus serving in the 18th to 19th centuries. Since heat and time of exposure are likely to increase the amount of Pb leeched, this is an additional factor to consider in the vectors of exposure.

The second limitation in the comparison of ceramics and people is that the ore with available Pb isotope data are not necessarily representative of the Pb used within materials available to these populations. There are multiple factors contributing to this limitation of relevant Pb isotope comparative data. First, it must be known what ore fields were active at the time, or what ore fields the Pb in recirculation would be coming from. While there is an interest in England's and America's industrial mining history (e.g., Burt, 1984; and citations within Appendix Table 19), no similar studies are available for France on a regional level. Therefore, it is unclear what mines were active during the 17th to 19th centuries. In turn, generalizations about deposits in the country must be made from available Pb isotope data. This leads to an additional limitation, that Pb ore isotope data are often acquired as part of prospecting rather than examining mines that may have been active during the period in question (see references within Table 45). Therefore, there is ambiguity as to whether these deposits would be similar to those materials possibly available during the period of study, particularly when it is not clear what areas were active.

Meaningful groupings of the geological deposits within Britain and France were approximated using locations from geological maps and historical mining regions. In England and Wales, this started with known mining areas (Burt, 1969; Burt, 1984) that

were further consolidated using longitude and latitude of ore samples overlayed with bedrock geology maps and Evans et al. (2022) bioavailable Pb map. For France, a similar bedrock geology approach was taken, but less information on historical mining regions was available. In general, geologically based interpretations are preferred rather than by region (Killick et al., 2020). In both cases there the number of ore groupings was problematic for data visualization. The summarized outlines of data were used for presentation not interpretation, but this loses any sense of data density. Since not all groups had sufficient data to characterize a deposit, this may also not fully represent the materials available within a region.

5.2 Future Work

The studies within this dissertation have opened or highlighted multiple areas of future research. One of the main conclusions from the fisheries context is a questioning of the applicability of cultural focusing within the Atlantic World. There are two parts to this question; first is the timing of the introduction of American Pb within American-made ceramics, then exploring whether non-locally expected Pb values occur within individuals who were born locally.

For Pb isotope ratios to be used in American colonies, even in the 19th-century industrial areas, the geological source of Pb-containing objects must be better understood. As seen in the ceramic Pb isotope values (Chapter 4), production in one region does not mean locally mined ore would be used in the glaze. Ceramic production in American colonies was active by the 17th century, and some small-scale Pb mines were also present, but it needs to be clarified that the Pb would be used in the manufacture of ceramics and

glaze. The source of Pb likely differed between potteries and over time. England did export large amounts of Pb to America, but this likely varied during times of war, as would production and need for Pb in other sectors. So, examining American ceramics available within a region and potentially over time would help address this question.

Additionally, this dissertation has also expressed concern about how representative Pb isotope values from ore bodies are in Europe and North America. Areas with mines during the 17th to early-18th centuries are known, such as within the massif of Vosges in the French Alps (Forel et al., 2010) and the Loudville Pb mines in Massachusetts, USA (Marshall and Dunn, 1976). However, the 12 ore samples from the massif of Vosges (Blichert-Toft et al., 2016) are likely insufficient to characterize the deposit area (Stos-Gale and Gale, 2009), and no data are available for mines northeast of New York in the United States. Future research would benefit from an interdisciplinary study of the history of Pb mining within regions of France and America and how this is associated with pottery production. Additionally, further ore sampling from areas of interest would help to fill in holes in our understanding of bioavailable Pb isotope values.

Related to this lack of ore data is the need for further Pb concentration and isotope values of individuals within American contexts during other periods to explore the variability of expected values within these populations and how this possibly changed over time. Further data are being processed from Louisbourg, though these are associated with the same period as the adolescents from Rochefort Point (Chapter 4.5.5.1), so given the time period represented, they were unlikely exposed to significant American Pb, or at least Pb ore with distinct values from Western Europe and Britain. In association with Dr. Varney, I am processing Pb concentration and isotope ratio data for populations in

Caribbean and Illinois during the 18th 19th centuries, which may help to illuminate the variability. Values from different regions would be beneficial to address this question of how much variability in Pb isotope values exists within historic populations.

Finally, Pb isotope analyses are commonly applied to identify first generation immigrants to a region, including the United States (i.e., Fitch et al. 2012; Aronsen et al., 2019; Quinn et al. 2020). Since this dissertation has questioned the applicability of Pb isotope ratios within the Atlantic World, it leaves the issue of what isotope systems may help in identifying migrants unclear. Unfortunately, there is an overlap in expected $\delta^{18}O$ and $^{87}Sr/^{86}Sr$ values between England and New England, so it is difficult to differentiate between first-generation immigrants to New England and those that were born in the colonies that (Bowen and Wilkinson, 2002; Bowen et al., 2007; Bataille and Bowen, 2012; Bataille et al., 2018). While there is an understanding that, at least in southern American colonies during the 17th–18th centuries, there was significant C_4 foods consumed, as seen by France et al. (2014). Additionally, pork from New York during the 19th century had C_4 values (Guiry et al., 2017). However, data from Louisbourg has seen variability in the diets of individuals believed to be from New England from C_3 to mixed-C_3/C_4 diets (Ellerbrok, 2014; Scott et al., 2023). The question arises of the variability of C_3 versus C_4 foods available to the individuals within New England during the 17th and 18th centuries. If animals in the early 1700s were fed C_4 foods (likely maize), then immigrants would be expected to be distinguishable from those born locally. A faunal baseline within urban and rural contexts of Massachusetts and New England more broadly should be established for the 17th to 18th centuries. This would act as another tool

to compare with Pb isotope values to address migration and whether Pb isotope values can be attributed to the movement of people rather than objects during this period.

5.3 Concluding Thoughts

This research addressed several key objectives related to improving our understanding of historical Pb exposures during the early European settlement of Northeastern North America. The first objective was to review recent studies of Pb concentration and isotope ratios in teeth to identify the challenges, limitations, and assumptions associated with understanding historical Pb exposures. The review highlighted a major limitation of the foundational assumption in populations outside of Britain that the environment was the primary source of Pb exposure and identified deficiencies in reporting of sample preparation and analysis that limit the ability to compare studies.

The second objective was to evaluate if historical Pb exposure was high enough in North American fishery communities to result in tooth enamel Pb concentrations higher than would be expected in the absence of local Pb environmental pollution. Childhood Pb exposure during the late-17th to mid-19th centuries were similar to medieval exposure levels in England and contemporary colonial contexts, despite the absence of local anthropogenic Pb environmental sources. This suggests that there are unidentified sources of Pb contamination that are not often considered in North American fisheries, and likely other colonial and European settlement contexts.

The third objective was to identify and compare potential geological sources of Pb between fishing communities and other early settlement populations in Northeastern

North America. Sources of Pb based on isotope ratios are broadly consistent with European sources, which are similar to contemporaneous English/Welsh contexts. However, there are outliers, and this variability is also seen in other recent studies on 19th-century populations, highlighting the need to consider what the potential sources of Pb exposures were, especially given the absence of local identifiable environmental exposures in 17th- to 19th- fishing communities.

The fourth objective evaluated whether the Pb levels in contemporary ceramics available to French and English fishing populations in Northeastern North America were sufficiently high to result in leaching that could contribute to anthropogenic exposure. Pb in the glaze contributed to a third to half of the glaze composition, similar to other studies of contemporary materials. The resulting leaching that would have occurred during cooking, storage, or serving with these vessels would have resulted in at least some anthropogenic Pb exposure.

The final objective compared the Pb isotope ratios in ceramics to tooth enamel associated with childhood exposure, as well as to values of European and American ore bodies. All the ceramic Pb isotope ratios from the ceramic glazes in this study were consistent with available Pb ores from Western Europe. In situations where the origin of the ceramic was known, evidence for modern local ore deposits was not necessarily consistent with that used in the glaze, suggesting that potteries occasionally imported Pb or the glazes from outside the region or country to areas with Pb.

Trade throughout the Atlantic World, and beyond, allowed individuals within communities access to various Pb-containing materials from a wide range of origins. This can be challenging for data interpretation given that the Pb within the ceramic glaze is not

always correlated with the location where the ceramic was produced, and while not studied directly here, was possibly the case with other Pb-containing materials available to these communities. Additionally, personal connections would influence the availability of ceramics to individual families. This is particularly poignant with materials using American Pb sources.

Cultural focusing assumes some similarities in materials and exposures within a community, but agency allows individuals access to other materials. Identifying outlying values within a population is usually interpreted as representing a recent first-generation immigrant, but outlying values such as these may otherwise indicate that the material sources of exposure have been transported. The availability of materials that include different isotope ratios of Pb due to different geological sources, such as ceramics glazed with American or Welsh Pb, could also contribute to uncertainty. Additional studies examining Pb isotopes of ores from mines active in France and USA during the medieval to mid-19th centuries and on the timing of American Pb use in ceramics are needed before we can reliably identify immigrants using outlying Pb isotope ratios.

6 REFERENCES

Åberg, G., Fosse, G., & Stray, H. (1998). Man, nutrition, and mobility: A comparison of teeth and bone from the Medieval era and the present from Pb and Sr isotopes. *Science of the Total Environment*, *224*, 109-119. https://doi.org/10.1016/s0048-9697(98)00347-7

Adriano, D.C. (1986). *Trace Elements in the Terrestrial Environment*. Springer-Verlag.

Agarwal, S.C., & Glencross, B.A. (Eds.). (2011). *Social Bioarchaeology*. Wiley-Blackwell.

Agency for Toxic Substances and Disease Registry (ATSDR) (2020). *Toxicological profile for lead*. ATSDR, Division of Toxicology and Human Health Sciences. https://www.atsdr.cdc.gov/ToxProfiles/tp13.pdf

Al-Ashban, R.M., Aslam, M., & Shaw, A.H. (2004). Kohl (surma): a toxic traditional eye cosmetic study in Saudi Arabia. *Public Health, 118*, 292-298. https://doi.org/10.1016/j.puhe.2003.05.001

Alfonso-Durruty, M.P., Thompson, J.L., & Crandall, J.J. (2014). Little Bodies, Big Voices: The Lives of Children in the Past. In, J.L. Thompson, J.J. Crandall, & M.P. Alfonso-Duruty (Eds.), *Tracing Childhood: Bioarchaeological Investigations of Early Lives in Antiquity* (pp. 248-257). University Press of Florida.

Al-Mosawi, M., G.R., Davis, A., Bushby, J., Montgomery, J., Beaumont, & Al-Jawad, M. (2018). Crystallographic texture and mineral concentration quantification of developing and mature human incisal enamel. *Scientific Reports*, *8*, e14449. https://doi.org/10.1038/s41598-018-32425-y

AlQahtani, S.J., Hector, M.P., & Liversidge, H.M. (2010). Brief communication: The London atlas of human tooth development and eruption. *American Journal of Physical Anthropology*, *142*, 481-490. https://doi.org/10.1002/ajpa.21258

Aronsen, G.P., Fehren-Schmitz, L., Krigbaum, J., Kamenov, G.D., Conlogue, G.J., Warinner, C., Ozga, A.T., Sankaranarayanan, K., Griego, A., DeLuca, D.W., Eckels, H.T., Byczkiewicz, R.K., Grgurich, T., Pelletier, N.A., Brownlee, S.A., Marichal, A., Williamson, K., Tonoike, Y., & Bellantoni, N.F. (2019). “The dead shall be raised”:

Multidisciplinary analysis of human skeletons reveals complexity in 19th century immigrant socioeconomic history and identity in New Haven, Connecticut. *PLOS One*, *14*, e0219279. https://doi.org/10.1371/journal.pone.0219279

Aufderheide, A.C., Angel, J.L., Kelley, J.O., Outlaw, A.C., Outlaw, M.A., Rapp, G., & Wittmers, L.E. (1985). Lead in bone III. Prediction of social correlates from skeletal lead content in four colonial American populations (Catoctin Furnace, College Landing, Governor's Land, and Irene Mound). *American Journal of Physical Anthropology*, *66*, 353-361. https://doi.org/10.1002/ajpa.1330660402

Aufderheide, A.C., Wittmers, L.E., Rapp, G., & Wallren, J. (1988). Anthropological applications of skeletal lead analysis. *American Anthropologist*, *90*, 931-936. https://doi.org/10.1525/aa.1988.90.4.02a00110

Austin, R.S., Giusca, C.L., Macaulay, G., Moazzez, R., & Bartlett, D.W. (2016). Confocal laser scanning microscopy and area-scale analysis used to quantify enamel surface textural changes from citric acid demineralization and salivary remineralization in vitro. *Dental Materials*, *32*, 278-284. https://doi.org/10.1016/j.dental.2015.11.016

Bacon, J.R., & Bain, D.C. (1995). Characterization of environmental water samples using strontium and lead stable-isotope compositions. *Environmental Geochemistry and Health*, *17*, 39-49. https://doi.org/10.1007/BF00188631

Baldwin, C.K. (1993). *Great and Noble Jar: Traditional Stoneware of South Carolina*. University of George Press.

Barker, D., & Crompton, S. (2007). Slipware in the Collection of the Potteries Museum and Art Gallery. A&C Black.

Barton, K.J. (1963), The Medieval pottery of the Saintonge. *Archaeological Journal*, *120*(1), 201-214. https://doi.org/10.1080/00665983.1963.10854240

Bataille, C.P., & Bowen, G.B. (2012). Mapping $^{87}Sr/^{86}Sr$ variations in bedrock and water for large scale provenance studies. *Chemical Geology*, *304-305*, 39–52. https://doi.org/10.1016/j.chemgeo.2012.01.028

Bataille, C.P., von Holstein, I.C.C., Laffoon, J.E., Willmes, M., Liu, X-M., & Davies, G.R. (2018). A bioavailable strontium isoscape for Western Europe: A machine

learning approach. *PLoS ONE*, *13*(5), e0197386. https://doi.org/10.1371/journal.pone.0197386

Bataille, C.P., Crowley, B.E., Wooler, M.J., & Bowen, G.J. (2020). Advances in global bioavailable strontium isoscapes. *Palaeogeography, Palaeoclimatology, Palaeoecology*, *555*, e109849. https://doi.org/10.1016/j.palaeo.2020.109849

Baxter, J.E. (2005). *The Archaeology of Childhood: Children, Gender, and Material*. AltaMira Press.

Baxter, M.J., & Gale, N.H. (1998). Testing for multivariate normality via univariate tests: A case study using lead isotope ratio data. *Journal of Applied Statistics*, *25*, 671–683. https://doi.org/10.1080/02664769822891

Baxter, M.J., Beardah, C.C., & Westwood, S. (2000) Sample size and related issues in the analysis of lead isotope data. *Journal of Archaeological Science*, *27*(10), 973-980. https://doi.org/10.1006/jasc.1999.0546

Bayley, W.S. (1910). *Iron Mines and mining in New Jersey*. MacCrellish & Quigley.

Beagrie, N. (1989). The Romano-British pewter industry. *Britannia*, *20*, 213-244. https://doi.org/10.2307/526161

Beaumont, J., Montgomery, J., Buckberry, J., & Mandy, J. (2015) Infant mortality and isotopic complexity: New approaches to stress, maternal health, and weaning. *American Journal of Physical Anthropology*, 157(3), 441-457. https://doi.org/10.1002/ajpa.22736

Beherec, M.A., Levy, T.E., Tirosh, O., Najjar, M., Knabb, K.A., & Erel, Y. (2016). Iron Age nomads and their relation to copper smelting in Fayan (Jordan): trace metal and Pb and Sr isotopic measurements from the Wadi Fidan 40 cemetery. *Journal of Archaeological Science*, *65,* 70-83. https://doi.org/10.1016/j.jas.2015.10.006

Bellis, D.J., Parsons, P.J., Jones, J., & Amarasiriwardena, D. (2009). Evaluation of Laser Ablation Inductively Coupled Plasma Mass Spectrometry for the Quantitative Determination of Lead in Different Parts of Archeological Human Teeth. *Spectroscopy Letters*, *42*, 491–496. https://doi.org/10.1080/00387010903267807

Bentley, A. (2006). Strontium isotopes from the earth to the archaeological skeleton: a review. *Journal of Archaeological Method and Theory*, *13*, 135-187. https://doi.org/10.1007/s10816-006-9009-x

Berkovitz, B.K.B., Holland, G.R., & Moxham, B.J. (2017). Oral anatomy, histology, and embryology (5th edition). Elsevier.

Bird, B.W., Wilson, J.J., Escobar, J., Kamenov, G.D., Pollard, H.J., & Monaghn, G.W. (2019). Pre-Columbian lead pollution from Native American galena processing and land use in the midcontinental United States. *Geology*, *47*, 1193–1197. https://doi.org/10.1130/G46673.1

Blackström, Y., Mispelaere, J., Ingvarsson, A., Fjellström, M., & Britton, K. (2018). Integrating in isotopes and documentary evidence: dietary concerns in a late medieval and early modern mining community, Sweden. *Archaeological and Anthropological Sciences*, *10*, 2075-2094. https://doi.org/10.1007/s12520-017-0518-1

Blanchette, J.-F. (1981). The role of artifacts in the study of foodways in New France, 1720-1760. Parks Canada, *History and Archaeology*, No. 52.

Blichert-Toft, J., Delile, H., Lee, C.-T., Stos-Gale, Z., Billström, K., Andersen, T., Hannu, H., & Albarède, F. (2016). Large-scale tectonic cycles in Europe revealed by distinct Pb isotope provinces. *Geochemistry, Geophysics, Geosystems*, *17*(10), 3854-3864. https://doi.org/10.1002/2016GC006524

Bogard, D. (2007). Turnpike Trusts and property income: New evidence on the effects of transport improvements and legislation in eighteenth-century England. *The Economic History Review*, *62*(1), 128-152. https://doi.org/10.1111/j.1468-0289.2008.00434.x

Bowen, G.J., & Wilkinson, B. (2002) Spatial distribution of $\delta^{18}O$ in meteoric precipitation. *Geology*, *30*(4), 315–318. https://doi.org/1130/0091-7613(2002)030%3C0315:SDOOIM%3E2.0.CO;2

Bowen, G.J., Ehleringer, J.R., Chesson, L.A., Stange, E., & Cerling, T.E. (2007). Stable isotope ratios of tap water in the contiguous United States. *Water Resources Research*, *43*(3), W03419. https://doi.org/10.1029/2006WR005186

Bower, N.W., Getty, S.R., Smith, C.P., & Hoffman, J.M. (2005). Lead isotope analysis of intra-skeletal variation in a 19th century mental asylum cemetery: Diagenesis versus

migration. *International Journal of Osteoarchaeology*, *15*, 360-370. https://doi.org/10.1002/oa.796

Bower, N.W., McCants, S.A., Custodio, J.N. Ketterer, M.E., Getty, S.R., & Hoffman, J.M. (2007). Human lead exposure in a late 19th century mental asylum population. *Science of the Total Environment*, *372*(2-3), 463-473. https://doi.org/10.1016/j.scitotenv.2006.10.019

Brännvall, M.-L., Bindler, R., Renberg, I., Emteryd, O., Bartnicki, J., & Billström, K. (1999). The medieval metal industry was the cradle of modern large-scale atmospheric Pb pollution in Northern Europe. *Environmental Science & Technology*, *33*, 4391-4395. https://doi.org/10.1021/es990279n

Bright, D.A., Richardson, G.M., & Dodd, M. (2006). Do Current Standards of Practice in Canada Measure What is Relevant to Human Exposure at Contaminated Sites? I: A Discussion of Soil Particle Size and Contaminant Partitioning in Soil. *Human and Ecological Risk Assessment*, *12*(3), 591-605. https://doi.org/10.1080/10807030600561816

Budd, P., Pollard, A.M., Scaife, B., & Thomas, R.G. (1995). The Possible Fractionation of Lead Isotopes in Ancient Metallurgical Processes. *Archaeometry*, *37*, 143–150. https://doi.org/10.1111/j.1475-4754.1995.tb00732.x

Budd, P., Montgomery, J., Cox, A., Krause, P., Barreiro, B., & Thomas, R.G. (1998). The distribution of lead within ancient and modern human teeth: implications for long-term and historical exposure monitoring. *Science of the Total Environment*, *220*, 121-136. https://doi.org/10.1016/s0048-9697(98)00244-7

Budd, P., Montgomery, J., Evans, J., & Barreiro, B. (2000a). Human tooth enamel as a record of the comparative lead exposure of prehistoric and modern people. *Science of The Total Environment*, *263*, 1–10. https://doi.org/10.1016/S0048-9697(00)00604-5

Budd, P., Montgomery, J., Barreiro, B., & Thomas, R.G. (2000b). Differential diagenesis of strontium in archaeological human dental tissues. *Applied Geochemistry*, *15*, 687-694. https://doi.org/10.1016/S0883-2927(99)00069-4

Budd, P., Montgomery, J., Evans, J., & Trickett, M. (2004). Human lead exposure in England from approximately 5500 BP to the 16th Century AD. *Science of the Total Environment*, *318,* 5-58. https://doi.org/10.1016/S0048-9697(03)00357-7

Burt, R. (1969). Lead production in England and Wales, 1700-1770. *The Economic History Review*, *22*(2), 249-268.

Burt, R. (1984). *The British Lead Mining Industry*. Dyllansow Tryran.

Cadigan, S. (1995). *Hope and deception in Conception Bay: Merchant-settler relations in Newfoundland, 1785-1855*. University of Toronto Press.

Candow, J.E. (2001). An overview of the Northwest Atlantic Fisheries, 1502-1904. In L. Francois and A.K. Isaacs (Eds.), *The Sea in European History* (pp. 163-184). Pisa 2001.

Candow, J.E. (2010). *The lookout: A history of Signal Hill*. Breakwater Books Ltd.

Carlson, A.K. (1996). Lead isotope analysis of human bone for addressing cultural affinity: A case study from Rocky Mountain House, Alberta. *Journal of Archaeological Science*, *23*, 557-567. https://doi.org/10.1006/jasc.1996.0052

Carvalho, M.L., Marques, A.F., Marques, J.P., & Casaca, C. (2007). Evaluation of the diffusion of Mn, Fe, Ba and Pb in Middle Ages human teeth by synchrotron microprobe X-ray fluorescence. *Spectrochimica Acta Part B: Atomic Spectroscopy, A Collection of Papers Presented at the 18th International Congress on X-Ray Optics and Microanalysis (ICXOM 2005)*, *62*, 702–706. https://doi.org/10.1016/j.sab.2007.02.011

Cell, G.L. (1969). *English enterprise in Newfoundland, 1577-1660*. University of Toronto Press.

Champagne, M. & Losier, C. (2021). Cod fish and cooking pots: Research on the trade routes of French North Atlantic. *Northeast Historical Archaeology*, *50*, e8. https://orb.binghamton.edu/neha/vol50/iss1/8

Charlier, B.L.A., Ginibre, C., Morgan, D., Nowell, G.M., Pearson, D.G., Davidson, J.P., & Ottley, C.J. (2006). Methods for the microsampling and high-precision analysis of strontium and rubidium isotopes at single crystal scale for petrological and

geochronological applications. *Chemical Geology*, *232*, 114–133. https://doi.org/10.1016/j.chemgeo.2006.02.015

Choudhury, S., Swanston, T.L., Varney, T.L., Cooper, D.M.L., George, G.N., Pickering, I.J., Grimes, V., Bewer, B., & Coulthard, I. (2016) Confocal x-ray fluorescence imaging facilitates high-resolution elemental mapping in fragile archaeological bone. *Archaeometry*, *58*(S1), 207-217. https://doi.org/10.1111/arcm.12232

Choudhury, S., Agyeman-Budu, D.N., Woll, A.R., Swanston, T., Varney, T., Cooper, D.M.L., Hallin, E., George, G.N., Pickering, I.J., & Coulthard, I. (2017) Superior spatial resolution in confocal x-ray techniques using collimating channel array optics: Elemental mapping and speciation in archaeological human bone. *Journal of Analytical Atomic Spectrometry*, *3*, 527-537. https://doi.org/10.1039/C6JA00297H

Christopher, T.K., (1999). Paleolimnology in an urban environment : the history of environmental change in St. John's, Newfoundland. (Publication No. 9445) [Doctoral thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Corruccini, R.S., Aufderheide, A.C., Handler, J.S., & Wittmers, L.E. Jr. (1987). Patterning of skeletal lead content in Barbados slaves. *Archaeometry*, *29*, 233-239. https://doi.org/10.2307/1171025

Crawford, M.D., & Clayton, D.G. (1973). Lead in bones and drinking water in towns with hard and soft water. *British Medical Journal*, *2*, 21-23. https://doi.org/10.1136%2Fbmj.2.5857.21

Crompton, A.J. (2012). The historical archaeology of a French fortification in the colony of Plaisance: The View Fort Site (ChAl-04), Placentia, Newfoundland (Publication No. 9656) [Doctoral dissertation, Memorial University of Newfoundland]. Memorial University Research Repository.

Cui, J., & Wu, X. (2010). An experimental investigation on lead isotopic fractionation during metallurgical processes. *Archaeometry*, *53*(1), 205-214. https://doi.org/10.1111/j.1475-4754.2010.00548.x

Dean, M.C., Le Cabec, A., van Malderen, S.J.M., & Garrevoet, J. (2020). Synchrotron X-ray fluorescence imaging of strontium incorporated into the enamel and dentine of

wild-shot orangutan canine teeth. *Archives of Oral Biology, 199*, e104879. https://doi.org/10.1016/j.archoralbio.2020.104879

Deetz, J. (1996). *In small things forgotten: An archaeology of early American life* (Revised ed.). Anchor.

DeMuynck, D., Cloquet, C., & Vanhaecke, F. (2008). Development of a new method for Pb isotopic analysis of archaeological artefacts using single-collector ICP-dynamic reaction cell-MS. *Journal of Analytical Atomic Spectrometry*, *23*, 62-71. https://doi.org/10.1039/B709461B

Descy, J-P., Floury, M., Moatar, F., & Sauvage, S. (2022). Continental Atlantic Rivers: The Meuse, Loire and Adour-Garonne Basins. In K. Tockner, C. Zarfl, & C.T. Robinson (Eds.), *Rivers of Europe* (225-228, 2nd ed.). Elsevier. https://doi.org/10.1016/B978-0-08-102612-0.00005-5

Directive 84/500/EEC. *Council Directive (EU) 84/500/EEC of 15 October 1984 on the approximation of the laws of the Member States relating to ceramic articles intended to come into contact with foodstuffs*. https://www.legislation.gov.uk/eudr/1984/500/adopted

Doe, R.B. (1970). *Lead Isotopes*. Springer-Verlag.

Douglas, M.A. (2000). An electron microprobe study of 19th century Staffordshire glazes and pigments including those of the Spode pottery worlds. [Masters thesis, University of Georgia, Athens].

Dudás, F.Ö., LeBlanc, S.A., Carter, S.W., & Bowring, S.A. (2016). Pb and Sr concentrations and isotopic compositions in prehistoric North American teeth: A Methodological Study. *Chemical Geology, 429*, 21-32. https://doi.org/10.1016/j.chemgeo.2016.03.003

Duffy, K.I., Carlson, J.H., & Swann, C.P. (2002). A study of green-glazed ware from England and South Carolina, USA (1760–1780). *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, *189*, 369–372. https://doi.org/10.1016/S0168-583X(01)01090-4

Dunn, O.J. (1964). Multiple comparisons using rank sums. *Technometrics, 6*(3), 241-252. https://doi.org/10.2307/1266041

Dupont, W.D., & Plummer, W.D. (1990). Power and sample size calculations: A review and computer program. *Controlled Clinical Trials*, *11*, 116-128. https://doi.org/10.1016/0197-2456(90)90005-m

Dupont, W.D., & Plummer, W.D. (1998). Power and sample size calculations for studies involving linear regression. *Controlled Clinical Trials*, *19*, 589-601. https://doi.org/10.1016/s0197-2456(98)00037-3

Dupont, W.D., & Plummer, W.D. (2018). PS: Power and sample size calculation. Version 3.1.6, October 2018. https://biostat.app.vumc.org/wiki/Main/PowerSampleSize

Elias, R.W., Hirao, Y., & Patterson, C.C. (1982). The circumvention of the natural biopurification of calcium along nutrients pathways by atmospheric inputs of industrial lead. *Geochimica et Cosmochimica Acta*, *46*, 2561-2580. https://doi.org/10.1016/0016-7037(82)90378-7

Ellerbrok, B.A. (2014). *An isotopic reconstruction of diet and origins in an 18th century mass burial site at the Fortress of Louisbourg, Nova Scotia*. Masters thesis Memorial University of Newfoundland.

Ericson, J.E., Shirahata, H., & Patterson, C. (1979). Skeletal concentrations of lead in ancient Peruvians. *New England Journal of Medicine, 300*, 946-951. https://doi.org/10.1056/NEJM197904263001703

Eshel, T., Yahalom-Mack, N., Tirosh, O., Maeir, A.M., & Harlavan, Y. (2020). Pollution and human mobility in the southern Levant during the Iron Age using chemical and isotopic analysis of human tooth enamel. *Journal of Archaeological Science*, *124*, e105262. https://doi.org/10.1016/j.jas.2020.105262

Evans, J.A., Montgomery, J., Wildman, G., & Boulton, N. (2010). Spatial variations in biosphere $^{87}Sr/^{86}Sr$ in Britain. *Journal of the Geological Society*, *164*, 1-4. https://doi.org/10.1144/0016-76492009-090

Evans, J.A., Chenery, C.A., & Montgomery, J. (2012). A summary of strontium and oxygen isotope variation in human tooth enamel excavated from Britain over the last 6000 years. *Journal of Analytical Atomic Spectrometry*, *5*, 754-764. https://doi.org/10.1039/C2JA10362A

Evans, J., Pashley, V., Madgwick, R., Neil, S., & Chenery, C. (2018a). Tracking natural and anthropogenic lead exposure to its geological source. *Scientific Reports*, *8,* e1969. https://doi.org/10.1038/s41598-018-20397-y

Evans, J.A., Pashley, V., Chenery, C.A., Loe, L., & Chenery, S.R. (2018b). Lead isotope analysis of tooth enamel from a Viking age mass grave in southern Britain and the constraints in places on the origin of the individuals. *Archaeometry*, *60*, 859-869. https://doi.org/10.1111/arcm.12361

Evans, J.A., Pashley, V., Mee, K., Wagner, D., Parker Pearson, M., Frenindeau, D., Albarella, U., Madgwick, R. (2022). Applying lead (Pb) isotopes to explore mobility in humans and animals. PLoS ONE 17(10), e0274831. https://doi.org/10.1371/journal.pone.0274831

Falcon-Lang, H.J. (2009). Earliest History of Coal Mining and Grindstone Quarrying at Joggins, Nova Scotia, and its Implications for the Meaning of the Place Name "Joggins". *Atlantic Geology*, *45*, 74-80. https://doi.org/10.4138/atlgeol.2009.001

Farell, J., Amarasiriwardena, D., Goodman, A.H., & Arriaza, B. (2013). Bioimaging of trace metals in ancient Chilean mummies and contemporary Egyptian teeth by laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS). *Microchemical Journal*, *106,* 340-346. https://doi.org/10.1016/j.microc.2012.09.005

Faure, G., & Mensing, T.M. (2005). *Isotopes: Principles and Applications* (3rd edition). John Wiley and Sons Inc.

Filon, F.L., Boeniger, M., Maina, G., Adami, G., Spinelli, P., & Damian, A. (2006). Skin absorption of inorganic lead (PbO) and the effect of skin cleansers. *Journal of Occupational and Environmental Medicine*, *48*, 392-699. https://doi.org/10.1097/01.jom.0000214474.61563.1c

Finucane, B.C. (2007). Mummies, maize, and manure: multi-tissue stable isotope analysis of late prehistoric human remains from the Ayacucho Valley, Peru. *Journal of Archaeological Science*, *34*, 2115-2124. https://doi.org/10.1016/j.jas.2007.02.006

Fitch, A., Grauer, A., & Augustine, L. (2012). Lead isotope ratios: Tracking the migration of European-Americans to Grafton, Illinois in the 19th century. *International Journal of Osteoarchaeology*, *22*, 305–319. https://doi.org/10.1002/oa.1207

Fleming, D.E.B., Chettle, D.R., Webber, C.E., & O'Flaherty, E.J. (1999). The O'Flaherty model of lead kinetics: An evaluation using data from a lead smelter population. *Toxicol. and Appl. Pharmacol.*, *161*, 100-109. https://doi.org/10.1006/taap.1999.8790

Flora, G., Gupta, D., & Tiwari, A. (2012). Toxicity of lead: A review with recent updates. *Interdisciplinary Toxicology*, *5*, 47–58. https://doi.org/10.2478/v10102-012-0009-2

Flora, S.J.S., Flora, G., & Sazena, G. (2006). Environmental occurrence, health effects and management of lead poisoning. In Casas, J.C., Sordo, J. (Eds.), *Pb: Chemistry, Analytical Aspects, Environmental Impact and Health Effects* (pp. 158-228). Elsevier.

Foley, N.K., Sinha, A.K., & Craig, J.R. (1981). Isotopic composition of lead in the Austinville-Ivanhoe Pb-Zn District, Virginia. *Economic Geology*, *76*, 2012-2017. https://doi.org/10.2113/gsecongeo.76.7.2012

Forel, B., Monna, F., Petit, C., Bruguier, O., Losno, R., Fluck, P., Begeot, C., Richard, H., Bichet, V., & Chateau, C. (2010). Historical mining and smelting in the Vosges Mountains (France) recorded in two ombrotrophic peat bogs. *Journal of Geochemical Exploration*, *107*(1), 9-20. https://doi.org/10.1016/j.gexplo.2010.05.004

Fosse, G., Wesenberg, G.R., Tvinnereim, H.M., Eide, R., Kristoffersen, Ø., Nag, O.H., Wierzbicka, M., Banoczy, J., de Oliveira, A.A., Srisopak, C., & Zamuido, A. (1995). Lead in deciduous teeth from larger cities of some countries. *International Journal of Environmental Studies*, *47*, 203-210. https://doi.org/10.1080/00207239508710960

France, C., Owsley, D.W., & Hayek, L-A.C. (2014). Stable isotope indicators of provenance and demographics in 18th and 19th century North Americans. *Journal of Archaeological Science*, *42*, 356-366. https://doi.org/10.1016/j.jas.2013.10.037

Frank, A.B., Frei, R., Moutafi, I., Voutsaki, S., Orgeolet, R., Kristiansen, K., & Frei, K.M. (2021). The geographic distribution of bioavailable strontium isotopes in Greece – A base for provenance studies in archaeology. *Science of the Total Environment*, *791*, e1148156. https://doi.org/10.1016/j.scitotenv.2021.148156

Franson, R.T., Lucas, A.R., Giroux, L., & Kenniff, P. (1977). *Canadian Law and the Control of Exposure to Hazards*. Science Council of Canada.

Garlie, M.E. (2022). *An Isotopic Investigation of the Diet and Origins of 18th and 19th Century Individuals from Newfoundland and Louisbourg, Nova Scotia* (Publication

No. 15764) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository. https://doi.org/10.48336/EX71-RY04

Gaulton, R. (2001). *Early historic Beothuk evidence from Ferryland, Newfoundland (CgAf-02)* [Honours thesis]. Department of Anthropology, Memorial University of Newfoundland.

Gaulton, B., & Losier, C. (2020). Recasting mobility and movement in eastern North America: A fisheries perspective. In C.E. Orser Jr., A. Zarankin, P. Funari, S. Lawrence, J. Symonds (Eds.), *The Routledge Handbook of Global Historical Archaeology* (pp. 828–850). Routledge.

Ghotbizadeh, M., Cuss, C.W., Grant-Weaver, I., Markov, A., Noernberg, T., Ulrich, A., & Shotyk, W. (2022). Spatiotemporal variations of total and dissolved trace elements and their distributions amongst major colloidal forms along and across the lower Athabasca River. *Journal of Hydrology: Regional Studies, 40*, e101029 https://doi.org/10.1016/j.ejrh.2022.101029

Giovas, C.M., Kamenov, G.D., Fitzpatrick, S.M., & Krigbaum, J. (2016). Sr and Pb isotopic investigations: Pre-Columbian zoogeographic records from the Lesser Antilles, West Indies. *Journal of Archaeological Science*, *69*, 39-53. https://doi.org/10.1016/j.jas.2016.03.006

Gomes, V.E., da Luz Rosário de Sousa, M., Barbosa Jr, F. Jr., Krug, F.J., da Conceição Pereira Saraiva, M., Cury, J.A., & Gerlach, R.F. (2004). In vivo studies on lead content of deciduous teeth superficial enamel of preschool children. *Science of the Total Environment*, *320*, 25-35. https://doi.org/10.1016/j.scitotenv.2003.08.013

González de Mejía, E., & Craigmill, A.L. (1996). Transfer of Lead from Lead-Glazed Ceramics to Food. *Archives of Environmental Contamination and Toxicology, 31*, 581-584.

Grandjean, P., & Holma, B.A. (1973). History of lead retention in the Danish population. *Environmental and Physiology & Biochemistry*, *3*, 268-273.

Grandjean, P., & Jørgenson, P.J. (1990). Retention of lead and cadmium in prehistoric and modern human teeth. *Environmental Research*, *53*, 6-15. https://doi.org/10.1016/s0013-9351(05)80126-4

Green, D.R., Smith, T.M., Green, G.M., Bidlak, F.B., Tafforeau, P., & Colman, A.S. (2018). Quantitative reconstruction of seasonality from stable isotopes in teeth. *Geochimica et Cosmochimica Acta*, *235*, 483-504. https://doi.org/10.1016/j.gca.2018.06.013

Greer, M.C., & MacDonald, B.L. (2022). Buying local: a multi-method approach to determining the provenance of locally made lead-glazed earthenwares. *Archaeological and Anthropological Sciences*, *14*, e136. https://doi.org/10.1007/s12520-022-01597-0

Greer, M.C., MacDonald, B.L., & Stalla, D. (2021). Cobalt, lead, and borax: Preliminary LA-ICP-MS and SEM-EDS analysis of Late-18th- to Mid-19th-century British refined earthenware glazes. *Journal of Archaeological Science: Reports*, *37*, e103013. https://doi.org/10.1016/j.jasrep.2021.103013

Grobler, S.R., Theunissen, F.S., & Kotze, T.J. (2000). The relation between lead concentrations in human dental tissues and in blood. *Archives of Oral Biology*, *45*, 607-609. https://doi.org/10.1016/s0003-9969(00)00024-8

Guede, I., Zuluaga, M.C., Ortega, L.A., Alonso-Olazabal, A., Murelaga, X., Pina, M., & Gutierrez, F.J. (2017). Analyses of human dentine and tooth enamel by laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) to study the diet of medieval muslim individuals from Tauste (Spain). *Microchemical Journal*, *130*, 287-294. https://doi.org/10.1016/j.microc.2016.10.005

Guiry, E., Szpak, P., & Richards, M.P. (2017). Isotopic analyses reveal geographical and socioeconomic patterns in historical domestic animal trade between predominantly wheat- and maize-growing agricultural regions in Eastern North America. *American Antiquity*, *82*(2), 341-352. https://doi.org/10.1017/aaq.2016.34

Gulson, B.L. (1986). *Lead isotope in mineral exploration, Developments in Economic Geology*. Elsevier.

Gulson, B.L. (1996). Tooth analyses of sources and intensity of lead exposure in children. *Environmental Health Perspectives*, *104*, 306-312. https://doi.org/10.1289/ehp.96104306

Gulson, B.L. (2008). Stable lead isotopes in environmental health with emphasis on human investigations. *Science of the Total Environment*, *400*, 75-79. https://doi.org/10.1016/j.scitotenv.2008.06.059

Gulson, B.L., & Wilson, D. (1994). History of lead exposure in children revealed from isotopic analyses of teeth. *Archives of Environmental Health*, *49*, 279-83. https://doi.org/10.1080/00039896.1994.9937480

Gulson, B.L., Mizon, K.J., Korsch, M.J., & Howarth, D. (1996a). Non-orebody sources are significant contributors to blood lead of some children with low to moderate lead exposure in a major lead mining community. *Science of the Total Environment*, *181*, 223-30. https://doi.org/ 10.1016/0048-9697(95)05015-9

Gulson, B.L., Mizon, K.J., Korsch, M.J., & Howarth, D. (1996b). Importance of monitoring family members in establishing sources and pathways of lead in blood. *Science of the Total Environment, 188*, 173-182. https://doi.org/10.1016/0048-9697(96)05170-4

Gulson, B.L., Mizon, K.J., Korsch, M.J., Palmer, J.M., & Donnelly, J.B. (2003). Mobilization of lead from human bone tissue during pregnancy and lactation – a summary of long-term research. *Science of the Total Environment*, *303*, 79-104. https://doi.org/0.1016/s0048-9697(02)00355-8

Gulson, B.L., Mizon, K.J., Davis, J.D., Palmer, J.M., & Vimpani, G. (2004). Identification of sources of lead in children in a primary zinc-lead smelter environment. *Environmental Health Perspectives*, *112*, 52-60. https://doi.org/ 10.1289/ehp.6465

Gwiazda, R., Campbell, C., & Smith, D. (2005). Noninvasive isotopic approach to estimate the bone lead contribution to blood in children: Implications for assessing the efficacy of lead abatement. *Environmental Health Perspectives*, *113*, 104-110. https://doi.org/10.1289/ehp.7241

Halcrow, S.E., & Tayles, N. (2011). The bioarchaeological investigation of children and childhood. In S.C Agarwal, B.A. Glencross (Eds.), *Social Bioarchaeology* (pp. 333-360). Wiley-Blackwell.

Handcock, W.G. (2003). *So longe as there comes no women: Origins of English settlement in Newfoundland*. Breakwater Books.

Harris, D. (1974*). Block 3 Skeletal Report (A E 58), Fortress of Louisbourg (No. A E 58)*. Parks Canada.

Harris, O.J.T., Cobb, H., Batey, C.E., Montgomery, J., Beaumont, J., Gray, H., Murtagh, P., & Richardson, P. (2017). Assembling places and persons: a tenth-century Viking boat burial from Swordle Bay on the Ardnamurchan peninsula, western Scotland. *Antiquity*, *91*(355), 191-206. https://doi.org/10.15184/aqy.2016.222

Head, C.G. (1976). *Eighteenth Century Newfoundland: A Geographer's Perspective*. McGill-Queen's Press.

