Hidden dangers?
An investigation of volcanic and environmental impacts on human health and life in historical Iceland

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Abstract

Volcanic eruptions can cause significant human health and environmental threats both during and after their event due to the hazardous materials and gases that are actively or passively released into the surrounding environment. Historical records suggest that severe historic eruptions in Iceland caused mass mortality to livestock, famine, altered weather and led to the contamination of water and air, all of which significantly impacted the health and living condition of people in the past. The aim of the project was to investigate the effects of volcanic eruptions on human health across Icelandic history, as well as the impacts of the anthropogenic use of heavy metals (e.g., Hg, As, Pb) and climate change (e.g., cooling weather during the Little Ice Age).

The study used a range of different methods but mainly analyses on human and animal bones and soil samples. Standard osteological analyses were conducted on skeletal individuals (n=186) from archaeological sites (n=7) across Iceland dated between the 10th and the 19th century. Samples were then collected for further analyses: human bone (n=36) and teeth samples (n=31), as well as animal bone samples (n=23) and soil samples (n=13), which were selected from the monastic-hospital site of Skriðuklaustur (AD 1493-1554). In addition to previously published comparative data, bone samples (n=14) and soil samples (n=9) were selected from a farm site, called Skeljastaðir, which was abandoned during the AD 1104 eruption of the nearby volcano Hekla. Standard osteological and palaeopathological methods were used for the skeletal analysis and anthropological descriptions. Microscopy, radiography, endoscopy, and other specialized techniques were used where necessary. Isotope ($\delta^{18}O$, $^{87}Sr/^{86}Sr$, $\delta^{13}C$) and trace element (Hg, Pb, Cd, As, Zn, Sb, Ba, Sr) analysis of dental enamel was undertaken to investigate geographic provenance and possible exposure to toxic emissions during childhood. At the same time, isotope ratio mass spectometry of bone collagen samples ($\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$) from humans and animals was used to reconstruct palaeodiet and provide indications about overall nutrition. Trace element analysis (ICP-MS and ISE) was also used on bone samples to investigate ante-mortem exposure to toxic elements of volcanogenic origins (F, Hg, Pb, Cd, As). Isotope analyses conducted for the reconstruction of geographic provenance of the people buried at Skriðuklaustur showed a local population born in Iceland that came to the monastery seeking treatment, hospice, trade, or religious activity from throughout the south-eastern quarter of the country. Dietary reconstruction showed a mixed marine and terrestrial diet with freshwater fish input at the monastic-hospital Skriðuklaustur, while a primarily terrestrial protein diet with freshwater fish input at the inland farm site Skeljastaðir.

Overall, the results indicated that the burden of skeletal fluorosis on the human population was low, perhaps because people, unlike the livestock, quickly fled from areas directly impacted by volcanic fallout. The skeletal burden of most other volcanogenic pollutants was also low, indicating that any slight elevations were the result of natural, passive background emissions or limited anthropogenic exposure. However, mercury was elevated in these skeletal assemblages, due to both anthropogenic uses (e.g., as a medicine) and volcanogenic exposure. The impacts of volcanic hazards in the past cannot be dismissed and they certainly caused complicated, life-threatening, and long-term effects upon the living conditions and health of people in the past. On the other hand, it seems that the immediate dangers (e.g., volcanic smoke, lava flow) were mostly circumvented, unlike previously hypothesized. People likely mitigated against volcanic disasters by temporarily moving away from areas undergoing eruptions.

Útdráttur

Eldgos geta ógnað heilsufari fólks og haft alvarleg áhrif á náttúruna, bæði meðan á þeim stendur og til langs tíma, sökum hættulegra efna og eitraðra lofttegunda sem þau losa út í umhverfið. Stór eldgos á Íslandi hafa áður verið talin hafa valdið stórfelldum búfjárdauða, hungri, breytingum á veðurfari og vatns- og loftmengun, sem hafði í kjölfarið mikil áhrif á heilsu og lífsskilyrði íbúa landsins fyrr á tíð. Markmið þessarar rannsóknar var að kanna hversu mikil áhrif eldsumbrotin höfðu á heilsufar fólks í gegnum aldirnar á Íslandi en um leið að skoða í sama skyni önnur mengunaráhrif í umhverfinu, til dæmis af mannavöldum (s.s. með notkun kvikasilfurs, arseniks og blýs) og vegna loftslagsbreytinga (s.s. kólnandi hitastigs á Litlu ísöld).

Í rannsókninni var margvíslegum aðferðum beitt en einkum stuðst við greiningar á beinum manna og dýra, auk jarðvegssýna. Í úrtaki hennar voru bein 186 einstaklinga frá sjö mismunandi stöðum á Íslandi sem voru í byggð allt frá 10. öld til 19. aldar. Þá voru sértækar greiningar gerðar á gögnum frá Skriðuklaustri í Fljótsdal, þar sem var klaustur og spítali á árabilinu 1493-1554, og frá Skeljastöðum í Þjórsárdal, sem lögðust í eyði í kjölfar Heklugoss árið 1104. Alls voru 36 sýni úr mannabeinum og 31 tannsýni, sem og 23 dýrabeinasýni og 13 jarðvegssýni valin til rannsóknarinnar frá Skriðuklaustri. Til samanburðar voru valin 14 beinasýni og níu jarðvegssýni frá Skeljastöðum. Hefðbundnum beina- og meinafræðilegum aðferðum var beitt við greiningar á beinunum. Smásjárskoðanir, röntgenmyndatökur, speglanir og aðrar sérhæfðar aðferðir voru nýttar til frekari rannsókna þegar þess þurfti. Ísótópagreiningar ($\delta^{18}O$, $^{87}Sr/^{86}Sr$, $\delta^{13}C$) og snefilefnarannsóknir (Hg, Pb, Cd, As, Zn, Sb, Ba, Sr) á tannglerungi voru framkvæmdar til að kanna landfræðilegan uppruna einstaklinga og mögulega útsetningu fyrir eitrun í æsku. Samhliða voru gerðar massagreiningar á samsætum úr kollagensýnum ($\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$) sem tekin voru úr beinum manna og dýra til að afla vísbendinga um mataræði og almenna næringu einstaklinganna. Snefilefnarannsóknir (ICP-MS og ISE) voru jafnframt gerðar á beinasýnum til að kanna útsetningu fyrir eiturefnum úr eldgosum (F, Hg, Pb, Cd, As) fyrir andlát. Niðurstöður ístópagreininga sýna að þau sem leituðu til Skriðuklausturs, til að sækja sér læknismeðferð, í viðskiptalegum erindagerðum eða af trúarlegum ástæðum, voru fædd á Íslandi og virðast hafa komið hvaðanæva að af suðausturhorni landsins. Niðurstöður rannsókna á mataræði íbúa þar leiddu ennfremur í ljós að meginuppistaða fæðuvals þeirra var blanda af sjávar- og landdýrum, auk þess sem merki voru um neyslu á ferskvatnsfiski. Íbúar Skeljastaða hafa hins vegar fyrst og fremst nærst á prótínríkri fæðu úr landdýrum, auk nokkurs ferskvatnsfisks.

Helstu niðurstöður rannsóknarinnar eru þær að tíðni flúoreitrunar í sýnaúrtakinu er almennt lág, hugsanlega sökum þess að fólk, ólíkt búfé, flúði svæðin fljótt í kjölfar eldsumbrota. Eituráhrif vegna flestra annarra gosefna mældust sömuleiðis lág, sem gefur til kynna að smávægileg eiturfrávik séu til komin vegna óbeinna, náttúrulegra orsaka eða takmarkaðra útsetninga af mannavöldum. Þó mældust gildin hærri þegar kom að útsetningu fyrir kvikasilfurseitrunum, bæði af völdum eldgosa og manna. Á meðan ekki verður horft fram hjá þeirri hættu sem eldgos ollu á Íslandi á fyrri tíð, virðist sem að beinir áhættuvaldar (s.s. gosgufur og gjóska) hafi haft takmarkaðri áhrif á heilsufar en áður hefur verið talið. Engu að síður hafa eldgos vissulega valdið flóknum, lífshættulegum langtímaáhrifum á framfærslu-, búsetu- og veðurfarsleg skilyrði sem síðan geta haft óbein áhrif á lifnaðarhætti og heilsu fólks. Líklegt er að fólk hafi komist hjá slíkum hamförum með tímabundnum búferlaflutningum frá gossvæðum. Í framtíðinni gæti verið mikilvægt að beina sjónum að viðbúnaði landsvæða á virkum gossvæðum gegn langtímaáhrifum félagslegra, pólitískra, umhverfislegra og efnahagslegra þátta fremur en að horfa til alvarleika skammtímaáhrifa sem eldsumbrot kunna að valda.

List of Publications

This doctoral thesis is based upon the following articles:

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II. **Walser III, J.W.**, Kristjánsdóttir, S., Gröcke, D., Gowland, R., Jakob, T., Nowell, G., Ottley, C., and 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. doi: 10.1002/ajpa.23973

III. **Walser III, J.W.**, Gowland, R.L., Desnica, N. and Kristjánsdóttir S. **2020**. Hidden dangers? Investigating the impact of volcanic eruptions and skeletal fluorosis in medieval Iceland. *Archaeological and Anthropological Sciences,* **12**: Article 77. doi: 10.1007/s12520-020-01026-0

All published papers were reproduced here with permission from the journals concerned: reproduction licenses were obtained from the *International Journal of Osteoarchaeology* and *Archaeological and Anthropological Sciences*. A license was not required from the *American Journal of Physical Anthropology* because it has been published as an open-access article.

Other publications that are referenced and relevant to this thesis, but were not included in it:

Ebenesersdóttir, S.S., Sandoval-Velasco, M., Gunnarsdóttir, E.D., Jagadeesan, A., Guðmundsdóttir, V.B., Thordardottir, E.L., Einarsdóttir, M.S., Moore, K.H.S., Sigurðsson, A., Magnúsdóttir, D.N., Jónsson, H., Snorradóttir, S., Hovig, E., Moller, P., Kockum, I. , Olsson, T., Alfredsson, L., Werge, T., Cavalleri, G.L., Gilbert, E., Lalueza-Fox, C., **Walser III, J.W.**, Kristjánsdóttir, S., Gopalakrishnan, S., Árnadóttir, L., Magnússon, O.P., Gilbert, M.T.P., Stefánsson, K. and Helgason, A. 2018. Ancient genomes from Iceland reveal the making of a human population. *Science* **360**(6392): 1028-1032. DOI 10.1126/science.aar2625

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This work is dedicated to R^2, my mom, H., Granny, dad, my brother, and the rest of my family and to the loving memory of Allison Plummer (1988-2017) and Joseph B. Ruth (1963-2021).

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"Since the outbreak began, the atmosphere of the whole country has been full of vapour, smoke and dust, so much so that the sun looked brownish red, and the fishermen could not find the banks... Old people, especially those with weak chests, suffered much from the smell of sulphur and the volcanic vapours, being affected with dyspnoea. Various persons in good health fell ill, and more would have suffered had not the air been cooled and refreshed from time to time by rains" (Holm pg.57-60 quoted in Creighton, 1965, pg. 414).