Hemphill, C.P., Ruby, M.V., Beck, B.D., Davis, A., & Bergstrom, P.D. (1991). The Bioavailability of Lead in Mining Wastes: Physical/Chemical Considerations. *Chemical Speciation & Bioavailability*, *3*(3-4), 135-148. https://doi.org/10.1080/09542299.1991.11083165

Hernández-Avila, M., Smith, D., Meneses, F., Sanin, L.H., & Hu, H. (1998). The influence of bone and blood lead on plasma lead levels in environmentally exposed adults. *Environmental Health Perspectives*, *106*, 473-477. https://doi.org/ 10.1289/ehp.106-1533211

Heyl, A.V., Lyons, E.J., Agnew, A.F., & Behre, C.H. Jr. (1955). Zinc-lead-copper resources and general geology of the Upper Mississippi Valley District. *Contributions to Economic Geology, Geological Survey Bulletin issue*, *1015*, 227-243.

Heyl, A.V., Delevaux, M.H., Zartman, R.E., & Brock, M.R. (1966). Isotopic study of galenas from the Upper Mississippi Valley, the Illinois-Kentucky, and some Appalachian Valley mineral districts. *Economic Geology*, *61*, 933-961. https://doi.org/10.2113/gsecongeo.61.5.933

Hiller, J.K. (1996). The Newfoundland fisheries issue in Anglo-French treaties, 1713-1904. *The Journal of Imperial and Commonwealth History*, *24*(1), 1-23. https://doi.org/10.1080/03086539608582966

Hiller, J. (2008). The Nineteenth Century, 1815-1914. p. 77-102 In *A Short History of Newfoundland and Labrador*. Edited by Newfoundland Historical Society, Portugal Cove-St. Philip's: Boulder Publications.

Hillson, S. (2014). *Tooth Development in Human Evolution and Bioarchaeology*. Cambridge University Press.

Hinners, T.A., Hughes, R., Outridge, P.M., Davis, W.J., Simon, K., & Woolard, D.R. (1998). Interlaboratory comparison of mass spectrometric methods for lead isotopes and trace elements in NIST SRM 1400 Bone Ash. *Journal of Analytical Atomic Spectrometry*, *13*, 963-970. https://doi.org/10.1039/A803373K

Holm, P., Ludlow, F., Scherer, C., Travis, C., Allaire, B., Brito, C., Hayes, P.W., Matthews, A.J., Rankin, Breen, R.J., Legg, R., Lougheed, K., & Nicholls, J. (2019). The North Atlantic fish revolution (ca. AD 1500). *Quaternary Research*, *108*, 1-5. https://doi.org/10.1017/qua.2018.153

Holt, E., Evans, J.A., & Madgwick, R. (2021). Strontium ($^{87}Sr/^{86}Sr$) mapping: A critical review of methods and approaches. *Earth-Science Reviews*, *216*, e103593. https://doi.org/10.1016/j.earscirev.2021.103593

Hornberg, C., Maciuleviciute, L., & Seemayer, N.H. (1998). Analysis of Cyto- and Genotoxic Effects of Atmospheric Pollutants on Rodent and Human Trachaeobronchial Epithelial cells in vitro. *Toxicology in Vitro*, *11*(5), 711-715. https://doi.org/10.1016/s0887-2333(97)00065-9.

Hursh JB, Schraub A, Sattler EL, & Hofmann, H.P. (1969). Fate of 212Pb inhaled by human subjects. *Health Physics Journal*, *16*, 257-267.

Ingram, S. (2014) Folklore of Wells in newfoundland. *Occasional Paper on Intangible Cultural Heritage, 5*, 1-8. Heritage Foundation of Newfoundland and Labrador.

James, A.C., Stahlhofen, W., Rudolf, G., Köbrich, R., Briant, J.K., Egan, M.J., Nixon, W., & Birchall, A. (1994). Annexe D. Deposition of inhaled particles. *Annals of the ICRP*, *24*(1-3), 231-299. https://doi.org/10.1016/0146-6453(94)90042-6

Janowitz, M.F. (2013). Decline in the Use and Production of Red-Earthenware Cooking Vessels in the Northeast, 1780-1880. *Northeast Historical Archaeology*, *42*, 92–110. https://doi.org/10.22191/neha/vol42/iss1/7

Janzen, O.U. (2008). The Long Eighteenth Century, 1697-1815. p. 49-86 In *A Short History of Newfoundland and Labrador*. Edited by Newfoundland Historical Society, Portugal Cove-St. Philip's: Boulder Publications.

Janzen, A., Bataille, C., Copeland, S., Quinn, R., Ambrose, S.H., Reed, D., Hamilton, M., Grimes, V., Richards, M.P., le Roux, P., & Roberts, P. (2020). Spatial variation in bioavailable strontium isotope ratios ($^{87}Sr/^{86}Sr$) in Kenya and northern Tanzania: Implications for ecology, paleoanthropology, and archaeology. *Palaeogeography, Palaeoclimatology, Palaeoecology*, *560*, e109957. https://doi.org/10.1016/j.palaeo.2020.109957

Jarcho, S. (1964). Lead in the bones of prehistoric lead-glaze potters. *American Antiquity*, *30*, 94-96. https://doi.org/10.2307/277640

Jaworowski, Z. (1968). Stable lead in fossil ice and bones. *Nature*, *217*, 152-153. https://doi.org/10.1038/217152a0

Jaworowski, Z., Barbalat, F., Blain, C., & Peyre, E. (1985). Heavy metals in human and animal bones from ancient and contemporary France. *Science of the Total Environment*, *43*, 103-126. https://doi.org/ 10.1016/0048-9697(85)90034-8

Johnston, A.J.B. (1984). *Religion in life at Louisbourg, 1713-1758*. McGill-Queen's University Press.

Johnston, A.J.B. (2004). Fortress, Seaport, Community: Three Faces of 18th-Century Louisbourg. *Journal of the Royal Nova Scotia Historical Society*, *7*, 82–101.

Jonasson, M.E., & Afshari, R. (2017). Historical documentation of lead toxicity prior to the 20th century in English literature. *Human & Experimental Toxicology*, *37*(8), 775-788. https://doi.org/10.1177/0960327117737146

Jones, D.S., Turner, B.L., Buikstra, J.E., & Kamenov, G.D. (2017). Investigating the identities of isolated crania in the Lower Illinois River Valley through multi-isotopic analysis. *Journal of Archaeological Science: Reports*, *13*, 312-321. https://doi.org/10.1016/j.jasrep.2017.02.030

Jones, J. (2015). *The Political Ecology of Early Childhood Pb Exposure at the New York African Burial Ground* (Publication No. 307) [Doctoral dissertation, University of Massachusetts, Amherst]. https://doi.org/10.7275/6466541.0

Kamenov, G.D. (2008). High-precision Pb isotopic measurements of teeth and environmental samples from Sofia (Bulgaria): Insights for regional Pb sources and possible pathways to the human body. *Environmental Geology*, *55*, 669-680. https://doi.org/10.1007/s00254-007-1017-y

Kamenov, G.D., Mueller, P.A., & Perfit, M.R. (2004). Optimization of mixed Pb–Tl solutions for high precision isotopic analyses by MC-ICP-MS. *Journal of Analytical Atomic Spectrometry*, *19*(9), 1262-1267. https://doi.org/10.1039/B403222E

Kamenov, G.D., & Gulson, B.L. (2014). The Pb isotopic record of historical to modern human lead exposure. *Science of the Total Environment*, *490*, 861-870. https://doi.org/10.1016/j.scitotenv.2014.05.085

Kamenov, G.D., Logaro, E.M., Goad, G., & Krigbaum, J. (2018). Trace elements in modern and archaeological human teeth: Implications for human metal exposure and enamel diagenetic changes. *Journal of Archaeological Science*, *99*, 27-34. https://doi.org/10.1016/j.jas.2018.09.002

Kamenov, G.D., & Krigbaum, J. (2023). Pb Isotopes and Human Mobility: Natural, Cultural, or Diagenetic Signal? p. 163-185, In Price, T.D. (Ed.) *Isotopic Proveniencing and Mobility.* Interdisciplinary Contributions to Archaeology, Springer.

Kamp, K.A. (2001). Where have all the children gone?: The archaeology of childhood. *Journal of Archaeological Method and Theory*, *8*, 1-34. https://doi.org/10.1023/A:1009562531188

Katzenberg, M.A. (2008). Stable isotope analysis: a tool for studying past diet, demography and life history. In M.A. Katzenberg & A.L. Grauer (Eds.), *Biological Anthropology of the Human Skeleton* (pp. 469-504). John Wiley and Sons, Inc.

Keenleyside, A., Stenton, D.R., & Newman, K. (2021). The integration of isotopic and historical data to investigate the identification of crewmembers of the 1845 Franklin expedition. *Journal of Archaeological Science: Reports*, *40*, e103200. https://doi.org/10.1016/j.jasrep.2021.103200

Keller, A.T., Regan, L.A., Lundstrom, C.C., & Bower, N.W. (2016). Evaluation of the efficacy of spatiotemporal Pb isoscapes for provenancing of human remains. *Forensic Science International*, *261*, 83–92. https://doi.org/10.1016/j.forsciint.2016.02.006

Kendall, C., Eriksen, A.M., Kontopoulos, I., Collins, M.J., & Turner-Walker, G. (2018). Diagenesis of archaeological bone and tooth. *Paleogeography, Palaeoclimatology, Palaeoecology*, *491*(1), 21-37. https://doi.org/10.1016/j.palaeo.2017.11.041

Kesler, S.E., Cumming, G.L., Krstic, D., & Appold, M.S. (1994a). Lead isotope geochemistry of Mississippi Valley-type deposits of the Southern Appalachians. *Economic Geology*, *89*, 307-321. https://doi.org/10.2113/gsecongeo.89.2.307

King, C.L., Petchey, P., Buckley, H.R., Girvan, E., Kinaston, R., & Nowell, G.M. (2020). Lead astray: The potentials and pitfalls of lead isotopes in a New Zealand colonial burial context. *Journal of Archaeological Science: Reports*, *30*, e102213. https://doi.org/10.1016/j.jasrep.2020.102213

King, C.L., Buckley, H.R., Petchey, P., Roberts, P., Zech, J., Kinaston, R., Collins, C., Kardailsky, O., Matisoo-Smith, E., & Nowell, G. (2021). An isotopic and genetic study of multi-cultural colonial New Zealand. *Journal of Archaeological Science*, *128*, e105337. https://doi.org/10.1016/j.jas.2021.105337

Killick, D.J., Stephens, J.A., & Fenn, T.R. (2020). Geological constraints on the use of lead isotopes for provenance in archaeometallurgy. *Archaeometry*, *61*(S1), 86-105. https://doi.org/10.1111/arcm.12573

Knott, J., Gilbert, J. Hoccom, D.G., & Green, R.E. (2010). Implications for wildlife and humans of dietary exposure to lead from fragments of lead rifle bullets in deer shot in the UK. *Science of the Total Environment*, *409*, 95-99. https://doi.org/10.1016/j.scitotenv.2010.08.053

Knüsel, C.J., Batt, C.M., Cook, G., Montgomery, J., Müldner, G., Ogden, A.R., Palmer, C., Stern, B., Todd, J., & Wilson, A.S. (2010). The identity of the St Bees Lady, Cumbria: An osteobiographical approach. *Medieval Archaeology*, *54*(1), 271-311. https://doi.org/10.1179/174581710X12790370815931

Kowal, W.A., Beattie, O.B., Baadsgaard, H., & Krahn, P.M. (1991). Source identification of lead found in tissues of sailors from the Franklin Arctic Expedition of 1845. *Journal of Archaeological Science*, *18*, 193-203. https://doi.org/10.1016/0305-4403(91)90048-T

Lacruz, R.S., Habelitz, S., Wright, J.T., & Paine, M.L. (2017). Dental enamel formation and implications for oral health and disease. *Physiological Reviews*, *97*, 939-993. https://doi.org/10.1152/physrev.00030.2016

Laffoon, J.E., Shuler, K.A. Millard, A.R., Connelly, J.N., & Schroeder, H. (2020). Isotopic evidence for anthropogenic lead exposure on a 17th/18th century Barbadian plantation. *American Journal of Physical Anthropology*, *171*, 529-538. https://doi.org/10.1002/ajpa.23938

Laidlaw, M.A.S., Mielke, H.W., Fillippelli, G.M., Johnson, D.L., & Gonzales, C.R. (2005). Seasonality and children's blood lead levels: developing a predictive model using climatic variables and blood lead data from Indianapolis, Indiana, Syracuse, New York, and New Orleans, Louisiana (USA). *Environmental Health Perspectives*, *113*, 793-800. https://doi.org/10.1289/ehp.7759

Lamb, A.L., Evans, J.E., Buckley, R., & Appleby, J. (2014). Multi-isotope analysis demonstrates significant lifestyle changes in King Richard III. *Journal of Archaeological Science*, *50*, 559-565. https://doi.org/10.1016/j.jas.2014.06.021

Landry, N. (2004). Les activités de course dans un port colonial français: Plaisance, Terre-Neuve, durant la guerre de Succession d'Espagne, 1702-1713. *Acadiensis*, *34*(1), 56-79.

Landry, N. (2008). *Plaisance: Terre-Neuve, 1650-1713: Une colonie française en Amérique*. Septentrion.

Landry, N. (2018). Relations between the Basques and the French administration of Plaisance, 1600-1713. *Newfoundland sand Labrador Studies*, *33*(1), 113-135. https://doi.org/10.7202/1055867ar

Lecos, C. (1987). *Pretty Poison: Lead and Ceramic Ware*. Department of Health and Human Services, Public Health Service, Food and Drug Administration, Office of Public Affairs.

Lessler, M.A. (1988). Lead and Lead Poisoning from Antiquity to Modern Times. *Ohio Journal of Science*, *88*, 78–84.

Lévesque, B., Duchesne, J.-F., Gariépy, C., Rhainds, M., Dumas, P., Scheuhammer, A.M., Proulx, J.-F., Déry, S., Muckle, G., Dallaire, F., & Dewailly, É. (2003).

Monitoring of umbilical cord blood lead levels and sources assessment among the Inuit. *Occupational and Environmental Medicine*, *60*, 693-695. http://dx.doi.org/10.1136/oem.60.9.693

Lillehammer, G. (2016). 25 Years with the 'Child' and the Archaeology of Childhood. *Childhood in the Past: An International Journal*, *8*(2),78-86. https://doi.org/10.1179/1758571615Z.00000000030

Lilley, S.G., Florence, T.M., & Stauber, J.L. (1988). The use of sweat to monitor lead absorption through the skin. *Science of the Total Environment*, *76*, 267-278. https://doi.org/10.1016/0048-9697(88)90112-X

Livingston, M., & Losier, C. (2021). "From the Sea, Work": Investigating historical French landscapes and lifeways at Anse à Bertrand, Saint-Pierre et Miquelon. *Northeast Historical Archaeology*, *50*, e7. https://orb.binghamton.edu/neha/vol50/iss1/7

Loewen, B., & Delmas, V. (2012). The Basques in the Gulf of St. Lawrence and adjacent shores. *Canadian Journal of Archaeology*, *36*(2), 213-266.

López-Costas, O., Kylander, M., Mattielli, N., Álvarez-Fernández, N., Pérez-Rodrígez, M., Mighall, T., Bindler, R., & Martínez Cortizas, A. (2020). Human bones tell the story of atmospheric mercury and lead exposure at the edge of Roman World. *Science of the Total Environment*, *710*, e136319. https://doi.org/10.1016/j.scitotenv.2019.136319

Losier, C., & Lear, M. (2018). *Final Report for Tors Cove 2016 (ChAf-01 and ChAf-02)* (No. 16.07). Provincial Archaeology Office.

Losier, C., Loewen, B., & Goya, M. E. (2018). In the Midst of Diversity: Recognizing the Seventeenth-Century Basque Cultural Landscape and Ceramic Identity in Southern Newfoundland and Saint-Pierre-et-Miquelon. *Newfoundland and Labrador Studies*, *33*(1), 200-236.

Losier, C., Liew, Z.M., Champagne, M., & Livingston, M. (2021). Le grand métier et la petite pêche: Archéologie des XIXe et XXe siècles à l'anse à Bertrand, Saint-Pierre et Miquelon. *Revue d'archeologie contemporaine*, *1*, 59-80.

Mackie, A., Townshend, A., & Waldron, H.A. (1975). Lead concentrations in bones from Roman York. *Journal of Archaeological Science*, *2*, 235-237. https://doi.org/10.1016/0305-4403(75)90062-X

MacKinnon, R. (1991). Farming the rock: The evolution of commercial agriculture around St. John's, Newfoundland, to 1945. *Acadiensis*, *20*(2), 32-61.

Mahaffey, K.R. (1978). Environmental exposure to lead. In J.O. Nriagu (Ed.), *The Biogeochemistry of Lead in the Environment* (pp. 1-36). Pat B: Biological Effects. Topics in Environmental Health, Part 1B.

Mannion, J. (2001). Irish migration and settlement in Newfoundland: The formative phase, 1697-1732. *Newfoundland and Labrador Studies*, *17*(2), 257-293.

Mannion, J. (2004). Tracing the Irish: A geographical guide. *The Newfoundland Ancestor*, *9*(1), 4-18. https://nlirishconnections.dreamhosters.com/wp-content/uploads/2020/08/TRACING-THE-IRISH.pdf

Marshall, J.H. Jr., & Dunn, P.J. (1976). The lead mines of Loudville. *Rocks & Minerals*, *51*(5), 250-255. https://doi.org/10.1080/00357529.1976.11762804

Martin, W. (1983). *Once Upon a Mine: Story of Pre-Confederation Mines on the Island of Newfoundland*. Canadian Institute of Mining and Metallurgy, vol. 26.

Matthews, K. (1973). *Lectures on the history of Newfoundland, 1500-1830*. Maritime History Group, Memorial University of Newfoundland.

Matthews, K. (2001). Historical fence building: A critique of the historiography of Newfoundland. *Newfoundland and Labrador Studies*, *17*(2), 143-165.

McIntosh, R. (2000). *Boys in the Pits: Child Labour in Coal Mines*. McGill-Queen's University Press.

McLeod-Lesie, H. (2014). Archaeology and Atlantic Canada's African Diaspora. *Acadiensis*, *43*(1), 137-146.

McMillan, R., Snoeck, C., de Winter, N., Claeys, P., & Weis, D. (2019). Evaluating the impact of acetic acid chemical pre-treatment on 'old' and cremated bone with the 'Perio-spot' technique and 'Perios-endos' profiles. *Palaeogeography, Palaeoclimatology, Palaeoecology*, *530*, 330-344. https://doi.org/10.1016/j.jas.2017.10.003

Métreau, L., Cattin, F., Villa, I.M., André, P., & Chateau-Smith, C. (2021). Lead provenance for medieval decorated tile glazes from Brittany and Anjou (13th-14th c.). *Journal of Archaeological Science: Reports*, *38*, e103037. https://doi.org/10.1016/j.jasrep.2021.103037

Mielke, H.W. & Reagan, P.L. (1998). Soil is an important pathway of human lead exposure. *Environmental Health Perspectives*, *106*, 217-229. https://doi.org/10.1289/ehp.98106s1217

Millard, A. (2006). Comment on Martínez-García et al., "Heavy metals in human bones in different historical epochs." *Science of the Total Environment*, *354*, 295-297. https://doi.org/10.1016/j.scitotenv.2005.11.010

Millard, A., Montgomery, J., Trickett, M., Beaumont, J., Evans, J., & Chenery, S. (2014). Childhood lead exposure in the British Isles during the Industrial Revolution. In M.K. Zuckerman (Ed.), *Modern environments and human health: Revisiting the second epidemiologic transition* (pp. 279-299). John Wiley and Sons.

Millen, T.M., Zartman, R.E., & Heyl, A.V. (1995). *Lead Isotopes from the Upper Mississippi Valley District: A Regional Perspective*. U.S. Geological Survey Bulletin 2094-B, pp. B1-B3.

Milot, J., Blichert-Toft, J., Sanz, M.A., Fetter, N., Télouk, P., & Albarède, F., (2021). The significance of galena Pb model ages and the formation of large Pb-Zn sedimentary deposits. *Chemical Geology*, *583*(20), e120444. https://doi.org/10.1016/j.chemgeo.2021.120444

Montgomery, J. (2002). *Lead and Strontium Isotope Compositions of Human Dental Tissues as an Indicator of Ancient Exposure and Population Dynamics* (Publication No. 453) [Doctoral dissertation, University of Bradford]. Archaeology Data Service. https://doi.org/10.5284/1000249

Montgomery, J., Budd, P., & Evans, J. (2000). Reconstructing the lifetime movements of ancient people: A Neolithic case study from Southern England. *European Journal of Archaeology*, *3*, 407-422. https://doi.org/10.1179/146195700807860828

Montgomery, J., Evans, J.A., Powlesland, D., & Roberts, C.A. (2005). Continuity or colonization in Anglo-Saxon England? Isotope evidence for mobility, subsistence

practice, and status at West Heslerton. *American Journal of Physical Anthropology*, *126*, 123-138. https://doi.org/10.1002/ajpa.20111

Montgomery, J., Evans, J., Chenery, S., Pashley, V., & Killgrove, K. (2010). "Gleaming, white and deadly": using lead to track human exposure and geographic origins in the Roman period in Britain. *Journal of Roman archaeology Supplements, 78*, 199–226.

Moore, J., Rose, A., Anderson, S., Evans, J., Nowell, G., Gröcke, D.R., Pashley, V., Kirby, M., & Montgomery, J. (2020). A multi-isotope (C, N, O, Sr, Pb) study of Iron Age and Roman period skeletons from east Edinburgh, Scotland exploring the relationship between decapitation burials and geographical origins. *Journal of Archaeological Science: Reports*, *29*, e102075. https://doi.org/10.1016/j.jasrep.2019.102075

Moore, J., Filipek, K., Kalenderian, V., Gowland, R., Hamilton, E., Evans, J., & Montgomery, J. (2021). Death metal: Evidence for the impact of lead poisoning on childhood health within the Roman Empire. *International Journal of Osteology*, *31*(5), 846-856. http://doi.org/10.1002/oa.3001

Morrow, P., Beiter, H., Amato, F., & Gibb, F.R. (1980). Pulmonary retention of lead: an experimental study in man. *Environmental Research*, *21*(2), 373-384. https://doi.org/10.1016/0013-9351(80)90040-7

Mosier, M. (1948). *Bamford zinc mine, Lancaster County, Pa.*, Report of investigations / United States Department of the Interior, Bureau of Mines, volume 4247. U.S. Department of the Interior, Bureau of Mines.

Müller, W., Fricke, H., Halliday, A.N., McCulloch, M.T., & Wartho, J.-A. (2003). Origin and migration of the alpine iceman. *Science*, *302*(5646), 862-866. https://doi.org/10.1126/science.1089837

Müller, W., Nava, A., Evans, D., Rossia, P.F., Alt, K.W., & Bondioli, L. (2019). Enamel mineralization and compositional time-resolution in human teeth evaluated via histologically-defined LA-ICPMS profiles. *Geochimica et Cosmochimica Acta*, *255*, 105-126. https://doi.org/10.1016/j.gca.2019.03.005.

Munkittrick, T.J.A. (2015). A nursery for seamen: life histories from the St. John's Royal Naval Hospital Cemetery, Newfoundland (Publication No. 11708) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Munkittrick, T.J.A., Varney, T.L., Pike, K.-A., & Grimes, V. (2019). Life histories from the Southside Cemetery, St. John's, Newfoundland: Insights into Royal Naval diet using stable isotopes. *Journal of Archaeological Science: Reports*, *24*, 815–828. https://doi.org/10.1016/j.jasrep.2019.02.016

Munkittrick, T.J.A., Varney, T.L., & Grimes, V. (2023). The use and abuse of Pb in bioarchaeological studies: A review of Pb concentration and isotope analyses of teeth. *Journal of Archaeological Science*, 156, e105803. https://doi.org/10.1016/j.jas.2023.105803

Mushak. P. (1991). Gastro-Intestinal Absorption of Lead in Children and Adults: Overview of Biological and Biophysico-chemical Aspects. *Chemical Speciation & Bioavailability*, *3*(3/4), 87-104. https://doi.org/10.1080/09542299.1991.11083160

Nadon, P. (2004). *La Baie du Grand Pabos. Une seigneurie gaspésienne en Nouvelle-France au XVIIIe siècle*. Association des archaéologues du Quebec.

Needleman, H. (2004). Lead Poisoning. *Annual Review of Medicine*, *55*, 209-222. https://doi.org/10.1146/annurev.med.55.091902.103653

Nef, J.U. (1987). Mining and Metallurgy in Medieval Civilisation. In C. Postan, E. Miller, & M.M. Postan (Eds.), *The Cambridge Economic History of Europe from the Decline of the Roman Empire: Volume 2, Trade and Industry in the Middle Ages* (pp. 691–761). Cambridge University Press. https://doi.org/10.1017/CHOL9780521087094.012

Nemec, T.F. (2006). Historic Adaptive Strategies of Euro-Newfoundland Forager-Fishers. In X.S.R. Campos, & J.M.S. Solla (Eds.), *Galicia & Terranova & Labrador: Comparative Studies on Economic, Political, and Social-cultural Processes* (pp. 169–179). Universidade de Santiage de Compostela.

Neuman WF, & Neuman, M.W. (1958). *The Chemical Dynamics of Bone Mineral*. University of Chicago Press.

O'Connell, T.C., & Hedges, R.E.M. (1999). Isotopic comparison of hair and bone: archaeological analyses. *Journal of Archaeological Science*, *26*, 661-665. https://doi.org/10.1006/jasc.1998.0383

O'Flaherty, E.J. (1998). A physiologically based kinetic model for lead in children and adults. *Environmental Health Perspectives*, *106*, 1495-1503. https://doi.org/10.1289/ehp.98106s61495

Oberdörster, G. (1993). Lung dosimetry: pulmonary clearance of inhaled particles. *Aerosol Science and Technology*, *18*(3), 279-289. https://doi.org/10.1080/02786829308959605

Ogle, D.H., Wheeler, P., & Dinno, A. (2020). *Fisheries Stock Analysis*. R package version 0.8.30. https://github.com/droglenc/FSA

Pan, T.-L., Wang, P.-W., Al-Suwayeh, S.A., Chen, C.-C., & Fang, J.-Y. (2010). Skin toxicology of lead species evaluated by their permeability and proteomic profiles: A comparison of organic and inorganic lead. *Toxicology Letters*, *197*, 19-28. https://doi.org/10.1016/j.toxlet.2010.04.019.

Pastore, R. (1989). The collapse of the Beothuk world. *Acadiensis*, *19*(1), 52-71.

Patterson, C.C., Shirahata, H., & Ericson, J.E. (1987). Lead in ancient human bones and its relevance to historical developments of social problems with lead. *Science of the Total Environment*, *61*, 167-200. https://doi.org/10.1016/0048-9697(87)90366-4

Patterson, C., Ericson, J. Manea-Krichten, M., & Shirahata, H. (1991). Natural skeletal levels of lead in *Homo sapien sapiens* uncontaminated by technological lead. *Science of the Total Environment*, *107*, 205-236. https://doi.org/10.1016/0048-9697(91)90260-l

Pendery, S.R. (1985). Ceramics and the colonial system: The Charlestown example. In S.P. Turnbaugh (Ed.), Domestic pottery of the Northeastern United States (pp. 67-80). Academic Press Inc.

Petrucci, J.F. (1999). *Les poteries et les potiers de Vallauris 1501-1945.* [Doctoral dissertation]. École des Hautes Études en Sciences Sociales.

Pike, K.-A. (2013). *Osteological Assessment of Human Remains from St. Paul's Anglican Church (CkAh-6), Harbour Grace, Newfoundland*. Provincial Archaeology Office.

Pillola, G.L. (2002). Mining activities and history. *Rend. Soc. Paleont. Ital.*, *1*, 235-236.

Pin, C., Gannoun, A., & Dupont, A. (2014). Rapid, simultaneous separation of Sr, Pb, and Nd by extraction chromatography prior to isotope ratios determination by TIMS and MC-ICP-MS. *Journal of Analytical Atomic Spectrometry*, *29*, 1858–1870. https://doi.org/10.1039/C4JA00169A

Pollard, A.M. (2009). What a long, strange trip it's been: lead isotopes and archaeology. In A.J. Shortland, I.C. Freestone, & T. Rehren (Eds.), *From Mine to Microscope: Advances in the Study of Ancient Technology* (pp. 181-189). Oxbow Books.

Pope, P. (1993). Scavengers and caretakers: Beothuk/European settlement dynamics in seventeenth-century Newfoundland. *Newfoundland and Labrador Studies, 9*(2), 279-293.

Pope, P. (2003). Modernization on hold: The traditional character of the Newfoundland cod fishery in the seventeenth century. *International Journal of Maritime History*, *15*(2), 233-264.

Pope, P. (2004). *Fish into wine: The Newfoundland plantation in the seventeenth century*. The University of North Carolina Press.

Pope, P. (2008). Historical archaeology and the Maritime cultural landscape of the Atlantic fishery. In A. MacEachern, W.J. Turkel (Eds.), *Method and meaning in Canadian environmental history* (pp. 36-54). Nelson Education Ltd.

Pope, P. (2009). Transformation of the Maritime cultural landscape of Atlantic Canada by migratory European fishermen, 1500-1800. In L. Sicking, & D. Abreu-Ferreira (Eds.), *On Beyond the Catch: Fisheries of the North Atlantic, the North Sea and the Baltic, 900-1850* (pp. 123-154). Brill.

Price, T.D., Knipper, C., Grupe, G., & Smrcka, V. (2004). Strontium isotopes and prehistoric human migration: The Bell Beaker period in central Europe. *European Journal of Archaeology*, *7*, 9-40. https://doi.org/10.1177/1461957104047992.

Price, T.D., Frei, R., Bäckström, Y., Frei, K.M., & Ingvarsson-Sundstrom, A. (2017). Origins of inhabitants from the 16th century Sala (Sweden) silver mine cemetery – A lead isotope perspective. *Journal of Archaeological Science*, *80*, 1–13. https://doi.org/10.1016/j.jas.2017.01.013

Price, T.D., Frei, R., Brinker, U., Lidke, G., Terberger, T., Frei, K.M., & Jantzen, D. (2019). Multi-isotope proveniencing of human remains from a Bronze Age battlefield in the Tollense Valley in northeast Germany. *Archaeological and Anthropological Science*, *11*, 33-49. https://doi.org/10.1007/s12520-017-0529-y

Price, T.D., Klassen, L., & Sjögren, K.-G. (2021). Pitted ware culture: Isotopic evidence for contact between Sweden and Denmark across the Kattegat in the Middle Neolithic, ca. 3000 BC. *Journal of Anthropological Archaeology*, *61*, e101254. https://doi.org/10.1016/j.jaa.2020.101254

Pritchard, J. (1999). "Le profit et la gloire": The French Navy's alliance with private enterprise in the defense of Newfoundland, 1691-1697. *Newfoundland and Labrador Studies*, *15*(2), 161–175.

Quinn, R.L., Warnasch, S.C., Watson, M., Godfrey, L., Setera, J.B., VanTongeren, J., Mortlock, R., & Wright, J. (2020). Biogeochemical evidence for residence, diet, and health of the Woman in the Iron Coffin (Queens, New York City). *International Journal of Osteoarchaeology*, *30*, 225-235. https://doi.org/10.1002/oa.2850

R Core Team (2022). *R: A language and environment for statistical computing*. R Foundation for Statistical Computing. https://www.R-project.org/

Rabinowitz, M.B., & Wetherill, G.W. (1972). Identifying sources of lead contamination by stable isotope techniques. *Environmental Science & Technology*, *6*, 705-709. https://doi.org/10.1021/es60067a003

Rabinowitz, M.B., Wetherill, G.W., & Kopple, J.D. (1976). Kinetic analysis of lead metabolism in healthy humans. *The Journal of Clinical Investigation*, *58*, 260–270. https://doi.org/10.1172/JCI108467

Rabinowitz, M.B., Kopple, J.D., & Wetherill, G.W. (1980). Effect of food intake and fasting on gastrointestinal Pb absorption in humans. *The American Journal of Clinical Nutrition*, *33*, 1784-1788. https://doi.org/10.1093/ajcn/33.8.1784

Radosevich, S.C. (1993). The six deadly sins of trace element analysis: a case of wishful thinking in science. In M. Sandford (Ed.), *Investigations of Ancient Human Tissue: Chemical Analyses in Anthropology* (pp. 269-332). Gordon & Breach.

Ram, R., Morrisroe, L., Etschmann, B., Vaughan, J., & Brugger, J. (2021). Lead (Pb) sorption and co-precipitation on natural sulfide, sulfate and oxide minerals under environmental conditions. *Minerals Engineering*, *163*(15), e106801. https://doi.org/10.1016/j.mineng.2021.106801

Rasmussen, K.L., Skyette, L., Jensen, A.J., & Boldsen, J.L. (2015). Comparison of mercury and lead levels in the bones of rural and urban populations in southern Denmark and northern Germany during the Middle Ages. *Journal of Archaeological Science: Reports*, *3*, 358-370. https://doi.org/10.1016/j.jasrep.2015.06.021

Rasmussen, K.L, Milner, G., Skytte, L., Lynnerup, N., Thomsen, J.L., & Boldsen, J.L. (2019). Mapping diagenesis in archaeological human bones. *Heritage Science*, *7*, e41. https://doi.org/10.1186/s40494-019-0285-7

Rasmussen, K.L., Milner, G.R., Delbey, T., Ivalu Jensen, L.K., Witte, F., Rehren, T., Kjaer, U., Grinder-Hansen, P. (2022) Release of lead from Renaissance lead-glazed ceramics from southern Denmark and northern Germany: Implications from acetic acid etching experiments. *Heritage Science*, *10*, e63. https://doi.org/10.1186/s40494-022-00703-8

Rawlyk, G. (1999). *Yankees at Louisbourg: The story of the first siege 1745*. Breton Books.

Reddigan, K. (2007, November). *Ferryland District*. 1857 census Ferryland District. http://ngb.chebucto.org/C1857/1857-ferryland-district-census-1.shtml

Renberg, I., Brännvall, M.-L., Bindler, R., & Emteryd, O. (2000). Atmospheric Lead Pollution History during Four Millennia (2000 BC to 2000 AD) in Sweden. *AMBIO: A Journal of the Human Environment*, *29*(3), 150–156. https://doi.org/10.1579/0044-7447-29.3.150

Riesmeier, M., Keute, J., Veall, M.-A., Borschneck, D., Stevenson, A., Garnett, A., Williams, A., Ragan, & M., & Devièse, T. (2022). Recipes of Ancient Egyptian kohls more diverse than previously thought. *Scientific Reports*, *12*, e5932. https://doi.org/10.1038/s41598-022-08669-0

Rieuwerts, J.S., Farago, M.E., Cikrt, M., & Bencko, V. (2000). Differences in lead bioavailability between a smelting and a mining area. *Water, Air, & Soil Pollution*, *122*, 203-229. https://doi.org/10.1023/A:1005251527946

Robbins, N., Zhang, Z.-F., Sun, J., Kettererm, M.E., Lalumandier, J.A., & Shulze, R.A. (2010). Childhood lead exposure and uptake in teeth in the Cleveland area during the era of leaded gasoline. *Science of the Total Environment*, *408*, 4118-4127. https://doi.org/10.1016/j.scitotenv.2010.04.060

Rohl, B.M. (1996). Lead isotope data from the Isotrace laboratory, Oxford: Archaeometry data base 2, galena from Britain and Ireland. *Archaeometry*, *38*(1), 165-180. https://doi.org/10.1111/j.1475-4754.1996.tb00769.x

Rohl, B., & Needham, S. (1998). *The circulation of metal in the British Bronze Age: the application of lead isotope analysis*. British Museum.

Ryan, S. (1971). *The Newfoundland Cod Fishery in the Nineteenth century*. (Publication No. 593) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Samuelsen, J.R., & Potra, A. (2020). Biologically available Pb: A method for ancient human sourcing using Pb isotopes from prehistoric animal tooth enamel. *Journal of Archaeological Science*, *115*, e105079. https://doi.org/10.1016/j.jas.2020.105079

Scaffidi, B.K., Kamenov, G.D., Sharpe, A.E., & Krigbaum, J. (2021). Non-local enemies or local subjects of violence?: Using strontium ($^{87}Sr/^{86}Sr$) and Lead ($^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$) isobiographies to reconstruct geographic origins and early childhood mobility of decapitated male heads from the Majes Velley, Peru. *Journal of Archaeological Method and Theory*, *29*, 426-479. https://doi.org/10.1007/s10816-021-09519-5

Schaule, B.K., & Patterson, C.C. (1981). Pb concentrations in the Northeast Pacific: Evidence for global anthropogenic perturbations. *Earth and Planetary Science Letters*, *54*, 97-116. https://doi.org/10.1016/0012-821X(81)90072-8

Schroeder, H., Shuler, K.A., & Chenery, S.R. (2013). Childhood Pb exposure in an enslaved African community in Barbados: Implications for birthplace and health

status. *American Journal of Physical Anthropology*, *150*, 203-209. https://doi.org/10.1002/ajpa.22193

Schurr, M.R., Donohue, P.H., Simonetti, A., & Dawson, E.L. (2018). Multi-element and lead isotope characterization of early nineteenth century pottery sherds from Native American and Euro-American sites. *Journal of Archaeological Science: Reports*, *20*, 390–399. https://doi.org/10.1016/j.jasrep.2018.05.014

Scott, A.B., MacInnes, S., Hughes, N., Munkittrick, T.J.A., Harris, A.J.T., & Grimes, V. (2023). A Bioarchaeological Exploration of Adolescent Males at the Eighteenth-Century Fortress of Louisbourg, Nova Scotia, Canada. *Bioarchaeology International, Emerging Adolescence in Bioarchaeology: Current Status and Potential Development*, 7(2), 146–172. https://doi.org/10.5744/bi.2022.0007

Sharma, P., & Dubey, R.S. (2005). Lead toxicity in plants. *Brazilian Journal of Plant Physiology*, *17*, 35-52. https://doi.org/10.1590/S1677-04202005000100004

Sharp, Z.D., Atudorei, V., Panarello, H.O., Fernández, J., & Douthitt, C. (2003). Hydrogen isotope systematics of hair: archaeological and forensic applications. *Journal of Archaeological Science*, *30*, 1709-1716. https://doi.org/10.1016/S0305-4403(03)00071-2

Sharpe, A.E., Kamenov, G.D., Gilli, A., Hodell, D.A., Emery, K.F. Brenner, M., & Krigbaum, J. (2016). Lead (Pb) isotope baselines for studies of ancient human migration and trade in the Maya Region. *PLOS One*, *11*, e0164871. https://doi.org/10.1371/journal.pone.0164871

Sharpe, A.E., Kamenov, G.D., & Krigbaum, J. (2022). Assessing the applicability of lead isotope data from animal teeth in the Maya region. In J. Krigbaum, A.E. Sharpe (Eds.), *Isotope Research in Zooarchaeology* (pp. 149-174). University of Press Florida.

Shaw, H., Montgomery, J., Redfern, R., Gowland, R., & Evans, J. (2016). Identifying migrants in Roman London using lead and strontium. *Journal of Archaeological Science*, *66*, 57-68. https://doi.org/10.1016/j.jas.2015.12.001

Shotyk, W., Weiss, D., Heisterkamp, M., Cheburkin, A.K., Appleby, P.G., & Adams, F.C. (2002). New Peat Bog Record of Atmospheric Lead Pollution in Switzerland: Pb Concentrations, Enrichment Factors, Isotopic Composition, and Organolead Species.

Environmental Science and Technology, *36*, 3893–3900. https://doi.org/10.1021/es010196i

Simmons, L.M., Montgomery, J., Beaumont, J., Davis, G.R., & Al-Jawad, M. (2013). Mapping the spatial and temporal progression of human dental enamel biomineralization using synchrotron X-ray diffraction. *Archives of Oral Biology*, *58*, 1726-1734. https://doi.org/10.1016/j.archoralbio.2013.08.012

Simonetti, A., Buzon, M.R., Corcoran, L., Breidenstein, A.M., & Emberling, G. (2021). Trace element and Pb and Sr isotope investigation of tooth enamel from archaeological remains at El-Kurru, Sudan: Evaluating the role of groundwater-related diagenetic alteration. *Applied Geochemistry*, *132*, e105068. https://doi.org/10.1016/j.apgeochem.2021.105068

Simpson, R., Varney, T.L., Coulthard, I., Swanston, T., Grimes, V., Munkittrick, T.J.A., Jankauskas, R., & Cooper, D.M.L. (2021). Insights into biogenic and diagenetic lead exposure in experimentally altered modern and archaeological bone: Synchrotron radiation X-ray fluorescence imaging. *Science of the Total Environment*, *790*, e148144. https://doi.org/10.1016/j.scitotenv.2021.148144

Smith, P.E. (1987). Transhumant Europeans overseas: The Newfoundland case. *Current Anthropology*, *28*, 241–250. https://doi.org/10.1086/203526.

Smith, P.E. (1995). Transhumance among European Settlers in Atlantic Canada. *The Geographical Journal*, *161*, 79–86. https://doi.org/10.2307/3059930

Smith, T.M., & Tafforeau, P. (2008). New Visions of Dental Tissue Research: Tooth Development, Chemistry, and Structure. *Evolutionary Anthropology*, *17*, 213-226. https://doi.org/10.1002/evan.20176

Smith, T.M., Austin, C., Green, D.R., Joannes-Boyau, R., Bailey, S., Dumitriu, D., Fallon, S., Grün, R., James, H.F., Moncel, M.-H., Williams, I.S., Wood, R., & Arora, M. (2018). Wintertime stress, nursing, and lead exposure in Neanderthal children. *Science Advances*, *4*, eaau9483. https://doi.org/10.1126/sciadv.aau9483

Sofaer, J. (2011). Towards a social bioarchaeology of age. In S.C. Agarwal, B.A. Glencross (Eds.), *Social Bioarchaeology* (pp. 285-311). Wiley-Blackwell.