1 Introduction

Environmental perils often remain concealed and overlooked despite the constant influences ecological conditions assert upon life and landscapes. Tectonic activity, earthquakes, and volcanic eruptions can cause severe natural disasters, such as those which have occurred throughout Icelandic history. Volcanoes are both destructive and productive and affect climatic, geologic, human, animal, vegetative, and other environmental constructs through their eruptions. They may cause significant human health and environmental threats both during and after their event due to the hazardous materials and gases that are actively or passively released into the surrounding environment (Hansell et al., 2006). Additionally, volcanic eruptions are known to cause the death of crops, livestock, inland fish and marine fauna near volcanic activity and the drying of wells following ground water level changes (Black, 1981, Grattan, 2006). In Iceland, climatic conditions during the Middle Ages complicated cultivation and losing many crops or animals at one time could be catastrophic to a population's subsistence strategy and overall health (Mehler, 2011; McGovern et al., 2014). Changes in climate, the use of land, the environment and availability of resources all directly affect public health (Semenza and Menne, 2009). The connection between the environment and human health has been observed since ancient times, such as in "Airs, Waters and Places" by Hippocrates (5th century BC) (Roberts, 2016). Nonetheless, archaeological research, has only infrequently considered the relationship between human health and pathological conditions resulting from environmental causes, such as volcanic emissions (see Petrone et al., 2013; Nelson et al., 2016).

Archaeological skeletal remains have been retained from archaeological excavations in Iceland as far back as the end of the 19th century. Only a few skeletal analyses, such as Bruun (1903) and Hooton (1918), were conducted prior to 1939 when the medical doctor Jón Steffensen (1905-1991) first began to analyse skeletons from the excavation of cemetery at Skeljastaðir in Þjórsárdalur valley. From this point forward, Jón Steffensen served as the only physical anthropologist working on skeletal remains in Iceland until his death in 1991 (Zoëga and Gestsdóttir, 2010). Meanwhile, physical anthropologist Jens Ólafur Páll Pálsson (1926-2002) established the Anthropological Institute of the University of Iceland (Mannfræðistofnun Háskóla Íslands) in 1969, working mainly with anthropometry, taking measurements of living people. Despite the systematic approach of skeletal analysis that began with Jón Steffensen's work, the principal aims of the analyses at that time were focused on using non-metric traits to assess the ancestry of the settlers of Iceland and identifying specific individuals from coffin inscriptions, historical records, burial locations, and comparisons of skulls to portraits (Steffensen, 1988; Zoëga and Gestsdóttir, 2010). These objectives were intertwined with the strong focus held by historians and archaeologist on identifying individuals and verifying events and locations described in the sagas at that time. Since then, the subject has changed considerably, moving away from case studies aimed at individual identification, with practitioners focusing instead on population analyses, palaeodemography, palaeopathology, migration studies, palaeodietary reconstructions and other multidisciplinary approaches to anthropological and bioarchaeological analyses (Zoëga and Gestsdóttir, 2010; Gestsdóttir, 2012). Recently, Icelanders, health researchers and many others have expressed concerns about the environmental and human health impacts of volcanic emissions (Gislason and Alfredsson, 2010).

The research presented here will provide statistically relevant findings regarding the relationship between health and the volcanic environment in the past that will deepen our understanding of volcanogenic emissions upon the human body and Icelandic environnment in the present. The overarching aim of this thesis was to examine the impact of volcanogenic earth elements on human health in historical Iceland through osteological and biochemical analyses of human skeletal remains. Human/environment interactions were contextualized by assessing skeletal assemblages representing populations historically residing near volcanic activity or during known volcanic events. Climatic changes, volcanic eruptions, natural resources and environmental disease burden were considered to be important agents to the shifting cultural and occupational behaviors occurring throughout history. Recent research studies have extensively addressed the relationship between pathological conditions and exposure to volcanogenic pollutants (Horwell et al., 2013). However, this type of study has only very rarely been conducted on archaeological populations. The diagnosis of elemental toxicity is otherwise extremely rare in bioarchaeological or palaeopathological cases (Littleton, 1999; Gestsdóttir et al., 2006). Finally, the study also considered the importance of and connection between environmental and genetic influences on human health. This thesis aimed, moreover, to expand upon the scientific, historical, and medical record of volcanic and environmental health conditions, which still afflict modern populations, with both Icelandic and international applicability.

1.1 Aims and Objectives

The overarching aim of this research – as noted above – was to use osteological and archaeometric analyses to examine the health impacts and disease burden sustained by the historical population of Iceland as a consequence of the volcanic environment. In addition to skeletal changes, archaeometric analyses using inductively coupled mass spectrometry (ICP-MS) and ion-selective electrode (ISE) were employed to directly examine the uptake of toxic elements in bone. The estimation of elemental exposure in past populations is limited by the lack of pathognomonic osteological markers for environmental toxicities. Thus, considering bone element concentrations in correlation with bone pathological changes can provide additional perspective, as well as assist in identifying toxic exposure in incomplete individuals or individuals that lack bone pathologies. Isotope analyses were used for dietary reconstruction and to identify the geographic provenance of the analysed individuals, as both provide detailed information about the life course and the potential for exposure to toxic elements or emissions.

The osteological component of this research aimed to explore environmental disease burden from the 10th-19th centuries through skeletal analyses of 186 adult individuals from seven sites across Iceland. The seven sites include Skeljastaðir (ÞSK), representing the 10th-12th centuries, Skriðuklaustur (SKR) and Haffjarðarey (HFE), representing the 13th-16th centuries, and Viðey (VEY), Bessastaðir (BES), Reykholt (RKH) and Reykjavík (RVK), representing the 17th-19th centuries. Due to funding limitations and ethical considerations regarding the destructive effects of isotopic and elemental analyses, which require human bone or dental tissues, only individuals from Skeljastaðir (10th-12th centuries) and Skriðuklaustur (15th-16th centuries) were sampled. Although it remains a limitation in this research, these two earlier periods were selected over the later sites, due to the additional confounding factors faced during the 17th-19th century (e.g., industrialization, urbanization, increased trade, and shift in occupations). For example, mercury was used in product manufacturing factories (e.g., textiles, amalgams, early electronics), lead became more common in goods and infrastructure, and cadmium exposure likely increased due to the increased use of tobacco products (Parsons and Percival, 2005; Brodziak-Dopierala et al., 2015). Deficiency diseases, particularly scurvy and rickets, are also believed to have increased significantly during this period (Jónsson, 1998), thereby implying that the risk of toxic elemental exposure would have also increased. Therefore, elemental exposure in the 17th-19th centuries should be analysed further in a future study, aimed at evaluating whether population density and the cultural changes occurring at that time increased elemental burden.

1.1.1 Investigating mercury exposure with ICP-MS and osteological analyses (Article I)

The objective of the mercury analysis was to investigate mercury exposure in medieval Iceland as a function of environmental exposure via volcanic eruptions or degassing and anthropogenic exposure via medical (e.g., as a treatment for infectious disease), cultural practices (e.g., as a pigment in religious manuscripts) or subsistence strategy (e.g., consumption of marine resources). The research aimed to use ICP-MS on bone and soil samples (n=50, human; n=23, animal; n=22, soil) to determine mercury concentrations. Skeletal analysis was also performed to attempt to correlate potentially elevated mercury concentrations with mercury induced bone or dental changes, as well as characterize the pathological conditions that may have been treated with mercury in the past. Since Skeljastaðir (ca. AD 1000-1104) was inhabited until the eruption of Mt. Hekla in AD 1104, it was hypothesized that the population residing there, represented by a skeletal assemblage (n=56), were likely chronically exposed to mercury through passive degassing. Some individuals may have been acutely exposed to mercury during the eruption. Meanwhile, it was hypothesized that at least certain individuals represented by the skeletal assemblage (n=295) from Skriðuklaustur (AD 1493-1554) were likely exposed to the mercurial formulations commonly used in Europe at that time to treat infectious diseases, such as venereal syphilis.

1.1.2 Investigating Diet and Geographic Provenance with Isotope Analyses (Article II)

The aim of the isotope analyses was to perform palaeodietary reconstruction ($\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$) using bone samples collected from Skeljastaðir (ca. AD 1000-1104) and Skriðuklaustur (AD 1493-1554) and to determine the geographic provenenance of the people from Skriðuklaustur using isotopic analyses ($\delta^{18}O$, $^{87}Sr/^{86}Sr$) on dental enamel samples. Additionally, the research aimed to evaluate ante-mortem exposure to lead (Pb) during childhood, considering its environmental, anthropogenic, health implications and as a marker of geographic origin. Barium (Ba), zinc (Zn), strontium (Sr), arsenic (As), antimony (Sb), and mercury (Hg) trace element concentrations were also determined, as they can likewise impart information about provenance, population composition, biocultural differences between individuals, and possibly palaeodiet or toxic exposure during childhood. Reconstructing palaeodiet and geographic origin was not only important to the overall context of the research; diet has important implications for overall health, which can also affect one's predisposition to elemental uptake. Geographic origin or residence has important implications about whether people would have been exposed to environmental emissions in the first place based on their residential proximity to volcanogenic emissions or activity. This aspect of the project also aimed to produce baselines and a novel dataset of trace elements measured in archaeological remains as well as attempt to examine the possible health impacts of toxic element accumulation. For example, can elevated toxic element concentrations be correlated with osteological indications of traumatic injury, metabolic disease, or other skeletal changes? A further aim of the dental enamel analysis was to investigate the use, efficacy, and value of trace elements in the application of determining geographic provenance or origin. The hypotheses were that despite its inland location, the population of Skriðuklaustur would represent a wide range of individuals including immigrants and exhibit a diet primarily derived from marine resources due to religious fasting. Palaeodiet at Skeljastaðir on the other hand was analysed in a previous study (Sveinbjörnsdóttir et al., 2010), indicating a primarily terrestrial protein diet, but the addition of sulphur isotope analysis provided new indications about migration to the site and freshwater fish consumption.

1.1.3 Investigating fluoride exposure with ICP-MS and osteological analyses (Article III)

The aim of the fluoride analyses was to diachronically investigate the health burden of fluoride emissions from volcanogenic sources on multiple skeletal assemblages from seven Icelandic sites listed above. The study examined bone changes potentially correlated with skeletal fluorosis, as well as determine fluoride concentrations in bone (n=50) and soil (n=4) samples from Skeljastaðir and Skriðuklaustur. As Skeljastaðir is located near the volcano Mt. Hekla, a known fluoride and mercury emitter, and is believed to have been abandoned around the time of the AD 1104 eruption, it was hypothesised that the individuals residing there may have been acutely or chronically exposed to fluoride emissions. Furthermore, several modern eruptions have proven lethal to livestock grazing in the same region as the site, indicating that fluoride emissions likely polluted the surrounding water sources and foliage. Similarly, the Veiðivötn-Bárðarbunga volcanic system, relatively close to Skriðuklaustur, underwent a momentous eruption in AD 1477, prior to the establishment of the monastery. Since the eruption caused severe destruction resulting in the abandonment of farms in the region, it was hypothesised that fluoride pollution to aquifers and foliage may have affected the individuals that were buried at Skriðuklaustur, who were all local to the area according to the isotopic analyses.

1.1.4 Investigating exposure to other trace elements in bone (Pb, Cd, As) with ICP-MS (Thesis Chapter 5)

An additional aim was to investigate other toxic heavy metals in bone and dental enamel samples from Skriðuklaustur and Skeljastaðir, potentially resulting from environmental emissions or anthropogenic activity. Lead (Pb), cadmium (Cd) and arsenic (As) were evaluated in addition to the previously discussed analysis of mercury (Hg) and fluoride (F) as toxic emissions that may be released during volcanic eruptions or degassing and their potential effects on the human population of medieval Iceland. The elements measured in bone samples aimed to draw conclusions about toxic exposure during childhood (dental enamel) and adulthood (bone) within a few years prior to death.

1.2 Research Questions

The general research questions addressed by the articles (I, II, III) and additional analyses included in this thesis are:

1.2.1 How did environmental conditions impact subsistence, landscapes, human health, and culture in past Icelandic populations?

1.2.2 What were the geographic origins of the people residing at Skeljastðir and Skriðuklaustur and does provenance relate to pathological conditions or exposure to toxic elements?

1.2.3 Did volcanogenic emissions of mercury (e.g., from the Hekla eruption of AD 1104) or other elements affect the health of the local population of Skeljastaðir? Were any toxic substances used medicinally at the monastery?

1.2.4 Regarding exposure to toxic elements, were there any identifiable differences (e.g., cultural, occupational, or behavioral) between men, women, children or individuals of different social status or age groups?

1.2.5 Were socio-cultural or environmental conditions responsible for dietary shifts between the populations living during the Medieval Warm Period (Skeljastaðir assemblage) and the Little Ice Age (Skriðuklaustur assemblage)?

1.2.6 Does osteological evidence for metabolic or nutritional distress show any relationship with pathological conditions, toxic element exposure or individual or population diet?

1.2.7 Did historic eruptions in Iceland result in mass human mortality such as occurred among livestock?

1.2.8 What osseous pathologies are present in the human skeletal assemblages and are they associated with heavy metal exposure or fluorosis, as revealed by ICP-MS and ISE?

1.3 Thesis Outline

This thesis was produced by publication rather than traditional format, with the major results presented in a series of three published manuscripts (Article I, II, III), which are republished here in Chapter 8: Manuscripts. Additional unpublished results are also described in Chapter 5: Results and Discussion.

Chapter 1. Introduction. Chapter 1 presents a general introduction to the project, followed by aims: assessing volcanic pollutants in bone and pathological skeletal changes associated with them, reconstructing palaeodiet and identifying geographic provenance of skeletal populations. The chapter also includes the research questions addressed in this thesis and finally the thesis outline.

Chapter 2. Background: Volcanoes, elements, and health in the past. Chapter 2 provides a background discussion covering volcanic activity and ecology in Iceland, the health effects of volcanic emissions, environmental and climate change in historic Iceland, the record of past eruptions, cultural or occupational factors leading to exposure and the outcomes of Icelandic volcanic eruptions on flora, fauna, and human populations. It also reviews pathological conditions and the history/bioarchaeology, biological mechanisms and skeletal implications of exposure to volcanogenic toxic emissions and toxic substances or materials derived from the same elements. It thus addresses how skeletal and chemical evidence may be used to attempt to identify cases of exposure to toxic earth elements in archaeological human remains.

Chapter 3. Background: Origins and Diet. Chapter 3 discusses the geographic provenance of archaeological individuals and the ancestral origins of the Icelandic population. This chapter also discusses past diet and themes related to dietary subsistence. Essentially, the chapter reviews previously published studies that have addressed questions of provenance and past diet.

Chapter 4. Materials and Methods. Chapter 4 presents the materials and methods used during this research. Firstly, the archaeological sites and populations represented by the analysed skeletal assemblages are described, followed by detailed descriptions of the osteological methods, skeletal changes associated with mercury and fluoride toxicity, the methods used in elemental analyses (ICP-MS, ISE) on cortical bone samples and finally the methods used for the isotope and trace element analyses conducted on bone collagen and dental enamel samples.

Chapter 5. Results and Discussion. Chapter 5 describes the results of the various analyses conducted in this project. Firstly, the results of the chemical analysis of animal bones and soil samples (ICP-MS) are presented, followed by the results of mercury (Hg) analysis in human bone samples and dental enamel. A discussion on the health burden of fluorosis (F) in the past as indicated by osteological analysis and ISE analysis of bone samples follows. The analytical results and discussion of lead (Pb), cadmium (Cd), arsenic (As) and antimony (Sb), are then respectively presented. Finally, the results of isotope analyses conducted on bone collagen and dental enamel for the reconstruction of geographic provenance and past diet, are then discussed. The chapter also provides a discussion, describing the relevance and meaning behind the results of this project and relationship or connection between the various strands of evidence described in Articles I, II and III and differing approaches of analyses which sought to answer questions about historical human health as correlated with environmental change and volcanic activity.

Chapter 6. Conclusions. Chapter 6 presents a summary of the project, the aims and findings of each article and the overall conclusions of the completed research.

Chapter 7. Bibliography. This chapter contains the bibliographic list of publications cited in this thesis summary and the thesis articles featured in Chapter 8.

Chapter 8. Manuscripts. This chapter contains the published articles (I, II, III) of the thesis.

2 'Black sun, high flame:' Volcanoes, elements, and health in the past

2.1 Volcanism in Iceland

Volcanoes and volcanic activity are represented by an array of differing structures constructed around geothermal vents that can be formed either by monogenetic (volcanoes erupting only once) or polygenetic eruptions (volcanoes that may erupt repeatedly) (Francis and Oppenheimer, 2004). Throughout history, Iceland has experienced all known forms of eruption and volcano types except diatremes, which have caused impacts on the hemispheric scale if not globally following some of the larger eruptions (Thorarinsson and Sæmundsson, 1979; Thorarinsson, 1981a, 1981b; Thordarson and Larsen, 2007). Types of volcanic eruptions, typified by effusive basaltic eruptions as well as explosive styles (e.g., felsic eruptions, mafic subglacial phreatomagmatic eruptions) occurring in Iceland range from classical conical shape stratovolcanos to archetypal mafic lava shields (Thordarson and Larsen, 2007). Volcanic systems in Iceland are composed either of a central volcano or fissure swarm, or both.

Iceland's basaltic plateau is situated upon the junction of the Mid-Atlantic Ridge and the Greenland-Iceland-Faroe Ridge, two extensive submarine physiographic structures, and rises more than 3000 m above the local sea floor (Gudmundsson, 2000; Thordarson and Larsen, 2007). Its formation likely began around 24 million years ago, resulting from interactions occurring between a mantle plume and a spreading plate boundary, but the oldest exposed rocks only date to 14-16 million years old (McDougall et al., 1984; Allen et al., 1999; Thordarsen and Larsen, 2007). This interaction is represented today through the geological architecture seen in Iceland, most apparently noticeable from the arrangement of its active volcanic zones and its elevation from the sea floor (Thordarsen and Larsen, 2007). There are 30 identified volcanic systems situated within the active volcanic zones of Iceland, which cover about one-third of the country and include the Reykjanes Ridge, Reykjanes Volcanic Belt, South Iceland Seismic Zone, West Volcanic Zone, Mid-Iceland Belt, East Volcanic Zone, North Volcanic Zone, Tjörnes Fracture Zone, Kolbeinsey Ridge, Öræfi Volcanic Belt and the Snæfellsnes Volcanic Belt (Jóhannesson and Sæmundsson, 1989; Thordarson and Höskuldsson, 2002; Thordarson and Larsen, 2007) (see Figure 4.1 or the volcanic system map at www.vedur.is). The East Volcanic Zone, which contains the most active volcanic systems – Veiðivötn, Hekla, Grímsvötn and Katla – have produced as much as 80% of all historically verified eruptions in Iceland (Thordarsen and Larsen, 2007).

2.1.1 Volcanic pollution

Aside from affecting human health, volcanic eruptions can cause local and global climate change, air pollution, environmental alteration, and a wide array of detrimental effects to humans, animals, and vegetation, both natural and cultivated (D'Alessandro, 2006; Grattan, 2006). Volcanoes emit numerous gases and other elements, during and between eruptions, including Carbon dioxide (CO_2), Sulphur dioxide (SO_2), Hydrogen chloride (HCL), Ammonia (NH_3), Hydrogen sulfide (H_2S), Hydrogen fluoride or Hydroflouric acid (HF), Carbon monoxide (CO), halides, tephra, silica, and radon (Weinstein and Cook, 2005; Hansell et al., 2006). These gases rapidly interact with the ashes and the atmospheric water in the volcanic plume forming acidic aerosols. Such volcanic emissions do not only occur during eruptions but also consistently appear between eruptions in volcanic and geothermal systems through passive degassing (Thordarson et al., 1996; Delmelle, 2002; Hansell et al., 2006; Tchounwou et al., 2012). Silica and radioactive radon are bound by falling ash and can, when inhaled, become lodged in the respiratory tract causing toxicity and respiratory complications (Hansell et al., 2006). The effects of exposure to volcanic particulates can occur either chronically or acutely. For example, fluoride poisoning, a chronic condition, is of primary concern in the examination of the health impacts of volcanic pollution (Weinstein and Cook, 2005). Acute conditions have been noted during various modern eruptions throughout the world (e.g., Mount St. Helens – Bernstein et al., 1986; Montserrat – Forbes et al., 2003; Mount Etna – Fano et al., 2010; Hawaii – Mannino et al., 1996; Miyake Volcano – Scojima et al., 2006). Rarely, volcanic eruptions can also cause mortality through extremely elevated temperatures resulting in thermal lung burns, deep cutaneous burns and asyphyxiation. For example, it has been reported that at Herculaneum, Italy, those who perished during the eruption of Vesuvius in AD 79 exhibited heat-related skeletal markers (Petrone et al., 2018; Martyn et al., 2020). Respiratory burns can result in permanent respiratory damage, fatal pharyngeal or pulmonary edema, or secondary respiratory infections and can even permeate protective gear or structures because of the extremely small size of many volcanic particles (Hansell et al., 2006).