Sponheimer, M., & Lee-Thorp, J.A. (2006). Enamel diagenesis at South African Australopith Sites: Implications for paleoecological reconstruction with trace elements. *Geochimica et Cosmochimica Acta*, *70*, 1644-1654. https://doi.org/10.1016/j.gca.2005.12.022

Steele, M.J., Beck, B.D., Murphy, B.L., & Strauss, H.S. (1990). Assessing the contribution from lead in mining wastes to blood lead. *Regulatory Toxicology and Pharamology*, *11*, 158-190. https://doi.org/10.1016/0273-2300(90)90019-8

Stos-Gale, Z.A., & Gale, N.H. (2009). Metal provenancing using isotopes and the Oxford archaeological lead isotope database (OXALID). *Archaeological and Anthropological Sciences*, *1*, 195–213. https://doi.org/10.1007/s12520-009-0011-6

Swanston, T., Varney, T., Coulthard, I., Feng, R., Bewer, B., Murphy, R., Hennig, C., & Cooper, D. (2012) Element localization in archaeological bone using synchrotron radiation X-ray fluorescence: identification of biogenic uptake. *Journal of Archaeological Science*, *39*, 2409-2413. https://doi.org/10.1016/j.jas.2012.01.041

Swanston, T., Varney, T.L., Kozachuk, M., Choudhury, S., Bewer, B., Coulthard, I., Keenleyside, A., Nelson, A., Martin, R.R., Stenton, D.R., & Cooper, D.M.L. (2018). Franklin expedition lead exposure: New insights from high resolution confocal x-ray fluorescence imaging of skeletal microstructure. *PLOS One*, *8*, e0202983. https://doi.org/10.1371/journal.prone.0202983

Szostek, K., & Głąb, H. (2001). Trace element concentrations in human teeth from a Neolithic common grave at Nakonowo (Central Poland). *Variability and Evolution*, *9*, 51-59.

Tapsoba, I., Arbault, S., Walter, P., & Amatore, C. (2010). Finding out Egyptian gods secret using analytical chemistry: Biomedical properties of Egyptian black makeup revealed by amperometry at single cells. *Analytical Chemistry*, *82*, 457-460. https://doi.org/10.1021/ac902348g

Teutsch, N., Erel, Y., Halicz, L., & Banin, A. (2001). Distribution of natural and anthropogenic lead in Mediterranean soils. *Geochimica et Cosmochimica Acta*, *65*, 2853–2864. https://doi.org/10.1016/S0016-7037(01)00607-X

Thomas, G. (1989). Functions of the Newfoundland Outhouse. *Western Folklore*, *48*(3), 221-243.

Thornton, I., Davies, D.J.A., Watt, J.M., & Quinn, M.J. (1990). Pb Exposure in young children from dust and soil in the United Kingdom. *Environmental Health Perspectives*, *89*, 55-60. https://doi.org/10.1289/ehp.908955

Tite, M.S., Freestone, I., Mason, R., Molera, J., Vendrell-Saz, M., & Wood, N. (1998). Lead glazes in antiquity – methods of production and reasons for use. *Archaeometry*, *40*, 241-260. https://doi.org/10.1111/j.1475-4754.1998.tb00836.x

Tite, M.S. (2008). Ceramic production, provenance and use–A review. *Archaeometry*, *50*(2), 216-231. https://doi.org/10.1111/j.1475-4754.2008.00391.x

Tschetsch, L., Mussauer, A., Mauder, M., Lohrer, J., Kröger, P., Mayr, C., Söllner, F., & Grupe, G. (2020). Multi-isotope fingerprints (O, Sr, and Pb) in archaeological animal bone bioapatite: Similarity search and the sustainability for provenance analysis in a geologically complex Alpine region. *Archaeometry*, *62*, 35-52. https://doi.org/10.1111/arcm.12550

Tunstall, S., & Amarasiriwardena, D. (2002). Characterization of lead and lead leaching properties of lead glazed ceramics from the Solis Valley, Mexico, using inductively coupled plasma-mass spectrometry (ICP-MS) and diffuse reflectance infrared Fourier transform spectroscopy (DRIFT). *Microchemical Journal*, *73*(3), 335-347. https://doi.org/10.1016/S0026-265X(02)00100-5

Turgeon, L. (2000). Pêches basques du Labourd en Atlantique nord (XVIe–XVIIIe siècle): ports, routes et traffics. *Itsas Memoria: Revista de Estudios Maritimos del Pais asco*, 3, 163-178.

Turner, B.L. (2021). Residential mobility in the Inka sacred valley: Oxygen, strontium, and lead isotopic analysis at Patallaqta, Peru. *Journal of Archaeological Science: Reports*, *37*, e102930. https://doi.org/10.1016/j.jasrep.2021.102930

Turner, B.L., & Armelagos, G.J. (2012). Diet, residential origin, and pathology at Machu Picchu, Peru. *American Journal of Physical Anthropology*, *149*(1), 71-83. https://doi.org/10.1002/ajpa.22096

Turner, B.L., Kamenov, G.D., Kingston, J.D., & Armelagos, G.J. (2009). Insights into immigration and social class at Machu Picchu, Peru based on oxygen, strontium, and Pb isotopic analysis. *Journal of Archaeological Science*, *36*, 317-332. https://doi.org/10.1016/j.jas.2008.09.018

Valentine, B., Kamenov, G.D., & Krigbaum, J. (2008). Reconstructing Neolithic groups in Sarawak, Malaysia through Pb and strontium isotope analysis. *Journal of Archaeological Science*, *35*, 1463-1473. https://doi.org/10.1016/j.jas.2007.10.016

Valentine, B., Kamenov, G.D., Kenoyer, J.M., Shinde, V., Mushrif-Tripathy, V., Otarola-Castillo, E., & Krigbaum. J. (2015). Evidence for patterns of selective urban migration in the Greater Indus Valley (2600-1900 BC): A Pb and strontium isotope mortuary analysis. *PLOS One*, *10*, 1-20. https://doi.org/10.1371/journal.pone.0123103

Veizer, J., Buhl, D., Diener, A., Ebneth, S., Podlaha, O.G., Bruckschen, P., Jasper, T., Korte, C., Schaaf, M., Ala, D., & Karem, A. (1997). Strontium isotope stratigraphy: Potential resolution and event correlation. *Palaeogeography, Palaeoclimatology, Palaeoecology*, *132*(1-4), 65-77. https://doi.org/10.1016/S0031-0182(97)00054-0

Venovcevs, A., & Gaulton, B. (2018). Winter Housing: Archaeological Perspectives on Newfoundland's Non-Pastoral Transhumant Tradition. In E. Costello, E. & Svensson (Eds.), *Historical Archaeologies of Transhumance across Europe, European Journal of Archaeology Monograph* (pp. 43-57). Routledge.

Verbeeck, R.M.H., Lassuyt, C.J., Heijligers, H.J.M., Driessens, F.C.M., & Vrolijk, J.W.G.A. (1981). Lattice parameters and cation distribution of solid solutions of calcium and lead hydroxyapatite. *Calcified Tissue International*, *33*, 243-247. https://doi.org/10.1007/BF02409444

Waldron, H.A. (1981). Postmortem absorption of Pb by the skeleton. *American Journal of Physical Anthropology*, *55*, 395-398. https://doi.org/10.1002/ajpa.1330550313

Waldron, H.A. (1983). On the Post-Mortem Accumulation of Pb by Skeletal Tissues. *Journal of Archaeological Science*, *10*, 35-40. https://doi.org/10.1016/0305-4403(83)90124-3

Waldron, H.A., Mackie, A., & Townshend, A. (1976). The Pb content of some Romano-British bones. *Archaeometry*, *18*, 221-227. http://dx.doi.org/10.1111/j.1475-4754.1976.tb00165.x

Waldron, H.A., Khera, A., Walker, G., Wibberley, G., & Green, C.J.S. (1979). Lead concentrations in bones and soil. *Journal of Archaeological Science*, *6*, 295-298. https://doi.org/10.1016/0305-4403(79)90008-6

Wall, R. (1964). Louisbourg, 1745. *The New England Quarterly*, *37*(1), 64–83. https://doi.org/10.2307/365332.

Walser III, J.W., Kristjánsdóttir, S., Gröcke, D.R., Gowland, R.L., Jakob, T., Nowell, G.M., Ottley, C.J., & Montgomery, J. (2019). At the world's edge: Reconstructing diet and geographic origins in medieval Iceland using isotope and trace element analyses. *American Journal of Biological Anthropology*, *171*(1), 142-163. https://doi.org/10.1002/ajpa.23973

Wang, X., & Tang, Z. (2020). The first large-scale bioavailable Sr isotope map of China and its implication for provenance studies. *Earth-Science Reviews*, *210*, e103353. https://doi.org/10.1016/j.earscirev.2020.103353.

Warren, C. (2000). *Brush with death: A social history of lead poisoning*. The John Hopkins University Press.

Washburn, E., Nesbitt, J., Ibarra, B., Fehren-Schmitz, L., & Oelze, V.M. (2021). A strontium isoscape for the Conchucos region of highland Peru and its application to Andean archaeology. *PLOS One*, *16*(3), e0248209. https://doi.org/10.1371/journal.pone.0248209

Wasserstein, R.L., & Lazar, N.A. (2016). The ASA statement on *p*-Vales: Context, process, and purpose. *The American Statistician*, *70*(2), 129-133. https://doi.org/10.1080/00031305.2016.1154108

Wasserstein, R.L., Schirm, A.L., & Lazar, N.A. (2019). Moving to a world beyond "*p*<0.05". *The American Statistician*, *73*(sup1), 1-19. https://doi.org/10.1080/00031305.2019.1583913

Webb, E., Amarasiriwardena, D., Tauch, S., Green, E.F., Jones, J., & Goodman, A.H. (2005). Inductively coupled plasma-mass (ICP-MS) and atomic emission spectrometry

(ICP-AES): Versatile analytical techniques to identify the archived elemental information in human teeth. *Microchemical Journal*, *81*, 201-208. https://doi.org/10.1016/j.microc.2005.04.002

Wells A, Venn J, & Heard M. (1975). Deposition in the lung and uptake to blood of motor exhaust labelled with ^{203}Pb. *Inhaled Particles*, *4*, 175-189.

Whittaker, D.K., & Stack, M.V. (1984). The Pb, cadmium and zinc content of some Romano-British teeth. *Archaeometry*, *26*, 37-42. https://doi.org/10.1111/j.1475-4754.1984.tb00315.x

WHO (2010). *Childhood lead poisoning*. World Health Organization.

Willmes, M., Bataille, C.P., James, H.F., Moffat, I., McMorrow, L., Kinsley, L., Armstrong, R.A., Eggins, S., & Grün, R. (2018). Mapping of bioavailable strontium isotope ratios in France for archaeological provenance studies. *Applied Geochemistry*, *90*, 75-86. https://doi.org/10.1016/j.apgeochem.2017.12.025

Wittmers, L.E., Aufderheide, A.C., Pounds, J.G., Jones, K.W., & Angel, J.L. (2008). Problems in determination of skeletal lead burden in archaeological samples: an example from the First African Baptist Church population. *American Journal of Physical Anthropology*, *136*, 379–386. https://doi.org/10.1002/ajpa.20819

Zapata, J., Pérez-Sirvent, C., Martínez-Sánchez, M.J., & Tovar, P. (2006). Diagenesis, not biogenesis: Two late Roman skeletal examples. *Science of the Total Environment*, *369*(1-3), 357-368. https://doi.org/10.1016/j.scitotenv.2006.05.021

Zhang, Z.W., Subida, R.D., Agetano, M.G., Nakatsuka, H., Inoguchim, N., Wanatabe, T., Shimbo, S., Higashikawa, K., & Ikeda, M. (1998). Non-occupational exposure of adult women in Manila, the Philippines, to lead and cadmium. *Science of the Total Environment*, *215*, 157-165. https://doi.org/10.1016/s0048-9697(98)00118-1

Zhou, S., Yuan, Q., Li, W., Lu, Y., Zhang, Y., & Wang, W. (2014). Trace metals in atmospheric fine particles in one industrial urban city: Spatial variations, sources, and health implications. *Journal of Environmental Sciences*, *26*(1), 205-213. https://doi.org/10.1016/S1001-0742(13)60399-X

7 APPENDICES

7.1 Supplemental Materials Chapter 2

7.1.1 Literature review citations

The following papers were examined in the literature review.

Åberg, G., Fosse, G., & Stray, H. (1998). Man, nutrition, and mobility: A comparison of teeth and bone from the Medieval era and the present from Pb and Sr isotopes. *Science of the Total Environment*, *224*, 109-119. https://doi.org/10.1016/s0048-9697(98)00347-7

Aronsen, G.P., Fehren-Schmitz, L., Krigbaum, J., Kamenov, G.D., Conlogue, G.J., Warinner, C., Ozga, A.T., Sankaranarayanan, K., Griego, A., DeLuca, D.W., Eckels, H.T., Byczkiewicz, R.K., Grgurich, T., Pelletier, N.A., Brownlee, S.A., Marichal, A., Williamson, K., Tonoike, Y., & Bellantoni, N.F. (2019). "The dead shall be raised": Multidisciplinary analysis of human skeletons reveals complexity in 19th century immigrant socioeconomic history and identity in New Haven, Connecticut. *PLOS One*, *14*, e0219279. https://doi.org/10.1371/journal.pone.0219279

Beherec, M.A., Levy, T.E., Tirosh, O., Najjar, M., Knabb, K.A., & Erel, Y. (2016). Iron Age nomads and their relation to copper smelting in Fayan (Jordan): trace metal and Pb and Sr isotopic measurements form the Wadi Fidan 40 cemetery. *Journal of Archaeological Science*, *65*, 70-83. https://doi.org/10.1016/j.jas.2015.10.006

Bellis, D.J., Parsons, P.J., Jones, J., & Amarasiriwardena, D. (2009). Evaluation of Laser Ablation Inductively Coupled Plasma Mass Spectrometry for the Quantitative Determination of Lead in Different Parts of Archeological Human Teeth. *Spectroscopy Letters*, *42*, 491–496. https://doi.org/10.1080/00387010903267807

Budd, P., Montgomery, J., Cox, A., Krause, P., Barreiro, B., & Thomas, R.G. (1998). The distribution of lead within ancient and modern human teeth: implications for long-term

and historical exposure monitoring. *Science of the Total Environment*, *220*, 121-136. https://doi.org/10.1016/s0048-9697(98)00244-7

Budd, P., Montgomery, J., Evans, J., & Barreiro, B. (2000). Human tooth enamel as a record of comparative Pb exposure of prehistoric and modern people. *Science of the Total Environment*, *263*, 1-10. https://doi.org/10.1016/s0048-9697(00)00604-5

Budd, P., Montgomery, J., Evans, J., & Trickett, M. (2004). Human lead exposure in England from approximately 5500 bp to the 16th century ad. *Science of the Total Environment*, *318*, 45–58. https://doi.org/10.1016/S0048-9697(03)00357-7

Carvalho, M.L., Casaca, C., Pinheiro, T., Marques, J.P., Chevallier, P., & Cunha, A.S. (2000). Analysis of human teeth and bones from the chalcolithic period by X-ray spectrometry. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, *168*, 559–565. https://doi.org/10.1016/S0168-583X(00)00049-5

Carvalho, M.L., Marques, A.F., Marques, J.P., & Casaca, C. (2007). Evaluation of the diffusion of Mn, Fe, Ba and Pb in Middle Ages human teeth by synchrotron microprobe X-ray fluorescence. *Spectrochimica Acta Part B: Atomic Spectroscopy, A Collection of Papers Presented at the 18th International Congress on X-Ray Optics and Microanalysis (ICXOM 2005)*, *62*, 702–706. https://doi.org/10.1016/j.sab.2007.02.011

Chiaradia, M., Gallay, A., & Todt, W. (2003). Different contamination styles of prehistoric human teeth at a Swiss necropolis (Sion, Valais) inferred from lead and strontium isotopes. *Applied Geochemistry*, *18*, 353–370. https://doi.org/10.1016/S0883-2927(02)00072-0

de Winter, N.J., Snoeck, C., Schulting, R., Fernández-Crespo, T., & Claeys, P. (2019). High-resolution trace element distributions and models of trace element diffusion in enamel of Late Neolithic/Early Chalcolithic human molars from the Rioja Alavesa region (north-central Spain) help to separate biogenic from diagenetic trends. *Palaeogeography, Palaeoclimatology, Palaeoecology*, *532*, e109260. https://doi.org/10.1016/j.palaeo.2019.109260

Dudás, F.Ö., LeBlanc, S.A., Carter, S.W., & Bowring, S.A. (2016). Pb and Sr concentrations and isotopic compositions in prehistoric North American teeth: A Methodological Study. *Chemical Geology*, *429*, 21-32. https://doi.org/10.1016/j.chemgeo.2016.03.003

Ericson, J.E., Shirahata, H., & Patterson, C. (1979). Skeletal concentrations of lead in ancient Peruvians. *New England Journal of Medicine*, *300*, 946-951. https://doi.org/10.1056/NEJM197904263001703

Eshel, T., Yahalom-Mack, N., Tirosh, O., Maeir, A.M., & Harlavan, Y. (2020). Pollution and human mobility in the southern Levant during the Iron Age using chemical and isotopic analysis of human tooth enamel. *Journal of Archaeological Science*, *124*, e105262. https://doi.org/10.1016/j.jas.2020.105262

Evans, J., Pashley, V., Madgwick, R., Neil, S., & Chenery, C. (2018a). Tracking natural and anthropogenic lead exposure to its geological source. *Scientific Reports*, *8*, e1969. https://doi.org/10.1038/s41598-018-20397-y

Evans, J.A., Pashley, V., Chenery, C.A., Loe, L., & Chenery, S.R. (2018b). Lead isotope analysis of tooth enamel from a Viking age mass grave in southern Britain and the constraints in places on the origin of the individuals. *Archaeometry*, *60*, 859-869. https://doi.org/10.1111/arcm.12361

Farell, J., Amarasiriwardena, D., Goodman, A.H., & Arriaza, B. (2013). Bioimaging of trace metals in ancient Chilean mummies and contemporary Egyptian teeth by laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS). *Microchemical Journal*, *106*, 340-346. https://doi.org/10.1016/j.microc.2012.09.005

Fitch, A., Grauer, A., & Augustine, L. (2012). Lead isotope ratios: Tracking the migration of European-Americans to Grafton, Illinois in the 19th century. *International Journal of Osteoarchaeology*, *22*, 305–319. https://doi.org/10.1002/oa.1207

Gleń-Haduch, E., Szostek, K., & Głąb, H. (1997). Cribra orbitalia and trace element content in human teeth from neolithic and early bronze age graves in southern Poland. *American Journal of Physical Anthropology*, *103*(2), 201-207. https://doi.org/10.1002/(SICI)1096-8644(199706)103:2<201::AID-AJPA5>3.0.CO;2-W

Grandjean, P., & Jørgenson, P.J. (1990). Retention of lead and cadmium in prehistoric and modern human teeth. *Environmental Research*, *53*, 6-15. https://doi.org/10.1016/s0013-9351(05)80126-4

Guede, I., Zuluaga, M.C., Ortega, L.A., Alonso-Olazabal, A., Murelaga, X., Pina, M., & Gutierrez, F.J. (2017). Analyses of human dentine and tooth enamel by laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) to study the diet of medieval muslim individuals from Tauste (Spain). *Microchemical Journal*, *130*, 287-294. https://doi.org/10.1016/j.microc.2016.10.005

Jones, D.S., Turner, B.L., Buikstra, J.E., & Kamenov, G.D. (2017). Investigating the identities of isolated crania in the Lower Illinois River Valley through multi-isotopic analysis. *Journal of Archaeological Science: Reports*, *13*, 312-321. https://doi.org/10.1016/j.jasrep.2017.02.030

Keenleyside, A., Stenton, D.R., & Newman, K. (2021). The integration of isotopic and historical data to investigate the identification of crewmembers of the 1845 Franklin expedition. *Journal of Archaeological Science: Reports*, *40*, e103200. https://doi.org/10.1016/j.jasrep.2021.103200

Keller, A.T., Regan, L.A., Lundstrom, C.C., & Bower, N.W. (2016). Evaluation of the efficacy of spatiotemporal Pb isoscapes for provenancing of human remains. *Forensic Science International*, *261*, 83-92. https://doi.org/10.1016/j.forsciint.2016.02.006

King, C.L., Petchey, P., Buckley, H.R., Girvan, E., Kinaston, R., Nowell, G.M. (2020). Lead astray: the potentials and pitfalls of lead isotopes in a New Zealand colonial burial context. *Journal of Archaeological Science: Reports*, *30*, e102213. https://doi.org/10.1016/j.jasrep.2020.102213.

King, C.L., Buckley, H.R., Petchey, P., Roberts, P., Zech, J., Kinaston, R., Collins, C., Kardailsky, O., Matisoo-Smith, E., & Nowell, G. (2021). An isotopic and genetic study of multi-cultural colonial New Zealand. *Journal of Archaeological Science*, *128*, e105337. https://doi.org/10.1016/j.jas.2021.105337

Laffoon, J.E., Shuler, K.A., Millard, A.R., Connelly, J.N., & Schroeder, H. (2020). Isotopic evidence for anthropogenic lead exposure on a 17th/18th century Barbadian plantation.

American Journal of Physical Anthropology, *171*, 529–538. https://doi.org/10.1002/ajpa.23938

Lamb, A.L., Evans, J.E., Buckley, R., & Appleby, J. (2014). Multi-isotope analysis demonstrates significant lifestyle changes in King Richard III. *Journal of Archaeological Science*, *50*, 559-565. https://doi.org/10.1016/j.jas.2014.06.021

Millard, A., Montgomery, J., Trickett, M., Beaumont, J., Evans, J., & Chenery, S. (2014). Childhood lead exposure in the British Isles during the Industrial Revolution. In M.K. Zuckerman (Ed.), *Modern Environments and Human Health: Revisiting the Second Epidemiologic Transition* (pp. 279-299). John Wiley and Sons. https://doi.org/10.1002/9781118504338.ch15

Montgomery, J., Budd, P., & Evans, J. (2000). Reconstructing the lifetime movements of ancient people: A Neolithic case study from Southern England. *European Journal of Archaeology*, *3*, 407-422. https://doi.org/10.1179/146195700807860828

Montgomery, J., Evans, J., Chenery, S., Pashley, V., & Killgrove, K. (2010). "Gleaming, white and deadly": using lead to track human exposure and geographic origins in the Roman period in Britain. *Journal of Roman archaeology Supplements, 78*, 199–226.

Montgomery, J., Evans, J.A., Powlesland, D., & Roberts, C.A. (2005). Continuity or colonization in Anglo-Saxon England? Isotope evidence for mobility, subsistence practice, and status at West Heslerton. *American Journal of Physical Anthropology*, *126*, 123-138. https://doi.org/10.1002/ajpa.20111

Montgomery, J., Knüsel, C.J., & Tucker, K. (2011). Identifying the origins of decapitated male skeletons from 3 Driffield Terrace, York, through isotope analysis. In M. Bonogofsky (Ed.), *The Bioarchaeology of the Human Head, Decapitation, Decoration and Deformation* (pp.141-178). University Press of Florida.

Moore, J., Filipek, K., Kalenderian, V., Gowland, R., Hamilton, E., Evans, J., & Montgomery, J. (2021). Death metal: Evidence for the impact of lead poisoning on childhood health within the Roman Empire. *International Journal of Osteoloarchaeology*, *31*(5), 846-856. http://doi.org/10.1002/oa.3001

Moore, J., Rose, A., Anderson, S., Evans, J., Nowell, G., Gröcke, D.R., Pashley, V., Kirby, M., & Montgomery, J., (2020). A multi-isotope (C, N, O, Sr, Pb) study of Iron Age and

Roman period skeletons from east Edinburgh, Scotland exploring the relationship between decapitation burials and geographical origins. *Journal of Archaeological Science: Reports*, *29*, e102075. https://doi.org/10.1016/j.jasrep.2019.102075

Müller, W., Fricke, H., Halliday, A.N., McCulloch, M.T., & Wartho, J-A. (2003). Origin and migration of the alpine iceman. *Science*, *302*(5646), 862-866. https://doi.org/10.1126/science.1089837

Patterson, C., Ericson, J. Manea-Krichten, M., & Shirahata, H. (1991). Natural skeletal levels of lead in *Homo sapien sapiens* uncontaminated by technological lead. *Science of the Total Environment*, *107*, 205-236. https://doi.org/ 10.1016/0048-9697(91)90260-l

Porter, M. (1985). "She was the skipper of the shore-crew:" Notes on the history of the sexual division of labour in Newfoundland. *Labour/Le Travailleur*, *15*, 105-124.

Price, T.D., Frei, R., Bäckstrom, Y., Frei, K.M., & Ingvarsson-Sundstrom, A. (2017). Origins of inhabitants from the 16th century Sala (Sweden) silver mine cemetery – A lead isotope perspective. *Journal of Archaeological Science*, *80*, 1-13. https://doi.org/10.1016/j.jas.2017.01.013

Price, T.D., Frei, R., Brinker, U., Lidke, G., Terberger, T., Frei, K.M., & Jantzen, D. (2019). Multi-isotope proveniencing of human remains from a Bronze Age battlefield in the Tollense Valley in northeast Germany. *Archaeological and Anthropological Science*, *11*, 33-49. https://doi.org/10.1007/s12520-017-0529-y

Quinn, R.L., Warnasch, S.C., Watson, M., Godfrey, L., Setera, J.B., VanTongeren, J., Mortlock, R., & Wright, J. (2020). Biogeochemical evidence for residence, diet, and health of the Woman in the Iron Coffin (Queens, New York City). *International Journal of Osteoarchaeology*, *30*, 225-235. https://doi.org/10.1002/oa.2850

Samuelsen, J.R., & Potra, A. (2020). Biologically available Pb: A method for ancient human sourcing using Pb isotopes from prehistoric animal tooth enamel. *Journal of Archaeological Science*, *115*, e105079. https://doi.org/10.1016/j.jas.2020.105079

Scaffidi, B.K., Kamenov, G.D., Sharpe, A.E., & Krigbaum, J. (2022). Non-local enemies or local subjects of violence?: Using strontium ($^{87}Sr/^{86}Sr$) and Lead ($^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$, $^{208}Pb/^{204}Pb$) isobiographies to reconstruct geographic origins and early childhood mobility

of decapitated male heads from the Majes Velley, Peru. *Journal of Archaeological Method and Theory*, *29*, 426-479. https://doi.org/10.1007/s10816-021-09519-5

Schroeder, H., Shuler, K.A., & Chenery, S.R. (2013). Childhood Pb exposure in an enslaved African community in Barbados: Implications for birthplace and health status. *American Journal of Physical Anthropology*, *150*, 203-209. https://doi.org/10.1002/ajpa.22193

Shaw, H., Montgomery, J., Redfern, R., Gowland, R., & Evans, J. (2016). Identifying migrants in Roman London using lead and strontium. *Journal of Archaeological Science*, *66*, 57-68. https://doi.org/10.1016/j.jas.2015.12.001

Simonetti, A., Buzon, M.R., Corcoran, L., Breidenstein, A.M., & Emberling, G. (2021). Trace element and Pb and Sr isotope investigation of tooth enamel from archaeological remains at El-Kurru, Sudan: Evaluating the role of groundwater-related diagenetic alteration. *Applied Geochemistry*, *132*, e105068. https://doi.org/10.1016/j.apgeochem.2021.105068

Smits, E., Millard, A.R., Nowell, G., & Pearson, D.G. (2010). Isotopic Investigation of Diet and Residential Mobility in the Neolithic of the Lower Rhine Basin. *European Journal of Archaeology*, *13*, 5–31. https://doi.org/10.1177/1461957109355040

Szostek, K., & Głąb, H. (2001). Trace element concentrations in human teeth from a Neolithic common grave at Nakonowo (Central Poland). *Variability and Evolution*, *9*, 51-59.

Turner, B.L. (2021). Residential mobility in the Inka sacred valley: Oxygen, strontium, and lead isotopic analysis at Patallaqta, Peru. *Journal of Archaeological Science: Reports*, 37, e102930. https://doi.org/10.1016/j.jasrep.2021.102930

Turner, B.L., Kamenov, G.D., Kingston, J.D., & Armelagos, G.J. (2009). Insights into immigration and social class at Machu Picchu, Peru based on oxygen, strontium, and Pb isotopic analysis. *Journal of Archaeological Science*, *36*, 317-332. https://doi.org/10.1016/j.jas.2008.09.018

Turner, B.L., & Armelagos, G.J. (2012). Diet, residential origin, and pathology at Machu Picchu, Peru. *American Journal of Physical Anthropology*, *149*(1), 71-83. https://doi.org/10.1002/ajpa.22096

Valentine, B., Kamenov, G.D., & Krigbaum, J. (2008). Reconstructing Neolithic groups in Sarawak, Malaysia through Pb and strontium isotope analysis. *Journal of Archaeological Science*, 35, 1463-1473. https://doi.org/10.1016/j.jas.2007.10.016

Valentine, B., Kamenov, G.D., Kenoyer, J.M., Shinde, V., Mushrif-Tripathy, V., Otarola-Castillo, E., & Krigbaum. J. (2015). Evidence for patterns of selective urban migration in the Greater Indus Valley (2600-1900 BC): A Pb and strontium isotope mortuary analysis. *PLOS One*, *10*, 1-20. https://doi.org/10.1371/journal.pone.0123103

Walser III, J.W., Kristjánsdóttir, S., Gröcke, D.R., Gowland, R.L., Jakob, T., Nowell, G.M., Ottley, C.J., & Montgomery, J. (2020). At the world's edge: Reconstructing diet and geographic origins in medieval Iceland using isotope and trace element analyses. *American Journal of Physical Anthropology*, *171*, 142–163. https://doi.org/10.1002/ajpa.23973

Webb, E., Amarasiriwardena, D., Tauch, S., Green, E.F., Jones, J., & Goodman, A.H. (2005). Inductively coupled plasma-mass (ICP-MS) and atomic emission spectrometry (ICP-AES): Versatile analytical techniques to identify the archived elemental information in human teeth. *Microchemical Journal*, *81*, 201-208. https://doi.org/10.1016/j.microc.2005.04.002Whittaker, D.K., & Stack, M.V. (1984). The Pb, cadmium and zinc content of some Romano-British teeth. *Archaeometry*, *26*, 37-42. https://doi.org/10.1111/j.1475-4754.1984.tb00315.x

7.1.2 Literature review data

The following tables detail the information collected during the literature review in terms of sample preparation.

Table 8. Sample information extracted from papers for literature review (NS: not specified)

Author	Year	Pb Conc.	Pb Isotope ratio	Multiple teeth per indiv. sampled	Systematic tooth choice specified	Systematic for what teeth	Systematic for Age	Preserv. considered	Preserv. defined
Ericson et al.	**1979**	yes	no	no	NS	NA	no	NS	no
Whittaker and Stack	**1984**	yes	no	yes	NS	NA	no	NS	no
Grandjean and Jørgenson	**1990**	yes	no	no	NS	NA	no	yes	no
Patterson et al.	**1991**	yes	yes	no	yes	M2, M3	no	yes	no
Gleń-Haduch et al.	**1997**	yes	no	no	yes	M1, M2, deciduous m2	no	yes	yes
Åberg et al.	**1998**	yes	yes	no	NS	NA	no	NS	no
Budd et al.	**1998**	yes	no	no	yes	C or PM1	NA	yes	no
Budd et al.	**2000**	yes	yes	yes	NS	NA	yes	yes	yes
Carvalho et al.	**2000**	yes	no	no	broadly	molars	no	NS	no
Montgomery et al.	**2000**	yes	yes	yes	NS	NA	yes	yes	yes
Szostek and Głąb	**2001**	yes	no	yes	NS	NA	no	yes	no
Chiaradia et al.	**2003**	yes	yes	yes	yes	I2 or C	no	NS	no
Muller et al.	**2003**	yes	yes	yes	yes	PM2	yes	NS	no
Budd et al.	**2004**	yes	yes	no	yes	PM2	yes	yes	no
Montgomery et al.	**2005**	yes	yes	no	yes	PM2 or M2, and M3	no	yes	yes
Webb et al.	**2005**	yes	yes	no	NS	NA	yes	NS	no
Carvalho et al.	**2007**	yes	no	no	broadly	molars	no	yes	no
Valentine et al.	**2008**	no	yes	yes	yes	M1 or M2, M3	no	NS	no
Bellis et al.	**2009**	yes	no	no	NS	NA	no	NS	no
Turner et al.,	**2009**	no	yes	no	yes	M1	yes	NS	no
Montgomery et al.	**2010**	yes	yes	no	yes	PM2	no	yes	yes
Smits et al.	**2010**	no	yes	no	NS	NA	no	NS	no

Montgomery et al.	**2011**	yes	yes	no	yes	PM2	yes	yes	yes
Fitch et al.	**2012**	no	yes	no	broadly	PM unspecified	yes	yes	no
Turner et al.,	**2012**	no	yes	yes	yes	M1, M2, or PM1	no	NS	no
Farell et al.	**2013**	yes	no	no	NS	NA	no	NS	no
Schroeder et al.	**2013**	yes	no	no	yes	M1	yes	yes	yes
Lamb et al.	**2014**	yes	yes	yes	yes	PM2 and M2	yes	NS	no
Millard et al.	**2014**	yes	yes	yes	yes	PM2 or M2	yes	yes	no
Valentine et al.	**2015**	no	yes	yes	yes	M1, M2, and M3	yes	NS	no
Beherec et al.	**2016**	yes	yes	yes	NS	NA	no	NS	no
Dudás et al.	**2016**	yes	yes	no	NS	NA	no	NS	no
Keller et al.,	**2016**	yes	yes	no	yes	I, C, or M1	no	NS	no
Shaw et al.	**2016**	yes	yes	no	NS	NA	yes	yes	no
Guede et al.,	**2017**	yes	no	no	broadly	molars	no	NS	no
Jones et al.	**2017**	no	yes	no	yes	M1, M2, M3	yes	NS	no
Price et al.	**2017**	yes	yes	no	NS	NA	no	NS	no
Evans et al.	**2018a**	yes	yes	no	NS	NA	yes	NS	no
Evans et al.	**2018b**	yes	yes	no	NS	NA	no	yes	no
Aronsen et al.	**2019**	no	yes	no	NS	NA	no	NS	no
de Winter et al.	**2019**	yes	no	no	yes	M1 or M2	yes	NS	no
Laffoon et al.	**2020**	yes	yes	no	yes	M1	yes	NS	no
Price et al.	**2019**	yes	yes	no	yes	PM1 or PM2	no	NS	no
Walser et al	**2020**	yes	no	no	yes	PM1 or PM2	yes	NS	no
Eshel et al.	**2020**	yes	yes	yes	yes	M2 or M3	yes	NS	no
King et al.	**2020**	yes	yes	no	yes	M1, or deciduous m1 and m2	no	NS	no
Moore et al.	**2020**	yes	yes	no	yes	PM2, M2	no	NS	no

Quinn et al.	**2020**	yes	yes	no	yes	PM2 or M2	yes	NS	no
Samuelsen and Potra	**2020**	yes	yes	no	yes	M2	yes	NS	no
Keenleyside et al.	**2021**	no	yes	no	NS	M1 or M2	yes	NS	no
King et al.	**2021**	yes	yes	no	yes	M2	no	NS	no
Moore et al.	**2021**	yes	no	no	NS	NA	no	NS	no
Scaffidi et al	**2022**	no	yes	yes	yes	M1/I and PM/M2	yes	NS	no
Simonetti et al.	**2021**	yes	yes	no	NS	NA	no	yes	yes
Turner	**2021**	no	yes	no	yes	M1	yes	NS	no

Table 9. Details on sample preparation information extracted from papers as part of the literature review.

Author	**Year**	**Portion(s)**	**Leaching used**	**Abrasion**	**cleaning tools**	**cut/powder tools**
Ericson et al.	**1979**	enamel	yes – dilute HNO_3	yes – silicon carbide	silicon carbide tool	silicon carbide tool
Whittaker and Stack	**1984**	whole tooth	yes – 2% HNO_3	yes	NS	NS
Grandjean and Jørgenson	**1990**	circumpulpar dentine	no	NA	cite Grandjean et al., 1984	cite Grandjean et al., 1984
Patterson et al.	**1991**	enamel	yes	yes	unspecified dental burr	unspecified rotary dental saw
Gleń-Haduch et al.	**1997**	whole tooth	no	no	unspecified brush	agate mortar
Åberg et al.	**1998**	whole tooth	no	no	NS	NS
Budd et al.	**1998**	longitudinal cross-section: enamel, dentine	yes – H2O2 and acid wash	NA – LA	tungsten carbide dental drill	diamond-impregnated stainless steel cutting disc
Budd et al.	**2000**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Carvalho et al.	**2000**	longitudinal cross-section: enamel, dentine	no	no – not surface, just cut surface for LA	NA-sync XRF	NS

Montgomery et al.	**2000**	enamel, dentine	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Szostek and Głąb	**2001**	whole tooth	no	no	NA	NA
Chiaradia et al.	**2003**	enamel, dentine	yes	yes	tungsten-carbide chisel	diamond saw
Muller et al.	**2003**	enamel, and enamel-dentine composite	yes	no	NS	unspecified chisel
Budd et al.	**2004**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Montgomery et al.	**2005**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Webb et al.	**2005**	whole tooth	yes	no	acid-cleaned toothbrush	mortar and pestle
Carvalho et al.	**2007**	longitudinal cross-section: enamel, dentine	no	no – not surface, just cut surface for LA	NA-sync XRF	diamond saw
Valentine et al.	**2008**	enamel	yes	yes	scalpel or unspecified drill	fissure carbide bit
Bellis et al.	**2009**	longitudinal cross-section: enamel, dentine, circumpulpal dentin	NA	NA	NA-LA	Buehler isomet diamond-saw
Turner et al.,	**2009**	enamel	no	yes	tungsten carbide	NS
Montgomery et al.	**2010**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Smits et al.	**2010**	enamel	no	yes	tungsten carbide burr	flexible diamond impregnated cutting disc
Montgomery et al.	**2011**	enamel, dentine	no	yes	tungsten carbide dental drill	unspecified dental saw
Fitch et al.	**2012**	enamel	yes	no	NS	vibrating metal pick Cavitron-Parkell, or diamond dental saw
Turner et al.,	**2012**	enamel	no	NS	NS	NS
Farell et al.	**2013**	longitudinal cross-section: enamel, dentine, pulp	NA	NA	NA-LA	Buehler isomet steel-bladed saw

Schroeder et al.	**2013**	enamel	yes	no	NS	NS
Lamb et al.	**2014**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Millard et al.	**2014**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Valentine et al.	**2015**	enamel	yes	yes	diamond bit	agate mortar and pestle
Beherec et al.	**2016**	enamel, dentine	yes	unknown	NS – cite Ericson et al., 1979, Manea-Krichten et al., 1991, and Patterson et al., 1991	unspecified – cite Ericson et al., 1979, Manea-Krichten et al., 1991, and Patterson et al., 1991
Dudás et al.	**2016**	enamel	yes	no	NA	diamond saw , abrasive wheel, carbide or diamond-tipped burs
Keller et al.,	**2016**	enamel	yes	yes	carbide burr dental drill	NS
Shaw et al.	**2016**	enamel	no	yes	tungsten carbide burr	flexible diamond impregnated cutting disc
Guede et al.,	**2017**	longitudinal cross-section: enamel, dentine	NA	NA	NA-LA	diamond disc
Jones et al.	**2017**	enamel	no	yes	tungsten carbide dental drill	NS
Price et al.	**2017**	enamel	yes	no	NS	NS
Evans et al.	**2018 a**	enamel, dentine	yes – 5min dilute HCl and HNO_3	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Evans et al.	**2018 b**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Aronsen et al.	**2019**	enamel	yes	yes	diamond tipped dental bit	diamond tipped dental bit
de Winter et al.	**2019**	longitudinal cross-section: enamel, dentine, pulp cavity	no	yes	NA-XRF	Buehler isomet diamond-saw
Laffoon et al.	**2020**	enamel	no	NS	NS	NS

Price et al.	**2019**	enamel	yes	no	carbide burr dental bit	unspecified dental drill circular saw
Walser et al	**2020**	enamel	no	yes	Montgomery 2002, NS	Montgomery 2002, NS
Eshel et al.	**2020**	enamel	yes	yes	NS	NS
King et al.	**2020**	enamel	no	yes	unspecified dental burr	NS
Moore et al.	**2020**	enamel	no	yes	tungsten carbide dental drill	NS
Quinn et al.	**2020**	enamel	no	yes	unspecified bit with foredom rotary tool	NA
Samuelsen and Potra	**2020**	enamel	yes	yes	unspecified drill bit	diamond wheel bit
Keenleyside et al.	**2021**	enamel	no	yes	diamond tip dental burr	NS
King et al.	**2021**	enamel	no	yes	diamond tip dental burr	diamond cutting blade dremel
Moore et al.	**2021**	enamel	no	yes	tungsten carbide dental drill	flexible diamond edged cutting disc
Scaffidi et al	**2022**	enamel	no	yes	diamond tip dental drill	NS
Simonetti et al.	**2021**	enamel	no	yes	NS	NS
Turner	**2021**	enamel	yes	yes	NS	agate mortar and pestle

Table 10. Details on concentration method information extracted from papers for literature review (NS = Not specified)

Author	Year	Concentration instrumentation	Concentration method	Procedural blank	error (%)	Concentration method reproducible with info given
Ericson et al.	**1979**	ID-TIMS	post column with 208Pb tracer	yes	yes	partially
Whittaker and Stack	**1984**	AAS	bulk digest solution	NS	NS	no
Grandjean and Jørgenson	**1990**	AAS	bulk digest solution	NS	yes	partially
Patterson et al.	**1991**	ID-TIMS	bulk digest solution	yes	yes	no
Gleń-Haduch et al.	**1997**	ASV	bulk digest solution	NS	NS	cite Karai et al., 1980 and Wang 1985
Åberg et al.	**1998**	FAAS	bulk digest solution	NS	yes	partially
Budd et al.	**1998**	LA-ICP-MS & ID-TIMS	laser ablation & bulk digest solution	yes	yes	yes
Budd et al.	**2000**	ID-TIMS	bulk digest solution	yes	yes	partially
Carvalho et al.	**2000**	Synchrotron microprobe XRFS	XRF	NA – XRF	yes	yes
Montgomery et al.	**2000**	ID-TIMS	bulk digest solution	yes	NS	partially
Szostek and Głąb	**2001**	AAS	bulk digest solution	NS	NS	no
Chiaradia et al.	**2003**	ID-TIMS	post column solution	yes	yes	partially
Muller et al.	**2003**	ID-TIMS	bulk digest solution	yes	yes	yes
Budd et al.	**2004**	ID-TIMS	bulk digest solution	yes	yes	partially
Montgomery et al.	**2005**	ID-TIMS	bulk digest solution	NS	NS	partially
Webb et al.	**2005**	Quadrupole ICP-MS	bulk digest solution	NS	NS	yes
Carvalho et al.	**2007**	Synchrotron microprobe XRFS	XRF	NA – XRF	yes	yes
Valentine et al.	**2008**	NA	NA	NA	NA	NA

Bellis et al.	**2009**	LA-ICP-MS	laser ablation	NA – laser	NS	yes
Turner et al.,	**2009**	NA	NA	NA	NA	NA
Montgomery et al.	**2010**	Quadrupole ICP-MS	bulk digest solution	NS	NS	yes
Smits et al.	**2010**	NA	NA	NA	NA	NA
Montgomery et al.	**2011**	NS	NS	yes	NS	**no**
Fitch et al.	**2012**	NA	NA	NA	NA	NA
Turner et al.,	**2012**	NA	NA	NA	NA	NA
Farell et al.	**2013**	LA-ICP-MS	laser ablation	NA – laser	NS	yes
Schroeder et al.	**2013**	Quadrupole ICP-MS	bulk digest solution	NS	yes	partially
Lamb et al.	**2014**	Quadrupole ICP-MS	bulk digest solution	yes	yes	partially
Millard et al.	**2014**	Quadrupole ICP-MS	bulk digest solution	NS	NS	yes -cite Montgomery et al., 2010
Valentine et al.	**2015**	NA	NA	NA	NA	NA
Beherec et al.	**2016**	Quadrupole ICP-MS	bulk digest solution	NS	yes	yes
Dudás et al.	**2016**	ID-TIMS	bulk digest solution	NS	yes	yes
Keller et al.,	**2016**	NS	leaching wash solution	NS	NS	no
Shaw et al.	**2016**	NS	bulk digest solution	NS	NS	**no**
Guede et al.,	**2017**	LA-ICP-MS	laser ablation	NA – laser	NS	yes
Jones et al.	**2017**	NA	NA	NA	NA	NA
Price et al.	**2017**	ID-TIMS	bulk digest solution	yes	yes	partially
Evans et al.	**2018a**	Quadrupole ICP-MS	bulk digest solution	NS	NS	partially
Evans et al.	**2018b**	Quadrupole ICP-MS	bulk digest solution	NS	yes	partially
Aronsen et al.	**2019**	NA	NA	NA	NA	NA
de Winter et al.	**2019**	micro XRF	XRF	NA – XRF	yes	yes
Laffoon et al.	**2020**	ID-TIMS	post column solution	yes	NS	yes

Price et al.	**2019**	ID-TIMS	bulk digest solution	yes	NS	partially
Walser et al	**2020**	Quadrupole ICP-MS	bulk digest solution	NS	yes	no
Eshel et al.	**2020**	Quadrupole ICP-MS	bulk digest solution	yes	yes	yes
King et al.	**2020**	MC-CIP-MS	bulk digest solution	NS	NS	partially
Moore et al.	**2020**	Quadrupole ICP-MS	bulk digest solution	NS	yes	yes
Quinn et al.	**2020**	LA-ICP-MS	laser ablation	NA – laser	NS	partially
Samuelsen and Potra	**2020**	MC-ICP-MS	post column solution	yes	NS	partially
Keenleyside et al.	**2021**	NA	NA	NA	NA	NA
King et al.	**2021**	NS	post column solution	NS	NS	**no**
Moore et al.	**2021**	Quadrupole ICP-MS	bulk digest solution	NS	NS	yes
Scaffidi et al	**2022**	NA	NA	NA	NA	NA
Simonetti et al.	**2021**	HR-ICP-MS	bulk digest solution	NS	NS	yes
Turner	**2021**	NA	NA	NA	NA	NA

Table 11. Concentration interpretation information extracted from literature review.

Author	**Year**	**Concentration examined**	**Comparison material**	**other**
Ericson et al.	**1979**	change over time	modern	no
Whittaker and Stack	**1984**	change over time, general pattern	both	compare arch bone of individual
Grandjean and Jørgenson	**1990**	change over time, general pattern	both	no
Patterson et al.	**1991**	change over time, general pattern, method, diagenesis	modern	no
Gleń-Haduch et al.	**1997**	health, cribria orbitalia vs Pb conc	none	cribra orbitalia vs Pb
Åberg et al.	**1998**	Geographic differences	both	no

Budd et al.	**1998**	method	none	no interpretation – focus on LA pattern
Budd et al.	**2000**	change over time, method	both	no
Carvalho et al.	**2000**	general pattern, method	modern	no
Montgomery et al.	**2000**	general pattern	modern	no
Szostek and Głąb	**2001**	"biological status"	none	no interpretation
Chiaradia et al.	**2003**	diagenesis	modern	no
Muller et al.	**2003**	life history/profile	none	no interpretation
Budd et al.	**2004**	change over time, method	modern	no
Montgomery et al.	**2005**	change over time, general pattern	archaeological	no
Webb et al.	**2005**	change over time, general pattern, geographic differences	archaeological	no
Carvalho et al.	**2007**	general pattern, method, diagenesis	modern	post mortem uptake at the surface
Valentine et al.	**2008**	NA	NA	NA
Bellis et al.	**2009**	method	both	no
Turner et al.,	**2009**	NA	NA	NA
Montgomery et al.	**2010**	change over time, general pattern, Atmospheric comparison and identification of immigrants	archaeological	no
Smits et al.	**2010**	NA	NA	NA
Montgomery et al.	**2011**	general pattern	both	no
Fitch et al.	**2012**	NA	NA	NA
Turner et al.,	**2012**	NA	NA	NA
Farell et al.	**2013**	method	modern	no
Schroeder et al.	**2013**	health, identify migrants	both	no
Lamb et al.	**2014**	life history	archaeological	no
Millard et al.	**2014**	change over time, general pattern, natural vs anthropogenic cut off	both	compare to cribra orbitalia
Valentine et al.	**2015**	NA	NA	NA
Beherec et al.	**2016**	diagenesis, identify nomads and smelters	archaeological	compare to archaeological bone from the region

Dudás et al.	**2016**	method, diagenesis	both	no
Keller et al.,	**2016**	general pattern	none	no interpretation
Shaw et al.	**2016**	general pattern	archaeological	no
Guede et al.,	**2017**	general pattern, method	none	no
Jones et al.	**2017**	NA	NA	NA
Price et al.	**2017**	general pattern	both	no
Evans et al.	**2018a**	general pattern, natural vs anthropogenic exposure	archaeological	no
Evans et al.	**2018b**	general pattern	archaeological	no
Aronsen et al.	**2019**	NA	NA	NA
de Winter et al.	**2019**	method, diagenesis	none	Pattern of the trace elements compared to a diffusion-advection model
Laffoon et al.	**2020**	general pattern	archaeological	no
Price et al.	**2019**	general pattern	archaeological	no
Walser et al	**2020**	general pattern, social status, migration	archaeological	no
Eshel et al.	**2020**	change over time	archaeological	no
King et al.	**2020**	general pattern, diagenesis	both	no
Moore et al.	**2020**	general pattern, identify anthropogenic exposure	archaeological	no
Quinn et al.	**2020**	life history	archaeological	Grobler et al., 2000 equation
Samuelsen and Potra	**2020**	general pattern, method, diagenesis	none	compare within the site
Keenleyside et al.	**2021**	NA	NA	NA
King et al.	**2021**	general pattern	archaeological	no
Moore et al.	**2021**	health	archaeological	Grobler et al., 2000 equation
Scaffidi et al	**2022**	NA	NA	NA
Simonetti et al.	**2021**	diagenesis	none	U/Pb and Pb Enrichment factor
Turner	**2021**	NA	NA	NA

Table 12. Information on isotopic methods extracted as part of literature review (part 1)

Author	Year	Isotopic instrument	Bracket vs Tl	Calibration citation or values	Blank specified in (pg) or (%)	Accuracy (% error) given	Repeatability (sd, se, other variability) given?	If no %error enough info. to back calculate?	Error given for individual samples
Ericson et al.	**1979**	NA	**NA**	NA	NA	NA	NA	NA	NA
Whittaker and Stack	**1984**	NA	**NA**	NA	NA	NA	NA	NA	NA
Grandjean and Jørgenson	**1990**	NA	**NA**	NA	NA	NA	NA	NA	NA
Patterson et al.	**1991**	TIMS	NS	NS	yes	yes	no	no	yes
Gleń-Haduch et al.	**1997**	NA	**NA**	NA	NA	NA	NA	NA	NA
Åberg et al.	**1998**	TIMS	**bracket**	NS	yes	yes	no	no	no
Budd et al.	**1998**	NA	**NA**	NA	NA	NA	NA	NA	NA
Budd et al.	**2000**	PIMMS	NS	NS	NS	yes	no	no	yes
Carvalho et al.	**2000**	NA	**NA**	NA	NA	NA	NA	NA	NA
Montgomery et al.	**2000**	PIMMS	**Tl**	Ketterer et al. 1991; Walder et al. 1993	NS	yes	no	no	yes
Szostek and Głąb	**2001**	NA	**NA**	NA	NA	NA	NA	NA	NA
Chiaradia et al.	**2003**	TIMS	NS	NS	yes	no	no	no	no
Muller et al.	**2003**	TIMS with Secondary Electron Multiplier	**Tl**	NS	yes	yes	no	no	yes
Budd et al.	**2004**	TIMS, PIMMS	NS	NS	yes	yes	no	no	yes
Montgomery et al.	**2005**	TIMS	**bracket**	NS	NS	yes	no	no	yes
Webb et al.	**2005**	Quadrupole ICP-MS	NS	no	NS	no	no	no	no
Carvalho et al.	**2007**	NA	**NA**	NA	NA	NA	NA	NA	NA

Valentine et al.	**2008**	MC-ICP-MS	**Tl**	values given	NS	no	no	no	no
Bellis et al.	**2009**	NA	**NA**	NA	NA	NA	NA	NA	NA
Turner et al.,	**2009**	MC-ICP-MS	**Tl**	Kamenov et al. 2004	NS	no	yes	yes	no
Montgomery et al.	**2010**	TIMS, PIMMS	**Tl**	Thirwall 2002	NS	yes	no	no	no
Smits et al.	**2010**	MC-ICP-MS	NS	values given	yes	no	yes	yes	no
Montgomery et al.	**2011**	TIMS	**bracket**	NS	NS	yes	no	no	yes
Fitch et al.	**2012**	HR-ICP-MS	NS	NS	yes	yes	no	no	no
Turner et al.	**2012**	MC-ICP-MS	**Tl**	Kamenov et al. 2004	NS	no	yes	yes	no
Farell et al.	**2013**	NA	**NA**	NA	NA	NA	NA	NA	NA
Schroeder et al.	**2013**	NA	**NA**	NA	NA	NA	NA	NA	NA
Lamb et al.	**2014**	MC-ICP-MS	**Tl**	Thirwall 2002	NS	yes	yes	NA	yes
Millard et al.	**2014**	MC-ICP-MS	**Tl**	Thirwall 2002	yes	yes	yes	NA	yes
Valentine et al.	**2015**	MC-ICP-MS	**Tl**	values given	NS	no	no	no	yes
Beherec et al.	**2016**	MC-ICP-MS	NS	not specified	NS	no	yes	yes	no
Dudás et al.	**2016**	TIMS	**Tl**	Baker et al. 2004	yes	yes	no	no	no
Keller et al.,	**2016**	MC-ICP-MS	**Tl**	NS	NS	no	yes	no	yes
Shaw et al.	**2016**	MC-ICP-MS	**Tl**	Thirwall 2002	yes	no	yes	no	yes
Guede et al.,	**2017**	NA	**NA**	NA	NA	NA	NA	NA	NA
Jones et al.	**2017**	MC-ICP-MS	**Tl**	Kamenov et al. 2004	NS	no	yes	yes	yes
Price et al.	**2017**	TIMS	NS	Todt et al. 1993	yes	yes	no	no	yes
Evans et al.	**2018a**	TIMS, MC-ICP-MS	**Tl**	Thirwall 2002	yes	yes	no	no	no
Evans et al.	**2018b**	MC-ICP-MS	**Tl**	Thirwall 2002	yes	yes	no	no	no
Aronsen et al.	**2019**	MC-ICP-MS	**Tl**	Kamenov et al. 2004	NS	no	no	no	no
de Winter et al.	**2019**	NA	**NA**	NA	NA	NA	NA	NA	NA

Laffoon et al.	**2020**	TIMS	NS	Todt et al. 1993	NS	no	no	no	no
Price et al.	**2019**	TIMS	**Tl**	NS	yes	no	yes	yes	no
Walser et al	**2020**	NA	**Tl**	Baker et al. 2004	NA	no	yes	yes	NA
Eshel et al.	**2020**	MC-ICP-MS	NS	NS	yes	no	no	no	yes
King et al.	**2020**	MC-ICP-MS	**Tl**	Thirwall 2002	yes	no	yes	yes	no
Moore et al.	**2020**	MC-ICP-MS	NS	NS	yes	yes	no	no	yes
Quinn et al.	**2020**	MC-ICP-MS	**Tl**	NS	NS	no	yes	yes	no
Samuelsen and Potra	**2020**	MC-ICP-MS	**NA**	NA	yes	NA	NA	NA	yes
Keenleyside et al.	**2021**	MC-ICP-MS	**Tl**	Taylor et al. 2014	yes	no	yes	yes	yes
King et al.	**2021**	MC-ICP-MS	**Tl**	Baker et al. 2004	NS	no	yes	yes	yes
Moore et al.	**2021**	NA	**NA**	NA	NA	NA	NA	NA	NA
Scaffidi et al	**2022**	MC-ICP-MS	NS	NS	NS	no	yes	yes	no
Simonetti et al.	**2021**	MC-ICP-MS	**Tl**	Baker et al. 2004	NS	no	yes	yes	no
Turner	**2021**	MC-ICP-MS	NS	NS	NS	no	yes	no	no

Table 13. Information on isotopic methods extracted as part of literature review (part 2)

Author	**Year**	**Pb resin**	**Pb elution methodology**	**Pretreatment citation**	**Pre-treatment method reproducible with info given – Pb elution etc**	**Iso analysis reproducible with info given**
Ericson et al.	**1979**	NA	NA	NA	NA	NA
Whittaker and Stack	**1984**	NA	NA	NA	NA	NA
Grandjean and Jørgenson	**1990**	NA	NA	NA	NA	NA

Patterson et al.	**1991**	Unspec. anion resin column	in citation – full wash, and elution given (no cleaning info)	Manea-Krichten et al., 1991	partially	no
Gleń-Haduch et al.	**1997**	NA	NA	NA	NA	NA
Åberg et al.	**1998**	Unspec. crown ether resin	not specified	Horowitz et al., 1992	no	partially
Budd et al.	**1998**	NA	NA	NA	NA	NA
Budd et al.	**2000**	NS	unspecified"conventional anion exchange column chemistry"	none given	no	partially
Carvalho et al.	**2000**	NA	NA	NA	NA	NA
Montgomery et al.	**2000**	NS	"conventional anion exchange methods"	none given	no	partially
Szostek and Głąb	**2001**	NA	NA	NA	NA	NA
Chiaradia et al.	**2003**	NS	HBr ion exchange chromatography	none given	partially	partially
Muller et al.	**2003**	Srspec and Prefilter resin	elution agents given – citations with conflicting methods	Horwitz et al., 1992; Vajda et al., 1997; Theriault and Davis 2000	partially	yes
Budd et al.	**2004**	NS	unspecified"conventional anion exchange column chemistry"	none given	no	partially
Montgomery et al.	**2005**	Dowex 1x8 200-400 mesh	not specified	none given	no	partially
Webb et al.	**2005**	none	none	none given	no	yes
Carvalho et al.	**2007**	NA	NA	NA	NA	NA
Valentine et al.	**2008**	Dowex 1X-8 resin	conventional hydrobromic acid procedure	none given	partially	yes
Bellis et al.	**2009**	NA	NA	NA	NA	NA
Turner et al.,	**2009**	Dowex 1X-8 100-200 mesh	full clean, wash, and elution given	none given	yes	yes
Montgomery et al.	**2010**	NS	"conventional anion exchange methods"	Dicken 1995 – no access	no	yes

Smits et al.	**2010**	SrSpec resin – crown ether basex exchange chromatography medium	not specified	none given	no	no
Montgomery et al.	**2011**	NS	not specified	none given	no	no
Fitch et al.	**2012**	AG1-X8 200-400mesh resin	full clean, wash, and elution given	none given	yes	no
Turner et al.,	**2012**	in citation – Dowex 1X-2 100-200 mesh	in citation – full clean, wash, and elution given	Turner et al., 2009 and 2010	yes	yes
Farell et al.	**2013**	NA	NA	NA	NA	NA
Schroeder et al.	**2013**	NA	NA	NA	NA	NA
Lamb et al.	**2014**	NS	not specified	none given	no	yes
Millard et al.	**2014**	Dowex 1x8 200-400 mesh	in citation – full clean, wash, and elution given	Tricket 2006	yes	yes
Valentine et al.	**2015**	in citation – Dowex 1X-8	in citation – conventional HBr	Valentine et al., 2008 (mistake says 2007)	partial	yes
Beherec et al.	**2016**	Dowex 1x-8	unspecified- citation doesn't state	Erel et al., 2006	no	yes cite Erel et al., 2006
Dudás et al.	**2016**	AG-1 resin (EiChrom)	HBr acid processed twice on columns	none given	partial	yes
Keller et al.,	**2016**	unspecified Pb resin	full clean, wash, and elution given	none given	yes	partially
Shaw et al.	**2016**	AG 1X8 anion exchange	Separated out using of anion exchange resin"	none given	partially	partially
Guede et al.,	**2017**	NA	NA	NA	NA	NA
Jones et al.	**2017**	Dowex 1X-8 mesh	full clean, wash, and elution given	none given	yes	partially
Price et al.	**2017**	AG-1 resin (BioRadTM)	not specified	missing (modified of an unspecified article)	no	partially
Evans et al.	**2018a**	AG-1 resin (EiChrom)	HBr acid ion exchange	none given	partially	yes
Evans et al.	**2018b**	NS	"standard ion exchange techniques"	none given	no	yes

Aronsen et al.	**2019**	Dowex 1x-8	HBr procedure – no further context in citation	Valentine et al., 2008	partially	yes
de Winter et al.	**2019**	NA	NA	NA	NA	NA
Laffoon et al.	**2020**	unspecified Eichron anion resin – og cit AG-1 x8	in citation – only some loading procedure given, they cite another which just says HBR-HNO_3 mixtures	Connelly & Bizzarro 2009 – cite Lugmair and Galer 1992	partially	yes
Price et al.	**2019**	AG-1 resin (BioRadTM)	in citation – partial elution information	modified Kalsbeek and Frei 2006	no	partially
Walser et al	**2020**	NA	NA	NA	NA	NA
Eshel et al.	**2020**	Dowex 1x-8	unspecified- citation doesn't state	Erel and Torrent 2010	partially	partially
King et al.	**2020**	SrSpec resin	in citation – partial clean, wash, and elution given	Font et al., 2008 – modified Charlier et al., 2006	yes	yes
Moore et al.	**2020**	AG-1 resin (EiChrom)	HBr acid ion exchange	Evans et al., 2018 (archaeometry)	partially	yes
Quinn et al.	**2020**	NS	not specified	not specified	no	no
Samuelsen and Potra	**2020**	Dowex 1x-8 Pb resin	full clean, wash, and elution given	none given	yes	partially
Keenleyside et al.	**2021**	in citation – Srspec resin	in citation – full clean, wash, and elution given	Weiss et al., 2004 modified	yes	partially
King et al.	**2021**	SrSpec resin	in citation – partial clean, wash, and elution given	Font et al., 2008 – modified Charlier et al., 2006	yes	yes
Moore et al.	**2021**	NA	NA	NA	NA	NA
Scaffidi et al	**2022**	Dowex 1X-8	wash and elution given	Valentine et al., 2008	partially	yes
Simonetti et al.	**2021**	AG 1X8 resin 75-150 mesh	full clean, wash, and elution given	Manhes et al., 1980 and Koeman et al., 2015	yes	yes
Turner	**2021**	Dowex Pb resin	full clean, wash, and elution given	no	yes	yes

Table 14. Information on isotopic interpretation extracted as part of the literature review

Author	Year	Isotopic interpretation		Isotopic conclusions		
		Exposure source assumed (natural, anthropogenic, both)	Natural determination questionable	Identify non-local/movement	Diagenesis hinders interpretation	Other
Ericson et al.	**1979**	NA	NA	NA	NA	NA
Whittaker and Stack	**1984**	NA	NA	NA	NA	NA
Grandjean and Jørgenson	**1990**	NA	NA	NA	NA	NA
Patterson et al.	**1991**	natural	yes – Pb glazed ceramics	no	yes	no
Gleń-Haduch et al.	**1997**	NA	NA	NA	NA	NA
Åberg et al.	**1998**	anthro	NA	not the purpose	no	different local sources medieval to modern
Budd et al.	**1998**	NA	NA	NA	NA	NA
Budd et al.	**2000**	natural	no	yes	no	no
Carvalho et al.	**2000**	NA	NA	NA	NA	NA
Montgomery et al.	**2000**	natural	no	yes	no	no
Szostek and Głąb	**2001**	NA	NA	NA	NA	NA
Chiaradia et al.	**2003**	both	no	yes	yes	no
Muller et al.	**2003**	natural	no	yes	no	no
Budd et al.	**2004**	both	no	no	no	metallurgic focusing
Montgomery et al.	**2005**	both	no	yes	no	no
Webb et al.	**2005**	both	NA	not the purpose	no	Compare sites, overlap between regions
Carvalho et al.	**2007**	NA	NA	NA	NA	NA

Valentine et al.	**2008**	natural	no	yes	no	no
Bellis et al.	**2009**	NA	NA	NA	NA	NA
Turner et al.,	**2009**	natural	yes	yes	no	no
Montgomery et al.	**2010**	both	no	yes	no	no
Smits et al.	**2010**	natural	no	yes	no	no
Montgomery et al.	**2011**	both	no	yes	no	no
Fitch et al.	**2012**	anthro	NA	yes	no	no
Turner et al.,	**2012**	natural	yes	yes	no	no
Farell et al.	**2013**	NA	NA	NA	NA	NA
Schroeder et al.	**2013**	NA	NA	NA	NA	NA
Lamb et al.	**2014**	anthro	NA	no	no	no
Millard et al.	**2014**	anthro	NA	no	no	not from coal – particles too large
Valentine et al.	**2015**	natural	no	yes	no	no
Beherec et al.	**2016**	both	no	yes – using Sr	yes	Polluted individuals identified
Dudás et al.	**2016**	natural	yes but question in the discussion	yes	unclear	Use both Sr and Pb iso
Keller et al.,	**2016**	anthro	NA	yes	no	isoscape using atmospheric
Shaw et al.	**2016**	anthro	NA	yes	no	no
Guede et al.,	**2017**	NA	NA	NA	NA	NA
Jones et al.	**2017**	natural	no	no	no	no
Price et al.	**2017**	anthro	NA	yes	no	no
Evans et al.	**2018a**	both	no	yes	no	natural vs anthropogenic separated using 238U/204Pb
Evans et al.	**2018b**	natural	no	yes – not sole purpose	no	natural vs anthropogenic

						separated using 238U/204Pb
Aronsen et al.	**2019**	anthro	NA	yes	no	no
de Winter et al.	**2019**	NA	NA	NA	NA	NA
Laffoon et al.	**2020**	both	Yes, but question in the discussion	yes	Yes – the low concentrations are at higher risk	Low concentration at high risk for contamination
Price et al.	**2019**	natural	no	yes	no	no
Walser et al	**2020**	NA	NA	NA	NA	NA
Eshel et al.	**2020**	both	no	yes	yes – some	no
King et al.	**2020**	anthro	NA	no	yes	no
Moore et al.	**2020**	both	no	yes	no	no
Quinn et al.	**2020**	anthro	NA	no	no	no
Samuelsen and Potra	**2020**	natural	unclear – high conc in one	yes	no	local baseline determination
Keenleyside et al.	**2021**	anthro	no	yes	no	no
King et al.	**2021**	anthro	NA	yes	unclear	cluster analysis
Moore et al.	**2021**	NA	NA	NA	NA	NA
Scaffidi et al	**2022**	natural	yes	yes	no	no
Simonetti et al.	**2021**	unclear	yes	yes	yes – some	no
Turner	**2021**	natural	yes	yes	no	no

Table 15. Information extracted from literature review on methods used to define Pb source.

Author	Year	Mention bioavailability	Define bioavailability	How do they characterize the source isotopically	Consider material culture as a potential source
Ericson et al.	**1979**	NA	NA	NA	NA
Whittaker and Stack	**1984**	NA	NA	NA	NA
Grandjean and Jørgenson	**1990**	NA	NA	NA	NA
Patterson et al.	**1991**	no	no	bioavailable from the environment	NA
Gleń-Haduch et al.	**1997**	NA	NA	NA	NA
Åberg et al.	**1998**	no	no	bioavailable from the environment, human comparative	consider
Budd et al.	**1998**	NA	NA	NA	NA
Budd et al.	**2000**	yes	yes	bioavailable from the environment	NA
Carvalho et al.	**2000**	NA	NA	NA	NA
Montgomery et al.	**2000**	yes	no	bioavailable from the environment	NA
Szostek and Głąb	**2001**	NA	NA	NA	NA
Chiaradia et al.	**2003**	no	no	bioavailable from the environment	NA
Muller et al.	**2003**	no	no	bioavailable from the environment	NA
Budd et al.	**2004**	no	no	bioavailable from the environment, human comparative, lead ore	consider
Montgomery et al.	**2005**	yes	no	bioavailable from the environment, lead ore	ignore
Webb et al.	**2005**	no	no	human comparative	ignore
Carvalho et al.	**2007**	NA	NA	NA	NA
Valentine et al.	**2008**	yes	yes	bioavailable from the environment	NA
Bellis et al.	**2009**	NA	NA	NA	consider
Turner et al.,	**2009**	yes	yes	bioavailable from the environment	NA
Montgomery et al.	**2010**	yes	broadly	human comparative, lead ore	consider

Smits et al.	**2010**	no	no	bioavailable from the environment	NA
Montgomery et al.	**2011**	no	broadly	bioavailable from the environment, human comparative, lead ore	consider
Fitch et al.	**2012**	no	no	lead ore	dismiss
Turner et al.,	**2012**	yes	yes	bioavailable from the environment	*ignore
Farell et al.	**2013**	NA	NA	NA	NA
Schroeder et al.	**2013**	NA	NA	NA	NA
Lamb et al.	**2014**	no	no	human comparative	consider
Millard et al.	**2014**	no	no	bioavailable from the environment, human comparative, lead ore	consider
Valentine et al.	**2015**	yes	yes	bioavailable from the environment	NA
Beherec et al.	**2016**	yes	no	bioavailable from the environment, lead ore	consider
Dudás et al.	**2016**	no	no	bioavailable from the environment, human comparative, lead ore	consider
Keller et al.,	**2016**	no	no	bioavailable from the environment	ignore
Shaw et al.	**2016**	no	no	human comparative	consider
Guede et al.,	**2017**	NA	NA	NA	NA
Jones et al.	**2017**	yes	yes	bioavailable from the environment, lead ore	NA
Price et al.	**2017**	yes	yes	bioavailable from the environment, human comparative, lead ore	unclear
Evans et al.	**2018a**	yes	no	bioavailable from the environment, human comparative, lead ore	consider
Evans et al.	**2018b**	yes	no	bioavailable from the environment, human comparative, lead ore	consider
Aronsen et al.	**2019**	no	no	not specified	ignore
de Winter et al.	**2019**	NA	NA	NA	NA
Laffoon et al.	**2020**	yes	no	bioavailable from the environment, human comparative, lead ore	consider
Price et al.	**2019**	yes	yes	bioavailable from the environment	NA
Walser et al	**2020**	NA	NA	NA	consider
Eshel et al.	**2020**	yes	no	bioavailable from the environment, human comparative, lead ore	consider

King et al.	**2020**	no	no	lead ore	consider
Moore et al.	**2020**	yes	yes – cultural focusing	human comparative, lead ore	ignore
Quinn et al.	**2020**	no	yes	bioavailable from the environment, human comparative, lead ore	dismiss
Samuelsen and Potra	**2020**	yes	yes	bioavailable from the environment	NA
Keenleyside et al.	**2021**	yes	yes	human comparative, lead ore	consider
King et al.	**2021**	no	yes	bioavailable from the environment, human comparative, lead ore	ignore
Moore et al.	**2021**	NA	NA	NA	NA
Scaffidi et al	**2022**	no	no	human data itself	*NA
Simonetti et al.	**2021**	yes	broadly	lead ore	consider
Turner	**2021**	yes	broadly	bioavailable from the environment	*NA

Table 16. Information extracted during the literature review on consideration of diagenesis (NS: Not specified).

Author	**Year**	**Diagenesis considered**	**Enamel is stable**	**Conclude diagenesis occurred in enamel or main sample portion**	**Method to identify diagenesis**
Ericson et al.	**1979**	yes	no	possibly	not examined
Whittaker and Stack	**1984**	yes	no	yes	not examined
Grandjean and Jørgenson	**1990**	yes	no	no	not examined
Patterson et al.	**1991**	yes	no	yes	Pb/Ca, Ba/Ca in enamel
Gleń-Haduch et al.	**1997**	yes	no	no	not examined
Åberg et al.	**1998**	yes	yes	no	not examined
Budd et al.	**1998**	yes	yes	no	Pb/Ca between tissues
Budd et al.	**2000**	yes	yes	no	general Pb isotope variability compared to Pb iso of soil leachates

Carvalho et al.	**2000**	yes	no	no	trace element concentration
Montgomery et al.	**2000**	yes	yes	no	Pb concentration and iso ratios dentin vs enamel
Szostek and Głąb	**2001**	yes	no	yes	trace element conc burial soil correlation to teeth
Chiaradia et al.	**2003**	yes	no	yes	trace element concentration of teeth pattern
Muller et al.	**2003**	yes	yes	no	not examined
Budd et al.	**2004**	yes	yes	no	Pb isotope ratios soil compared to enamel
Montgomery et al.	**2005**	yes	yes	no	Pb iso of soil compared to enamel
Webb et al.	**2005**	yes	yes	no	not examined
Carvalho et al.	**2007**	yes	no	yes	trace element concentration
Valentine et al.	**2008**	yes	yes	no	bone Pb iso vs enamel, soil leachate Pb iso
Bellis et al.	**2009**	NS	no	no	NA
Turner et al.,	**2009**	yes	yes	no	not examined
Montgomery et al.	**2010**	yes	yes	no	not examined
Smits et al.	**2010**	yes	no	no	Sr iso in soils, not examined in Pb
Montgomery et al.	**2011**	yes	yes	no	not examined
Fitch et al.	**2012**	yes	no	yes	Pb concentration isotope ratio soil leachate
Turner et al.,	**2012**	NS	no	no	NA
Farell et al.	**2013**	NS	no	no	NA
Schroeder et al.	**2013**	yes	yes	no	enamel Pb conc pattern across site
Lamb et al.	**2014**	NS	no	no	NA
Millard et al.	**2014**	yes	yes	no	not examined
Valentine et al.	**2015**	yes	yes	no	not examined
Beherec et al.	**2016**	yes	testing	yes	trace element concentrations compared dentin and enamel, and Pb iso of leachates and ore compared to enamel
Dudás et al.	**2016**	yes	no	possibly	Pb concentration
Keller et al.,	**2016**	yes	no	no	not examined

Shaw et al.	**2016**	yes	yes	no	not examined
Guede et al.,	**2017**	yes	no	yes	REE and U concentration
Jones et al.	**2017**	yes	yes	no	not examined
Price et al.	**2017**	yes	yes	no	linear array of Pb iso ratios
Evans et al.	**2018a**	NS	no	no	NA
Evans et al.	**2018b**	NS	no	no	NA
Aronsen et al.	**2019**	yes	yes	possibly	Pb conc. MTC – Kamenov et al., 2018
de Winter et al.	**2019**	yes	no	yes	Diffusion-advection model with trace element concentrations
Laffoon et al.	**2020**	yes	no	yes	mixing model Pb iso values
Price et al.	**2019**	yes	yes	no	not examined
Walser et al	**2020**	NS	no	no	NA
Eshel et al.	**2020**	yes	yes	yes	Ba/Ca vs Pb/Ca via Patterson et al., 1991
King et al.	**2020**	yes	no	yes	Pb conc. MTC – Kamenov et al., 2018, Pb iso compared to lead coffin and soil leachates
Moore et al.	**2020**	NS	no	no	NA
Quinn et al.	**2020**	yes	no	no	Pb iso of Pb coffin
Samuelsen and Potra	**2020**	yes	no	possibly	MTC – Kamenov et al., 2018
Keenleyside et al.	**2021**	NS	no	no	NA
King et al.	**2021**	yes	no	yes	Pb conc. MTC – Kamenov et al., 2018
Moore et al.	**2021**	yes	yes	no	not examined
Scaffidi et al	**2022**	yes	yes	no	MTC – Kamenov et al., 2018
Simonetti et al.	**2021**	yes	yes	yes	MTC – Kamenov et al., 2018, Enrichment factor, Pb iso compared to geology linear array
Turner	**2021**	yes	no	no	not examined

7.2 Supplemental Materials Chapter 3

7.2.1 Site Archaeological Context

7.2.1.1 St. Paul's Anglican Church Cemetery, Harbour Grace, NL (CkAh-06)

In 1991, construction at the St. Paul's Anglican Church uncovered human remains. The provincial archaeology office (PAO) excavated a 30.5 m by 15 m trench over the course of a month to remove as many burials as possible (Pike, 2013:10). Based on the location of the remains and knowledge of the church building's construction, this cemetery section was used sometime between 1764 and 1820 (Pike, 2013). During this period, Harbour Grace was a large town in Conception Bay, with planters, higher-class merchants, and servants. The community had both Anglican and Catholic individuals, but since this was an Anglican cemetery, it was likely exclusively English or English-descent individuals. However, it is possible that there were regulations against Catholic burials similar to in St. John's, Irish or Irish-descent individuals may be present as well. The remains were reburied during a ceremony in the fall of 2019.

There are a minimum of 19 individuals, of which, 14 were excavated in context and five were found in back dirt piles from the construction (Pike, 2013:12). Ages ranged between mid-fetal life (24-28 weeks) and old adult (50+), and both males and females were present (Pike, 2013:15). A previous bone collagen and enamel carbonate isotopic study showed a mixed C_3 and marine diet during childhood, consistent with what would be expected growing up in a fishing community (Munkittrick et al., 2019). Their $\delta^{18}O_{carb}$ values were lower than in England, as expected locally (Munkittrick et al., 2019). Enamel

$^{87}Sr/^{86}Sr$ data suggests broadly coastal origins but are not able to distinguish from Newfoundland versus western European origins (Garlie, 2022).

7.2.1.2 Wester Point Cemetery, Portugal Cove, NL (CjAf-08)

In 2004, construction in a new subdivision uncovered human remains. The purpose of the archaeological survey was to recover those disturbed by the excavation and delineate the cemetery. In 1794, Portugal Cove had 34 families with servants, totalling 200 inhabitants (Reddigan, 2007; Harris, 2015). By 1853 the town grew significantly, with 651 inhabitants. Of these, almost all were born in Newfoundland, with five from England and 19 from Ireland. Historical records suggest West Country England, Ireland and Jersey were the main origins of individuals from this town (Pitt, 2015). The fishery was the main occupation throughout this period, but in the mid-1800s, there were clergy, farmers, and mechanics. There are no precise dates for this cemetery; therefore, it can only be assumed that it dates between the 18th and 19th-centuries. The remains were reburied during a ceremony in the summer of 2013.

An MNI of 13 individuals were found, but only nine were removed. There was one male, two females, and one possible male; the rest of the remains had insufficient material for sex estimation. All individuals were adults, with at least one old adult (50+) (Harris, 2015). Bone collagen isotope analyses showed a mixed C_3 and marine diet (Harris, 2015). However, a couple of individuals had exclusively C_3 diets suggesting either a distinct diet of some families or a change in geography (Harris, 2015).

7.2.1.3 Foxtrap-2, Foxtrap, NL (CjAf-10)

Land in a previous farmer's field was developed into a small-scale industrial business. While the small burial ground was known to the local community of Foxtrap, the site was initially surveyed by provincial archaeologist Steve Hull (Hull, 2007), but the decision was later made to move and re-inter the remains nearby. This was the first fully excavated burial ground in Newfoundland. A ground penetrating radar (GPR) survey was performed, and some anomalies were consistent with later ground-truthed burials, stones, or roots (Grimes et al., 2018). Areas with head- and foot-stones were targeted first to determine the stratigraphy. Excavations were carried out over several weeks spread across two seasons (2016 and 2017). All burials were in coffins, and artifacts included copper pins used with fabric to keep the jaw closed, an 1862 Nova Scotia halfpenny, and some silver or gold-plated wires. The only possible identification of an individual to be buried in the cemetery was Bridget Greeley, who died in 1896 of typhoid fever. However, this is based on an incomplete headstone that was moved since internment, so it cannot be confirmed which burial was hers (Grimes et al., 2018).

The dates of the burial ground are not clear. The town did have its own clergyman as of 1846, and the local cemetery for the All Saints Anglican Church was in use by 1853 (FitzGerald, 2000), and family plots were common before central cemeteries. While this family plot was used until at least the time of Bridget Greeley's death in 1896, a start date is unknown.

There were 31 separate burials excavated but only 18 contained preserved human remains. Based on the grave cut sizes and skeletal remains, there were eight adults, 13 subadults, and three individuals of indeterminate age. Gross morphological preservation

was not good. Many adult burials contained only diaphyses and skulls, and many subadult teeth had only the enamel. This included two possible males, one possible female, and four of indeterminate sex. Due to the fragmentary nature of preserved remains, the subadult ages were estimated by assessing tooth development and included infants to adolescents (Grimes et al., 2018). Unpublished data $\delta^{13}C_{coll}$ and $\delta^{15}N_{coll}$ data from Garlie (2022) suggests a mixed terrestrial marine diet with non-specifical coastal $^{87}Sr/^{86}Sr$ from enamel bioapatite values for all individuals.

7.2.1.4 Tors Cove burial ground, Tors Cove, NL (ChAf-01)

The Tors Cove burial ground is located on a hill overlooking the harbour, and the remains are actively eroding out of the bank. Archaeologists with permits and concerned citizens have recovered the bones from the bank over the last ~15 years. The burial ground itself has 18 to 23 burials with head- and foot-stones (Hull, 2006). There may have been more that have eroded or are covered by soil. While some headstones have inscriptions, many do not, including the three closest to the bank that are believed to represent at least some of the individuals recovered (Lear and Grimes, 2016). Hull (2006) believes that the earliest burials were likely towards the bank, and dates of 1812 or 1826 are the earliest from the cemetery (Hull, 2006; Lear and Grimes, 2016). The site certainly dates to the 19th century, possibly before the church cemetery was established in the later 1870s, but there is no known start date for the cemetery.

The town of Tors Cove is continuously associated with the fishery. The English established the town before 1675 (Losier and Lear, 2018), but Irish immigration was frequent in the late-18th to early-19th centuries. Between 1836 and 1871, the population

fluctuated between 300 and 430 (Reddigan, 2007; Losier and Lear, 2018). In 1853 almost all inhabitants were identified as being born in Newfoundland; only 24 were from Ireland and two from England. All were identified as Catholic.

Archaeology honours student Alicia Morry performed an initial survey of the remains. An additional examination by Munkittrick and Garlie in 2020 found an MNI of five based on the left parietal bones. Of the three individuals that have teeth, there are two adults, one male and one female. An additional subadult is at least 12 years old based on dental development.

7.2.1.5 St. Luke's Anglican Church Cemetery, Placentia (Plaisance), NL (ChAl-17)

Remains were removed from the St. Luke's Anglican Church Cemetery in 1969 during an excavation to find previous church foundations. There were four gravestones, assumed during excavation to be head and footstones. While now an Anglican Church Cemetery, it was previously used during the French occupation by the Catholic church and prior to this by the Basque. If these were Basque or French, the town of Plaisance was very involved with the migratory fishery, though fortifications in 1665 meant that there were other occupations and permanent settlement. The 1690s census suggests that some individuals were born locally, but many were from France, with a few individuals from Ireland or England. The town of Plaisance was on land granted to England during the Treaty of Utrect (1713) and renamed Placentia by the English. There are a minimum of six individuals that were highly fragmentary, all were adults but of indeterminate sex. Dietary isotopes from bone collagen ($\delta^{13}C$ and $\delta^{15}N$) from Garlie (2022) suggest mixed terrestrial and marine diets for most individuals. Additional enamel $^{87}Sr/^{86}Sr$ samples

suggest broadly coastal origins but are not able to distinguish from Newfoundland versus western European origins (Garlie, 2022).

7.2.1.6 Block 3 Cemetery, Fortress of Louisbourg, NS (3L)

The Block 3 cemetery was associated with the local parish but was only used from 1713 to 1722. After this, the town adopted a grid-style organization, garnering its archaeological name Block 3, so other buildings were built on the land. At that time many of the burials were moved, but some remained. Since this was from the earliest part of the occupation, none of the individuals could have been born locally. The Fortress of Louisbourg was established by the original occupants of Plaisance, Newfoundland, in 1714. However, there was extensive immigration during the early occupation, particularly from La Rochelle and Île de Ré. The 1724 Louisbourg census names the place of birth for those males or females who were heads of households. While most were from France, usually Normandy, Brittany, and Basque country, others were from New France (Quebec and Acadia) or a few from Switzerland, Belgium, and Germany (Johnston, 1995a;b). Around half of the females were from New France, with officers tending to marry Acadians (Johnston, 1995a, 2001). There were also a number of Black individuals, both enslaved and freed, that made up around 3% of the population throughout the period between 1724–1758 (Donovan, 2004). During the period that the cemetery was in use, there was only even one enslaved individual (Donovan, 2004). Louisbourg was a very mixed town with wealthy merchants, boat owners, indentured servants, enslaved labourers, and other tradespeople (Johnston, 2001).