Tephra is toxic if inhaled, consumed, or masticated and exists in phases including vesiculated glass, plagioclase, orthopyroxene, clinopyroxene, titanomagnetite and olivine (Hansell et al., 2006; Bergþórsdóttir, 2018). Tephra, and many other volcanic emissions including hydrochloric acid (HCl), halides and fluoride, are highly soluble and are easily incorporated into the soil of a volcanic region after the particles leach into the surface of the soil. The fluoride bearing compounds are present in the formed aerosols and adhere to tephra particles during and after eruptions. Additionally, because of the water-solubility of tephra, halides, HCl and fluoride, they are rapidly absorbed into the nose, oral mucosa and respiratory tract causing poisoning and respiratory disorders, especially bronchitis and asthma (Weinstein and Cook, 2005; Hansell et al., 2006). Mass fluoride poisoning of animals following volcanic eruptions throughout the world and the attrition effects of animals masticating glass-laden tephra deposited upon their grazing vegetation have been seen extensively in Iceland and elsewhere (Thorarinsson, 1981a; D'Alessandro, 2006; Grattan, 2006). The toll on human health of Icelandic volcanic eruptions has also been noted throughout Europe as the gases, ash and tephra are carried from Iceland on winds across the British Isles and onto continental Europe (Elliot et al., 2010; Offer et al., 2012; Carlsen et al., 2012a, 2012b).

Aside from respiratory conditions and fluoride poisoning, volcanic eruptions also directly cause other health conditions, such as asphyxiation, carbon dioxide poisoning, and crop, groundwater, and residential contamination (for example, with the Hekla eruptions in 1947-1948 and 1970). In fact, autopsies of volcanic eruption victims often determine the primary cause of death as asphyxiation. In addition to chronic and acute diseases, smoke, ash, tephra, and silica inhalation due to volcanic emissions have been linked to neoplastic diseases (cancer), particularly malignant mesothelioma and radon lung cancer, following long-term exposure (Mauderly, 1997; Horwell and Baxter, 2006). Respirated silica, perhaps the most toxic mineral found in volcanic ash deposits, or radicals, may also lead to DNA mutation and cancer by reacting directly with DNA by causing strands to break. Other respiratory pathologies, such as chronic obstructive pulmonary disease (COPD), silicosis (fibrotic lung scarification), and resurgence of dormant pulmonary tuberculosis or pneumoconiosis, may also occur from respirated silica (Harrison et al., 1994; Horwell et al., 2003; Horwell and Baxter, 2006; Hansell et al., 2006).

During volcanic eruptions, many gases and other volatile elements are deposited into the soil, upon crops, or into watersources thereby contaminating it and to a degree the crops growing in it. Soil contamination and acidification results in difficulties in feeding livestock and growing crops for years after an eruption (Young et al., 2004; Stewart et al., 2006). Ash can remain in the environment for decades after an eruption and the wind and human activity can remobilize it (Hansell et al., 2006). When inhaled, these gases cause irritation, toxicity, inflammation, and other damage to the respiratory system and alters the oxygen environment of the maxillary sinuses allowing the proliferation of anaerobic bacterial growth. Thus, regular exposure to volcanic emissions can cause chronic maxillary sinusitis, a condition which was prevalent in historical Iceland (see Collins, 2019). These emissions cause chlorotic and necrotic damage to leaves and fruits or vegetables of wild or cultivated flora, which can lead to reduced crop and produce yields (Delmelle et al., 2002). Outdoor cultivation in Medieval Iceland was limited to the short summer period and losing many crops at one time could be significantly detrimental to a population's subsistence and thus result in nutritional stress, decreased pathogen resistance and overall health. Most importantly, volcanic emissions contaminate (e.g., with fluoride) nearby water sources, as well as clean crops, and cause pH changes that increase the acidity of the water, thus endangering any organisms drinking from or living in it (Delmelle et al., 2002; Horwell and Baxter, 2006; Hansell et al., 2006). Wet climates that experience regular and heavy rainfall compound the already high solubility of the fluoride bound tephra particles, and potentially other particulates, into the surrounding soils (Young et al., 2004). Fluoride can be released into the surrounding environment almost immediately, as occurred during the 1970 Hekla eruption for example, while other eruptions release fluoride in successive extractions, often in highly soluble phases (Baxter et al., 1982; Cronin et al., 2003; Horwell et al., 2006; Baxter, 2009).

In Iceland, throughout the Medieval Warm Period (ca. AD 9th to13th century) (Cronin et al., 2003), subsistence strategies were based upon agriculture, fishing, marine mammal hunting (e.g., seals and walrus) and imported domestic livestock, including cattle, sheep, goats, horses, and pigs (Júlíusson, 2013; McGovern et al., 2014). Following eruptions, toxicity risk is especially high for free roaming livestock due to daily surface water consumption and grazing upon foliage embedded within ash-laden vegetation and topsoil, especially in wet areas altered by erosion or animals and people treading through the land (Shupe et al., 1963; Cronin et al., 2003; D'Alessandro, 2006; Gísladóttir et al., 2010). Grazing animals have most obviously demonstrated fluorine toxicity (chronic and acute) particularly following volcanic eruptions, such as at Mt Hekla, Iceland and Mt. Ruapehu, New Zealand (see Figure 2.1; D'Alessandro, 2006). While most animal fatalities were due to acute fluorosis following these eruptions, some survived long enough to exhibit dental enamel pitting and discoloration, inappetence (loss of appetite) and ataxia (loss of control of muscles or voluntary movements) (Thorarinsson and Sigvaldason, 1972; Cronin et al., 2003; Grattan, 2006; D'Alessandro, 2006). According to historical records, nearly all 23 historical eruptions of Hekla produced toxic fallouts, many of which would have caused the mass mortality of livestock (Dugmore and Véststeinsson, 2012) and potentially of humans (Grattan, 2006). Aside from volcanic and geothermal activity, modern exposure to fluoride and heavy metals in Iceland occurs with residential proximity to aluminum smelting factories, especially among terrestrial grazing mammals (Schlegel, 1974; Krater and Rose, 2009).

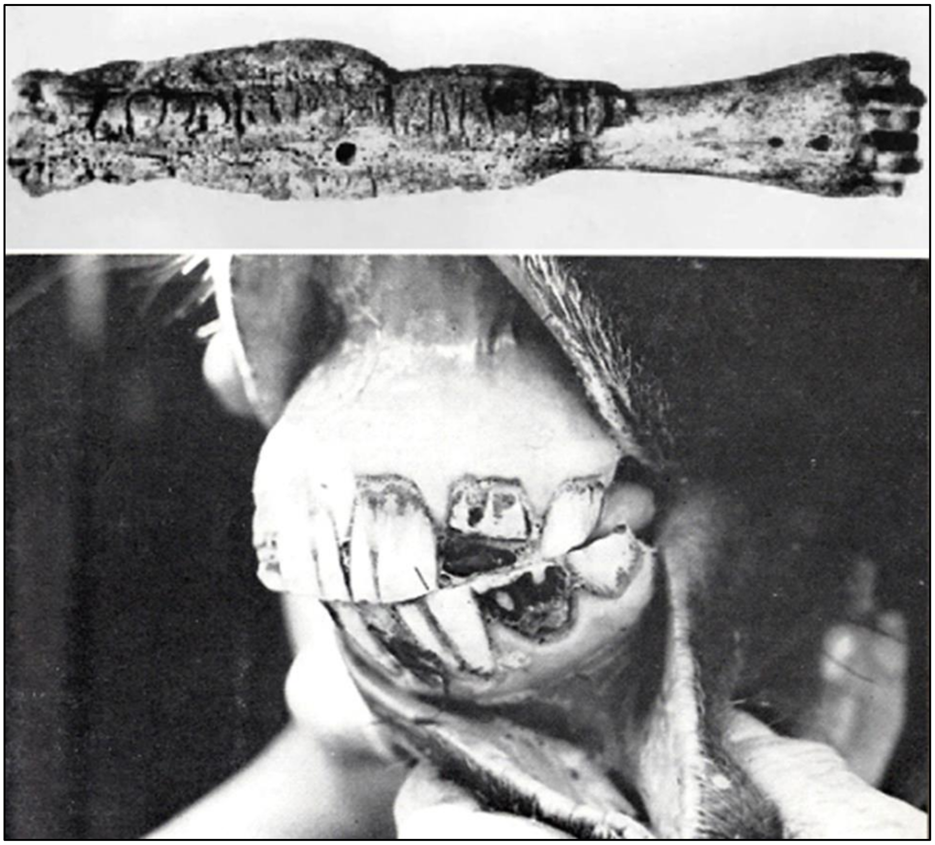

Figure 2.1 Top, sheep metatarsal with a bone fluoride concentration of 20,600 ppm (parts per million) and bone changes caused by skeletal fluorosis during the Mt. Hekla eruption of 1845. Reprinted from Roholm (1939). Bottom, dental fluorosis ("ash-teeth") seen in horse dentition following the Hekla eruption of 1970. © Keldur, the Institute for Experimental Pathology at the University of Iceland

2.1.2 Historical volcanic eruptions and climate change

The volcanic eruption of Mount Samalas, located in modern-day Indonesia, that occurred in AD 1257-1258, is thought to have initiated the dramatic cooling event throughout the North Atlantic known as the Little Ice Age (ca. 13^{th}-19^{th} centuries), succeeding the Medieval Warm Period (ca. 9^{th}-13^{th} centuries). At least seven other volcanic eruptions occurred throughout the world altering environmental conditions, increasing sea ice, storms, and changes in currents, and leading to a decline in farming production, marine mammal populations and navigation in Iceland during these times (Cronin et al., 2003; Matthews and Briffa, 2005; McGovern et al., 2014). Farms were often abandoned long-term or permanently as a result, as in the south at Skeljastaðir during the eruption of Hekla in AD 1104 (Þórðarson, 1943; Dugmore and Vésteinsson, 2012). One of the largest known historical eruptions in Iceland occurred in Veiðivötn in the east near Skriðuklaustur in AD 1477; a monastic-hospital site heavily featured in this study. The impacts of this violent eruption were felt throughout Iceland, caused crop failure, devastated the landscape, human and animal populations and produced one of the largest tephra falls of historical times (Larsen, 1988; Rafnsson, 1990; Thordarson and Larsen, 2007; Global Volcanism Program, 2013). For a modern comparison, the 2014-2015 Bárðarbunga-Veiðivötn eruption released toxic gases and metals in the atmosphere, including over 60,000t of sulphur dioxide (SO_2) and 500t of hydrogen chloride (HCl) into the atmosphere per day, causing respiratory illnesses across Iceland (Galeczka et al., 2016). Sulphur deposits into ground soils, upon crops but also into the surrounding water supplies and sources, which can be a severe environmental hazard for human and animal health and agricultural systems (Cronin et al., 2003; Horwell and Baxter, 2006).

Sulphur dioxide is a highly reactive irritant in the human body even at low concentrations impacting people both near and distant from the volcanic activity. Lung function may decrease within a few minutes of exposure, particularly if pre-existing conditions exists (e.g., asthma) (Delmelle et al., 2002; Hansell et al., 2006; Horwell and Baxter, 2006; Roberts, 2007).