Excavations of the Block 3 cemetery began in 1973 by Jason Henderson and in later 1974 by Donald Harris, uncovering a total of 26 individuals. Only three were coffin burials, and the rest were shrouded. Burial 1 was found with a button. All individuals were adults, except for four who had age ranges between 16 and 25. Most individuals are identified as male (n = 14) or possible male (n = 4), though there were females (n = 1) and possible females (n = 2). There were also individuals of indeterminate sex (n = 4). Researchers have examined these remains finding lots of pathological conditions, including cribra orbitalia (Scott et al., 2020). Several individuals (n = 12) also had vitamin D deficiency according to their rachitic pulp morphologies, one of whose bones had evidence of residual rickets (Hinton, 2019). Unpublished dietary isotopic data ($\delta^{13}C$ and $\delta^{15}N$) suggests variable dietary patterns, including mostly C_3 terrestrial, mixed C_3 terrestrial and marine foods, and mixed C_3 and C_4 diets. In addition to varied diets, their enamel and $^{87}Sr/^{86}Sr$ values suggest a wider range of variation, outside of solely coastal origins, suggesting that they were not all coming from Plaisance, but likely from France and New France as well (Garlie, 2022).

7.2.1.7 Southside Cemetery, St. John's, NL (CjAe-54)

The Southside Cemetery was partially excavated in 1979 when construction workers uncovered remains while digging sewer pipes. The Royal Newfoundland Constabulary oversaw the construction workers digging the remains using a backhoe; archaeologists were not permitted to assist. At least some of these individuals were buried in coffins, and one individual had two copper half pennies from the last quarter of 18th century placed on their eyes. While initially assumed to be associated with the nearby St. Mary the Virgin

Anglican Church, demographic and isotopic research established that this was an unlikely explanation (Munkittrick et al., 2019). Instead, at least the portions excavated, were associated with the St. John's Royal Naval Hospital. The British Royal Navy used this cemetery from as early as the 1750s until 1825, when their local station closed.

The RN role in NL was as local government and to protect the fishing fleet. Seasonal role, though there was an over wintering force – the exact date of that starting is unclear. The hospital records are sparce, but at least in the 1790s onwards the hospital was in use year-round, though this was mainly serving those that were too ill to return to England rather than having many new additions.

The number of Royal Navy sailors varied drastically during the 18th century from less 20,000 during peace time, but over 100,000 during the Napoleonic Wars. Within the Royal Navy as a whole there were varied origins. England was the main area for enlistment, followed by Scotland. While many did join willingly, press gangs were also a common means to fill numbers, forcing individuals into service rather than a national conscription. It was often that a vessel would lose numbers throughout their voyages, either through disease, run aways, or less commonly, battle. To make up for these numbers the navy used impressment to fill their numbers. Newfoundland specifically was known as a "nursery for seamen" because of the abundance of trained sailors which the navy frequently pressed men into service from. While not exclusively so, the West Country was highly involved with the fishery (Head, 1976). As such, many, at least during the 18th century, many of those that were pressed into service from the NL English fishery would have been English citizens. Or, from the small but growing permanent population in Newfoundland itself.

To examine the origins of sailors, present in the navy at the NL squadron, a records from five vessels were examined as part of the historical research for my masters thesis on the St. John's Naval Hospital and the Southside Cemetery (Munkittrick, 2015: 83–85). These vessels were from varying sizes and specifically of those indicated within the hospital records as depositing sailors for treatment. The numbers of which were taken from the closest available records to the time of reference in the hospital records (1786–1806). The results of these found that the largest vessels were most variable (and not necessarily stationed fulltime there). Most vessels had individuals of unknown or un-recorded origins, this made up between 0 and 34% of the sailors (0 to 64 individuals). Origins unsurprisingly included England (20–57%), Ireland (0–16%), and Scotland (0–12%), but also British colonies such as Nova Scotia (0–12%), Newfoundland (0–14%). Note that the smaller vessels which tended to be those stationed in NL year-round had high percentages of those from NS and NL. Yet, there were also small numbers of individuals from America, other European countries, and Africa. The distribution seen in Newfoundland are similar to records from a slightly earlier period (1774) of those stationed in Halifax. The (minimal) variability of these origins is consistent with the isotope data from those buried in the Southside Cemetery (Munkittrick, 2015; Munkittrick et al., 2019).

There was a minimum of 21 individuals, but due to excavation practices, the assemblage is comingled (von Hunnius, 1998). All individuals were adults, except for one teenager (von Hunnius, 1998). Sex identification was difficult due to co-mingling and the poor preservation of gross morphology, but where possible, all but one (based on *os coxa*) individual could be identified as male or possible male (Munkittrick et al., 2019;

von Hunnius, 1998). Previous isotopic analyses ($\delta\delta^{15}$Nand δ^{15}N) suggested that their diet was exclusively C_3 foods, except for a couple of individuals (NP 163 A3 and B7) who has a mixed marine and terrestrial C_3 diet (Munkittrick et al., 2019). Additionally, δ^{13}C and δ^{18}O values from dental carbonate for all but two individuals (NP 163 A3 and B8; no tooth available for B7) suggest that they are inconsistent with having grown up locally, with no or little marine food contribution and drinking water consistent with a lower latitude (Munkittrick et al., 2019). The exception to this for carbonate isotope values was A3 and B8 (no tooth was available for B7) who had higher δ^{13}C values, suggesting more marine and or C_4 food in their diets. While A3 had δ^{18}O values consistent with those buried at other Newfoundland cemeteries, B8 had much higher value suggesting a childhood spent at a much lower latitude closer to the southeastern United States or the Caribbean (Munkittrick et al., 2019). Taken with historical context of the Royal Naval impressment practices, it is suggested that at least most of these individuals were likely born in the Great Britain (Munkittrick et al., 2019).

7.2.2 Comparative populations for Pb concentration and isotope ratio values

See tables Table 17 and Table 18 for all comparative site information and citations.

Table 17. Site names, locations, and dates for Pb concentration comparative data. Sites are grouped by the region and period comparative used in text with the number of samples and citations indicated.

Region and period group	Period	Location	Site name	Pb conc.	Publication
Royal Navy	1750s-1825 CE	St. John's, Newfoundland, CA	Southside Cemetery	19	This Publication
Prehistoric England, Ireland, and Scotland	Neolithic	Shetland, Scotland	Sumburgh	8	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Neolithic	N. Yorkshire, England	West Heslerton	2	Montgomery, 2002
Prehistoric England, Ireland, and Scotland	Bronze Age	N. Yorkshire, England	West Heslerton	3	Montgomery, 2002
Prehistoric England, Ireland, and Scotland	Bronze Age	Yorkshire, England	Callis Wold	2	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	Yorkshire, England	Towthorpe	2	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	Yorkshire, England	Aldro	5	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	Mull, Scotland	Ardachy	1	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	Yorkshier Wolds, England	Garton Slack	2	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	Yorkshier Wolds, England	Acklam Wold	1	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	South Uist, Scotland	Cladh Hallan	3	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	Yorkshire, England	Gristhorpe	1	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Bronze Age	Lewis, Scotland	Cnip	1	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Prehistoric	Shepperton, England	Shepperton	1	Budd et al., 2004

Prehistoric England, Ireland, and Scotland	Prehistoric	Monkton, England	Monkton	4	Budd et al., 2000b in Budd et al., 2004; Montgomery 2002 ($^{20n}Pb/^{206}Pb$)
Prehistoric England, Ireland, and Scotland	Bronze Age	Monkton, England	Monkton	1	Budd et al., 2004
Prehistoric England, Ireland, and Scotland	Iron Age	N. Yorkshire, England	West Heslerton	3	Montgomery, 2002
Prehistoric England, Ireland, and Scotland	Iron Age	Yorkshire, England	Wetwang	8	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Iron Age	Musselburgh, Scotland	Musselburgh	1	Moore et al., 2020
Prehistoric England, Ireland, and Scotland	Scottish/Irish Iron Age	Dublin, Ireland	Rathoath	1	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Scottish/Irish Iron Age	Dublin, Ireland	Rath	1	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Scottish/Irish Iron Age	Orkney, Scotland	Mine Howe	2	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Scottish/Irish Iron Age	Isle of Skye, Scotland	High Pasture Cave	1	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Scottish/Irish Iron Age	Lewis, Scotland	Galson	5	Montgomery et al., 2010
Prehistoric England, Ireland, and Scotland	Scottish/Irish Iron Age	South Uist, Scotland	Kilpheder	2	Montgomery et al., 2010
Roman Britain	1^{st}-4^{th} c CE	York, England	3/6 Driffield Terrace	4	Montgomery et al., 2010
Roman Britain	Romano-British	London, England	Southwark	1	Budd et al., 2004
Roman Britain	Romano-British	Winchester, England	Eagle Hotel, Winchester	4	Budd et al., 2004; Montgomery, 2002 ($^{20n}Pb/^{206}Pb$)
Roman Britain	Romano-British	Bristol, England	Mangotsfield	2	Budd et al., 2004; Montgomery, 2002 ($^{20n}Pb/^{206}Pb$)
Roman Britain	Romano-British	London, England	Spitalfields	1	Budd et al., 2004; Montgomery, 2002 ($^{20n}Pb/^{206}Pb$)
Roman Britain	Romano-British	London, England	various London sites	20	Shaw et al., 2016

Roman Britain	Romano-British	Musselburgh, Scotland	Musselburgh	6	Moore et al., 2020
Early Medieval England and Scotland	5-7thc CE – Anglo-Saxon	Berinsfield, England	Berinsfield, Oxfordshire	19	Evans et al., 2018a
Early Medieval England and Scotland	5-7thc CE – Anglo-Saxon	Eastbourne, England	Eastbourne, East Sussex	21	Evans et al., 2018a
Early Medieval England and Scotland	5-7th c CE – Anglo-Saxon	Wasperton, England	Wasperton, Warwickshire	17	Montgomery et al., 2010
Early Medieval England and Scotland	5-7th c CE – Anglian	West Heslerton, England	West Heslerton	33	Montgomery 2002 in Evans et al., 2018a
Early Medieval England and Scotland	7thc CE – Anglo-Saxon	Wiltshire, England	Stonehenge	1	Budd et al., 2004
Early Medieval England and Scotland	7-9th c CE – Anglo-Saxon	Bamburgh, England	Bambrugh	2	Budd et al., 2004
Early Medieval England and Scotland	7-8th c CE	Repton, England	Monastic and Charnel areas, Repton	5	Budd et al., 2004
Early Medieval England and Scotland	10-12th c CE – Anglo-Saxon	Riccal, England	Riccall	6	Budd et al., 2004
Early Medieval England and Scotland	8th-11th c CE-pict	Orkney, Scotland	Westness	3	Montgomery et al., 2010
Viking Scotland, Ireland, and England	8th-11th c CE	Orkney, Scotland	Westness	2	Montgomery et al., 2010
Viking Scotland, Ireland, and England	8th-11th c CE	Dublin, Ireland	Great St. George St	3	Montgomery et al., 2010
Viking Scotland, Ireland, and England	872-875 CE	Repton, England	Viking burial, Repton	3	Budd et al., 2004
Viking Scotland, Ireland, and England	970-1025 CE	Weymouth, Dorset, England	Weymouth	31	Evans et al., 2018b
Viking Scotland, Ireland, and England	Viking	Ardnamurchan, Scotland	Ardnamurchan boat burial	1	Harris et al., 2017 in Evans et al., 2018a
Viking Scotland, Ireland, and England	8th-11th c CE	Lewis, Outer Hebrides	Cnip	3	Montgomery et al., 2010
Late Medieval Atlantic Archipelago	1301-1407 CE	Cumbria, England	St.Bees Priory	1	Knüsel et al., 2010

Late Medieval Atlantic Archipelago	Late Medieval	Hereford, England	Hereford Catheral	20	Montgomery et al., 2010
Late Medieval Atlantic Archipelago	Late Medieval	Orkney, Scotland	Graemsay	1	Montgomery et al., 2010
Late Medieval Atlantic Archipelago	1250 CE	Lihou, Guernsey	Lihou	1	Budd et al., 2004
Late Medieval Atlantic Archipelago	13th c CE	Essex, England	Rivenhall	1	Budd et al., 2004
Late Medieval Atlantic Archipelago	Late Medieval	Glouster, England	Blackfriars	3	Budd et al., 2004; Montgomery, 2002 (^{20n}Pb/^{206}Pb)
Newton Plantation, Barbados	1660-1820 CE	Christ Church, Barbadoes	Newton Planation Cemetery	24	Laffoon et al., 2020
Otago, New Zealand	1860-1890 CE	Milton, New Zealand	St. John's Milton	5	King et al., 2020
Otago, New Zealand	1861-1866 CE	Lawrence, New Zealand	Androssan St. Cemetery, Lawrence	14	King et al., 2020
New Haven, Connecticut	1833-1851 CE	New Haven, Connecticut, USA	Christ Church, Catholic cemetery	3	Aronsen et al., 2020
Industrial England	1825-1847 CE	Coventry, England	Coventry	9	Millard et al., 2014
Industrial England	1833-1853 CE	London, England	Golden Lane	5	Millard et al., 2014
Industrial England	1843-1854 CE	London, England	Lukin Street	45	Millard et al., 2014
Industrial England	18th-19th c CE	London, England	Chelsea Old Church	24	Millard et al., 2014
Industrial USA	1879-1899 CE	Pueblo, Colorado, USA	Colorado Mental Health Institute cemetery	27	Keller et al., 2016

Table 18. Site names, locations, and dates for $^{20n}Pb/^{204}Pb$ and $^{20n}Pb/^{206}Pb$ comparative data. Sites are grouped by the region and period comparative used in text with the number of samples and citations indicated.

Region and period group	Period (CE)	Location	Site name	$^{20n}Pb/^{204}Pb$	$^{20n}Pb/^{206}Pb$	Publication
Royal Navy	1750s-1825	St. John's, Newfoundland, CA	Southside Cemetery	19	19	This Publication
Late Medieval Atlantic Archipelago	1301-1407	Cumbria, England	St.Bees Priory	1	0	Knüsel et al., 2010
Late Medieval Atlantic Archipelago	Late Mediaeval	Orkney, Scotland	Graemsay	1	1	Montgomery et al., 2010
Late Medieval Atlantic Archipelago	1250	Lihou, Guernsey	Lihou	1	1	Budd et al., 2004
Late Medieval Atlantic Archipelago	13th c.	Essex, England	Rivenhall	1	1	Budd et al., 2004
Late Medieval Atlantic Archipelago	Late Mediaeval	Glouster, England	Blackfriars	4	4	Budd et al., 2004; Montgomery, 2002 ($^{20n}Pb/^{206}Pb$)
Newton Plantation Barbados	1660-1820	Christ Church, Barbados	Newton Plantation Cemetery	17	17	Laffoon et al., 2020
Otago, New Zealand	1860-1890	Milton, New Zealand	St. John's Milton	5	5	King et al., 2020
Otago, New Zealand	1861-1866	Lawrence, New Zealand	Androssan St. Cemetery	14	14	King et al., 2020
New Haven, Connecticut	1833-1851	New Haven, Connecticut, USA	Christ Church, Catholic cemetery	4	4	Aronsen et al., 2019
Industrial England	1825-1847	Coventry, England	Coventry	10	10	Millard et al., 2014
Industrial England	18th-19th c	London, England	Chelsea Old Church	24	24	Millard et al., 2014
Industrial USA	1832-1873	Grafton, Illinois, USA	Grafton Cemetery	0	16	Fitch et al., 2012
Industrial USA	1879-1899	Pueblo, Colorado, USA	Colorado Mental Health Institute cemetery	31	0	Keller et al., 2016

7.2.3 Galena comparative Pb isotope groups

Pb isotope ratios of ore were compiled in Blichert-Toft et al. (2016) for much of Western Europe and was broadly grouped by country (Figure 25; Figure 29). Not all data was originally published giving all Pb isotope ratios, but they were calculated by Blichert-Toft et al. (2016) when missing. It should be noted that while some of these data relate directly to ore from historical contexts (Baron et al., 2006), most are taken from mines in use, recently used, or taken for the purposes of prospecting. Therefore, these do not necessarily relate to specific comparative materials that would be in use contemporaneously to that of the human populations examined in this study.

For American ore bodies, there has been no similar large comparative paper, but data were previously compiled in Bird et al. (2019), though individual data points were not published. Pb was grouped into regions by the original authors of the articles (Figure 7) but are also shown as compiled Pb values (Figure 30). These were typically defined by specific named ore districts. The citations for these data are found in Table 19. As with the European Pb ore deposits, not all areas were in use during the lives of the fishery populations in this study, so not all directly are relevant.

Table 19. Groupings of Pb ore data from deposits in the United States. Number of ore samples are denoted along with the citations of the original data. The date that mining started in the region or specific site along with a reference for that date are also indicated.

Pb deposits	States	Ore #	Citations	Date of mining start	Date citation
Upper Mississippi Valley	IA, WI, IL, MN	39	Millen et al., 1995; Heyl et al., 1966 corrected in Millen et al., 1995	late 1680s Indigenous people trading with French	Henry, 1976; Millhouse, 2010
Piedmont Massive Sulfide deposit	VA, NC	25	LeHuray 1982	1770s	Poole, 1974
Austinville-Ivanhoe	VA	10	Foley et al., 1981	1756	Whisonant, 1996
East Tennessee – Southern Appalachians	TN	83	Kesler et al., 1994a	1780s or Revolutionary war	Whyman, 2018
Central Missouri barite	MO	12	Goldhaber et al., 1995	1830	Bundtin, 2021
Central Illinois and Northeast Kentucky	IL, KY	3	Heyl et al., 1966	1835	Holibaugh, 1895
Illinois-Kentucky fluorite district	IL, KY	8	Heyl et al., 1966	1830s discovery	Lasemi, 2010
Friedensville district – Central Appalachians	NJ	4	Kesler et al., 1994b	1840s	Kaas, 2016
Appalachian Valley	PA, NJ	3	Heyl et al., 1966	by 1850s	Price, 1947; Mosier, 1948
Old Lead Belt	Southeast MO	133	Goldhaber et al., 1995, Deloule et al., 1986; Svergensky et al., 1979	1864-1972	Hevrnk & Radford, 1997
Southeast Kansas	KS	2	Heyl et al., 1966	by 1870s	Manders & Aber, 2014
Colorado	CO	8	Lüders et al., 2009	1870s	Irey, 1951
Tristate district	OK	125	Deloule et al., 1986	1891 OK, 1850s MO	Everett, n.d.
Timberville	VA	11	Kesler et al., 1994a	early 1900s	Herbert, 1956

7.2.4 Pb concentration and isotope analyses methodologies

7.2.4.1 Tooth choice: type and preservation

One tooth, preferably the second molar, from each individual was taken for sampling. This minimizes the likelihood of exposure to maternal or caregiver's Pb, and its size maximizes the potential of other isotopic data from a single tooth. However, if it was not available, the fourth premolar, third premolar, or first molar were taken instead. We chose teeth with minimal cracks and staining and avoided areas of teeth that had caries. Additionally, we did not sample deciduous or unerupted teeth since under-mineralized teeth are more prone to diagenetic alteration.

Amount of sample necessary for analysis was estimated based on previous studies sample size of ~50 mg for Pb concentration and isotope analyses (Montgomery, 2002). Estimates on the implication of this were considered given the analytical procedures defined below, where a standard SRM 981 solution of 100 ppb resulted in ~4V in the faraday cup at mass 208 (or 0.04 ppb/V) on the MC-ICP-MS. A hypothetical enamel concentration of 0.5 ppm was considered since it was likely that the samples would be affected by at least some anthropogenic Pb. The density of the sample within 1 mL of 0.3 M HNO_3 was considered. Since ¼ of the sample digest solution would be taken for concentration analyses, the density was calculated as ~1.0375 g/mL with a modified sample mass of 0.0375 g. The resulting 0.018 ppb was calculated into estimated voltage at 0.72 V. While this was under the 4 V of the SRM 981 solution, it was close to 1 V which should still be sufficient for analyses as it is well above the voltage from blanks during protocol establishment (0.019 V).

Tooth enamel was cleaned with a tungsten carbide bit (Brasseler round carbide) to at least 100 μm or until surface staining was removed. An enamel sample of ~50 mg (6.5–68.7 mg; Table 20) was separated using a diamond dental saw (Brasseler DS Miniflex). Since these blades have a potential for contamination (Montgomery, 2002), all cut surfaces were further cleaned with a clean tungsten carbide bit (Brasseler round carbide), and any adhering dentin was removed under 5X magnification. The dental tools were cleaned between each sample by ultrasonication in deionized water, rinsed, dipped in 2M HNO_3 and allowed to dry. Following cutting, enamel was ultrasonicated three times in deionized water for five minutes each. After rinsing with ~200 μL of acetone, the samples were transferred to the clean lab and left to air dry in the laminar air flow clean boxes.

Samples were transferred to clean 3 mL or 7 mL PFA vials (Savillex) and weighed. Two mL of distilled 8 M HNO_3 were added to each sample and digested on a hot plate for approximately one hour. A blank and standard (NIST SRM 1400; ~25 mg) were included in each group of approximately eight samples.

7.2.4.2 Concentration methods and errors

An aliquot of 0.5 mL was taken from the digest solution for concentration analysis carried out at one of two laboratories, The Earth Resources Research and Analysis facility at Memorial University (TERRA) or the Geochemistry laboratory at the University of Ottawa (uOttawa). Here an aliquot of the 0.5mL sample was taken up, and 0.3 M HNO_3 solution was added using gravitation measurement to ensure that it was taken up into 10 mL of solution.

Since the 2 mL for digestion was added to the samples using volumetric, not gravimetric, measurement, the solution's density must be corrected. To do this, the weight of the vial was removed from the weight of the final solution. This was then divided by the mass of the sample. The sample concentration was determined using the calibrated ppm of the solution that went through the Q-ICP-MS. For some of the early samples (indicated in Table 20) we did not have the mass of the solution, so an average of the values used in subsequent analyses were used instead. This gave a mean and 1σ of 2.4753 ± 0.1040 ($n = 124$). This correction is not necessary if the solution is dried down and brought up into the final analyzed solution, as measured through gravitational measurement. However, to the authors' knowledge, this correction factor is not mentioned in any previous archaeological Pb enamel publications, but it can make a significant difference to the final value. For example, the SRM 1400 standards go from a relative error of almost 40% to less than 10%. The trace element laboratories may be making this correction for the researchers, but more detail must be given in trace element studies to ensure accurate data.

For data analyzed at the TERRA facility (indicated in Table 20), analyses were carried out on acidified sample solutions of ~0.2 M HNO_3 at a dilution factor of ~6000X. To achieve this, aliquots of samples in 8 M HNO_3, based on their starting weight and first dilution with the 8 M HNO_3, were diluted to ~10 g with de-ionized 18.2 MΩ water if the aliquot was bigger than 0.25 g, or with 0.2 M HNO_3 if less. The concentration of 35 elements including Pb was determined with solution ICP-Q-MS (inductively coupled plasma mass spectrometry) on a Perkin-Elmer Elan DRCII with a Cetac ASX-520 Autosampler. The RF frequency is 1200 W using Nickel cones. The plasma gas was

Table 20. Biological information and Pb sample context for individuals from the following burial sites: Foxtrap-2; St. Paul's Anglican Church cemetery, Harbour Grace (St. Paul's); Tors Cove burial ground, Tors Cove (Tors Cove); Wester Point burial ground, Portugal Cove-St. Philip's (Wester Point); St. Luke's Anglican Church cemetery, Placentia (St. Luke's); Block 3 burial ground, Louisbourg (Block 3) Southside Cemetery, St. John's (Southside). Concentration analysis was performed at the University of Ottawa using an ICP-QQQ at, or at Memorial University of Newfoundland using either a QICMS or an MC-ICP-MS. Correction for volumetric vs gravimetric mass of sample in solution indicated as using the actual mass or an average.

Site	Borden number	Individual	Sex	Age	Tooth	Sample size mg	Pb conc.	Instrument	correction	V	Blank % of iso
Foxtrap-2	CjAf-10	4	IND	> 9 y.o.	LRM2	64.28	4.6	MC-ICP-MS	actual	4.9	0
Foxtrap-2	CjAf-10	7	IND	Adult	ULM2	56.42	4.9	MC-ICP-MS	actual	4.6	0
Foxtrap-2	CjAf-10	9	IND	Adult	URM2/3	42.24	3.1	MC-ICP-MS	actual	2.2	0
Foxtrap-2	CjAf-10	10	IND	Adult	ULM2	56.85	7.9	MC-ICP-MS	actual	3.7	0
Foxtrap-2	CjAf-10	11	PM	Adult	LLM2	55.19	7.7	MC-ICP-MS	actual	3.7	0
Foxtrap-2	CjAf-10	12	PF	Adult	LRM2	51.86	7.4	MC-ICP-MS	actual	6.8	0
Foxtrap-2	CjAf-10	13	IND	Adult	URM2	55.01	4.5	MC-ICP-MS	actual	4.4	0
Foxtrap-2	CjAf-10	14	PM	Adult	URM2	50.20	4.7	MC-ICP-MS	actual	4.2	0
Foxtrap-2	CjAf-10	30	IND	Adult	LRM2	51.15	5.6	MC-ICP-MS	actual	5.2	0
St. Paul's	CkAh-06	NP250/F3	M	middle adult (30-45 y.o.)	LRM2	10.88	7.4	ICP-QQQ	average	2.0	1
St. Paul's	CkAh-06	NP251/F4	M	middle adult (38-50 y.o.)	LLPM4	52.82	6.2	MC-ICP-MS	actual	5.8	0
St. Paul's	CkAh-06	NP254/F7	F	young adult (20-30 y.o.)	M2	12.24	28.9	ICP-QQQ	average	1.7	1
St. Paul's	CkAh-06	NP258/F13	M	middle adult (35-50 y.o.)	URM2	60.65	6.1	MC-ICP-MS	actual	3.4	0
St. Paul's	CkAh-06	NP262/F21	IND[1]	adolescent (12-14y.o.)	M2	13.92	6.0	ICP-QQQ	average	4.8	0
St. Paul's	CkAh-06	NP263A/F22	F	young adult (25-35 y.o.)	M2	17.22	3.4	ICP-QQQ	average	12.0	0
St. Paul's	CkAh-06	NP264 A/3	IND	young adult (20-35y.o.)	LLM2	14.20	2.8	ICP-QQQ	average	12.8	0
St. Paul's	CkAh-06	NP264 B/2	IND	young adult (20-35y.o.)	LLM1	37.12	10.3	MC-ICP-MS	actual	3.5	0
Tors Cove	ChAf-01	NP 2	IND	IND	LRM2	40.54	2.6	MC-ICP-MS	actual	1.9	1
Tors Cove	ChAf-01	NP 54	IND	adolescent	LRM2	82.69	4.5	MC-ICP-MS	actual	6.7	0

Tors Cove	ChAf-01	NP 68		IND	LLM2	55.37	0.9	MC-ICP-MS	actual	0.9	1
Wester Pt.	CjAf-08	2	F	18-25	LLM2	16.22	5.0	ICP-QQQ	average	4.5	0
Wester Pt.	CjAf-08	4	F	50+	ULPM1	13.36	18.6	ICP-QQQ	average	6.5	0
Wester Pt.	CjAf-08	5	IND	21+	URM3	14.98	0.1	ICP-QQQ	average	NA	NA
St. Luke's	ChAl-20	62A	IND	IND	URM2	45.47	4.3	QICPMS	average	3.9	0
St. Luke's	ChAl-20	63	IND	IND	URM2	45.31	14.4	QICPMS	average	6.7	0
St. Luke's	ChAl-20	66B	IND	IND	LRM2	44.63	8.4	QICPMS	average	3.5	0
St. Luke's	ChAl-20	67	IND	IND	URM2	38.73	13.2	QICPMS	average	4.6	0
Block 3	3L	1	M	35-40	LLM2or3	50.59	7.0	QICPMS	average	1.5	1
Block 3	3L	2	M	20-25	URM2	55.45	0.8	QICPMS	average	0.8	2
Block 3	3L	3	PM	18-21	ULM2	65.51	14.1	QICPMS	average	3.4	0
Block 3	3L	4	M	18-25	URPM4	55.88	20.9	QICPMS	average	4.8	0
Block 3	3L	7	M	30-35	LLPM4	55.45	1.5	QICPMS	average	3.7	0
Block 3	3L	8	F	35-45	LLM2	33.87	4.6	QICPMS	average	0.9	2
Block 3	3L	9	M	25-40	ULPM4	46.67	9.1	QICPMS	average	2.3	1
Block 3	3L	10	M	30-34	LLM2	38.94	0.3	QICPMS	average	3.0	0
Block 3	3L	12	M	20-40	LLM2	41.33	0.9	QICPMS	average	0.5	3
Block 3	3L	13	IND	16-19	ULM2	53.03	7.3	QICPMS	average	1.2	1
Block 3	3L	15	PF	25-30	ULM2	28.96	4.0	QICPMS	average	2.4	1
Block 3	3L	16	M	25-39	ULM2	58.96	2.8	QICPMS	average	0.6	2
Block 3	3L	17	M	30-39	LRM2	11.72	1.1	QICPMS	average	3.4	0
Block 3	3L	18	M	<45	LRM2	48.36	2.1	QICPMS	average	1.5	1
Block 3	3L	19	M	17-18	URPM4	49.25	2.0	QICPMS	average	0.6	2
Block 3	3L	20	M	20-35	URM2	52.09	0.2	QICPMS	average	0.3**	3
Block 3	3L	23	M	30-45	ULM2	6.51	3.1	QICPMS	average	0.4	3
Block 3	3L	25	PM	20-40	URPM3	8.45	28.3	QICPMS	average	5.7	0
Block 3	3L	44	M	27-35	LLPM4	46.59	11.8	QICPMS	average	2.1	1
Southside	CjAe-54	A1	PF	MA	LLM2	20.00	0.6	QICPMS	actual	0.3	3
Southside	CjAe-54	A2	PM	MA	LLM2	48.07	18.7	QICPMS	actual	5.9	0
Southside	CjAe-54	A3	PM	MA	LLM2	46.41	12.4	QICPMS	actual	6.5	0
Southside	CjAe-54	A4	M	MA	LRM2	59.77	1.5	QICPMS	actual	1.6	0
Southside	CjAe-54	A5	IND	IND	LLM1*	67.18	5.6	QICPMS	actual	1.9	0

Southside	CjAe-54	A6	PM	IND	LLM2	32.28	24.6	QICPMS	actual	3.7	0
Southside	CjAe-54	A7	IND	IND	LRM2	68.72	1.3	QICPMS	actual	1.0	1
Southside	CjAe-54	A8	IND	IND	LRM2	58.49	2.9	QICPMS	actual	3.8	0
Southside	CjAe-54	A9	IND	IND	LLM2	37.13	0.9	QICPMS	actual	0.7	1
Southside	CjAe-54	A10	PM	IND	LLM2	49.27	5.3	QICPMS	actual	5.8	0
Southside	CjAe-54	B1	PM	IND	LLM2	12.45	3.7	QICPMS	actual	1.0	1
Southside	CjAe-54	B2	IND	IND	LLM2	45.15	1.8	QICPMS	actual	2.2	0
Southside	CjAe-54	B4	PM	IND	LLM2	33.04	10.3	QICPMS	actual	6.7	0
Southside	CjAe-54	B5	IND	IND	LLM2	51.75	1.1	QICPMS	actual	1.1	1
Southside	CjAe-54	B6	IND	IND	LRM2	47.78	1.0	QICPMS	actual	0.9	1
Southside	CjAe-54	B8	IND	IND	LRM1	24.57	4.8	QICPMS	actual	3.2	0
Southside	CjAe-54	B9	PM	IND	LLM2	35.82	15	QICPMS	actual	3.5	0
Southside	CjAe-54	B10	IND	IND	LLM2	42.29	0.2	QICPMS	actual	0.3	4
Southside	CjAe-54	B11	IND	IND	LRM2	33.93	1.8	QICPMS	actual	1.5	1

IND: Indeterminate – but for age, only those with at least second molars fully erupted were included for analyses; ** small voltage; PM or PF: possible male or possible female sex estimation.

16 L/min, the nebulizer gas flow was 1.16 L/min, and the auxiliary was 1.2 L/min. Pb calibration and concentration methods were a modified version of Friel et al. (1990) where different multi-element solutions are used for external calibration and Sc, Re, Rh and Th are used to monitor instrumental drift. Pb concentration calibration solution was 10.11 ppb and was run at the beginning and end of the run, and between every eight unknown samples. USGS T-143 is a standard water reference used as a check standard.

For data acquired at uOttawa, the solution was diluted 10X and 50X due to the high concentration of the initial solution. The concentration of Pb was determined with solution ICP-Q-MS on an Agilent triple quadrupole ICP-MS based on the sum of Pb isotope counts. Approximately every 11 samples a check standard and blank were performed. The limit of detection for Pb was 0.0022 ppb and the sample blank had a maximum of 1% of to sample concentration. Sample concentrations were determined using ^{204}Pb, ^{206}Pb, ^{207}Pb, and ^{208}Pb, but sum counts of the isotopes were used as the final concentration. Calibration curve was determined at the beginning of the run using solutions of six different concentrations (blank, 6.25, 12.5, 25, and 100 ppb), with a formula was $y=76165.15x-10809.37$ ($r^2 = 0.998$). The solution of 12.5 ppb was used as a check standard during the run.

Due to instrument problems, concentrations were estimated for some of the more recently analyzed samples. This was done using the ^{208}Pb voltage from the MC-ICP-MS of each sample. Each run had a known concentration of the SRM 981 solution. The known concentration was divided by the averaged voltage of the standard solution on either side of the bracket. This gave the ppb/V which was multiplied by the voltage of the

sample to give the raw ppb value of the solution. This was then converted to concentration of the sample using the known sample mass and density of solution.

For Pb concentration analyses at the TERRA facility, the procedural blanks were usually below the method detection limit or just above (0.03–0.05 ppb). Since the final concentrations are a product of the sample weight, a better sense of how large of an impact the blank had on the samples. Using the highest blank per analysis, the median of the 72 samples was 0.06%, but the highest value was 1.82%. The blank correction was not done for the TERRA samples since it did not make an interpretable difference to the data (<2% of final concentrations). For those analyzed at uOttawa, the limit of detection was 0.0022 ppb and the mean blank was 0.024 ppb (maximum of 0.026 ppb). For the nine samples analyzed, the procedural blank contributed to a median of 1.1%, with a maximum of 23% for Burial 5 of Wester Point; no other samples were above 2.1%. All samples from uOttawa were blank corrected. Depending on what data was used (dilution factor of 10X or 50X), the concentration was between 0.09 ppm and 0.19 ppm. Since this was the lowest concentration found during these analyses, it was deemed inappropriate to ignore that such a low exposure level was found. Instead, caution was used during the interpretation of this data. While the blank was a similar concentration to those analyzed in the TERRA facility, less enamel was sampled (~10 mg rather than ~50 mg), so it contributed more to the final sample concentrations.

Replicates were run on ~25% (n = 15) of the samples at 100x dilution rather than 10x, and the within assay variability (coefficient of variation) was 2.99%. Replicates were run on all analyses (n = 10) at uOttawa using 10X and 50X dilution and the within assay variability was 3.47%. The between assay variability was determined using two

standard reference materials at the Memorial University TERRA facility, USGS T-143 and SRM 1400. The accuracy obtained against the standard values in this study were 105.6% for USGS T-143 (mean ± 2sd; 88.08 ± 2.94, n = 8), 90.5% for SRM 1400 (8.21 ± 0.12, n = 5), and 100.3% for uOttawa's internal standard (12.54 ± 0.68, n = 4) (Table 21; Table 22). The between assay variability for the SRM 1400 concentrations determined using MC-ICP-MS were almost three times higher than those determined with the Q-ICPMS (Table 23). This increased error is likely caused in part by the solution being the elution of the column chemistry. There is significant variability possible from sample loss during the column chemistry, and the solution density is based on the initial sample loaded, not the final analysis.

Table 21. The summarized Pb concentration values for USGS T-143 used as the TERRA check standard and the 12.5 ppb solution used by University of Ottawa. The TERRA values run over seven days of analyses and the University of Ottawa on one day.

	USGS T-143	**uOttawa 12.5 ppb**
Most probable Pb concentration (ppb)	83.4	12.5
Mean Pb concentration (ppb)	88.08	12.54
count	8	4
1σ	2.21	0.34
% error	5.61	0.34

Table 22. The summarized Pb concentration values for all SRM 1400 analyses. This includes the replicate as well as the summary statistics ran over four days of Q-ICPMS analyses. For MC-ICP-MS, this was ran over two days.

SRM 1400	**Q-ICPMS**	**MC-ICP-MS**
Certified Pb concentration (ppm)	9.07 ± 0.12	9.07 ± 0.12
Mean Pb concentration (ppm)	8.23	12.50
count	11	3
1σ	0.28	2.31
% error	10.26	27.46

7.2.4.3 Pb isotope methodology and errors

The remaining 1.5 mL of solution was used for Pb isotope analyses. Using fabricated columns, Pb was isolated by extraction chromatography using Sr-spec resin (Eichrom Technologies) (Charlier et al., 2006). Samples were prepared within groups of ten, including a blank and SRM 1400 as an internal standard. The column chemistry was based on Pin et al. (2014; see Figure 23 for flowchart) and allowed for the simultaneous extraction of Sr (Garlie, 2022) and Pb. Pb is eluted with 1.5 mL of 6.2 M HCl, dried down on the hotplate, then taken up in 0.8 mL of 0.3 M HNO_3 and transferred to a centrifuge tube for analysis.

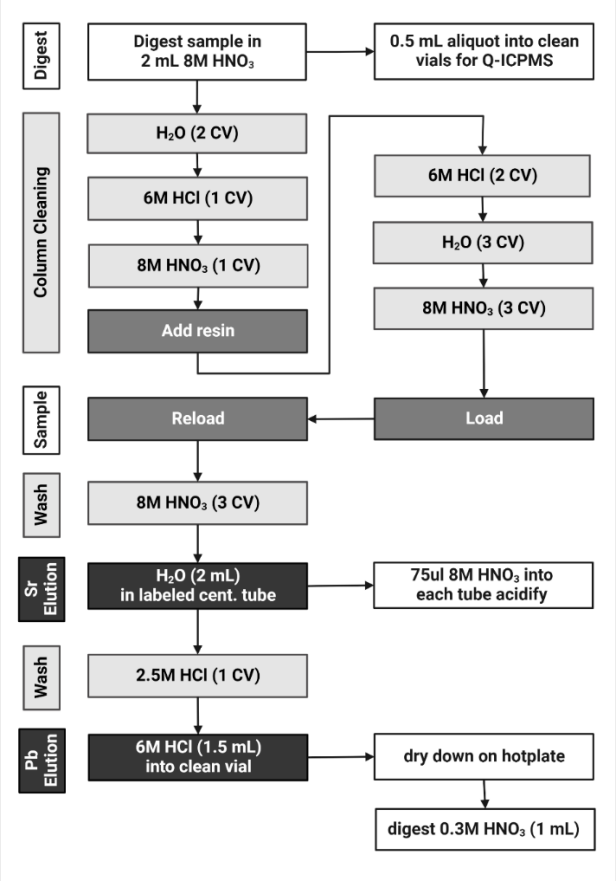

Figure 23. Flow chart of Pb elution column chemistry based on parameters from Pin et al. (2014).

The sample analysis parameters are found in Table 23. The baseline correction used a defocused beam measured for a 30 second (30 cycles with 1 second integration time) The analytical measurement was 50 cycles with an 8.389 second integration time. To allow for calibration, a ~100 ppb solution of NIST SRM 981 (Common Pb isotopic standard) bracketing every three samples. Samples and standards were preceded by an acid wash using 0.3 N HNO_3 for until the ^{208}Pb signal came down to baseline (~5 min – but timing depended on the sample concentration) to minimise Pb memory from the previous sample. Data were corrected for Hg using 202 to calculate the amount of ^{204}Hg to subtract from the overall 204 signal, but the Hg correction was negligible in most cases. Samples were blank corrected (unless identified otherwise), and ideally, one carousel worth of data was run per day to account for changes in blanks. Data was then calibrated using the most probable values of SRM 981 from Baker et al. (2004) and De Munyck (2008) using a linear equation to account for any drift during analysis. Some samples with low solution concentrations were analyzed using a bracket of SRM 981 solution of ~12 ppb to reduce error during calibration corrections. These samples are indicated in Table 20.

The target voltage during analysis of ^{208}Pb was 4V, but not all solutions had sufficient concentration for this to be met. Since the blank to sample voltage did not exceed 4%, none of the samples were removed for consideration (Table 20). Two of the samples were bracket analyzed with an SRM 981 solution of ~12 ppb rather than 100 ppb since they were of a very low concentration.

Table 23. MC-ICP-MS analysis parameters on Thermo Neptune during eight days of analyses

MC-ICP-MS	
Spray chamber	Sis cleaned with sitranox and rinsed with 0.3 N nitric
Nebulizer	50 μm flushed with 0.3 N nitric
Cones	Nickel sampler cone and nickel "H" skimmer cone
Faraday array	202, 204, 205, 206, 207, 208
Auxillary gas flow rate	1.00 L/min
Sample argon gas flow rate	1.020 to 1.045 L/min
Zoom focus	+1.5 to +3
Source offset	-7 to -15
Baseline – defocused beam	30-cycles 1 sec. integration time
Analytical measurement	50 cycles, 8.389 sec. integration time
Software suite	Neptune Software, version 3.2.0.14

The isotopic data is based on runs over 11 non-consecutive days. The blanks had a mean voltage of ^{208}Pb 0.0135 ± 0.0072 (n = 23). The between assay variability is determined using SRM 1400. The accepted values are the most probable values as published by Hinners et al. (1998). Since Tl correction was not performed for the isotopic data, the standard errors are important for understanding accuracy. The summary values are found for each ratio analysed in Table 24.

Table 24. The summary statistics of the Pb isotope ratios for all SRM 1400 analyses ran over eight days, n = 20, including the most probable values according to Hinners et al. (1998).

	^{208}Pb/ ^{206}Pb	**^{206}Pb/ ^{207}Pb**	**^{208}Pb/ ^{204}Pb**	**^{207}Pb/ ^{204}Pb**	**^{206}Pb/ ^{204}Pb**	**^{208}Pb/ ^{207}Pb**	**^{207}Pb/ ^{206}Pb**
Most probable value	2.10209	NA	38.610	15.6702	18.3676	2.46389	0.85314
Count	20	20	20	20	20	20	20
Mean	2.10289	1.17162	38.61738	15.67407	18.36394	2.46427	0.85352
2σ	0.00176	0.00092	0.05852	0.01847	0.02176	0.00102	0.00067
% error	0.0439	NA	0.0191	0.0247	0.0199	0.0154	0.0446

Table 25. The summarized within assay variability determined through propagated error (2σ) of all enamel samples.

	$^{208}Pb/^{206}Pb$	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$	$^{208}Pb/^{207}Pb$
Count	64	64	64	64	64	64
Average	0.000132	0.000422	0.006501	0.003064	0.002964	0.000321
Min	0.000120	0.000417	0.03903	0.00156	0.001767	0.00031
Max	0.000193	0.000434	0.025071	0.015064	0.011914	0.000381

7.2.5 R code for statistical analyses and plots

Code used in R Statistical Software (v4.2.2; R Core Team, 2022) to perform statistical analyses are as follows using an example of $^{208}Pb/^{206}Pb$ data analysis and Pb concentration for the boxplot. Column name of comparative groupings is comp.

```
## ---- load data ----
Pbcomp <-read.csv("filename.csv")
#comp is the regional and or temporal groupings heading
#x208Pb.206Pb is the 208Pb/206Pb data heading
#Pb is the Pb concentration data heading

## ---- instal/load data ----
#The program dplyr is necessary for the data summary (Wickham et al., 2022), FSA for the Dunn test (Ogle et al., 2022), ggstatsplot (Patil, 2021) and for the boxplot, and tidyverse (Wickham et al., 2019), showtext (Qiu, 2023), and ggplot2 (Wickham, 2016) for additional changes to the plot.

#install.packages("put program name here") #use to instal, otherwise use code below

library (dyplyr)
library(FSA)
library(ggstatsplot)
library(tidyverse)
library(showtext)
library(ggplot2)

## ---- summarize data ----
group_by(Pbcomp, comp)%>%
```

```
  summarise(count=n (), mean= mean(x208Pb.206Pb, na.rm = 208PB/206PB
sd(X208Pb.206Pb, na.rm =y TRUE), median=median(x208Pb.206Pb, na.rm = TRUE),
IQR=IQR(208Pb/206Pb b, na.rm=TRUE))

## ---- Kruskal-wallis test ----
kruskal.test(x208Pb.206Pb ~ comp, data = Pbcomp)

## ---- Dunn Test ----
x208.206 =dunnTest(x208Pb.206Pb ~ comp, data=Pbcomp, method = "bh")
x208.206

## ----Boxplot----
## Prep data before plotting – gets rid of the over max overlaps issue, then add fonts
options(ggrepel.max.overlaps = Inf)

font_add_google("Roboto", "Roboto")
font_add_google("Open Sans", "Open Sans")
showtext_auto()

plt <- ggbetweenstats(data = Pbcomp, x = comp, y = Pb, type = "np",
  pairwise.comparisons = FALSE, results.subtitle = FALSE, centrality.plotting = FALSE,
)+
  labs(x = "omparative region or context",  y = "Pb concentration (ppm)"
)+
  theme(text = element_text(family = "Roboto", size = 10, color = "black"),
    plot.title = element_text(family = "Open Sans", size = 18, face = "bold", color =
"#1b2838"),
    plot.subtitle = element_text(family = "Roboto", size = 10, face = "bold", color="black"
    ),
    plot.title.position = "plot",
    axis.text = element_text(size = 12, color = "black"),
    axis.title = element_text(size = 14),
    axis.ticks = element_blank(),
    axis.line = element_line(colour = "grey50"),
    panel.grid = element_line(color = "#b4aea9"),
    panel.grid.minor = element_blank(),
    panel.grid.major.x = element_blank(),
    panel.grid.major.y = element_line(linetype = "dashed"),
)+
  coord_cartesian(ylim = c(0,125))+
  scale_color_manual(values = c("#8E0022", "#103356","#FEDA8B","#7889CF",
"#2D8CAC", "#C18402", "#F67E4B", "#9AA03C", "#FF7594",
"#000000","#B2182B", "#2166AC")
)
plt
```

7.2.6 Results

7.2.6.1 Concentration and isotope results

Initial Pb concentration results are found in Table 20 of the Appendix. Pb isotope ratio data and propagated errors are available in Table 26 and Table 27

7.2.6.2 Diagenesis

Kamenov et al. (2018) suggested that diagenetic alteration of the enamel is identifiable using concentrations of elements (Ca, V, Mn, Fe, La, Ce, Nd, Dy, Yb, Th, and U) compared to a maximum threshold concentration created using data from modern individuals. This specific method suggests that if the concentration to maximum threshold concentration (MTC) ratio is greater than one, some alteration had occurred; if many element's MTC were high, there was a concern for major alteration (Kamenov et al., 2018). Memorial University's TERRA facility's methodology analyses seven of the elements (Ca, V, Mn, Fe, La, Ce, and U). Data from Placentia, Louisbourg, and Southside cemeteries were available for this comparison, and full results are available in the Appendix as Table 28. Interpretation of the MTC values is within text in Chapter 3.4.1.

Table 26. Pb isotope values for teeth from Fishery and Royal Naval burials in Newfoundland and Nova Scotia. Sites include: Foxtrap-2; St. Paul's Anglican Church cemetery, Harbour Grace (St. Paul's); Tors Cove burial ground, Tors Cove (Tors Cove); Wester Point burial ground, Portugal Cove-St. Philip's (Wester Pt.); St. Luke's Anglican Church cemetery, Placentia (St. Luke's); Block 3 burial ground,$^{208}Pb/^{206}Pb$ Louisbourg (Block 3) Southside Cemetery, St. John's (Southside) (Part 1)

Site	Individual	$^{208}Pb/^{206}Pb$	prop. 2σ	$^{206}Pb/^{207}Pb$	prop. 2σ	$^{208}Pb/^{204}Pb$	prop. 2σ	$^{207}Pb/^{204}Pb$	prop. 2σ
Foxtrap-2	4	2.08686	0.00014	1.17898	0.00042	38.45487	0.00524	15.62972	0.00202
Foxtrap-2	7	2.08733	0.00013	1.17678	0.00042	38.39326	0.00499	15.63054	0.00189
Foxtrap-2	9	2.09501	0.00013	1.17119	0.00042	38.34300	0.00627	15.62704	0.00283
Foxtrap-2	10	2.0884	0.00013	1.17577	0.00042	38.36829	0.00529	15.62547	0.00198
Foxtrap-2	11	2.08558	0.00012	1.17931	0.00042	38.43219	0.00407	15.62597	0.00168
Foxtrap-2	12	2.08951	0.00014	1.18069	0.00042	38.65205	0.00498	15.66721	0.00204
Foxtrap-2	13	2.08794	0.00013	1.17718	0.00042	38.42706	0.00469	15.63428	0.00196
Foxtrap-2	14	2.08878	0.00013	1.17815	0.00042	38.52744	0.00466	15.6558	0.00194
Foxtrap-2	30	2.08629	0.00012	1.17722	0.00042	38.35905	0.00404	15.61847	0.00161
St. Paul's	NP250/F3	2.08578	0.00013	1.17811	0.00042	38.39700	0.00510	15.62612	0.00215
St. Paul's	NP251/F4	2.08652	0.00013	1.17751	0.00042	38.38500	0.00427	15.62335	0.00176
St. Paul's	NP254/F7	2.05484	0.00013	1.20651	0.00043	38.85648	0.00593	15.67314	0.00268
St. Paul's	NP258/F13	2.08702	0.00013	1.17695	0.00042	38.38174	0.00488	15.62557	0.00218
St. Paul's	NP262/F21	2.08475	0.00013	1.18002	0.00042	38.45518	0.00455	15.63177	0.00174
St. Paul's	NP263A/F22	2.08601	0.00013	1.17754	0.00042	38.37211	0.00429	15.62157	0.00166
St. Paul's	NP264 A/3	2.08682	0.00012	1.17856	0.00042	38.45690	0.00410	15.63635	0.00160
St. Paul's	NP264 B/2	2.08954	0.00012	1.17539	0.00042	38.39133	0.00447	15.63155	0.00184
Tors Cove	NP 2	2.08659	0.00012	1.17631	0.00042	38.32422	0.00516	15.61404	0.00258
Tors Cove	NP 54	2.05841	0.00012	1.20350	0.00043	38.82001	0.00406	15.67021	0.00163
Tors Cove	NP 68	2.08755	0.00016	1.17440	0.00042	38.29020	0.01250	15.61838	0.00440
Wester Pt.	2	2.08651	0.00012	1.17734	0.00042	38.37790	0.00420	15.62238	0.00159
Wester Pt.	4	2.08851	0.00012	1.17559	0.00042	38.37380	0.00420	15.62946	0.00158
St. Luke's	62A	2.09182	0.00012	1.17255	0.000419	38.32839	0.00419	15.62666	0.00170
St. Luke's	63	2.08763	0.00012	1.17625	0.000421	38.36161	0.00444	15.62223	0.00173

St. Luke's	66B	2.08751	0.00012	1.17626	0.000421	38.36218	0.00439	15.62327	0.00184
St. Luke's	67	2.08730	0.00012	1.17646	0.00042	38.36861	0.00408	15.62486	0.00160
Block 3	1	2.08561	0.00013	1.17813	0.000422	38.41763	0.00624	15.63503	0.00301
Block 3	2	2.08168	0.00013	1.18101	0.000424	38.38660	0.00844	15.61384	0.004670
Block 3	3	2.08264	0.00014	1.17997	0.000422	38.40719	0.00525	15.62887	0.00194
Block 3	4	2.08223	0.00015	1.18075	0.000422	38.43261	0.00548	15.63230	0.00190
Block 3	7	2.08252	0.00012	1.18079	0.00042	38.42265	0.00451	15.62496	0.00196
Block 3	8	2.08400	0.00014	1.17988	0.000423	38.46574	0.00860	15.64372	0.00462
Block 3	9	2.08442	0.00013	1.17884	0.000421	38.41788	0.00475	15.63481	0.00221
Block 3	10	2.08418	0.00013	1.17903	0.00042	38.40874	0.00475	15.63033	0.00210
Block 3	12	2.08236	0.00014	1.18025	0.000424	38.35517	0.01311	15.60614	0.00759
Block 3	13	2.08627	0.00013	1.17811	0.000422	38.40934	0.00570	15.62713	0.00274
Block 3	15	2.08322	0.00013	1.17937	0.000422	38.37172	0.00523	15.61729	0.00243
Block 3	16	2.08508	0.00014	1.17859	0.000423	38.45326	0.01028	15.64781	0.00565
Block 3	17	2.08362	0.00012	1.18041	0.00042	38.44018	0.00438	15.62910	0.00188
Block 3	18	2.08296	0.00013	1.18037	0.000422	38.41864	0.00549	15.62568	0.00256
Block 3	19	2.08356	0.00014	1.18002	0.000424	38.41548	0.00916	15.62435	0.00484
Block 3	20	2.07889	0.00018	1.18588	0.00043	38.57847	0.01935	15.64841	0.00911
Block 3	23	2.08503	0.00015	1.17852	0.000425	38.37199	0.01318	15.61541	0.00708
Block 3	25	2.08278	0.00012	1.17981	0.00042	38.38276	0.00409	15.6204	0.00159
Block 3	44	2.08571	0.00013	1.17864	0.00042	38.45411	0.00556	15.6430	0.00239
Southside	A1	2.08883	0.00017	1.17454	0.00043	38.33220	0.02099	15.6241	0.01206
Southside	A2	2.08326	0.00012	1.18003	0.00042	38.41089	0.00457	15.62489	0.00176
Southside	A3	2.08450	0.00012	1.17981	0.00042	38.42675	0.00390	15.62480	0.00161
Southside	A4	2.08399	0.00013	1.18020	0.00042	38.42977	0.00583	15.62498	0.00287
Southside	A5	2.08938	0.00013	1.17468	0.00042	38.35897	0.00485	15.62862	0.00226
Southside	A6	2.08092	0.00012	1.18230	0.00042	38.44483	0.00439	15.62623	0.00199
Southside	A7	2.09003	0.00013	1.17401	0.00042	38.31852	0.00695	15.61616	0.00354
Southside	A8	2.09930	0.00012	1.16673	0.00041	38.26809	0.00433	15.62404	0.00188
Southside	A9	2.08694	0.00013	1.17556	0.00042	38.34177	0.00883	15.62780	0.00502

Southside	A10	2.08735	0.00012	1.17655	0.00042	38.37336	0.00421	15.62541	0.00169
Southside	B1	2.08184	0.00013	1.18060	0.00042	38.40799	0.00699	15.62721	0.00366
Southside	B2	2.08119	0.00012	1.18100	0.00042	38.39987	0.00479	15.62288	0.00228
Southside	B4	2.08465	0.00012	1.17806	0.00042	38.37475	0.00407	15.62596	0.00156
Southside	B5	2.08943	0.00013	1.17507	0.00042	38.31989	0.00681	15.60703	0.00356
Southside	B6	2.08692	0.00013	1.17637	0.00042	38.35982	0.00728	15.62554	0.00374
Southside	B8	2.08646	0.00019	1.17733	0.00042	38.39309	0.00736	15.62972	0.00313
Southside	B9	2.08438	0.00013	1.17875	0.00042	38.42352	0.00442	15.63839	0.00188
Southside	B10	2.08346	0.00019	1.18005	0.00043	38.45102	0.02507	15.64074	0.01506
Southside	B11	2.08872	0.00013	1.17424	0.00042	38.33937	0.00581	15.63172	0.00279

Table 27. Pb isotope values for teeth from Fishery and Royal Naval burials in Newfoundland and Nova Scotia (Part 2)

Site	individual	$^{206}Pb/^{204}Pb$	prop. 2σ	$^{208}Pb/^{207}Pb$	prop. 2σ	$^{207}Pb/^{206}Pb$	prop. 2σ
Foxtrap-2	4	18.42689	0.00209	2.46089	0.00032	0.84819	0.00033
Foxtrap-2	7	18.39379	0.00210	2.45680	0.00032	0.84977	0.00033
Foxtrap-2	9	18.30174	0.00271	2.45415	0.00032	0.85383	0.00033
Foxtrap-2	10	18.37206	0.00234	2.45595	0.00032	0.85051	0.00033
Foxtrap-2	11	18.42769	0.00184	2.46000	0.00031	0.84795	0.00033
Foxtrap-2	12	18.49818	0.00198	2.46757	0.00032	0.84696	0.00033
Foxtrap-2	13	18.40463	0.00200	2.45833	0.00032	0.84949	0.00033
Foxtrap-2	14	18.44541	0.00206	2.46135	0.00032	0.84879	0.00034
Foxtrap-2	30	18.38619	0.00194	2.45650	0.00031	0.84946	0.00033
St. Paul's	NP250/F3	18.40910	0.002327	2.45774	0.00032	0.84882	0.00034
St. Paul's	NP251/F4	18.39665	0.00190	2.45736	0.00031	0.84925	0.00033
St. Paul's	NP254/F7	18.90988	0.00259	2.47965	0.00032	0.82883	0.00032
St. Paul's	NP258/F13	18.39043	0.00224	2.45682	0.00032	0.84966	0.00034
St. Paul's	NP262/F21	18.44587	0.00195	2.46052	0.00032	0.84744	0.00032
St. Paul's	NP263A/F22	18.39500	0.00183	2.45687	0.00032	0.84922	0.00032

St. Paul's	NP264 A/3	18.42810	0.00184	2.45992	0.00031	0.84849	0.00033
St. Paul's	NP264 B/2	18.37327	0.00195	2.45652	0.00031	0.85078	0.00032
Tors Cove	NP 2	18.36697	0.00240	2.45497	0.00032	0.85012	0.00033
Tors Cove	NP 54	18.85928	0.00191	2.47779	0.00032	0.83091	0.00031
Tors Cove	NP 68	18.34251	0.00587	2.45216	0.00033	0.85150	0.00042
Wester Pt.	2	18.39309	0.00188	2.45701	0.00031	0.84937	0.00032
Wester Pt.	4	18.37370	0.00182	2.45568	0.00031	0.85064	0.00032
St. Luke's	62A	18.32291	0.00189	2.45323	0.00031	0.85284	0.00033
St. Luke's	63	18.37558	0.00200	2.45608	0.00031	0.85016	0.00033
St. Luke's	66B	18.37675	0.00198	2.45593	0.00031	0.85013	0.00033
St. Luke's	67	18.38235	0.00185	2.45609	0.00031	0.85001	0.00032
Block 3	1	18.42042	0.00282	2.45762	0.00032	0.84881	0.00035
Block 3	2	18.43951	0.00391	2.45899	0.00033	0.84673	0.00037
Block 3	3	18.44175	0.00219	2.45792	0.00032	0.84748	0.00034
Block 3	4	18.45808	0.00221	2.45904	0.00032	0.84692	0.00034
Block 3	7	18.45001	0.00215	2.45952	0.00032	0.84689	0.00033
Block 3	8	18.45771	0.00411	2.45933	0.00032	0.84755	0.00037
Block 3	9	18.43134	0.00215	2.45768	0.00032	0.84829	0.00033
Block 3	10	18.42825	0.00220	2.45781	0.00032	0.84815	0.00032
Block 3	12	18.41918	0.00646	2.45819	0.00033	0.84728	0.00039
Block 3	13	18.41117	0.00258	2.45835	0.00032	0.84881	0.00036
Block 3	15	18.41951	0.00236	2.45733	0.00032	0.84791	0.00033
Block 3	16	18.44244	0.00474	2.45791	0.00033	0.84847	0.00037
Block 3	17	18.44863	0.00203	2.46001	0.00031	0.84716	0.00032
Block 3	18	18.44387	0.00258	2.45912	0.00032	0.84719	0.00034
Block 3	19	18.43759	0.00444	2.45910	0.00033	0.84745	0.00039
Block 3	20	18.55886	0.00904	2.46582	0.00038	0.84326	0.00050
Block 3	23	18.40371	0.00598	2.45767	0.00035	0.84852	0.00042
Block 3	25	18.42908	0.00184	2.45775	0.00031	0.84760	0.00032
Block 3	44	18.43737	0.00238	2.45878	0.00032	0.84844	0.00034

Southside	A1	18.35133	0.01001	2.45385	0.00035	0.85139	0.00049
Southside	A2	18.43782	0.00207	2.45878	0.00031	0.84744	0.00033
Southside	A3	18.43435	0.00177	2.45980	0.00031	0.84760	0.00031
Southside	A4	18.44015	0.00272	2.46000	0.00032	0.84731	0.00035
Southside	A5	18.35902	0.00228	2.45487	0.00032	0.85130	0.00033
Southside	A6	18.47474	0.00204	2.46076	0.00032	0.84581	0.00032
Southside	A7	18.33402	0.00335	2.45421	0.00032	0.85178	0.00036
Southside	A8	18.22898	0.00195	2.44979	0.00032	0.85710	0.00033
Southside	A9	18.37252	0.00398	2.45383	0.00032	0.85066	0.00037
Southside	A10	18.38400	0.00191	2.45634	0.00031	0.84994	0.00032
Southside	B1	18.44952	0.00336	2.45830	0.00032	0.84702	0.00035
Southside	B2	18.45074	0.00226	2.45839	0.00032	0.84674	0.00033
Southside	B4	18.40850	0.00186	2.45631	0.00031	0.84885	0.00032
Southside	B5	18.33941	0.00327	2.45571	0.00032	0.85101	0.00035
Southside	B6	18.38120	0.00344	2.45543	0.00032	0.85007	0.00036
Southside	B8	18.40132	0.00249	2.45693	0.00034	0.84938	0.00038
Southside	B9	18.43404	0.00200	2.45743	0.00032	0.84836	0.00033
Southside	B10	18.45554	0.01191	2.45910	0.00038	0.84742	0.00059
Southside	B11	18.35545	0.00258	2.45315	0.000315	0.85162	0.00035

Table 28. The ratio of concentration to Maximum Threshold Concentration (MTC) of six elements as suggested by Kamenov et al. (2018 *JAS* 99) and the Pb concentration of each individual. Values above a ratio of 1, which suggests diagenetic alteration, are bolded.

		Ca (ppm/ MTC)	V (ppm/ MTC)	Mn (ppm/ MTC)	Fe (ppm/ MTC)	La (ppm/ MTC)	Ce (ppm/ MTC)	U (ppm/ MTC)	Pb (ppm)
MTC ppm			**0.11**	**15.4**	**143**	**0.1**	**0.12**	**0.05**	
Site	**Individual**								
St. Luke's	62A		<DL	0.17	<DL	0.12	0.05	0.01	4.3
St. Luke's	63		**1.83**	0.10	<DL	0.29	0.05	0.01	14.4
St. Luke's	66B		<DL	0.08	<DL	0.39	0.15	0.02	8.4
St. Luke's	67		**1.26**	0.11	<DL	0.58	0.13	0.02	13.2
Block 3	1		<DL	0.37	<DL	0.10	0.12	0.01	7.0
Block 3	2		<DL	0.56	<DL	0.41	0.31	0.37	0.8
Block 3	3		<DL	**1.39**	<DL	0.17	0.17	0.10	14.1
Block 3	4		<DL	0.99	<DL	0.17	0.21	0.04	20.9
Block 3	7		**1.66**	**1.10**	<DL	0.07	0.05	0.02	1.5
Block 3	8		<DL	0.21	<DL	0.05	0.05	<DL	4.6
Block 3	9		<DL	0.20	<DL	0.03	0.02	0.01	9.1
Block 3	10		**3.96**	**1.49**	<DL	0.07	0.06	0.70	0.3
Block 3	12		<DL	0.65	<DL	0.23	0.23	<DL	0.9
Block 3	13		<DL	**1.76**	<DL	0.07	0.06	0.02	7.3
Block 3	15		<DL	**2.59**	<DL	0.10	0.07	<DL	4.0
Block 3	16		<DL	0.27	<DL	0.32	0.21	0.27	2.8
Block 3	17		<DL	0.38	<DL	0.05	0.04	<DL	1.1
Block 3	18		<DL	**1.43**	<DL	0.09	0.14	<DL	2.1
Block 3	19		<DL	**2.81**	<DL	0.66	**1.03**	0.03	2.0
Southside	A1		<DL	0.34	<DL	0.05	0.04	<DL	0.6
Southside	A2		<DL	0.06	<DL	0.08	0.10	<DL	19.0

Southside	A3		<DL	0.06	<DL	0.02	0.01	<DL	12.3
Southside	A4		<DL	0.64	<DL	0.09	0.09	<DL	1.5
Southside	A5		<DL	0.11	<DL	0.18	0.19	<DL	5.7
Southside	A6		<DL	0.13	<DL	0.24	0.21	<DL	24.6
Southside	A7		<DL	0.18	<DL	0.04	0.03	<DL	1.3
Southside	A8		<DL	0.12	<DL	0.02	0.02	<DL	2.9
Southside	A9		<DL	0.42	<DL	0.12	0.07	<DL	0.9
Southside	A10		<DL	0.18	<DL	0.06	0.03	<DL	5.3
Southside	B1		<DL	0.59	<DL	**1.78**	**1.28**	<DL	3.7
Southside	B2		<DL	0.05	<DL	0.16	0.08	<DL	1.8
Southside	B4		<DL	0.89	<DL	0.22	0.21	<DL	10.3
Southside	B5		<DL	0.22	<DL	0.22	0.18	<DL	1.1
Southside	B6		<DL	0.07	<DL	0.01	<DL	<DL	1.0
Southside	B8		<DL	0.20	<DL	0.12	0.11	<DL	4.8
Southside	B9		<DL	**1.61**	<DL	0.11	0.07	<DL	15.0
Southside	B10		<DL	0.10	<DL	0.06	0.04	<DL	0.2
Southside	B11		<DL	**1.02**	<DL	0.26	0.08	<DL	1.8

7.2.6.3 Statistical results

There was no difference in concentrations between the fishery populations ($p = 0.201$, Kruskal-Wallis chi-squared = 7.2874, $df = 5$). The results of comparisons of Pb isotope data in the fisheries using a Kruskal-Wallis chi-squared and Dunn-test are in Table 29 and Table 30, respectively. There were differences between these populations ($p < 0.001$, Kruskal-Wallis chi-squared = 320.43, $df = 11$), but since these were populations from different periods, we could reject that these were coming from the same exposure situations. In turn, a Dunn Test was performed to identify what pairs of sites had different exposures (Table 5). Comparison of all Pb isotope ratios using a Kruskal-Wallis and then Dunn test are in Table 31 and Table 32, respectively .

Table 29. Results of Kruskal-Wallis chi-squared test comparing the Pb isotope ratios of the fishery sites.

	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$
Kruskal-Wallis	25.861	18.186	9.4857	3.4922	15.507
df	5	5	5	5	5
***p*-value**	<0.001	0.003	0.091	0.625	0.008

Table 30. *p-value* results of a Dunn-test comparing the Pb isotope ratios (denoted by their isotopes only) of the fishery sites.

	Foxtrap-2			St. Luke's			St. Paul's			Tors Cove			Wester Point		
	208/206	**207/206**	**206/204**	**208/206**	**207/206**	**206/204**	**208/206**	**207/206**	**206/204**	**208/206**	**207/206**	**206/204**	**208/206**	**207/206**	**206/204**
Block 3	0.000	0.068	0.181	0.005	0.009	0.018	0.140	0.239	0.292	0.369	0.280	0.281	0.111	0.133	0.256
Foxtrap-2				0.901	0.347	0.255	0.313	0.716	0.810	0.284	0.919	0.825	0.899	0.673	0.528
St. Luke's							0.296	0.206	0.250	0.303	0.440	0.552	0.948	0.830	0.809
St. Paul's										0.863	0.878	0.717	0.522	0.455	0.465
Tors Cove													0.522	0.640	0.763

Table 31. Results of Kruskal-Wallis test comparing the Pb isotope ratios of the Block 3 cemetery (Louisbourg) and remaining Newfoundland fishery sites to the Southside Cemetery and other previously published temporally contemporary sites detailed in Table 17.

	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$
Kruskal-Wallis chi-squared	86.43	74.226	72.082	47.935	75.452
df	7	7	8	8	8
***p*-value**	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001

Table 32. *p-value* results of a Dunn-test comparing the Pb isotope ratios of the Block 3 cemetery (Louisbourg) and remaining Newfoundland (NL) fishery sites to the Southside Cemetery and other previously published temporally contemporary sites detailed in Table 17..

Comparison sites		***p*-value**				
		$^{208}Pb/^{206}Pb$	**$^{207}Pb/^{206}Pb$**	**$^{208}Pb/^{204}Pb$**	**$^{207}Pb/^{204}Pb$**	**$^{206}Pb/^{204}Pb$**
NL fishery cemetery sites	**Block 3**	0.005	0.023	0.349	0.904	0.067
	Royal Navy	0.357	0.708	0.422	0.488	0.840
	Late Medieval	0.038	0.036	0.123	0.023	0.739
	Barbados	<0.001	<0.001	0.106	0.390	<0.001
	Connecticut	NA	NA	0.472	0.821	0.170
	New Zealand	0.560	0.180	0.008	0.023	0.169
	Indust. USA	<0.001	<0.001	<0.001	0.024	<0.001
	Indust. Eng.	<0.001	0.004	0.956	0.168	0.065
Block 3 cemetery, Louisbourg NS	**Royal Navy**	0.075	0.079	0.108	0.486	0.135
	Late Medieval	0.815	0.735	0.029	0.022	0.370
	Barbados	0.038	0.047	0.508	0.496	0.051
	Connecticut	NA	NA	0.891	0.821	0.717
	New Zealand	0.040	0.379	0.127	0.046	0.730
	Indust. USA	<0.001	<0.001	0.001	0.048	<0.001
	Indust. Eng.	0.802	0.864	0.356	0.169	0.804

7.2.6.4 Supplemental graphs

Not all graphical representations of the data were appropriate within the text, but many add meaningful representation of the data (Figure 24) or show baseline data used in production of the Pb ore ranges (Figure 25).

7.3 Supplementary Materials Chapter 4

7.3.1 Pb isotopes

Pb isotope ratios have long been applied to determine the geographic origin of Pb used within cultural materials and human remains (Kowal et al., 1991; Carlson, 1996; Budd et al., 2004; Killick, 2020). Four isotopes of Pb are typically measured as part of bioarchaeological applications of this technique: ^{206}Pb, ^{207}Pb, and ^{208}Pb, which are radiogenic, and ^{204}Pb, which is primordial (Faure and Mensing, 2005). The different original concentrations of parent isotopes in rocks (^{238}U, ^{235}U, ^{232}Th), the age since formation (half-lives), and mixing during formation create the variability of Pb isotope ratios within geological formations (Adriano, 1986; Gulson, 1986; Teutsch et al., 2001; Faure and Mensing, 2005). There is minute measurable but not meaningful isotopic fractionation of Pb isotopes during smelting and processing into cultural materials (Budd et al., 1995; Stos-Gale and Gale, 2009; Cui and Wu, 2010). Nor is there measurable fractionation when incorporated into humans where it is stored in their bones and teeth (Rabinowitz and Wetherill, 1972; Gulson, 1986).

Pb is incorporated into human tissue through exposure mainly by consumption of inhalation of Pb from the very small amounts of the metal that naturally occur within the

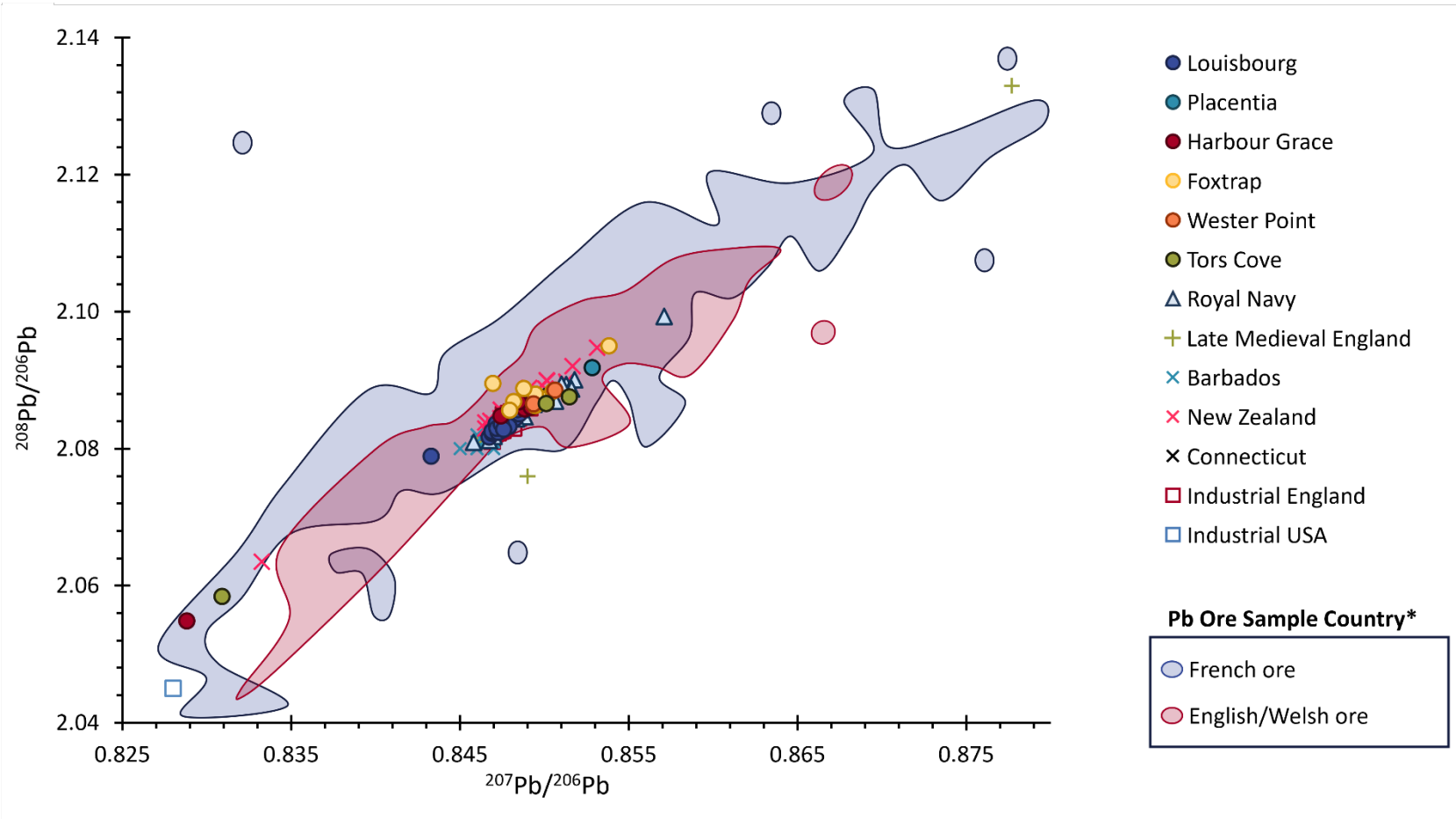

Figure 24. Comparison of $^{208}Pb/^{206}Pb$ and $^{207}Pb/^{206}Pb$ values between Newfoundland fisheries, Royal Navy, and other previously published contemporaneous archaeological sites. Pb ore field isotope value ranges for France and Britain are outlined. All comparative citations are available in Table 4 and Pb ore citations are in Table 5.

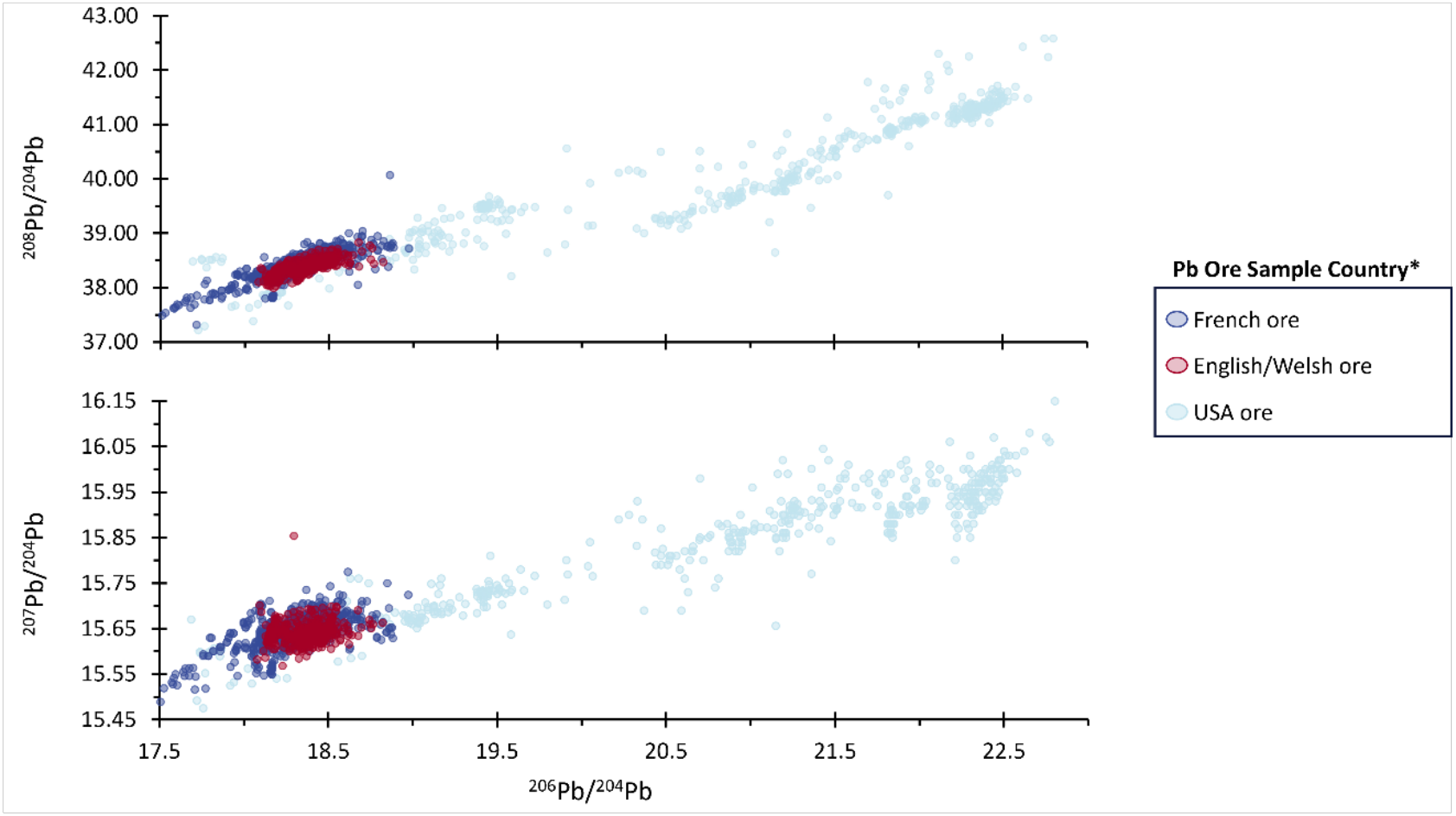

Figure 25. Pb isotope ratios of compiled ore data from Western Europe (Blichert-Toft et al., 2016) and USA (see Table 19 for full citations). Relevant comparative Pb ore from USA, France, England, and Wales are presented

local geology (Adriano, 1986; Brännvall et al., 1999). However, anthropogenic practices such as mining and smelting can contribute to the Pb burden within the environment (Gulson et al., 2004). By using the Pb within cultural materials, such as ceramic glazes, the metal can leach into other materials, such as the food or drink consumed from those materials. While humans are still exposed to the small amounts that naturally occur in the environment, they are also exposed to this anthropogenic Pb resulting in much higher Pb concentrations and isotope values that reflect a weighted average of those sources. In turn, the anthropogenic will often mask any of the natural exposure, particularly if concentration in teeth is above 0.6–0.8ppm (Millard et al., 2014). Rather than directly linking to the underlying geology, Pb isotope ratios may reflect an average of available sources due to cultural focusing, a concept established by Montgomery et al. (2005) for the similarity of ratios in individuals within a cultural region. In turn, the ratios within materials can be compared to those of likely ore bodies to exclude sources with which they are inconsistent.

Key to this sourcing is having good baselines of expected values and understanding where they may have originated. However, determining what geological or environmental Pb sources to consider when delimiting the expected 'local' range of Pb isotope ratios is not straightforward, and there is disagreement on what approach to take (see Chapter 2 for further discussion). Bioarchaeologists employ this process using human teeth identify if a child was not raised locally and suggest possible origins (e.g., Montgomery et al., 2005; Fitch et al., 2012; Valentine et al., 2015; Laffoon et al., 2020). Pb isotope ratios can help identify non-local individuals in cases where their values are outside the expected local ranges and or value from underlying geology, which are

determined when exposure occurred in non-anthropogenic, or 'natural' environments by using faunal remains. In anthropogenic contexts – as is relevant in this case – the ranges are determined by characterizing isotope ratios of local ore sources (Montgomery et al., 2005; Evans et al. 2018a; Samuelsen and Potra, 2020).

Most of the research regarding anthropogenic Pb sourcing within bioarchaeology is from England. While many of these are Romano-British studies (Montgomery et al., 2010; Shaw et al., 2016), there are also a number of studies from the medieval or industrial periods (e.g., Montgomery et al., 2005; Millard et al., 2014). Since England was a prominent producer of Pb and only began importing substantial amounts of Pb in the mid- to late-1800s (Burt, 1984), it is logical to use Pb isotopes from ores within England to create their baselines. In either case, Pb isotope ratios can be used to identify outliers that do not fit within the available Pb to a region.

Except for the Franklin expedition (Kowal et al., 1991), studies have not examined the Pb isotope ratios of materials available to a cultural group. Some Roman studies have used Pb isotope ratios in coins to suggest regions of origin for individuals (Montgomery et al., 2010; Shaw et al., 2016), with the argument being that the geographic origin of a coin is within the same region as the geological source of Pb used in that coin. Additionally, that the same Pb ore source was used in other cultural materials that contributed directly to human Pb exposure. While this may be reasonable, it is a way that researchers have approached typifying Pb values for different occupied regions in areas where Pb isotopes of ore bodies were not readily available. A handful of studies have suggested the specific materials that may have resulted in exposure. Laffoon et al. (2020)

even linked values consistent with England to locally born Barbadian enslaved labourers, but this approach is the minority.

7.3.2 Imported ceramics in 17th- to 19th- century Atlantic World fishing communities

Throughout the 16th to late-18th centuries, the migratory fishery was the primary industry within what is now Newfoundland and the French territory of Saint-Pierre and Miquelon. Although some early permanent English settlements were in the area, including the colony of Avalon (est. 1621, now Ferryland), Newfoundland's larger scale permanent English settlement occurred in the 18th century. However, settlements remained focused on the fishing industry, with large mercantile towns developing throughout the region to supply these settlements (e.g., St. John's, Harbour Grace, and Ferryland). These areas were unlikely to have local anthropogenic environmental Pb exposures as the mining and smelting of Pb-containing ores were not performed in this area until the mid to late 1800s. Coal, which does contain some Pb, was not commonly used, at least in the St. John's, NL, area, until the mid-19th century (Christopher, 1999), and its been suggested that the compounds are too large to be inhaled and result in Pb exposure (Millard et al., 2014) (for more specific discussion, see Chapter 3).