British folklore and historical literature mention that the effects of the Hekla eruptions were felt across Europe and left cold, dark winters that lasted for decades (Grattan, 2006). The impacts of eruptions of Mt. Hekla echoed across Europe, causing long, dark, and cold winters. For example, the 1970 eruption of Hekla emitted very large amounts of fluorine (and mercury) causing the death of up to 8000 animals in Iceland as grazing pastures were permeated with lethal doses of fluoride (see Figure 2.2; Thordarson and Self, 2003; Gudmundsson et al., 2008). During the Laki eruption of AD 1783-1784, a high concentration of sulphur, fluorine and chlorine was released into the air resulting in the death of 20% of the population and up to 75% of the livestock in Iceland (Thordarson and Self, 2003; D'Alessandro, 2006; Guðmundsson et al., 2008; Grattan, 2012). Contemporaneous descriptions note that the population faced deficiency diseases (e.g., scurvy, rickets), serious malnourishment and plagues (e.g., smallpox) that raged across the country (Pétursson et al., 1984; Halldórsson, 2013; Sigurðardóttir, 2017).

Figure 2.2 Left, a farmer and his son from Húnavatnssýsla stand beside a grave of their sheep that died of fluoride poisoning following the 1970 eruption of Mt. Hekla. © Magnús Finnsson. Right, sheep that died of fluoride poisoning following the Hekla eruption of 1947-1948. Reprinted from Hekla on Fire by Sigurður Þórarinsson (1956)

From the time of the Settlement (ca. AD 874) until AD 1200, there were approximately ten volcanic eruptions per century recorded. During the Late Middle Ages, the number of eruptions increased to twelve per century. It is probable that far more eruptions occurred than were recorded as their documentation may be incomplete. There are at least 205 recorded “fires,” or volcanic eruptions, mentioned in historical documents. In the post-Medieval era (after AD 1600), there were approximately 27 eruptions per century, significantly increasing the importance of understanding the health and environmental impacts of volcanic activities (Thordarson and Larsen, 2006). Climatic conditions began to worsen leading up to the Little Ice Age (ca. AD 13th-19th centuries), which likely resulted from the combination of a series of volcanic eruptions (initiated by the AD 1257 eruption of Mt. Salamas) throughout the world (Ogilvie and Jónsson, 2001; Matthews and Briffa, 2005; McGovern et al., 2014) and the terrestrial carbon sink (increased accumulation of carbon on the land surface) that occurred due to the sharp rise in mass fatalities worldwide during the spread of the Black Death (van Hoof et al., 2006; Yeloff and van Geel, 2007; Keenan and Williams, 2018). The Little Ice Age contributed heavily to the deterioration of grazing lands, living conditions, navigability, and the viability of farming in Iceland (McKinzey et al., 2005).

2.2 Health in the past

In this section, pathological conditions relevant to this research project, or conditions that will be discussed later in the results (see Chapter 5), are described. The descriptions presented here cover skeletal pathologies recorded in this study from individuals from seven Icelandic archaeological sites: Skeljastaðir, Haffjarðarey, Skriðuklaustur, Reykjavík, Bessastaðir, Reykholt and Viðey (see section 4.1). Treponemal disease, such as venereal syphilis, and hydatic disease (cystic echinococcus), are relevant to this study due to the historical use of mercury as a medical treatment for these conditions. Paget’s disease and orofacial clefts are described here because individuals featuring these conditions were identified and provided unique or outlying results, likely correlated with their pathological conditions. Although not found in a high frequency in this study, rickets and osteomalacia are described here because these conditions were thought to be common in historical Iceland and in light of their connections with heavy metal toxicity and skeletal fluorosis. Caries, calculus, and periodontal disease are discussed both because of their relationship and interactions with each other and with fluoride exposure.

2.2.1 Treponemal disease

Treponemal disease was an important infectious disease observed during this research due to its high frequency among the Skriðuklaustur cemetery population (ca. AD 1493-1554) and because of the connection between this infection and mercury as a medicinal treatment. At Skriðuklaustur at least nine individuals showed skeletal changes indicative of treponematoses, specifically probable venereal syphilis, according to the criteria outlined by Hackett (1976) and Ortner (2003) (e.g., see Figure 2.3; Walser III et al., 2019). Thus, infections with treponemal disease caused by the spirochete *treponema pallidum* spread around Iceland contemporaneously with the venereal syphilis epidemic in western Europe (Kristjánsdóttir, 2011; Walser III et al., 2019). Two other possible cases of treponemal disease have been described from Haffjarðarey and Viðey, although both are based only upon small, isolated, frontal lesions (Gestsdóttir, 2004; Hoffman, 2018; Walser III et al., 2019). While the appearance of both lesions is consistent with *caries sicca* resulting from treponemal disease, such diagnoses should be based on a combination of characteristic bone changes found throughout the entire skeleton (see e.g. Hackett, 1976; Ortner, 2003) and cannot be definitively diagnosed from isolated bone fragments with solitary frontal lesions (Cook and Powell, 2012). Such lesions could arise from other causes, such as scalping, neoplastic disease or tuberculosis, for example (Ortner, 2003). Another issue worth considering is that cases of treponemal infections consistent with endemic syphilis (treponarid), which may not be able to be differentiated from venereal syphilis in dry bone, have been described from medieval (16th century) Norway (Anderson et al., 1986). Although multiple hypotheses of the origin of venereal syphilis in the Old World persist today (Baker et al., 2020), a 2011 review found no solid evidence of a Pre-Columbian transmission of venereal syphilis in the Old World (Harper et al., 2011). Thus, in the case of the individual (HFE 34) from Haffjarðarey (occupied until AD 1563), a venereal syphilis infection was only likely to occur in the final ca. 70 years of the site’s use, although the occurence of endemic syphilis in Iceland prior to this date cannot be ruled out. So far, the only confidently diagnosable cases of treponemal disease were found among individuals buried at Skriðuklaustur.

The health impact of venereal syphilis was extensive throughout Europe and required a renegotiation of cultural practices, such as placing sanctions on prostitution, closing public baths and emphasizing the importance of religious devoutness. Divine punishment, bad air and bad blood were commonly thought to be the causative factors for contracting serious illnesses or impairments, such as syphilis or blindness for example (Meyer et al., 2002; Woolgar, 2006). These cultural changes also contributed to the “prudeness” associated with the Reformation. Other cultural manifestations, such as the wearing of gloves and wigs may have been used to conceal the visible syphilitic lesions that occur with long-term infections. Venereal syphilis is caused by contact with syphilitic lesions or bodily fluids, primarily during sexual contact, however it is also possible to become infected through poor hygiene and by using contaminated utensils or objects, albeit very rarely today. Venereal syphilis can also be acquired congenitally (i.e. transplacental infection *in utero*) (Meyer et al., 2002; Zuckerman, 2016, 2017a, 2017b).

Mercury concentrations in skeletal remains showing changes associated with venereal syphilis can reflect the biogenic uptake of it during life, providing secondary evidence of medical treatment for various conditions (e.g syphilis, leprosy, gonnorhea, lice) conducted at hospitals or monasteries (Rasmussen et al., 2015; Zuckerman, 2016, 2017). While syphilis eventually causes severe dermal lesions, skin conditions such as acrodynia, systemic allergic dermatitis, granulomas and others also occur due to mercury exposure (Boyd et al., 2000). Both venereal syphilis and mercury can also cause potentially irreversible psychological impairments (Pranjić et al., 2003; Crozatti et al., 2015). While mercury could be used to treat disease in the past, the ensuing mercury poisoning could not be treated (Guzzi and La Porta, 2008; Zuckerman, 2016, 2017a).

Figure 2.3 Cranium and tibia of young adult female individual (SKR 23) with bone changes suggestive of treponemal disease (probable venereal syphilis)

2.2.2 Hydatid disease

Hydatid disease, caused by a parasitic organism (echinococcus granulosus), was probably introduced to Iceland during the Settlement period towards the end of the 9th century and quickly became endemic throughout the country by AD 1200. Today the condition is believed to be completely eradicated, following experimentation to determine its lifecycle in 1863 and extensive public education and outreach thereafter (Kristjánsdóttir and Collins, 2011). Jón Hjaltalín (John Hjaltelin), the Inspecting Medical Officer of Iceland (1855-1881) of the time, used calomel and mercury based medicinals to treat some cases of hydatid disease (and syphilis), reportedly with success (Hjaltelin, 1868; Hjaltelin, 2013). Skeletal analyses of the individuals buried at Skriðuklaustur revealed at least 13 individuals with calcified hydatid cysts (e.g. see Figure 2.4; Kristjánsdóttir and Collins, 2011). The parasite has a lifecycle involving canids (e.g., dogs) as definitive hosts and ungulates as intermediate hosts. In the past, the traditional Icelandic practice of home butchering sheep and feeding their offal to dogs promoted the transmission of the parasite (Moro and Schantz, 2009). *Echinoccocuss granulosus* infections can form in the interosseous membrane of bone or viscera and remain active for decades as the parasites reproduce. The condition often causes pain, shock, allergic reactions, respiratory conditions, increased fracture rate, osteosclerosis, itching and several other signs and symptoms. Hydatid cysts may take up to a decade to grow substantially and do not always preserve well in archaeological contexts, implying that the disease burden of hydatidosis could have been much higher (Kristjánsdóttir and Collins, 2011). Aside from the cases found at Skriðuklaustur, a few other examples of cystic echinococcus have been discovered in 17th to 19th century individuals from Viðey (Gestsdóttir, 2004) and at Landssímareitur in Reykjavík (Zoëga, 2018).

Figure 2.4 Individual (SKR 126) from Skriðuklaustur with possible comorbidities of hydatid and treponemal disease. The black arrow indicates the hydatid cyst (© Steinunn Kristjánsdóttir)

2.2.3 Cleft lip and palate

Orofacial clefts are caused by the defective embryological development of the premaxillary, maxillary or palatal structures (Bhattacharya et al., 2009). Palatal clefts or perforations have several origins, (e.g., congenital, late-stage venereal syphilis), and generally result in substantial problems with drinking, eating, and speaking in uncorrected cases (see Figure 2.5 and 2.6; Patil, 2016; Ilczuk-Rypula et al., 2017). Breastfeeding is also challenging and compared with other children, people born with orofacial clefts generally take longer to adapt to consuming solid foods (Müldner et al., 2009; Wiet et al., 2017). In the past, infants with cleft lip had to be hand-fed, meaning that significant social care was required, especially for the first year of life, for there to be any chance of the child's survival (Bragg, 1994). In the 13th century, orofacial clefts were finally recognised as congenital conditions, although the first recorded successful surgical correction of a cleft palate was not performed until the 19th century. However, cleft lip surgeries or interventions have been recorded from as early as 390 BC (in China) (Bhattacharya et al., 2009). In *Þorgils saga skarða* from *Sturlunga saga*, a man called Þorgils skarði Böðvarsson is described as handsome, strong, attractive, and trustworthy despite being born with a cleft lip, from where his name "skarði" originates (Bragg, 1994). These descriptions indicate that, at the time the saga was written (13th-14th centuries), not all individuals with disfiguring conditions were socially marginalised and some could even be revered in Icelandic society. Recent research in the bioarchaeology of care further indicates that individuals with cleft palate may have been honoured and respected in the past, in light of their survival into adulthood and the typically non-deviant burials they received within their communities (Curry, 2019).