The first French colony directly managed by the French crown was in Plaisance (est. 1654 — permanent settlers arrived in 1662), which had strong connections back to LaRochelle, France. This colony and surrounding small permanent and migratory settlements, including at the nearby island of Saint-Pierre, were ceded to Great Britain in 1713 (Crompton, 2012). While the French did not regain Plaisance (which the English renamed Placentia), there were nine governance changes in the archipelago of Saint-

Pierre and Miquelon, with the French maintaining control since 1815. When the military and settlers of Plaisance initially lost their territory, they moved to Ile Royal (modern-day Cape Breton, Nova Scotia). They established the Fortress of Louisbourg in 1714, which became the French *entrepôt* with the local non-military population still engaged in the fishery. During the spring of 1745, the Fortress of Louisbourg was seized by the British with the help of many New England provincial forces, then returned to the French in 1748. The British seized it for a final time in 1758. Be it migratory fisher people or permanent settlers, these French and English settlements relied on imported goods, including ceramics or foodstuffs shipped in ceramics, that were then reused.

While behavioural and functional categories such as domestic, architectural, personal, etc., are more commonly used in historical archaeology (Brooks, 2005), they are less meaningful when referring to Pb exposure. Functional use subcategories of ceramics such as serving, cooking, and storage are probably most useful to address the likelihood of Pb-glazed ceramics resulting in Pb exposure. The amount of Pb leaching would vary with Pb content of the glaze, the purpose of the container, time of contact, and acidity of the contents. Pb isotope values would vary with the geological source of Pb used to make the glazes. While some ceramics were produced very widely within a region or country (e.g., pearlware, ironstone, faïence), specific styles can be attributed to smaller locals (e.g., Vallauris, Staffordshire). While the creation of the object is attributed to a region, the materials within can be imported from further abroad.

The specific ceramics predominant in each settlement or fishing stage depended on numerous factors. In Placentia, unlike many other French colonies, they continued accessing French ceramics – though not exclusively (Crompton, 2012). In most

settlements, the origin of ceramics depended on the last port of call before departure, what other merchants were active in the region, and the financial ability of the household to acquire imported goods from other sources. Regardless of European cultural association, materials from numerous countries and regions were used within a single household in all excavations.

Unfortunately, there has yet to be an analysis of primary source documents or archaeological excavations that allow for the assessment of differences in patterns throughout all English/Irish and French settlements in Newfoundland and Saint-Pierre. There are some compiled in French contexts (see citations in Losier et al., 2018). In both Placentia and Anse à Bertrand, SPM, Normandy stoneware is mainly used for foodstuffs, though this was not Pb-glazed. According to both studies, common coarse earthenware (CEW) materials included Saintonge, Sadriac style, Cox-Lomagne, and small amounts of Beauvais. In refined earthenware (REW), faïence was not identifiable to specific French provinces, but they are present. However, non-French ceramics were also present in small numbers, including Albisola from Northern Italy, Staffordshire slip from England, and small numbers of New England pottery. This pattern resembles materials found in a French occupation destruction layer from 1745 at the Fortress of Louisbourg (Drakich, 1982), where Plaisance occupants were moved to after 1714.

There is no similar amalgamation study for English- or Irish-associated sites in Newfoundland. Due to the seasonal occupations, either through migration or winter transhumance, it is difficult to specifically link the studies, which vary in ware types over the 17th to 19th centuries. While a complete comparison is outside the scope of this paper, it is possible to make some generalizations based on published research and Masters

theses (Burke, 1991; Venovcevs, 2017; Venovcevs and Gaulton, 2018; Williams, 2019; De La Plante, 2022). North Devon coarse grain is the most common CEW on English sites, though notably, not all sites had an abundance of CEW. Creamware, pearlware, and whiteware were ubiquitous tablewares. Staffordshire and North Devon smooth were some of the most common REW with known origins. While many other types were common, they were smaller in number. Regarding foreign made earthenwares, French faïence, Portuguese Redware, and Spanish Heavy/Iberian were most common, with some Northern Italian wares.

The diversity of ceramics available should be contextualized within the Atlantic World framework. This framework is the transnational study of European and colonial connections from the 1500s to the mid-19th century (Canny, 1999; Vidal, 2012). While there were complex transnational networks of influence between colonies and European centres, not all relationships were the same (Bailyn, 2009); some colonies retained a significant impact from metropoles and vice versa (Hornsby, 2005). This framework focuses on social history, thereby highlighting the multi-directional complex web of connections within the Atlantic during the establishment and settlement of the colonies during the early modern period (Canny, 2001; Bailyn, 2009).

Even though there were trade restrictions in place, colonies developed and maintained relationships more broadly. This can be used in reference to ceramic production and trade – since there were more diverse access than just the associated country. Also, those settlements that developed their own pottery traditions which were subsequently exported – the timing of these developments depended on their access to knowledge (immigration of potters particularly from England and Germany), materials,

and the focus of their local industries and implied time available (Hume, 1969; Myers, 1977). With no hard ending dates to the Atlantic World, since most are American centered (Vidal, 2012), it can be argued that the Atlantic World concepts of interconnected network of influence still applied into the mid-19th century, particularly in the North Atlantic. In considering this, the import and export of Pb-containing materials would either have swamped or averaged the Pb isotope ratios of individuals interacting with such objects.

7.3.3 Ceramic analyses

7.3.3.1 LASS analysis methodology and errors

Data was acquired using laser ablation split-stream over three days. Samples were ablated using the GeoLas Laser UV-193nm (ArF Excimer) with concurrent trace element and Pb isotope ratio analyses using an Element single-collector ICP-MS and Neptune (Thermo Scientific Neptune MC-ICP-MS). The tuning and analyses parameters can be found in Table 33 and Table 34. The Neptune was tuned to maximize the ^{208}Pb intensity but backed slightly to avoid too much oxide formation.

The glaze and, if exposed, the paste of each ceramic were analyzed at least twice. Multiple ceramics were placed into the analysis cell depending on their size. Two analyses of NIST 610 were performed after the insertion of a new cell and at the end of those analyses, this was done every two to ten analyses (an average of 6.5).

For the ICPMS, concentrations were normalized to a total of 96% of the oxides for the major elements, this was chosen to compensate for the Na_2O not measured. For MC-

ICP-MS, the first and last NIST 610 standard was used for calibration and the others as QC checks.

Calibration of concentration and Pb isotope data of ceramics was done using sample bracketing between NIST 610 with linear equations accounting for drift in standards. Data were normalized to values from ICPMS NIST 610. The errors for these analyses are found in Table 35 and Table 36.

Table 33. GeoLas Laser parameters for all analyses

Material type	**NIST 610 & Ceramic body**	**Ceramic glaze**
Shots	600	120
J/cm^2	5	3
Hz	10	2
µm	99	20

Table 34. Neptune (MC-ICP-MS) tuning parameters for three days of analyses

Neptune Tuning Parameters	
Ar gas	0.9 to 0.95 l/min
N_2 gas	'0.2' ~10 ml/min
He gas	'0.53' ~1.06 ml/min
Cone configuration	202, 204, 206, 207, 208
X	-0.48
Y	-1.21 to -0.97
Z	-1.6
Source offset	-7 V
Zoom focus	+3 V

Table 35. Errors on the NIST 610 standards not used for calibration (n = 19) over three days of analyses.

	$^{208}Pb/^{206}Pb$	**$^{207}Pb/^{206}Pb$**	**$^{208}Pb/^{204}Pb$**	**$^{207}Pb/^{204}Pb$**	**$^{206}Pb/^{204}Pb$**
Most probable value	2.1694	0.90986	36.991	15.515	17.052
Mean	2.1696	0.9100	37.005	15.516	17.050
1σ	0.0007	0.0006	0.056	0.007	0.012
% error	0.0074	0.0198	0.037	0.006	0.011

Table 36. The percent difference between replicate samples (n = 32)

	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$
Mean %diff	0.0179	0.0133	0.033	0.021	0.015
1σ %diff	0.0178	0.0106	0.03	0.019	0.010
Max %diff	0.0917	0.0527	0.148	0.100	0.047

7.3.3.2 Ceramic Results

Table 37. Concentration of lead (Pb), tin (Sn), manganese (Mn), and iron (Fe), and Pb isotope ratios within ceramic glaze

marc/ sample	Type	Pb weight %	Sn ppm	Mn ppm	Fe ppm	$^{208}Pb/^{206}Pb$	$^{207}Pb/^{206}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$
French										
807-4152	Pabu-Guingamp	48.3	8190	*46*	13810	2.08606	0.84865	38.45397	15.63943	18.43121
807-4148	Vallauris	56.3	<DL	*38*	9742	2.10661	0.85825	38.50093	15.68174	18.27480
5552	Faïance blanche	28.4	174985	*57*	*1199*	2.08473	0.84797	38.57661	15.65375	18.46302
708-3700	Faïance blanche	33.1	132795	*132*	1516	2.08327	0.84789	38.44408	15.63919	18.45226
710-3809	Faïance blanche	36.1	69585	*150*	1793	2.08837	0.84934	38.44182	**15.6458**	18.40790
508-2430	Faïance brune	48.5	104	14560	11575	2.08149	0.84654	38.44556	15.63639	18.47086
5549	17^{th}–18^{th} c - REW	56.6	67	*62*	4515	2.09057	0.85320	38.48618	15.63908	18.33638
708-3704	Saintonge	58.9	487	1105	2657	2.09009	0.85035	38.46108	15.63863	18.39922
5547	Saintonge	57.2	1267	*47*	2860	2.08347	0.84789	38.41054	15.63249	18.43658
5551	17^{th}–18^{th} c - REW	56.5	581	11610	2574	2.08722	0.85029	38.46425	15.64173	18.39233
English										
5545	N. Devon Gravel	54.9	147	*135*	1681-	2.08135	0.84674	38.42231	15.63096	18.46111
5546	N. Devon Smooth	57.6	3	223	24233	**2.08745**	**0.84976**	**38.44566**	**15.65096**	**18.41774**
5560	Staffordshire	35.8	<DL	*77*	8931	2.08306	0.84593	**38.50475**	15.64822	18.49690

5557	17th–18thc - CEW (Midlands Purple or Manganese Mottled?)	31.7	7	28125	14525	2.10480	0.85951	38.30017	15.64189	18.20021
5558	18th c - REW	39.5	3	368	24415	2.09137	0.85120	38.35942	15.62438	18.35353
5556	Creamware 18th c	41.0	6	<DL	3099	2.08261	0.84676	38.46076	15.63795	18.46838
5559	Whieldonware 18th c	54.7	19	220	2384	2.08233	0.84640	38.62298	15.63429	18.46212
Dutch										
5540	slipware	52.0	14	65	5919	2.09404	0.85366	38.26587	15.60086	18.26696
5540	slipware	50.7	7	*111*	9334	2.09479	0.85423	38.24726	15.59757	18.25597
5540 mean	slipware	51.3	10	88	7626	2.09441	0.85395	38.25657	15.59921	18.26146
5541	slipware	52.1	<DL	<DL	*9128*	2.09351	0.85327	38.25896	15.59532	18.27432
Italian										
804-4054	Albisola	43.9	89	16250	11250	2.10488	0.85667	38.55284	15.68542	18.31608
803-3968	Ligurian type	41.4	2	22785	10675	2.09206	0.85106	38.43594	15.63750	18.37342
Spanish										
5539	Spanish Heavy	58.4	293	<DL	*5376*	2.08244	0.84721	38.48917	15.64796	18.47021
707-3648	Spanish Heavy	50.7	278	462	3992	2.09280	0.85261	38.40074	15.64073	18.34717

Table 38. Pb isotope ratios of ceramic glaze with propagated error of each analysis

marc/ sample	$^{208}Pb/^{206}Pb$	prop. 2σ	$^{207}Pb/^{206}Pb$	prop. 2σ	$^{208}Pb/^{204}Pb$	prop. 2σ	$^{207}Pb/^{204}Pb$	prop. 2σ	$^{206}Pb/^{204}Pb$	prop. 2σ
French										
807-4152	2.08606	0.00051	0.84865	0.00048	38.45397	0.01951	15.63943	0.00466	18.43121	0.01268
807-4148	2.10661	0.00067	0.85825	0.00123	38.50093	0.02653	15.68174	0.00662	18.27480	0.01691
5552	2.08473	0.00074	0.84797	0.00125	38.57661	0.27399	15.65375	0.02104	18.46302	0.05505
708-3700	2.08327	0.00066	0.84789	0.00070	38.44408	0.02831	15.63919	0.00871	18.45226	0.01707
710-3809	2.08837	0.00030	0.84934	0.00017	38.44182	0.01703	15.63458	0.00680	18.40790	0.00799
508-2430	2.08149	0.00027	0.84654	0.00015	38.44556	0.01424	15.63639	0.00569	18.47086	0.00653
5549	2.09057	0.00063	0.85320	0.00109	38.48618	0.23773	15.63908	0.01798	18.33638	0.04728
707-3648	2.09280	0.00056	0.85261	0.00080	38.40074	0.02193	15.64073	0.00536	18.34717	0.01415
708-3704	2.09009	0.00063	0.85035	0.00066	38.46108	0.02628	15.63863	0.00788	18.39922	0.01587
5547	2.08347	0.00021	0.84789	0.00009	38.41054	0.01174	15.63249	0.00476	18.43658	0.00517
5551	2.08722	0.00032	0.85029	0.00049	38.46425	0.05642	15.64173	0.00540	18.39233	0.01353
English										
5545	2.08135	0.00045	0.84674	0.00032	38.42231	0.01798	15.63096	0.00534	18.46111	0.01005
5546	2.08745	0.00019	0.84976	0.00009	38.44566	0.01695	15.65096	0.00637	18.41774	0.00842
5560	2.08306	0.00020	0.84593	0.00010	38.50475	0.01797	15.64822	0.00766	18.49690	0.00816
5557	2.10480	0.00038	0.85951	0.00027	38.30017	0.01597	15.64189	0.00465	18.20021	0.00830
5558	2.09137	0.00016	0.85120	0.00008	38.35942	0.01453	15.62438	0.00618	18.35353	0.00641
5556	2.08261	0.00033	0.84676	0.00019	38.46076	0.01917	15.63795	0.00762	18.46838	0.00906
5559	2.08233	0.00053	0.84640	0.00075	38.62298	0.14218	15.63429	0.00692	18.46212	0.02288
Dutch										
5540	2.09404	0.00033	0.85366	0.00020	38.26587	0.02205	15.60086	0.00781	18.26696	0.01057
5540	2.09479	0.00049	0.85423	0.00041	38.24726	0.03405	15.59757	0.01072	18.25597	0.01680
5541	2.09351	0.00031	0.85327	0.00019	38.25896	0.01948	15.59532	0.00704	18.27432	0.01015
Italian										
804-4054	2.10488	0.00070	0.85667	0.00074	38.55284	0.03042	15.68542	0.00950	18.31608	0.01811
803-3968	2.09206	0.00027	0.85106	0.00011	38.43594	0.01493	15.63750	0.00611	18.37342	0.00690
Spanish										
5539	2.08244	0.00031	0.84721	0.00022	38.48917	0.05305	15.64796	0.00659	18.47021	0.00902

7.3.4 Dental analyses

7.3.4.1 Tooth choice: type and preservation

Additional dental enamel samples were taken for analysis from two sites. The first, St. Paul's Anglican Church Cemetery (CkAh-06, 1764–1820) from Harbour Grace, Newfoundland (Chapter 3) sampled the second molars of all individuals. However, one individual (Feature 7/NP265) who had outlying Pb concentration and isotope ratios was resampled using first and third molars, representing infancy (4.5 months to 3.5 years of age) and adolescence (8.5 to 14.5 years of age) (AlQahtani et al., 2010).

For further exploration of the implications of imported Pb-containing goods in the Northern Atlantic, individuals from Rochefort Point (55L34; 1730s to 1758), a later Louisbourg, NS cemetery were sampled. These are eight adolescent individuals who were previously identified based on historical and mortuary contexts and dietary isotope values to have originated from America, likely New England (Scott et al., 2023). Therefore, these individuals were buried between 1745 and 1748 during the New England occupation of Louisbourg. Second molars, representing childhood were sampled from each individual.

One tooth, preferably the second molar, from each of the eight individuals from Rochefort Point was taken for sampling. This minimizes the likelihood of exposure to maternal or caregiver's Pb, and its size maximizes the potential of other isotopic data from a single tooth for future research. A first and third molar were selected from the young adult female from St. Paul's Anglican Church cemetery, Harbour Grace (Feature 7/NP254). These represent different periods of social and physical development. Since Pb

can transfer from mother/caregiver to baby via breastmilk, there is some potential for Pb exposure that does not represent the physical or cultural environment that the baby was living, but rather Pb that may have been released from skeletal materials representing the caregiver's earlier life. For individuals at both sites, we selected teeth with the goal of avoiding areas with caries, large cracks, or staining.

Tooth enamel was cleaned with a tungsten carbide bit (Brasseler round carbide) to at least 100 μm or until surface staining was removed. An enamel sample of ~50 mg was separated using a diamond dental saw (Brasseler DS Miniflex). Since these blades have a potential for contamination (Montgomery, 2002), all cut surfaces were further cleaned with a clean tungsten carbide bit, and any adhering dentin was removed under 5X magnification. The dental tools were cleaned between each sample by ultrasonication in deionized water, rinsed, dipped in 2M HNO_3 and allowed to dry. Following cutting, enamel was ultrasonicated three times in deionized water for five minutes each. After rinsing with ~200 μL of acetone, the samples were transferred to the clean lab and left to air dry in the laminar air flow clean boxes.

Samples were transferred to clean 3 mL or 7 mL Savillex vials and weighed. Two mL of distilled 8 M HNO_3 were added to each sample and digested on a hot plate for approximately one hour. A blank and SRM 1400 (~25 mg) were included in each group of approximately eight samples.

7.3.4.2 Concentration methods and errors

An aliquot of 0.5 mL was taken for concentration analysis and transferred to the TERRA facility at Memorial University. Here a further aliquot was taken up, and 0.3 M

HNO_3 solution was added using gravitation measurement to ensure that it was taken up into 10 mL of solution.

Since the 2 mL for digestion was added to the samples using volumetric, not gravimetric measurement, the solution's density must be corrected. To do this, the weight of the vial was removed from the weight of the final solution. This was then divided by the mass of the sample. The sample concentration was determined using the calibrated ppm of the solution that went through the Q-ICP-MS.

For data analyzed at the Terra facility, analyses were carried out on acidified sample solutions of ~0.2 M HNO_3 at a dilution factor of ~6000X. To achieve this, aliquots of samples in 8 M HNO_3, based on their starting weight and first dilution with the 8 M HNO_3, were diluted to ~10 g with de-ionized 18.2 MΩ water if the aliquot was bigger than 0.25 g, or with 0.2 M HNO_3 if less. The concentration of 35 elements including Pb was determined with solution ICP-Q-MS (inductively coupled plasma mass spectrometry) on a Perkin-Elmer Elan DRCII with a Cetac ASX-520 Autosampler. The RF frequency is 1200 W using Nickel cones. The plasma gas was 16 L/min, the nebulizer gas flow was 1.16 L/min and the auxiliary was 1.2 L/min. Pb calibration and concentration methods were a modified version of Friel et al. (1990) where different multi-element solutions are used for external calibration and Sc, Re, Rh and Th are used to monitor instrumental drift. Pb concentration calibration solution was 10.11 ppb and was run at the beginning and end of the run, and between every eight unknown samples. USGS T-143 and USGS T-193 are standard water references used as check standards.

Due to instrument problems, concentrations were estimated for the first molar (M1) and third molar (M3) teeth from SPACC. This was done using the ^{208}Pb voltage from the

MC-ICP-MS of each sample. Each run had a known concentration of the SRM 981 solution. The known concentration was divided by the averaged voltage of the standard solution on either side of the bracket. This gave the ppb/V which was multiplied by the voltage of the sample to give the raw ppb value of the solution. This was then converted to concentration of the sample using the known sample mass and density of solution.

For Pb concentration analyses at the TERRA facility, the procedural blanks were usually below the limit of detection or just above. Of the six blanks that were above detection limit, they averaged 0.01 ppm (n = 3) Since the final concentrations are a product of the sample weight, a better sense of how large of an impact the blank had on the samples. The median of the 72 analyses was 0.06%, but the highest value was 1.82%. The blank correction was not done for the TERRA samples since it did not make an interpretable difference to the data.

Replicates were run on ~12% (n = 6) of the samples, and the mean within-assay variability (coefficient of variation) was 1.30%. The between assay variability was determined using three reference standards at the Memorial University TERRA facility, USGS T-143 (83.40 ppb), USGS T-193 (0.290 ppb), and NIST SRM 1400 (9.07 ppb). The accuracy obtained against the standard values in this study were 106.15%, 110.74%, and 106.55%, respectively (Table 39 and Table 40). The between assay variability for the SRM 1400 concentrations determined using MC-ICP-MS almost three times higher than those from the TERRA facility (Table 40). This increased error is likely caused in part by the solution being the elution of the column chemistry. There is significant variability possible from sample loss during the column chemistry, and the solution density is based on the initial sample loaded, not the final analysis.

Table 39. The summarized Pb concentration values for USGS T-143 and USGS T-193 used as the TERRA check standards. The TERRA values run over two analysis days.

	USGS T-143	**USGS T-193**
Most probable Pb concentration (ppb)	83.4	0.290
Mean Pb concentration (ppb)	88.53	0.321
Count	2	2
Mean/MPV	1.062	1.107
% error (% diff to MPV)	6.15	10.74

Table 40. The summarized Pb concentration values for all SRM 1400 analyses ran over two days of Q-ICPMS analyses, $n = 6$ and four days for MC-ICP-MS, $n = 5$.

SRM 1400	**Q-ICPMS**	**MC-ICP-MS**
Certified Pb concentration (ppm)	9.07 ± 0.12	9.07 ± 0.12
Mean Pb concentration (ppm)	8.48	8.70
count	6	5
1σ	0.65	2.52
% error	6.55	4.05

7.3.4.3 Pb isotope methodology and errors

The remaining 1.5 mL of solution was used for Pb isotope analyses. Using fabricated columns, Pb was isolated by extraction chromatography using Sr-spec resin (Eichrom Technologies) (Charlier et al., 2006). Samples were prepared within groups of ten, including a blank and SRM 1400 as an internal standard. The column chemistry was based on Pin et al. (2014; see Figure 20 for flowchart) and allowed for the simultaneous extraction of Sr (Garlie, 2022) and Pb. Pb is eluted with 1.5 mL of 6.2 M HCl, dried down on the hotplate, then taken up in 0.8 mL of 0.3 M HNO_3 and transferred to a centrifuge tube for analysis.

The sample analysis parameters are found in Table 41. Samples were analyzed using a 30-second baseline and 50 cycles of data per sample. To allow for calibration, a ~100

ppb solution of SRM 981 bracketing every three samples. Data was then calibrated using the most probable values of SRM 981 from Baker et al. (2004) and De Munyck (2008) using a linear equation to account for any drift during analysis. Samples were blank corrected, and one carousel worth of data was run per day to account for changes in blanks. Since both the St. Paul's and Rochefort Point are part of larger data collections, the data were acquired over four non-consecutive days to keep them associated with the original carousel of data.

Table 41. MC-ICP-MS analysis parameters

MC-ICP-MS	
Spray chamber	Sis cleaned with sitranox and rinsed with 0.3 N nitric
Nebulizer	50 µm flushed with 0.3 N nitric
Cones	Nickle
Faraday array	202, 204, 205, 206, 207, 208
Auxillary gas flow rate	1.00 l/min
Sample argon gas flow rate	1.020 to 1.045 l/min
Zoom focus	+1.5 to +3 V
Source offset	-7 to -15 V
Baseline – defocused beam	30-cycles, 1 sec. integration time
Blocks	50 cycles, 8.389 sec. integration time
Software suite	Neptune Software, version 3.2.0.14

The target voltage during analysis of ^{208}Pb was 4 V, but not all solutions had sufficient concentration for this to be met, other times dilution with 0.3 M HNO_3 was necessary at the time of analysis. Since the blank to sample voltage did not exceed 5% (max of 1%), none of the samples were removed for consideration.

The isotopic data is based on runs over 11 non-consecutive days. The blanks had a mean voltage of ^{208}Pb 0.0135 ± 0.0072 (n = 23). The between assay variability was

determined using SRM 1400. The accepted values are the most probable values as published by Hinners et al. (1998). Since Tl correction was not performed for the isotopic data, the standard errors are important for understanding accuracy. The summary errors for each isotope ratio analyzed are found in Table 42 and Table 43.

Table 42. The summarized Pb isotope ratios for all SRM 1400 analyses ran over three days including the most probable values as published by Hinners et al. (1998).

	$^{208}Pb/^{206}Pb$	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$	$^{208}Pb/^{207}Pb$
Most probable value	2.10209	NA	38.610	15.6702	18.3676	2.46389
Accepted RDS (%)	0.016	NA	0.052	0.038	0.038	0.0064
Count	4	4	4	4	4	4
Mean	2.1037	1.1716	38.6708	15.6891	18.3816	2.4653
1σ	0.0007	0.0002	0.0261	0.0074	0.0064	0.0005
% error	0.0787	NA	0.1576	0.1205	0.0763	0.0553

Table 43. The summarized within assay variability determined through propagated error (2σ) of all enamel samples.

	$^{208}Pb/^{206}Pb$	$^{206}Pb/^{207}Pb$	$^{208}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{206}Pb/^{204}Pb$	$^{208}Pb/^{207}Pb$
Count	11	11	11	11	11	11
Mean	0.00013	0.00042	0.00529	0.00239	0.00239	0.00032
1σ	0.00361	0.06573	0.00916	0.06276	0.00324	0.00258
Min	0.00013	0.00042	0.00420	0.00167	0.00190	0.00032
Max	0.00014	0.00043	0.00758	0.00417	0.00363	0.00033

7.3.4.4 Diagenesis

Kamenov et al. (2018) developed a method to identify possible cases of diagenetic alteration of human tooth enamel. Using concentrations of other heavy metals, TE and REEs from the sample tooth and comparing them to a maximum threshold concentration

(MTC) that they developed based on modern individuals, it is suggested that diagenetic alteration is identifiable. Alteration is suggested when the concentration to MTC ratio is greater than one; if many elements were high, there is a concern for major alteration (Kamenov et al., 2018). While they initially sampled many elements, they concluded that V, Mn, Fe, La, Ce, Nd, Dy, Yb, Th, and U are best.

Memorial University's TERRA facility's methodology analyses only six of the recommended elements (V, Mn, Fe, La, Ce, and U). Concentrations were available for the Rochefort Point individuals. In none of these cases were the concentration to MTC ratios above a 1.0 (Table 44); therefore, there is no reason to suggest diagenetic alteration of the enamel.

7.3.4.5 Human comparative groups

Populations used for comparisons include the fisheries populations from Chapter 3 (defined in text) and previously published groups from 19th century Otago, New Zealand (King et al., 2020), Connecticut, USA (Aronsen et al., 2019), and Colorado, USA (Keller et al., 2016). Site details for the previously published materials are found in Table 18. Note that only individuals with values originating from permanent teeth were included in the analysis from King et al. (2020).

7.3.5 Pb ore comparative

Some data or ore values compiled. While some of these were acquired as part of archaeological or environmental history research, much were associated with modern geology or mineral exploration studies and therefore do not necessarily directly represent the ore used during the 17th to 19th centuries.

Table 44. The ratio of enamel sample concentration in ppm to the Maximum Threshold Concentration (MTC) of six elements. The MTC in ppm is given for each element as established by Kamenov et al. (2018 *JAS* 99). Samples with a * denote replicate analyses. Pb concentrations not compared to the MTC are provided in ppm.

Individual	V (ppm/ MTC)	Mn (ppm/ MTC)	Fe (ppm/ MTC)	La (ppm/ MTC)	Ce (ppm/ MTC)	U (ppm/ MTC)	Pb (ppm)
MTC ppm	**0.11**	**15.4**	**143**	**0.1**	**0.12**	**0.05**	
1/2017	<DL	0.05	<DL	<DL	<DL	<DL	3.5
4/2017	<DL	0.14	<DL	<DL	<DL	0.09	2.8
6/2017	<DL	0.14	<DL	0.14	0.05	<DL	5.0
7/2017	<DL	0.78	<DL	0.13	0.04	<DL	0.6
20/2017	<DL	0.09	<DL	0.05	0.04	0.05	1.1
20/2017*	<DL	0.09	<DL	0.06	0.03	<DL	1.1
22/2017	<DL	0.09	<DL	0.08	0.09	<DL	1.5
28/2018	<DL	0.12	<DL	0.14	0.06	0.06	2.0
29/2018	<DL	0.05	<DL	<DL	0.02	<DL	2.6
29/2018*	<DL	0.06	<DL	0.06	0.03	<DL	2.7

Pb isotope ratios of ore were compiled in Blichert-Toft et al. (2016) for much of Western Europe and was broadly grouped by country. They included given or approximated latitude and longitude which were used to further define meaningful groups for analysis. Not all data was originally published giving all Pb isotope ratios, but they were calculated by Blichert-Toft et al. (2016) when missing. Also provided were the longitude and latitude of each sample. These were compared to maps of bedrock geology to create more meaningful groupings for analysis for England, Wales, and France.

It should be noted that while some of these data relate directly to ore from historical contexts (Métreau et al., 2021), most are taken from mines in use, recently used, or taken for the purposes of prospecting. Therefore, these do not necessarily relate to specific comparative materials that would be in use contemporaneously to that of the human populations examined in this study.

For American ore bodies, there has been no similar large comparative paper, but data were previously compiled in Bird et al. (2019), though individual data points were not published. Pb was grouped into regions by the original authors of the articles. These were typically defined by specific named ore districts. As with the European Pb ore deposits, not all areas were in use during the lives of the fishery populations in this study, so not all directly are relevant. First dates of use of the American mines are found in Table 19.

Table 45. Groupings of Pb ore isotope data for England, Wales, France, and USA used in this study along with the citations of original data. European, English, and Welsh Pb were compiled in Blichert-Toft et al., 2016.

Country	Grouping	Counties/States represented	No.	citations
England	Cornwall & Devon	Cornwall, Devon	52	Michard Vitrac et al., 1981; Rohl, 1996; Rohl and Needham, 1998
	Mendips	Somerset, Bristol	47	Rohl, 1996
	Derbyshire	Derbyshire, Cheshire, Leicestershire	54	Rohl, 1996; Rohl and Needham, 1998
	Northern Pennines & North and West Yorkshire	Durham, Northumberland, Cumbria, North Yorkshire	71	Rohl, 1996; Rohl and Needham, 1998
	Shropshire	Shropshire	16	Rohl, 1996; Rohl and Needham, 1998
Wales	Ceredigon & Montgomery (Central Wales)	Ceredigion, Gynedd; Anglesey; Conwy; Carmarth; Powys	116	Rohl, 1996; Rohl and Needham, 1998
	Flintshire & Denbyshire (Northern Wales)	Denbighshire; Flintshire; Wrexham, Conwy	27	Rohl, 1996; Joel et al., 1997; Rohl and Needham, 1998
France	Central Massif	Nouvelle-Aquitaine, Auvergne-Rhone Alps, Centre-val de Loire, Occitanie	271	Brevart et al., 1982; Marcoux, 1986; Marcoux et al., 1988; Downes, 1991; Lescuyer et al., 1999
	Cévennes	Occitanie	194	Brevart et al., 1982; Marcoux, 1986; Le Guen et al., 1991; Baron et al., 2006; Bode et al., 2009
	Montagne Noire	Occitanie	73	Brevart et al., 1982; Marcoux, 1986

	Armorican Massif	Brittany; Pays de la Loire	152	Marcoux, 1986; Lescuyer et al., 1999 and Jourdan, 2000 in Blichert-Toft et al. 2016
	Alps (including Maures Massif)	Provence alps cote d'azur; Auvergne-Rhone Alps	33	Marcoux, 1986
	Pyrennes	Occitanie	31	Marcoux, 1986
	Vosges	Grand-Est, Bourgogne-Franche-Comte,	22	Marcoux, 1986
	Guernsey & Jersey	Guernsey, Jersey	5	Marcoux, 1986
USA	Austinville-Ivanhoe	Virginia	10	Foley et al., 1981
	Central Missouri barite	Missouri	12	Goldhaber et al., 1995
	Colorado	Colorado	8	Lüders et al., 2009
	East Tennessee – Southern Appalachians	Tennessee	83	Kesler et al., 1994a
	Friedensville district – Central Appalachians	New Jersey	4	Kesler et al., 1994b
	Illinois-Kentucky fluorite district	Illinois, Kentucky	8	Heyl et al., 1966
	Southeast Kansas	Kansas	2	Heyl et al., 1966
	Central Illinois and Northeast Kentucky	Illinois, Kentucky	3	Heyl et al., 1966
	Appalachian Valley	Pennsylvania, New Jersey	3	Heyl et al., 1966
	Piedmont Massive Sulfide deposits	Virginia, North Carolina	25	LeHuray, 1982
	Old Lead Belt	SE Missouri	133	Svergensky et al., 1979 in Blichert-Toft et al. 2016 Deloule et al., 1986; Goldhaber et al., 1995
	Timberville	Virginia	11	Kesler et al., 1994b
	Tristate district	Oklahoma	125	Deloule et al., 1986

	Upper Mississippi Valley	Iowa, Wisconsin, Illinois, Minnesota	39	Heyl et al., 1966 corrected in Millen et al., 1995; Millen et al., 1995

7.3.6 Supplemental Results and figures

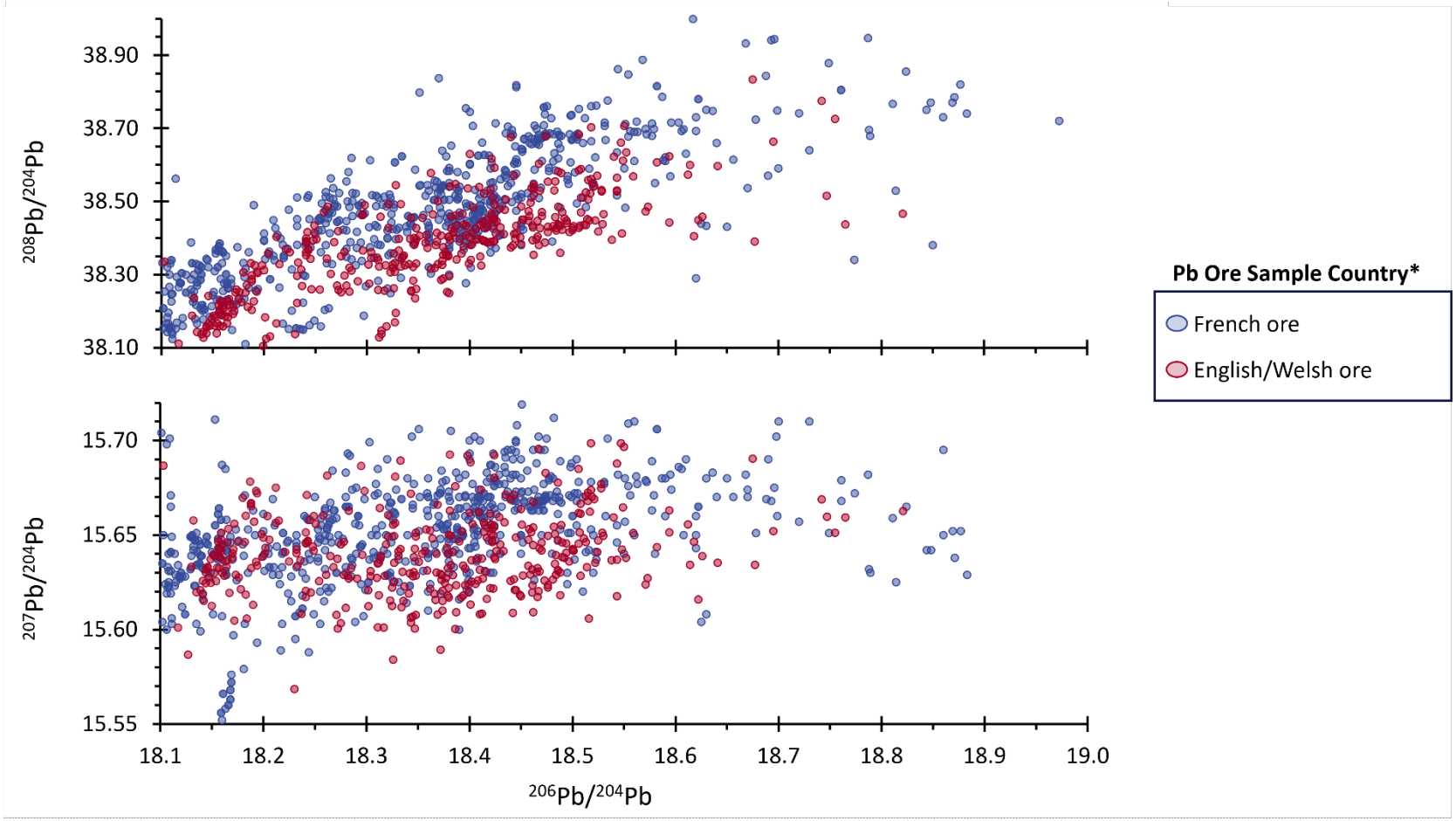

Figure 26. Pb ore isotope values for deposits in English/Welsh and French ore from data compiled in Blichert-Toft et al., 2016.

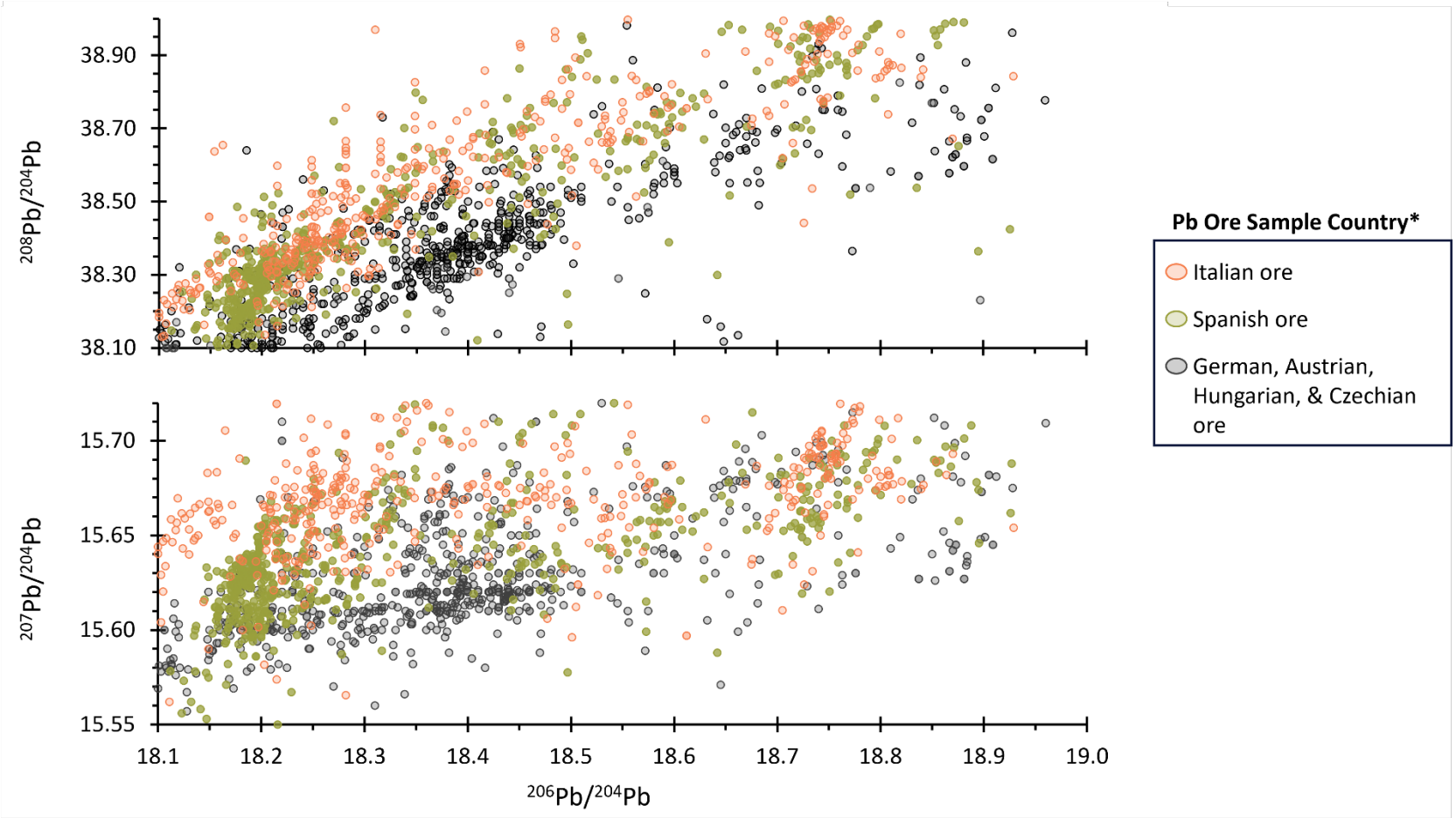

Figure 27. Pb ore isotope values for deposits in Italian, Spanish, and compiled German, Austrian, Hungarian, and Czechian ore from data compiled in Blichert-Toft et al., 2016.