Only a few cases of cleft lip, cleft palate and other maxillofacial anomalies have been discovered in Iceland (e.g., Figure 2.5 and 2.6). One individual (HFE 34, Figure 2.6) was diagnosed with Facio-Auriculo-Vertebral sequence (FAVs) (see Hoffman et al., 2019), a congenital condition with probable genetic aetiology which is known to have extreme variability in range and severity between cases (Hartsfield, 2007). The pathognomic malformations related to this condition are microtia (underdevelopment of external ear soft tissues), with other major deformities of the mandible, orbits, and vertebrae (Mathog and Leonard, 1980; Hartsfield, 2007; Muñoz-Pedroza and Arenas-Sordo, 2013). There are no orbital deformations or asymmetry in the mandible and the vertebrae and soft tissues – the anatomical elements where pathognomonic features may occur – are not archaeologically preserved or available for study. The primary skeletal pathology potentially associated with FAVs is the incomplete right cleft premaxilla with asymmetry of the nasal bones and aperture.

Figure 2.5 Left, cleft maxilla and premaxilla from SKR 22, inferior view. Right, inferior view of maxilla showing palatal perforations (SKR 201) resulting from an infectious process, such as treponemal disease, rather than a congenital or developmental defect

Figure 2.6 Anterior view of orofacial anomaly, an incomplete right unilateral cleft premaxilla (lip) possibly caused by FAVs, in an adult female (HFE 34). Anterior view of complete right unilateral cleft premaxilla (lip) in an adult female (SKR 9)

2.2.4 Paget's disease of bone

Paget's disease of bone appears to be caused by environmental factors acting upon a genetically susceptible individual. The condition predominately affects the skeletal system and is characterized by the abnormal and excessive remodeling of bone and deposition of irregular new bone (see Figures 2.7 and 2.8; Whyte, 2006; Mays, 2010). The skull is usually involved, showing cortical bone thickening, a "cotton wool" appearance in the skull caused by areas of sclerosis and expansion of the diploë (Bhargava and Maki, 2010). The condition is usually initially asymptomatic, but a range of complications may arise throughout the clinical course, including bone pain, achiness, increased rate of fractures, osteoarthritis, osteosarcomas, neurological problems (e.g., hearing loss, headaches/migraines, dementia), spinal compression or ischemia, arthropathies and myopathies (Whyte, 2006). Paget's disease of bone is very rare, although it is likely underrepresented in the palaeopathological record because not all bones of the body are always affected; some bones may exhibit the characteristic changes of Paget's disease, while other bones in the same skeleton may appear radiographically and microscopically normal (Mays and Turner-Walker, 1999). In bioarchaeological research, individuals with Paget's disease of bone often exhibit other skeletal complications such as bone deformity, osteosarcoma, fractures, and secondary osteoarthritis (Mays, 2010). The condition is statistically significantly more frequent in males than in females, according to studies conducted on archaeological skeletal remains and on modern populations (Mays, 2010). In a worldwide literature review of archaeological cases of Paget's disease of bone, Mays (2010) found that 94% of cases came from England, with no cases coming from beyond western Europe. This strong geographic distribution indicates an origin of Paget's disease of bone in western Europe, or even in England (Mays, 2010). However, it is important to note that other factors could also explain this phenomenon, such as misdiagnosis, observer error, poor preservation of pathological bone, the "Osteological Paradox", or insufficient diagnostic recognition in other regions. These findings are relevant to this thesis because the condition persist today in a family residing in eastern Iceland and because an individual with Paget's disease of bone was excavated from the cemetery at Skriðuklaustur also located in eastern Iceland (Walser III et al., 2020a, 2020b). A second possible case has since been discovered from the cemetery at Landssímareitur in Reykjavík (Zoëga, 2018). The genetic heritage of the Icelandic settlement population consisting predominately of the Norse (from Scandinavia) and Gaelic populations of the British Isles (Ebenesersdóttir et al., 2018), could possibly indicate the origin of Paget's disease in Iceland. The condition is also relevant to this study as a differential diagnosis of skeletal fluorosis.

Figure 2.7 Right radius, ulna and humerus and left femur from an older adult male (SKR 174) with bone changes consistent with Paget's disease – anterior view

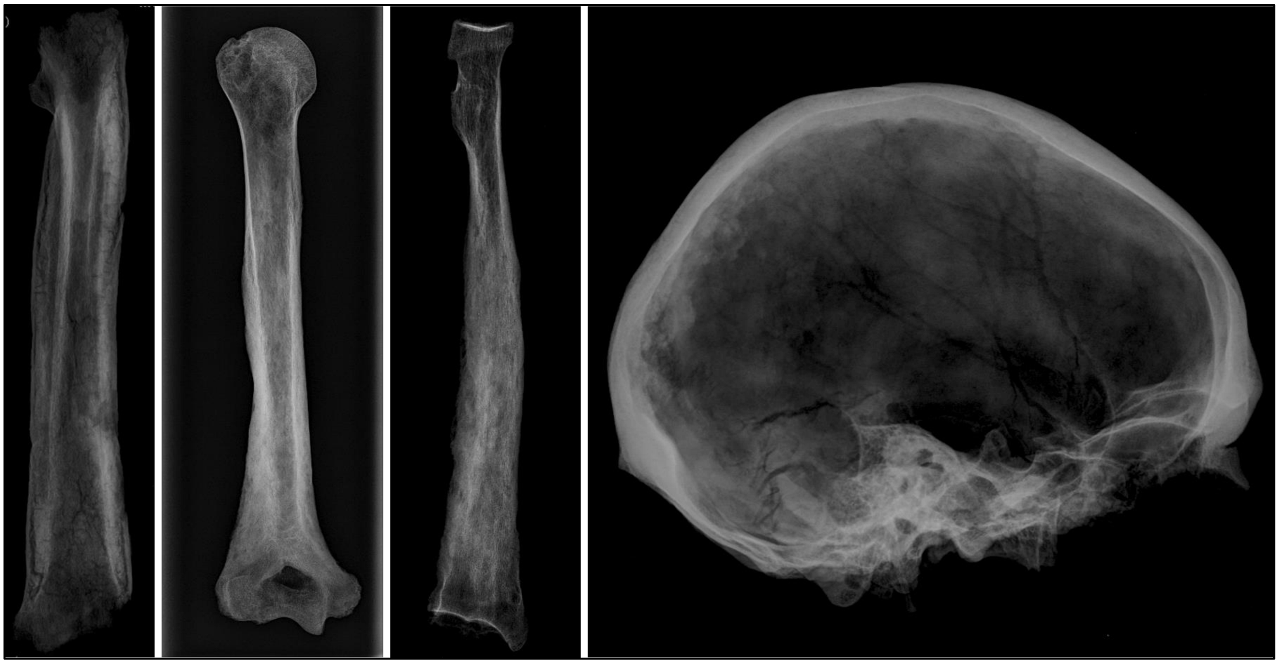

Figure 2.8 Radiographs showing skeletal changes observed in the older adult male (SKR 174) with Paget's disease, a differential diagnosis of skeletal fluorosis, from Skriðuklaustur. From left: left femur (anterior), right humerus (anterior), left radius (anterior) and cranium (right lateral aspect)

2.2.5 Rickets and osteomalacia

Rickets (childhood) and osteomalacia (adulthood) are conditions resulting from vitamin D deficiency, which causes reduced concentrations of calcium and phosphorous thereby inhibiting the mineralization of bone. Rickets has been recognized since antiquity but increased significantly in the post-Medieval period and during the Industrial Revolution in Europe. Other inherited or acquired conditions may also cause osteomalacia, but these cases are unusual compared to vitamin D deficiency osteomalacia (Ives and Brickley, 2014). Vitamin D is an important hormone that is best obtained from skin exposure to ultraviolet sunlight, but it can also be obtained from ingesting food sources containing it, such as eggs, oily fish, milk, and butter (Ives and Brickley, 2014), which were all staples of the past diet in Iceland (Gestsdóttir, 1998; Gísladóttir, 1999; Mehler, 2011). Apart from poor skeletal mineralization, the condition increases fracture risk, deforms bones (e.g., mandible, ribs, and long bones), inhibits the development of epiphyses and causes numerous health problems, although only infrequently results in death (Brickley et al., 2010; Ives and Brickley, 2014). The condition became more common in the medieval period due to overcrowding, poor hygiene, and urban sanitation, work indoors, environmental factors (e.g., pollution, industry) and cultural behaviors (e.g., swaddling infants, wearing clothes that prohibit sunlight from reaching the skin) (Brickley et al., 2010; Ives and Brickley, 2014).

Vitamin D deficiency (rickets and osteomalacia), iron anaemia and malnutrition were believed to be common in Iceland in the past due to the dark winter months, the latitude, and because of statements made in historical records and folklore (Gestsdóttir, 1998; Jónsson, 1998; Sigurðardóttir, 2017). However, vitamin D deficiency has only been rarely observed in Icelandic archaeological skeletal remains (Steffensen, 1943; Gestsdóttir, 1998, 1999; Sundman, 2011; Zoëga and Murphy, 2016). For example, early research by Jón Steffensen suggested that there were three individuals from Skeljastaðir with rickets or residual rickets (bone changes of childhood rickets persisting into adulthood), but a more recent reevaluation showed that only one of them had skeletal changes suggestive of vitamin D deficiency (Gestsdóttir, 1998). Perhaps the long summer months, the cultivation of angelica, the foraging of mushrooms, onions and wild berries, ample supply of freshwater fish, the consumption of seaweed, Iceland moss and imported foodstuffs meant that vitamin D deficiency and malnutrition were likely only encountered seasonally when these resources were limited. Nonetheless, vitamin D deficiency is described here due to its relationship with bone mineralization and the impacts that fluorosis and other toxic elements have on bone microstructure.

2.2.6 Caries

The prevalence of carious lesions (see Figure 2.9) is low in archaeological individuals from Iceland prior to the 18^{th}-20^{th} centuries (Walser III et al., 2020b). Research conducted in India noted that dental caries and fluorosis occur in combination, with some researchers explaining that it is important to recognise that fluorosed teeth are poorly mineralized teeth and not necessarily protected from dental disease (Yoder et al., 1998; Wondwossen et al., 2004; Susheela, 2007). Although it is known that fluoride impedes caries formation, the scale of its effects is unpredictable, and its functional concentrations are not uniform in all people. The level for reducing dental decay without causing detrimental side effects is normally indicated around 0.5 ppm (Littleton, 1999). While low-level fluoridated water can decrease caries prevalence, hypomineralisation caused by dental fluorosis increases caries risk. This relationship demonstrates the fragility of the ideal balance of fluoride intake. While the prevalence of caries is decreasing overall, the prevalence of fluorosis is increasing worldwide (Wondwossen et al., 2004; D'Alessandro, 2006; Petrone et al., 2013). In 2005, for example, at least 23% of people in the United States between 6-39 years of age exhibited at least mild enamel fluorosis (Everett, 2011). In a study conducted on Tanzanian children, it was shown that those exhibiting the least fluorotic teeth had the highest caries rate followed by those exhibiting the most fluorotic teeth. The lowest caries rate was seen in individuals with only moderately fluorotic teeth (Yoder et al., 1998).