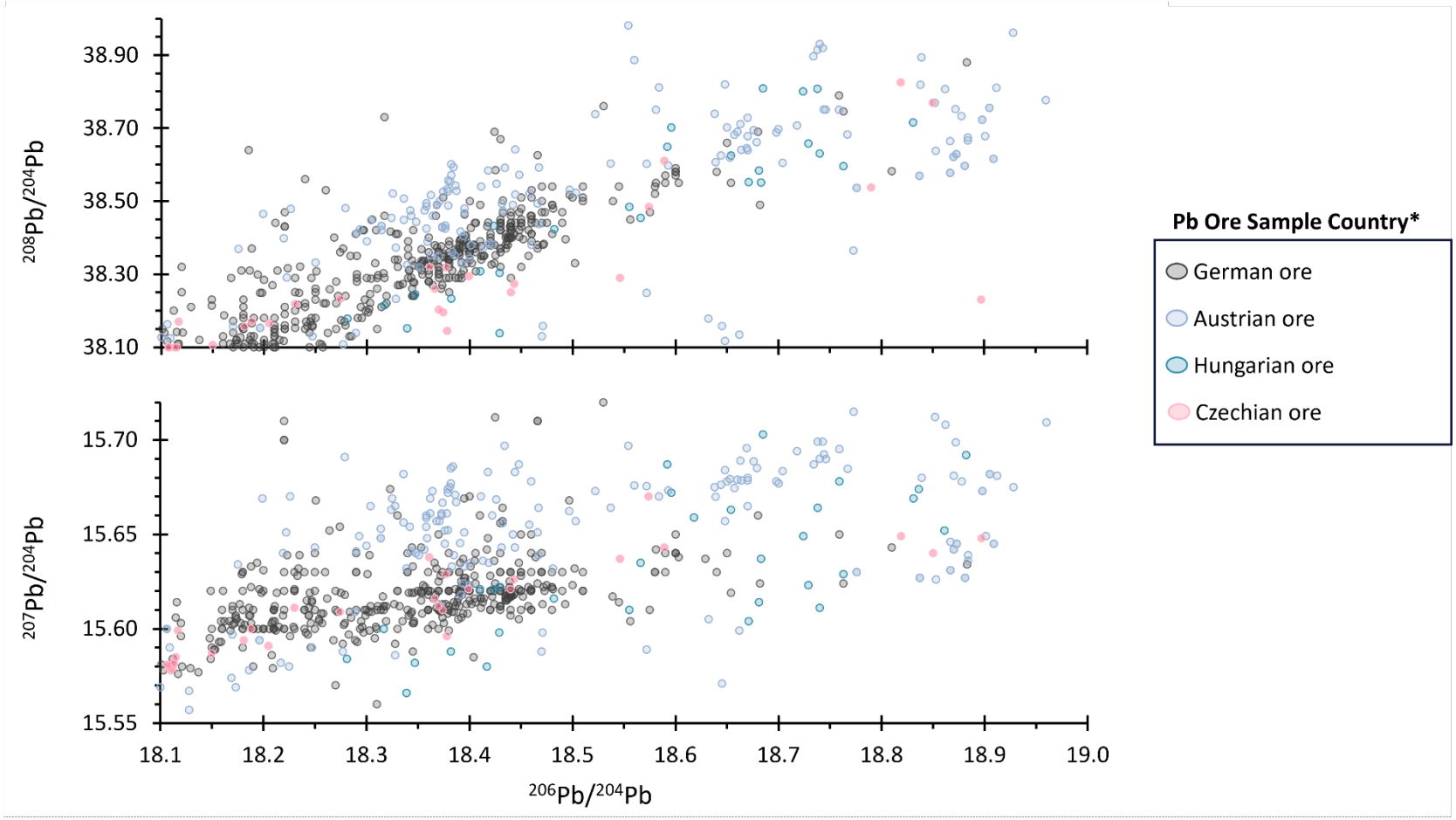

Figure 28. Pb ore isotope values for deposits combined in Figure 27. German, Austrian, Hungarian, and Czechian ore. Data are compiled in Blichert-Toft et al., 2016.

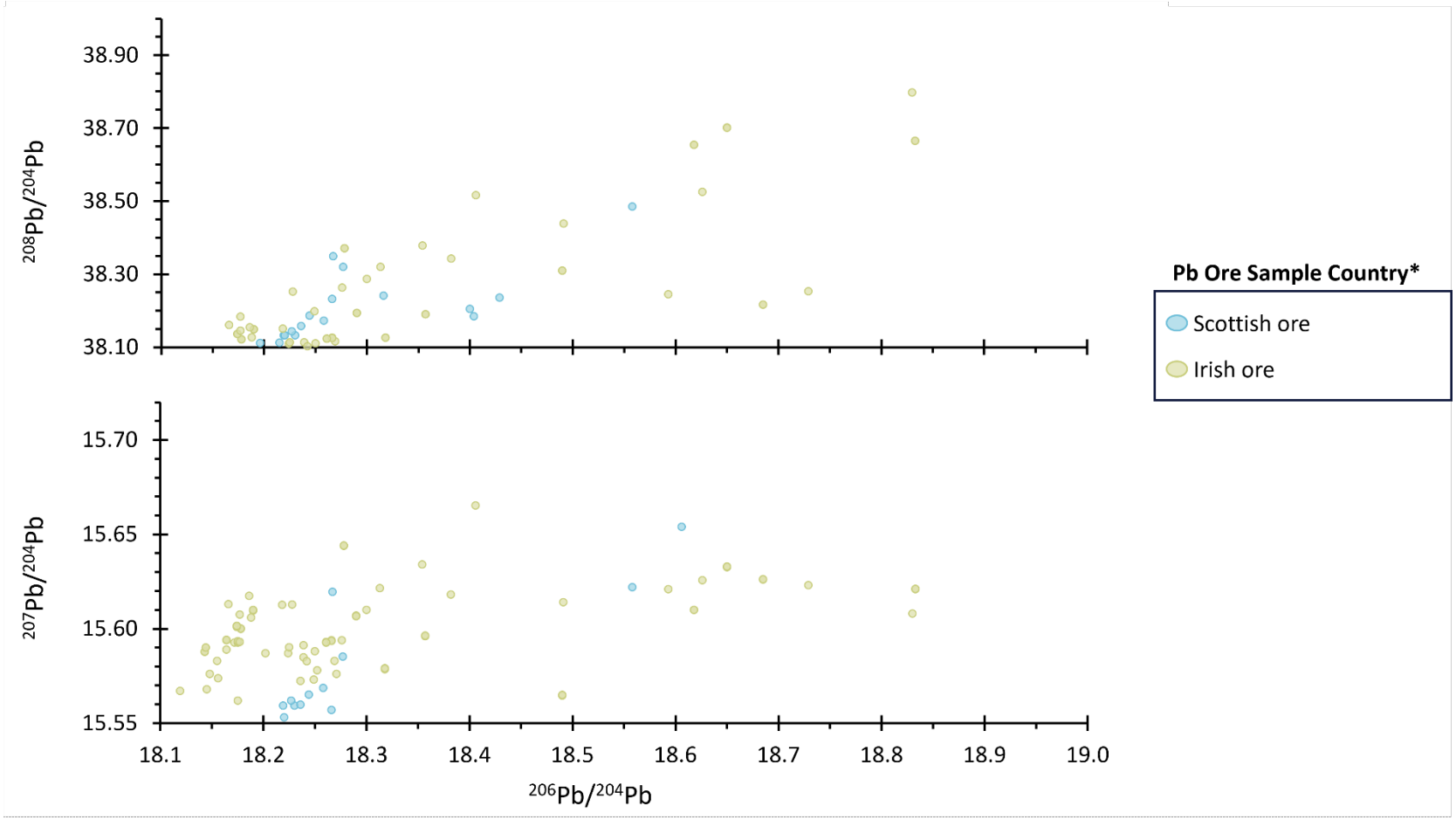

Figure 29. Pb ore isotope values for deposits in the Scottish and Irish from data compiled in Blichert-Toft et al., 2016.

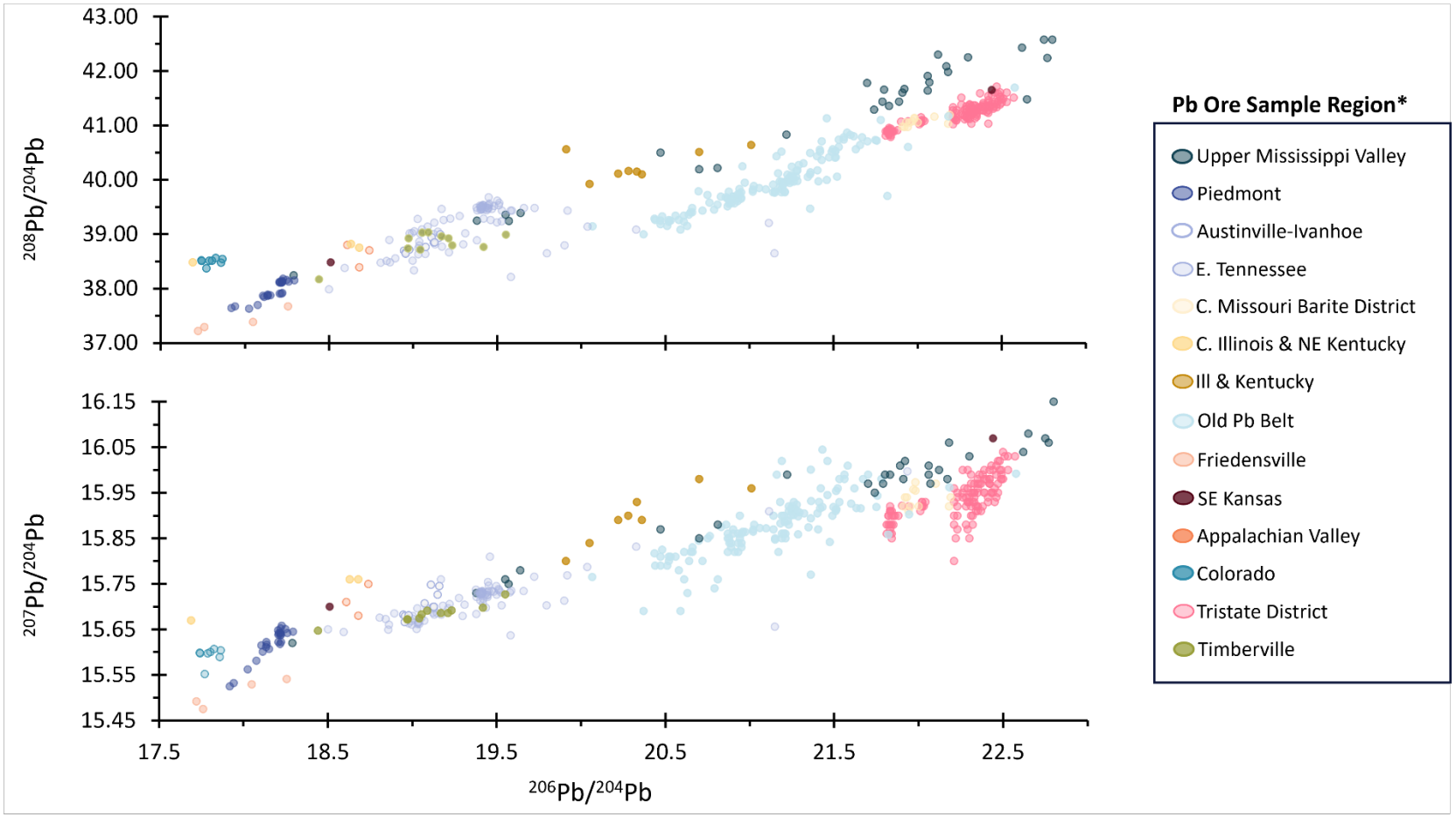

Figure 30. Pb ore isotope values for deposits in the USA. Full citations are available in Table 45.

7.4 Supplemental Materials References Cited

Adriano, D.C. (1986). *Trace elements in the terrestrial environment*. Springer, New York.

AlQahtani, S.J., Hector, M.P., & Liversidge, H.M. (2010). Brief communication: The London atlas of human tooth development and eruption. *American Journal of Physical Anthropology*, *142*, 481–490. https://doi.org/10.1002/ajpa.21258

Aronsen, G.P., Fehren-Schmitz, L., Krigbaum, J., Kamenov, G.D., Conlogue, G.J., Warinner, C., Ozga, A.T., Sankaranarayanan, K., Griego, A., DeLuca, D.W., Eckels, H.T., Byczkiewicz, R.K., Grgurich, T., Pelletier, N.A., Brownlee, S.A., Marichal, A., Williamson, K., Tonoike, Y., & Bellantoni, N.F. (2019). "The dead shall be raised": Multidisciplinary analysis of human skeletons reveals complexity in 19th century immigrant socioeconomic history and identity in New Haven, Connecticut. *PLOS One*, *14*, e0219279. https://doi.org/10.1371/journal.pone.0219279

Bailyn, B. (2009). Introduction. Reflections on some major themes. In B. Bernard & P.L. Denault (Eds.), *Soundings in Atlantic History* (pp. 1-43). Harvard University Press. https://doi.org/10.4159/9780674053533-intro

Baker, J., Peate, D., Waight, T., & Meyzen, C. (2004). Pb isotopic analysis of standards and samples using a ^{207}Pb–^{204}Pb double spike and thallium to correct for mass bias with a double-focusing MC-ICP-MS. *Chemical Geology*, *211*, 275–303. https://doi.org/10.1016/j.chemgeo.2004.06.030

Baron, S., Carignan, J., Laurent, S., & Ploquin, A. (2006). Medieval lead making on Mont-Lozère Massif (Cévennes-France): Tracing ore sources using Pb isotopes. *Applied Geochemistry*, *21*, 241–252. https://doi.org/10.1016/j.apgeochem.2005.09.005

Bird, B.W., Wilson, J.J., Escobar, J., Kamenov, G.D., Pollard, H.J., & Monaghn, G.W. (2019). Pre-Columbian lead pollution from Native American galena processing and land use in the midcontinental United States. *Geology*, *47*, 1193–1197. https://doi.org/10.1130/G46673.1

Blichert-Toft, J., Delile, H., Lee, C.-T., Stos-Gale, Z., Billström, K., Andersen, T., Hannu, H., & Albarède, F. (2016). Large-scale tectonic cycles in Europe revealed by distinct Pb

isotope provinces. *Geochemistry, Geophysics, Geosystems*, *17*(10), 3854-3864. https://doi.org/10.1002/2016GC006524

Bode, M., Hauptmann, A., & Mezger, K. (2009). Tracing Roman lead sources using lead isotope analyses in conjunction with archaeological and epigraphic evidence—a case study from Augustan/Tiberian Germania. *Archaeological and Anthropological Sciences*, *1*, 177-194. https://doi.org/10.1007/s12520-009-0017-0

Brännvall, M.-L., Bindler, R., Renberg, I., Emteryd, O., Bartnicki, J., & Billström, K. (1999). The medieval metal industry was the cradle of modern large-scale atmospheric Pb pollution in Northern Europe. *Environmental Science & Technology*, *33*, 4391-4395. https://doi.org/10.1021/es990279n

Brevart, O., Dupre, B., & Allegre, C.J. (1982). Metallogenic provinces and the remobilization process studied by lead isotopes; lead-zinc ore deposits from the southern Massif Central, France. *Economic Geology*, *77*, 564–575. https://doi.org/10.2113/gsecongeo.77.3.564

Brooks, A. (2005). Observing Formalities – the Use of Functional Artefact Categories in Australian Historical Archaeology. *Australian Historical Archaeology*, *23*, 7–14.

Budd, P., Pollard, A.M., Scaife, B., Thomas, R.G. (1995). The Possible Fractionation of Lead Isotopes in Ancient Metallurgical Processes. *Archaeometry*, *37*, 143–150. https://doi.org/10.1111/j.1475-4754.1995.tb00732.x

Budd, P., Montgomery, J., Barreiro, B., & Thomas, R.G. (2000b). Differential diagenesis of strontium in archaeological human dental tissues. *Applied Geochemistry*, *15*, 687-694. https://doi.org/10.1016/S0883-2927(99)00069-4

Budd, P., Montgomery, J., Evans, J., & Trickett, M. (2004). Human lead exposure in England from approximately 5500 bp to the 16th century ad. *Science of the Total Environment*, *318*, 45–58. https://doi.org/10.1016/S0048-9697(03)00357-7

Bundtin, D. (2021). *History of lead mining in Missouri by County or District* (PUB2979). Missouri Department of Natural Resources.

Burke, C.A. (1991). *Nineteenth century ceramic artifacts from a seasonally occupied fishing station on Saddle Island, Red Bay, Labrador* (Publication No. 4009) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Burt, R. (1984). *The British Lead Mining Industry*. Dyllansow Tryran.

Canny, N. (1999). Writing Atlantic History; or, Reconfiguring the history of colonial British America. *The Journal of American History*, *86*(3), 1093-1114. https://doi.org/10.2307/2568607

Canny, N. (2001). Atlantic history: What and why? *European Review*, *9*(4), 399-411. https://doi.org/10.1017/S1062798701000370

Carlson, A.K. (1996). Lead isotope analysis of human bone for addressing cultural affinity: A case study from Rocky Mountain House, Alberta. *Journal of Archaeological Science*, *23*, 557–567. https://doi.org/10.1006/jasc.1996.0052

Charlier, B.L.A., Ginibre, C., Morgan, D., Nowell, G.M., Pearson, D.G., Davidson, J.P., & Ottley, C.J. (2006). Methods for the microsampling and high-precision analysis of strontium and rubidium isotopes at single crystal scale for petrological and geochronological applications. *Chemical Geology*, *232*, 114–133. https://doi.org/10.1016/j.chemgeo.2006.02.015

Christopher, T.K., (1999). Paleolimnology in an urban environment: The history of environmental change in St. John's, Newfoundland. (Publication No. 9445) [Doctoral thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Crompton, A.J. (2012). *The historical archaeology of a French fortification in the colony of Plaisance: The View Fort Site (ChAl-04), Placentia, Newfoundland* (Publication No. 9656) [Doctoral dissertation, Memorial University of Newfoundland]. Memorial University Research Repository.

Cui, J., & Wu, X. (2010). An experimental investigation on lead isotopic fractionation during metallurgical processes. *Archaeometry*, *53*(1), 205-214. https://doi.org/10.1111/j.1475-4754.2010.00548.x

De La Plante, J. (2022). *Re-examining the lives of an 18th century garrison at Admiral's/Fort Point, Trinity, Newfoundland* (Publication No. 15453) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Deloule, E., Allegre, C., & Doe, B.R. (1986). Lead and sulfur isotope microstratigraphy in Galena crystals from Mississippi Valley-type deposits. *Economic Geology*, *81*, 1307-1321. https://doi.org/10.2113/gsecongeo.81.6.1307

De Muynck, D., Cloquet, C., & Vanhaecke, F. (2008). Development of a new method for Pb isotopic analysis of archaeological artefacts using single-collector ICP-dynamic reaction cell-MS. *Journal of Analytical Atomic Spectrometry*, *23*, 62–71. https://doi.org/10.1039/B709461B

de Winter, N.J., Snoeck, C., Schulting, R., Fernández-Crespo, T., & Claeys, P. (2019). High-resolution trace element distributions and models of trace element diffusion in enamel of Late Neolithic/Early Chalcolithic human molars from the Rioja Alavesa region (north-central Spain) help to separate biogenic from diagenetic trends. *Palaeogeography, Palaeoclimatology, Palaeoecology*, *532*, e109260. https://doi.org/10.1016/j.palaeo.2019.109260

Donovan, K. (2004). Slaves in Île Royale, 1713-1758. *French Colonial History*, *5*, 25-42.

Downes, H., Kempton, P.D., Briot, D., Harmon, R.S., & Leyreloup, A.F. (1991). Pb and O isotope systematics in granulite facies xenoliths, French Massif Centrimplications for crustal processes. *Earth and Planetary Science Letters*, *102*, 342–357. https://doi.org/10.1016/0012-821X(91)90028-G

Drakich, S. (1982). Eighteenth-Century Coarse Earthenwares Imported into Louisbourg. *Material Culture Review*, *16*, 83-98.

Evans, J., Pashley, V., Madgwick, R., Neil, S., & Chenery, C. (2018a). Tracking natural and anthropogenic lead exposure to its geological source. *Scientific Reports*, *8,* e1969. https://doi.org/10.1038/s41598-018-20397-y

Evans, J.A., Pashley, V., Chenery, C.A., Loe, L., & Chenery, S.R. (2018b). Lead isotope analysis of tooth enamel from a Viking age mass grave in southern Britain and the constraints in places on the origin of the individuals. *Archaeometry*, *60*, 859-869. https://doi.org/10.1111/arcm.12361Everett, D. (n.d.) Tri-State Lead and Zinc District. In *The Encyclopedia of Oklahoma History and Culture*. https://www.okhistory.org/publications/enc/entry.php?entry=TR014.

Faure, H., & Mensing, T. (2005). *Isotopes: Principles and Applications* (3rd edition). John Wiley & Sons.

Fitch, A., Grauer, A., & Augustine, L. (2012). Lead isotope ratios: Tracking the migration of European-Americans to Grafton, Illinois in the 19th century. *International Journal of Osteoarchaeology*, *22*, 305–319. https://doi.org/10.1002/oa.1207

FitzGerald, C. (2000). *All Saint's Anglican cemetery #1*. http://ngb.chebucto.org/Cemetery/cem-foxtrap-ang-hm.shtml

Foley, N.K., Sinha, A.K., & Craig, J.R. (1981). Isotopic composition of lead in the Austinville-Ivanhoe Pb-Zn District, Virginia. *Economic Geology*, *76*, 2012-2017. https://doi.org/10.2113/gsecongeo.76.7.2012

Friel, J.K., Skinner, C.S., Jackson, S.E., & Longerich, H.P. (1990). Analysis of biological reference materials, prepared by microwave dissolution, using inductively coupled plasma mass spectrometry. *Analyst*, *115*, 269–273. https://doi.org/10.1039/AN9901500269

Garlie, M.E. (2022). *An Isotopic Investigation of the Diet and Origins of 18th and 19th Century Individuals from Newfoundland and Louisbourg, Nova Scotia* (Publication No. 15764) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Goldhaber, M.B., Church, A.E., Doe, B.R., Aleinikoff, J.N., Brannon, J.C., Podosek, F.A., Mosier, E.L., Taylor, C.D., & Gent, C.A. (1995). Lead and sulfur isotope investigation of paleozoic sedimentary rock from the southern midcontinent of the United States: Implications for paleohydrology and ore genesis of the Southeast Missouri lead belts. *Economic Geology*, *90*, 1875-1910. https://doi.org/10.2113/gsecongeo.90.7.1875

Grimes, V., Lear, M., Munkittrick, J., & Lacy, R.S. (2018). *Excavation and preliminary analysis of a historical burial ground at Foxtrap-2 (CjAf-10), Foxtrap, Newfoundland*. In S. Hull & M. Delphina (Eds.), *Provincial Archaeology Office Annual Review for 2017 Field Season* (pp. 131-140). Provincial Archaeology Office Department of Tourism, Culture, Industry and Innovation Government of Newfoundland and Labrador.

Gulson, B.L. (1986). *Lead isotope in mineral exploration, Developments in Economic Geology*. Elsevier.

Gulson, B.L., Mizon, K.J., Davis, J.D., Palmer, J.M., & Vimpani, G. (2004). Identification of sources of lead in children in a primary zinc-lead smelter environment. *Environmental Health Perspectives*, *112*, 52-60. https://doi.org/ 10.1289/ehp.6465

Harris, A.J.T. (2015). *A Bioarchaeological Study of the Human Remains Recovered from Wester Point Cemetery (CjAf-08), Portugal Cove, NL.* In S. Hull & M. Delphina (Eds.), *Provincial Archaeology Office Annual Review for 2014 Field Season* (pp. 65-70). Provincial Archaeology Office Department of Tourism, Culture, Industry and Innovation Government of Newfoundland and Labrador.

Harris, O.J.T., Cobb, H., Batey, C.E., Montgomery, J., Beaumont, J., Gray, H., Murtagh, P., & Richardson, P. (2017). Assembling places and persons: a tenth-century Viking boat burial from Swordle Bay on the Ardnamurchan peninsula, western Scotland. *Antiquity*, 91(355), 191-206. https://doi.org/10.15184/aqy.2016.222

Henry, G. (1976). Galena, Illinois during the lead mine era (Publication No. 3442). [Masters thesis, Eastern Illinois University]. The Keep. https://thekeep.eiu.edu/theses/3442

Herbert, P. Jr., & Young, R.S. (1956). Sulfide Mineralization in the Shenandoah Valley of Virginia. *Department of Conservation and Development Virginia Division of Geology Bulltein* 70.

Hevrnk, A.W., Radford, C.S. (1997). Missouri Mines state historic site: Park Hills, Missouri. *Rocks & Minerals*, *72*(6), 387-392. https://doi.org/10.1080/00357529709605071

Heyl, A.V., Delevaux, M.H., Zartman, R.E., & Brock, M.R. (1966). Isotopic study of galenas from the Upper Mississippi Valley, the Illinois-Kentucky, and some Appalachian Valley mineral districts. *Economic Geology*, *61*, 933-961. https://doi.org/10.2113/gsecongeo.61.5.933

Hinners, T.A., Hughes, R., Outridge, P.M., Davis, W.J., Simon, K., & Woolard, D.R. (1998). Interlaboratory comparison of mass spectrometric methods for lead isotopes and trace elements in NIST SRM 1400 Bone Ash. *Journal of Analytical Atomic Spectrometry*, *13*, 963–970. https://doi.org/10.1039/A803373K

Hinton, J. (2019). *A radiographic exploration of vitamin D deficiency at the eighteenth-century Fortress of Louisbourg, NS* [Masters thesis]. University of New Brunswick.

Holibaugh, J.R. (1895). *The Lead and Zinc Mining Industry of Southwest Missouri and Southeast Kansas*. The Scientific Publishing Company.

Hornsby, S.J. (2005). *British Atlantic, American frontier: Spaces of power in early modern British America*. University Press of New England.

Hull, S.H. (2006). *Tors Cove Cemetery - Permit 06.37*. Report on file at the Newfoundland and Labrador Provincial Archaeology Office.

Hull, S.H. (2007). *Foxtrap 2, Delaney's Road: Site Inspection*. Report on file at the Newfoundland and Labrador Provincial Archaeology Office.

Hume, I.N. (1969). *A guide to the artifacts of colonial America*. University of Pennsylvania Press.

Irey, E.F. (1951). A social history of Leadville, Colorado, during the boom days, 1877-1881 (Publication No. 003163) [Doctoral dissertation, University of Minnesota]. Proquest Dissertations Publishing.

Joel, E.C., Taylor, J.J., Ixer, R.A., & Goodway, M. (1997). Lead isotope analysis and the Great Orme Mine, In A. Sinclair, E. Slater, & J. Gowlett (Eds.), *Archaeological Sciences 1995: Proceedings of a conference on the application of scientific techniques to the study of archaeology* (pp. 123-131). Oxbow Archaeological Monograph.

Johnston, J. (1995a). From port de pêche to ville fortifiée: The evolution of urban Louisbourg, 1713-1758. In E. Krause, C. Corbin, and W. O'Shea (Eds.), *Aspects of Louisbourg* (pp. 3-18). The University College of Cape Breton Press.

Johnston, J. (1995b). The fishermen of eighteenth-century Cape Breton: Numbers and origins. In E. Krause, C. Corbin, and W. O'Shea (Eds.), *Aspects of Louisbourg* (pp. 198-208). The University College of Cape Breton Press.

Johnston, A.J.B. (2001). *Control and Order in French Colonial Louisbourg, 1713-1758*. Michigan State University Press.

Kaas, L.M. (2016). The history of zinc mining in Friedensville, Pennsylvania. *2016 Mining History Journal*, 17-42.

Kamenov, G.D., Lofaro, E.M., Goad, G., & Krigbaum, J. (2018). Trace elements in modern and archaeological human teeth: Implications for human metal exposure and enamel

diagenetic changes. *Journal of Archaeological Science*, *99*, 27–34. https://doi.org/10.1016/j.jas.2018.09.002

Keller, A.T., Regan, L.A., Lundstrom, C.C., & Bower, N.W. (2016). Evaluation of the efficacy of spatiotemporal Pb isoscapes for provenancing of human remains. *Forensic Science International*, *261*, 83–92. https://doi.org/10.1016/j.forsciint.2016.02.006

Kesler, S.E., Cumming, G.L., Krstic, D., & Appold, M.S. (1994a). Lead isotope geochemistry of Mississippi Valley-type deposits of the Southern Appalachians. *Economic Geology*, *89*, 307-321. https://doi.org/10.2113/gsecongeo.89.2.307

Kesler, S.E., Appold, M.S., Cumming, G.L., & Krstic, D. (1994b). Lead isotope geochemistry of Mississippi Valley-type mineralization in the Central Appalachians. *Economic Geology*, *89*, 1492-1500. https://doi.org/10.2113/gsecongeo.89.7.1492

Killick, D.J., Stephens, J.A., & Fenn, T.R. (2020). Geological constraints on the use of lead isotopes for provenance in archaeometallurgy. *Archaeometry*, *61*(S1), 86-105. https://doi.org/10.1111/arcm.12573

Knüsel, C.J., Batt, C.M., Cook, G., Montgomery, J., Müldner, G., Ogden, A.R., Palmer, C., Stern, B., Todd, J., & Wilson, A.S. (2010). The identity of the St Bees Lady, Cumbria: An osteobiographical approach. *Medieval Archaeology*, *54*(1), 271-311. https://doi.org/10.1179/174581710X12790370815931

Kowal, W., Beattie, O.B., Baadsgaard, H., & Krahn, P.M. (1991). Source identification of lead found in tissues of sailors from the Franklin Arctic Expedition of 1845. *Journal of Archaeological Science*, *18*, 193–203. https://doi.org/10.1016/0305-4403(91)90048-T

Laffoon, J.E., Shuler, K.A., Millard, A.R., Connelly, J.N., & Schroeder, H. (2020). Isotopic evidence for anthropogenic lead exposure on a 17th/18th century Barbadian plantation. *American Journal of Physical Anthropology*, *171*, 529–538. https://doi.org/10.1002/ajpa.23938

Lasemi, Z. (2010). Lead, zinc, and fluorite mining. In *Geology of Illinois* (pp. 299-308). University of Illinois, Institute of Natural Resource Sustainability, Illinois State Geological Survey Urbana-Champaign.

Le Guen, M., Orgeval, J.-J., & Lancelot, J. (1991). Lead isotope behaviour in a polyphased Pb-Zn ore deposit: Les Malines (Cévennes, France). *Mineral. Deposita*, *26*, 180–188. https://doi.org/10.1007/BF00209256

Lear, M., & Grimes, V. (2016). From airplanes to clandestine burials: A preliminary report on archaeogeophysical (GPR) surveys during 2016. In S. Hull & M. Delphina (Eds.), *Provincial Archaeology Office Annual Review for 2015 Field Season* (pp. 151-157). Provincial Archaeology Office Department of Tourism, Culture, Industry and Innovation Government of Newfoundland and Labrador.

LeHuray, A.P. (1982). Lead isotopic patterns of galena in the Piedmont and Blue Ridge ore deposits, Southern Appalachians. *Economic Geology*, *77*, 335-351. https://doi.org/10.2113/gsecongeo.77.2.335

Losier, C., & Lear, M. (2018). *Final report fir Tors Cove 2016 (ChAf-01 and ChAf-02) - Permit 16.07*. Report on file at the Newfoundland and Labrador Provincial Archaeology Office.

Losier, C., Loewen, B., & Goya, M.E. (2018). In the Midst of Diversity: Recognizing the Seventeenth-Century Basque Cultural Landscape and Ceramic Identity in Southern Newfoundland and Saint-Pierre-et-Miquelon. *Newfoundland and Labrador Studies*, *33*, 200–236. https://doi.org/10.7202/1055870ar

Lüders, V., Romer, R.L., Gilg, H.A., Bodnar, R.J., Pettke, T., & Misantoni, D. (2009). A geochemical study of the Sweet Home Mine, Colorado Mineral Belt, USA: Hydrothermal fluid evolution above a hypothesized granite cupola. *Mineralium Deposita*, *44*(4), 415-434.

Manders, G.C., & Aber, J.S. (2014). Tri-state mining district legacy in northeastern Oklahoma. *Emporia state research studies*, *49*(2), 29-51.

Marcoux, E. (1986). Isotopes du plomb et paragenèses métalliques, traceurs de l'histoire des gîtes minéraux (Publication No. 117). [Doctoral dissertation, Université Clermont-Ferrand II].

Marcoux, E., Moëlo, Y., Picot, P., & Baubron, J.-C. (1988). Évolution minéralogique et isotopique (Pb) du filon sulfuré complexe des Borderies (Massif central français) —

Implications métallogéniques. *Mineralium Deposita*, *23*, 58–70. https://doi.org/10.1007/BF00204229

Métreau, L., Cattin, F., Villa, I.M., André, P., & Chateau-Smith, C. (2021). Lead provenance for medieval decorated tile glazes from Brittany and Anjou (13th-14th c.). *Journal of Archaeological Science: Reports*, *38*, e103037. https://doi.org/10.1016/j.jasrep.2021.103037

Michard Vitrac, M., Albarede, F., Allègre, C.J. (1981). Lead isotopic composition of Hercynian granitic K-feldspars constrains continental genesis. *Nature*, *291*, 460-464. https://doi.org/10.1038/291460a0

Millard, A., Montgomery, J., Trickett, M., Beaumont, J., Evans, J., & Chenery, S. (2014). Childhood lead exposure in the British Isles during the Industrial Revolution. In M.K. Zuckerman (Ed.), *Modern Environments and Human Health: Revisiting the Second Epidemiologic Transition* (pp. 279-299). John Wiley and Sons. https://doi.org/10.1002/9781118504338.ch15

Millen, T.M., Zartman, R.E., & Heyl, A.V. (1995). *Lead Isotopes from the Upper Mississippi Valley District: A Regional Perspective*. U.S. Geological Survey Bulletin 2094-B, pp. B1-B3.

Millhouse, P.G. (2010). Native American lead mining in the Galena River Valley and the potential for archaeological research. *Journal of the Illinois Archaeology Survey*, *22*(1), 318-357.

Montgomery, J. (2002). *Lead and Strontium Isotope Compositions of Human Dental Tissues as an Indicator of Ancient Exposure and Population Dynamics* (Publication No. 453) [Doctoral dissertation, University of Bradford]. Archaeology Data Service. https://doi.org/10.5284/1000249

Montgomery, J., Evans, J.A., Powlesland, D., & Roberts, C.A. (2005). Continuity or colonization in Anglo-Saxon England? Isotope evidence for mobility, subsistence practice, and status at West Heslerton. *American Journal of Physical Anthropology*, *126*, 123-138. https://doi.org/10.1002/ajpa.20111

Montgomery, J., Evans, J., Chenery, S., Pashley, V., & Killgrove, K. (2010). “Gleaming, white and deadly”: using lead to track human exposure and geographic origins in the Roman period in Britain. *Journal of Roman archaeology Supplements, 78*, 199–226.

Mosier, M. (1948). *Bamford zinc mine, Lancaster County, Pa*. US Department of the Interior, Bureau of Mines.

Munkittrick, T.J.A., Varney, T.L., Pike, K.-A., & Grimes, V. (2019). Life histories from the Southside Cemetery, St. John’s, Newfoundland: Insights into Royal Naval diet using stable isotopes. *Journal of Archaeological Science: Reports*, *24*, 815–828. https://doi.org/10.1016/j.jasrep.2019.02.016

Myers, S.H. (1977). A survey of the traditional pottery manufacture in the Mid-Atlantic and Northeastern United States. *Northeast Historical Archaeology*, 6(6), article 2. https://doi.org/10.22191/neha/vol6/iss1/2

Ogle, D.H., Doll, J.C., Wheeler, P., & Dinno, A. (2022). *FSA: Fisheries Stock Analysis. R package version 0.9.3*. https://github.com/fishR-Core-Team/FSA.

Patil, I. (2021). Visualizations with statistical details: The 'ggstatsplot' approach. *Journal of Open Source Software*, *6*(61), 3167. https://doi.org/10.21105/joss.03167

Pike, K.-A. (2013). *Bearing identity: a biocultural analysis of human remains from Old Mission Point (C1Dq-1), New Brunswick* (Publication No. 6356) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Pin, C., Gannoun, A., & Dupont, A. (2014). Rapid, simultaneous separation of Sr, Pb, and Nd by extraction chromatography prior to isotope ratios determination by TIMS and MC-ICP-MS. *Journal of Analytical Atomic Spectrometry*, *29*, 1858–1870. https://doi.org/10.1039/C4JA00169A

Pitt, R.D. (2015). Portugal Cove-St. Philip’s. In *Canadian Encyclopedia*, https://www.thecanadianencyclopedia.ca/en/article/portugal-cove-st-philips

Poole, J.L. (1974). Notes on some abandoned copper, lead, and zinc mines in the Piedmont of Virginia. *Virginia Minerals*, *20*(2), 9-14.

Price, J.W. (1947). A brief history of the Pequea silver mine, Lancaster County, Pennsylvania. *Proceedings of the Pennsylvania Academy of Science*, *21*, 52-57.

Qiu, Y. (2023, May 03). *showtext: Using fonts more easily in R graphs*. https://cran.rstudio.com/web/packages/showtext/vignettes/introduction.html

R Core Team (2022). *R: A language and environment for statistical computing*. R Foundation for Statistical Computing. https://www.R-project.org/.

Rabinowitz, M.B., & Wetherill, G.W. (1972). Identifying sources of lead contamination by stable isotope techniques. *Environmental Science and Technology*, *6*, 705–709. https://doi.org/10.1021/es60067a003

Reddigan, K. (2007, November). *Ferryland District*. 1857 census Ferryland District. http://ngb.chebucto.org/C1857/1857-ferryland-district-census-1.shtml

Rohl, B.M. (1996). Lead isotope data from the Isotrace laboratory, Oxford: Archaeometry data base 2, galena from Britain and Ireland. *Archaeometry*, *38*(1), 165-180. https://doi.org/10.1111/j.1475-4754.1996.tb00769.x

Rohl, B., & Needham, S. (1998). *The circulation of metal in the British Bronze Age: the application of lead isotope analysis*. British Museum.

Samuelsen, J.R., & Potra, A. (2020). Biologically available Pb: A method for ancient human sourcing using Pb isotopes from prehistoric animal tooth enamel. *Journal of Archaeological Science*, *115*, e105079. https://doi.org/10.1016/j.jas.2020.105079

Scott, A.B., Danforth, M., MacInnes, S., Hughes, N., & Fonzo, M. (2020). Colonial Urbanism: A Comparative Exploration of Skeletal Stress in Two Eighteenth Century North American French Colonies. In T.K. Betsinger, S.N. DeWitte (Eds.), *The Bioarchaeology of Urbanization: The Biological, Demographic, and Social Consequences of Living in Cities, Bioarchaeology and Social Theory* (pp. 275-294). Springer International Publishing. https://doi.org/10.1007/978-3-030-53417-2_11

Scott, A.B., MacInnes, S., Hughes, N., Munkittrick, T.J.A., Harris, A.J.T., & Grimes, V. (2023). A Bioarchaeological Exploration of Adolescent Males at the Eighteenth-Century Fortress of Louisbourg, Nova Scotia, Canada. *Bioarchaeology International, Emerging Adolescence in Bioarchaeology: Current Status and Potential Development*, *7*(2), 146–172. https://doi.org/10.5744/bi.2022.0007

Shaw, H., Montgomery, J., Redfern, R., Gowland, R., & Evans, J. (2016). Identifying migrants in Roman London using lead and strontium. *Journal of Archaeological Science*, *66*, 57-68. https://doi.org/10.1016/j.jas.2015.12.001

Stos-Gale, Z.A., & Gale, N.H. (2009). Metal provenancing using isotopes and the Oxford archaeological lead isotope database (OXALID). *Archaeological and Anthropological Science*, *1*, 195–213. https://doi.org/10.1007/s12520-009-0011-6

Svergensky, D.A., Rye, D.M., & Doe, R.B. (1979). The lead and sulfur isotopic compositions of galena from a Mississippi Valley-type deposit in the New Lead Belt, Southeast Missouri. *Economic Geology*, *74*, 149-153.

Teutsch, N., Erel, Y., Halicz, L., & Banin, A. (2001). Distribution of natural and anthropogenic lead in Mediterranean soils. *Geochimica et Cosmochimica Acta*, *65*, 2853–2864. https://doi.org/10.1016/S0016-7037(01)00607-X

Valentine, B., Kamenov, G.D., Kenoyer, J.M., Shinde, V., Mushrif-Tripathy, V., Otarola-Castillo, E., & Krigbaum. J. (2015). Evidence for patterns of selective urban migration in the Greater Indus Valley (2600-1900 BC): A Pb and strontium isotope mortuary analysis. *PLOS One*, *10*, 1-20. https://doi.org/10.1371/journal.pone.0123103

Venovcevs, A. (2017). *Not just fisherfolk: winter housing and the seasonal lifeways of rural Euro-Newfoundlanders* (Publication No. 12636) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Venovcevs, A., & Gaulton, B.C. (2018). Winter Housing: Archaeological Perspectives on Newfoundland's Non-Pastoral Transhumant Tradition. In E. Costello, E. & Svensson (Eds.), *Historical Archaeologies of Transhumance across Europe, European Journal of Archaeology Monograph* (pp. 43-57). Routledge.

Vidal, C. (2012). For a comprehensive history of the Atlantic World or histories connected in and beyond the Atlantic World. *Annales: Histoire, Sciences Sociales*, 67(2), 279-300. https://doi.org/10.1017/S2398568200000674

von Hunnius, T. (1998). *South Side Naval Cemetery Remains from St. John's, Newfoundland: 1725-1825*. Ms. on file at Newfoundland and Labrador Provincial Archaeology Office, St. John's, Newfoundland.

Whisonant, R.C. (1996). Geology and the civil war in southwestern Virginia: The Wythe county lead mines. *Virginia Minerals*, *42*(2), 13-19.

Wickham, H. (2016). *ggplot2: Elegant Graphics for Data Analysis*. Springer-Verlag. https://ggplot2.tidyverse.org.

Wickham, H., Averick, M., Bryan, J., Chang, W., McGowan, L.D., François. R., Grolemund, G., Hayes, A., Henry, L., Hester, J., Kuhn, M., Pedersen, T.L., Miller, E., Bache, S.M., Müller, K., Ooms, J., Robinson, D., Seidel, D.P., Spinu, V., Takahashi, K., Vaughan, D., Wilke, C., Woo, K., & Yutani, H. (2019). Welcome to the tidyverse. *Journal of Open Source Software*, *4*(43), e1686. doi:10.21105/joss.01686.

Wickham, H., François, R., Henry, L., & Müller, K. (2022). *dplyr: A grammar of data manipulation*. R package version 1.0.10. https://dplyr.tidyverse.org, https://github.com/tidyverse/dplyr

Williams, D. (2019). "A large house on the downs": household archaeology and middle-class gentility in early 19th-century Ferryland, Newfoundland (Publication No. 13777) [Masters thesis, Memorial University of Newfoundland]. Memorial University Research Repository.

Wyman, T.S. (2018). Embreeville Mines. Tennessee Encyclopedia. [published by Tennessee Historical Society]. https://tennesseeencyclopedia.net/entries/embreeville-mines/