The cariogenic bacteria *Staphylococcus mutans* has increased significantly since the Medieval period, becoming more dominant after the onset of the Industrial Revolution approximately 250 years ago (Warinner et al., 2014; Weyrich et al., 2015). Changes in food production and technology during this period allowed for the widespread production of processed, refined, and concentrated grains and sugars resulting in an increase of mono- and disaccharide consumption, which are primary actors in lowering plaque pH and in enamel demineralization (Adler et al., 2013; Warinner et al., 2014; Weyrich et al., 2015). Studies conducted on skeletal remains recovered from high fluoride regions such as Bahrain, Pakistan, and Abu Dhabi, for example, demonstrated that high carbohydrate and sugar consumption will result to some extent in carious lesions regardless of the protection that fluoride consumption may provide (Littleton, 1993; Yoshimura et al., 2006; Petrone et al. 2013). Another study of 2^{nd} and 3^{rd}-century individuals from Palmyra, Syria, connected the high fluoride levels in the water found in this arid region to fluorosis by measuring fluoride content in normal and suspected fluorotic teeth. The teeth with enamel hypomineralisation and staining contained a higher fluoride content than the teeth that were considered normal (Yoshimura et al., 2006). While rare in Iceland, fluorosis is still common in these arid regions today. For example, chronic fluoride toxicity remains endemic among the population of modern-day India and is associated with the local geology and dietary calcium deficiency (Teotia et al., 1998). Fluoride is not supplemented and is thus naturally low in Icelandic drinking water (median <0.1; range <0.1-0.6), except following eruptions, although it is used in toothpaste and other dental treatments today (Gunnarsdóttir, 2016). Modern caries rates in Iceland are generally considered high for European standards and are higher than in other Nordic countries (Ágústsdóttir et al., 2010). Still, dental fluorosis appears to be uncommon: past research found that most modern cases of enamel opacities were significantly associated with childhood illness, especially middle ear infections (otitis media), although some cases of diffuse enamel opacities could possibly be correlated with dental fluorosis (Árnadóttir et al., 2005). Previous studies show that caries rates reflect dietary conditions regardless of fluoride intake. However, caries rates may also be lower due to fluoride intake, thus complicating our understanding of the relationship between fluoride, caries, and diet, particularly regarding sugar and carbohydrates (Littleton and Frohlich, 1993).

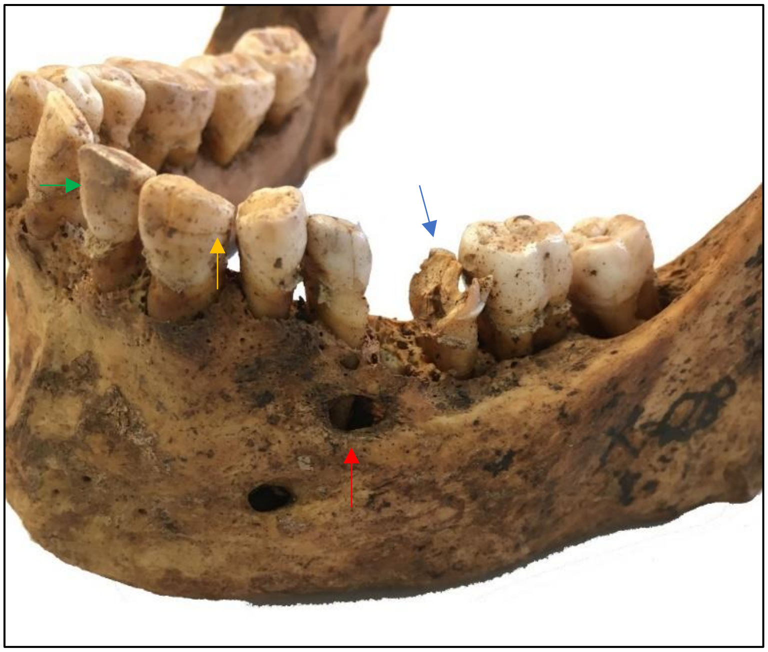

Figure 2.9 Mandibular dentition (BES 19) with carious lesion (blue), periapical lesion (red), linear enamel hypoplasia (yellow) and hypomineralisation (green; opacities in dental enamel) – anterolateral view

2.2.7 Calculus and periodontal disease

Dental calculus (and periodontal disease) was common in historical Iceland (see Figure 2.10; Sigurðardóttir, 2017). Calculus is essentially mineralized dental plaque, which sometimes includes other matter such as textile fragments, parasites, feathers, microparticles (e.g., protein, DNA) and botanical remains (e.g., phytoliths, pollen) (Weyrich et al., 2015; Juhola et al., 2019). The analysis of dental calculus can impart information about dietary resources, transitions in subsistence, cultural parafunctional behaviors (e.g., use of teeth as tools in weaving), ancient disease and the evolution of the microbiome (Hillson, 2003; Warinner et al., 2014; Juhola et al., 2019). Differences in calculus deposition between individuals can occur due to parafunctional or behavioral factors, such as preparing reeds for sewing by sliding them across the anterior mandibular teeth thereby removing calculus deposits (Greene et al., 2005, Aspiras et al., 2010). Sexually dimorphic caries rates can occur due to dietary differences, such as one sex in a population consuming more carbohydrates than the other (Greene et al., 2005). Inadequate dietary protein increases caries, especially when combined with high carbohydrate and sugar intake. Meanwhile, calculus rates increase with high starch, grain and protein diets (Hillson, 1979; Littleton and Frohlich, 1993; Johnson et al., 1995). Some studies suggest that high fluoride levels in dental calculus may also aid to inhibit the manifestation of carious lesions (Tatevossian, 1990; Aspiras et al., 2010).

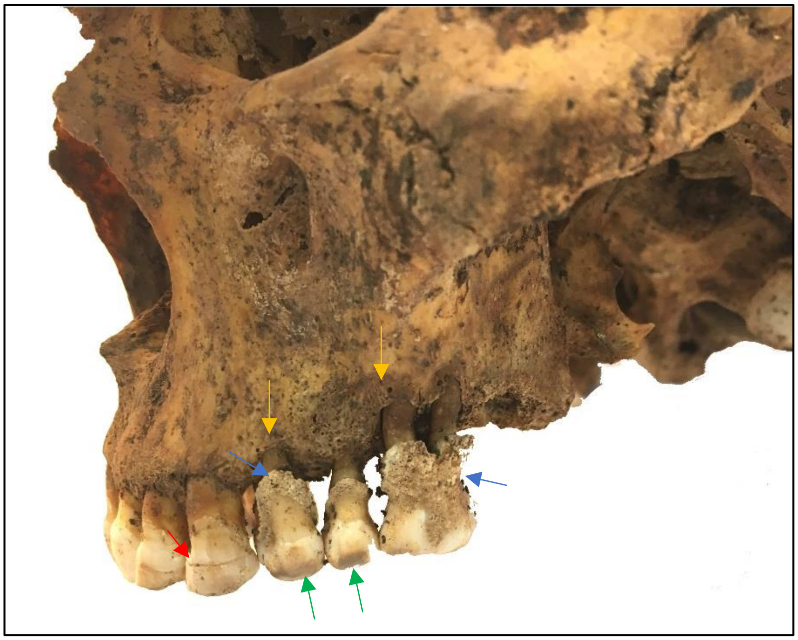

Figure 2.10 Maxillary dentition (BES 19) with calculus (blue), linear enamel hypoplasia (red), alveolar resorption (yellow; periodontal disease) and hypomineralisation (enamel opacities) – lateral view

Historical records and oral history indicate that dental calculus and periodontal disease were common in the past in Iceland and more prevalent than in neighbouring countries (Sigurðardóttir, 2017). Archaeological findings are in line with this, for example, at the early Christian cemetery of Keldudalur in northern Iceland, 100% of the individuals (n=21) presented with calculus (Zoëga and Murphy, 2016). As high protein diets increase oral alkalinity (Moynihan, 2000; Hillson, 2008; Roberts and Manchester, 2010), the high prevalence of calculus deposition is likely correlated with the historical staple diet that was predominately composed of meat, fish, eggs and dairy (Gísladóttir, 1999; Mehler, 2011) and low in grains, flour, and sugar until the 20th century (Jónsson, 1998; Gísladóttir, 1999; Sigurðsson, 2010; Mehler, 2011; Bjarnadóttir, 2016). Diets high in dairy or milk proteins can further contribute to the hardening of dental plaque and raise oral alkalinity thereby increasing calculus deposition; inversely, the calcium, phosphate and casein found in milk and cheese are cariostatic and therefore provide some prevention from caries, particularly in the absence of sugar (Moynihan, 2000; Johansson, 2002). Genetic variation, dental hygiene, local pH, and salivary flow also contribute to calculus formation (Hardy et al., 2009). Although toothpicks were used in the past, toothbrushes were not common until well into the 20th century (Sigurðardóttir, 2017).

2.3 Toxic Elements

In this section, toxic elements are described in terms of their connection with volcanic activity, anthropogenic uses, history, skeletal implications, biological mechanisms, previous archaeological findings, and their connections with Iceland. Exposure to high concentrations of heavy metals released during volcanic eruptions or anthropogenic activities or via prolonged exposure to objects containing them can lead to serious illness or death (González-Reimers et al., 2003; Swift et al., 2015; Rasmussen et al. 2015; Jónsdóttir and Smáradóttir, 2015). Unlike advanced skeletal fluorosis, mercury, lead, cadmium, and arsenic exposure do not appear to cause specific skeletal changes, however, these toxic metals enter soft tissues and are retained in skeletal tissues, causing innumerable pathological conditions while interacting with bone metabolism in various ways (Ericson et al., 1991; Suzuki et al., 2004; Tchounwou et al. 2012; Brito et al., 2014; Wu et al., 2014; Brodziak-Dopierala, 2015). Toxic metal pollution is ca. 100 times higher today than it was in the past (Settle and Patterson, 1980; González-Reimers et al., 2003).

2.3.1 Mercury (Hg)

a. History

Mercury is found in many forms, such as elemental (the purest form), ore (cinnabar), methylated, inorganic and organic mercury (Holmes et al., 2009). Unprocessed cinnabar ore, the natural scarlet colored rock-form of mercury (mercury sulfide) is known to have been used as far back as the Neolithic period (Gajić-Kvaščev et al., 2012; Emslie et al., 2019). For example, the Vinča culture (6th-5th millennium BC) of Serbia used cinnabar to decorate figurines, vessels, and altars (Gajić-Kvaščev et al., 2012). Skeletal remains found in Spain and Italy exhibit cinnabar staining possibly due to use as an embalming preservative or as a ritual decoration upon the remains (Parsons and Percival, 2005; Emslie et al., 2019). Cinnabar was also utilized to produce pigments used in scholarly work (e.g., vermilion) to illuminate manuscripts and in paint used to decorate objects or make rock art (Parsons and Percival, 2005; Mehler, 2015). Today, approximately 20,000 tons of mercury is released into the air per year as a direct result of anthropogenic activities, such as goldmining, causing global pollution to organisms and natural resources especially in cinnabar mining regions (Parsons and Percival, 2005; Zahir et al., 2005; Guzzi and LaPorta, 2008; Hylander and Meili, 2003). However, significant mining of cinnabar in the largest known mine only began in Spain ca. 430 BC and throughout the world thereafter. One of the only exporting mercury mines remaining today, called Khaidarkan in Kyrgystan, has been exploited since at least AD 982, as mentioned in the Persian written work *Hudūd al-ʻālam (“Limits of the Earth”)* (Thomann, 2015).

However, the extraction of mercury from ore may not have begun until around the 3rd century BC when Theophrastus of Eresus, a student of Aristotle, described how rubbing cinnabar with vinegar, inside a brass or copper mortar and pestle, could produce mercury. Vitruvius and Pliny the Elder documented the replacement of this method by distillation in the 1st century BC (Parsons and Percival, 2005). Once mercury is converted into its pure form, it must be contained to prevent its movement or evaporation into the atmosphere, as mercury is the only metal that evaporates and exists in liquid form at room temperature (Boyd et al., 2000). The Romans described the health hazards faced by those who were working in the cinnabar mines: thousands of individuals, primarily slaves and criminals, were noted to have died from the inhalation of mercury vapor. In the 2nd century BC, the Romans also noted that mercury toxicity killed or drove away fish and animals from areas where it was processed (Parsons and Percival, 2005).

The uses and compositions of ancient medicines are primarily known from the written historical records of ancient Greece (e.g., Theophrastus, ca. 371-287 BC and Galen ca. AD 129-200 or 216), Rome (Pliny the Elder, AD 23-79), Egypt (medical papyri, ca. 3000 BC*)*, China (ca. 2500 BC), India (ca. 1200 BC), and Persia (ca. 10th century AD) (Graeme and Pollack, 1998; Hylander and Meili, 2003; Parsons and Percival, 2005; Giachi, 2013). In AD 850, Irish monks prescribed a mixture of mercury and old butter to treat lice. Later, it was suggested in the 11th-century Persian encyclopedia “Canon of Medicine” that mercury was useful for dermatological treatment (Ozuah, 2000). The Sanskrit manuscript, *Rasarnavakalpa* (11th century AD), extensively discusses the medical and alchemical uses of mercury, as well as combinations of mercury, sulphur and other metals and their toxic salts (Craddock, 2009). In the 9th-11th centuries, Islamic physicians used mercury in numerous medicinal treatments (e.g., for nits, lice, and scabies and possibly gonorrhea), although several noted the poisonous effects of mercury on their patients (Parson and Percival 2005; Thomann, 2015). John of St. Amand (14th century AD) and Paracelsus (15th century AD) described the uses and medicinal benefits of mercury while also noting that high doses were poisonous rather than therapeutic (Ozuah, 2000; Hajdu, 2005; Parson and Percival, 2005).

By the 16th century, mercury became widely used in medicine throughout the world, in amalgamation with low-grade silver ores, and for the treatment of anything from cuts to depression. These uses played a significant part in the sharp elevation of mercury concentrations appearing in the environment today (Graeme and Pollack, 1998; Boyd et al., 2000; Parsons and Percivcal, 2005; Fornaciari, 2011; Zuckerman, 2016). Mercuric chloride ($HgCl_2$), also known as calomel and sweet mercury, was used topically in medicinal salves and delivered orally, while a mixture of mercuric chloride, cinnabar ore (HgS) and elemental (liquid, metallic) mercury (Hg) were used during fumigation (O'Shea 1990; Zuckerman 2016). Medieval antisyphilitic treatments were long-term (months to years), were provided via fumigation (mercurial vapour fumigation) and through dermatological preparations, and were generally provided in warm, enclosed spaces. The patients were also supposed to sit near a fire for extended periods of time to facilitate sweating. Pills made of mercury and other natural ingredients were also prescribed (O'Shea, 1990; Beck, 1997; Dobson, 2007; Kępa et al., 2012; Ávila et al., 2014; Thomann, 2015; Zuckerman, 2016). The toxic effects of mercurial treatments were recognized, and some practitioners even insisted on protecting the eyes from contact with mercurial vapors. Other historical accounts describe the side effects and iatrogenic effects resulting from mercurial treatment, including death, "hatters shakes," oral ulcers, kidney failure, tooth loss and psychoses resulting from the treatment (Thomann, 2015; Zuckerman, 2016). Whether or not mercury worked to treat such conditions in the past is difficult to determine, however, mercury has strong biocidal properties. It is thus plausible that mercury successfully treated primary or tertiary stage venereal syphilis (when the bacterial load is low) at least to some degree. Since mercury has been proven to be biocidal to the type of bacteria responsible for syphilis (spirochetes), it is possible that mercurial treatments successfully cured infections in the primary and tertiary stages when the bacterial load is low. The appearance and severity of syphilis can fluctuate or remain in a latent stage which can last for decades, or even spontaneously resolve, likely making resolution appear connected with mercurial treatment. It is also possible that treatments with mercury exacerbated infections by causing an overall degradation in the patient's health and immune response (Ortner, 2003; Zuckerman, 2016).

Until the use of an arsenical called arsphenamine (1909) was developed, mercury remained the standard treatment for venereal syphilis. Then in 1940 the first antibiotic, penicillin, was introduced, thereby revolutionizing medical treatment for bacterial infections, including syphilis (Sartin and Perry, 1995; Ozuah, 2000;). Aside from the toxic properties of mercurial medicine, its inhibition of the immune system to fight off disease may also counteract any potential medicinal benefits. Although other treatments for syphilis were available, including the use of Epsom salt, many physicians considered them ineffective (Zahir, 2005; Hjaltelin, 2013). As previously mentioned, the medical doctor Jón Hjaltalín claimed that mercurial treatments could successfully cure some cases (Hjaltelin, 1868; Hjaltelin, 2013).

Aside from anthropogenic uses, volcanic eruptions are a serious source of mercury emissions and potential exposure (Graeme and Pollack, 1998; Syversen and Kaur, 2012), with peaks in the mercury profile of the Greenlandic ice cores attributed to specific (and non-specific) volcanic events throughout history (Schuster et al., 2002). Nearly 10,000 tons of mercury enters the air per year as it is released in vapor form from the Earth's crust (Zahir et al., 2005). As mercury is released into the air it is also swept into water sources, thereby contaminating bodies of water and rainwater (Guzzi and La Porta, 2008). The consumption of fish and sea mammals is a serious source of mercury (methylmercury) exposure today, but its impact on sea animals in the past remains unknown (Graeme and Pollack, 1998; Syversen and Kaur, 2012). Methylmercury is an organometallic compound that is formed during biogeochemical cycles, likely as an interaction with bacteria, and proceeds to accumulate in fish and sea mammals. As predatory organisms consume other organisms, they also accumulate the mercury within them. Methylmercury raises health concerns today, because of the consumption of fish and sea mammals, and in the recent past several events resulted in significant morbidity to large groups of people. The oceanic mercury hazards observed today are the result of modern environmental activity (e.g., volcanic eruptions) and industry (e.g., burning of fossil fuels and coal) as well as from mining endeavors that have been contaminating the earth, air, and water since at least the Roman period (Holmes et al., 2009). Another toxic methylated form of mercury, called dimethylmercury, caused the death of the prominent Professor of Chemistry, Dr. Karen Wetterhahn, at Dartmouth College when a few drops of the substance fell upon her latex glove during laboratory work. Dimethylmercury is extremely toxic because it easily crosses biological barriers (i.e., blood-brain barrier) and because it is not quickly expelled, it bioaccumulates easily (Graeme and Pollack, 1998; Parsons and Percival, 2005; Bradberry, 2012; Syversen and Kaur, 2012).

Evidence from ice cores and lake sediment cores in the Himalayas and Tibetan Plateau also shows that environmental mercury pollution began to rise substantially with the onset of the Industrial Revolution, followed by a dramatic increase in mercury deposition after World War II (Kang et al., 2016). Today, it has been noted that those working in cinnabar mines, unwittingly contaminate their food and living quarters when they return home from work (Hylander and Meili, 2003; Lombardi et al., 2012; Ávila et al., 2014). Exposure also occurs with old dental amalgam fillings, broken thermometers, and fluorescent lights, for example (Zahir et al., 2005; Syversen and Kaur, 2012). In the latter half of the 20th century, several mercury poisoning epidemics killed thousands of people in Japan and Iraq (Guzzi and La Porta, 2008; Syversen and Kaur, 2012). Recently, low-level exposure has proven to cause neurological and developmental birth defects (e.g., mental retardation, cerebral palsy, dysarthia, autism, deafness, and blindness), particularly in places were large fish and predatory sea mammals are consumed (e.g., Faroe Islands, Greenland, Japan) (Zahir et al., 2005; Guzzi and La Porta 2008; Holmes et al., 2009; Syversen and Kaur, 2012). Various potential sources of mercury exposure were important to consider in this research not only due to the extensive volcanic activity in Iceland, but also the historical traditions of its use as a medicine and in manuscript illumination. Additionally, methylmercury in some Icelandic seafood, including halibut and fermented shark (*hákarl*) test beyond the tolerable weekly intake, indicating that it is a substantial source of mercury exposure today and has possibly been in the past as well (Traustadóttir, 2016).

b. Mechanisms

Clinical research has shown that only negligible amounts of mercury are absorbed dermally or through the gastrointestinal tract. While mercury may not easily absorb dermally, it can quickly vaporize while handling. The inhalation of mercury vapor is extremely destructive as the body can retain at least 74%-80% as it is dispersed through the body, first targeting the brain, kidneys, lungs and gastrointenstinal tract. People exposed to mercurial vapors can develop numerous symptoms (e.g., chills, cough, fever, weakness) within just hours of exposure (Graeme and Pollack, 1998; Boyd et al., 2000; Ozuah, 2000; Guzzi and La Porta, 2008; Bradberry, 2012; Syversen and Kaur, 2012). Elemental mercury causes toxicity following its oxidation to mercuric ions (Hg2+), thereby inhibiting transport functions and membrane, enzymatic, and cellular activity (Ozuah, 2000; Bradberry, 2012). Significant damage to the skin, eyes, lungs, and gingiva can occur due to acute exposure to large doses of inhaled mercury vapors. Low dose chronic exposure to mercury also results in systemic toxicity; it focuses within the nervous system resulting in tremors throughout the body, emotional and psychological conditions, as well as a multitude of conditions including pain, dental infections, sensory impairment, memory loss, delusions, psychosis, and weakness (Boyd et al., 2000; Ozuah, 2000; Risher et al., 2003; Guzzi and La Porta, 2008; Holmes et al., 2009; Bradberry, 2012; Syversen and Kaur, 2012). Low-dose mercury toxicity also affects the renal, reproductive, immune, and cardiovascular systems and may lead to genotoxicity. Additionally, mercury and other heavy metal toxicity have been suggested to be contributing agents in some adult disorders, such as Alzheimer´s, Lupus and Parkinson's disease (Ozuah, 2000; Risher et al., 2003; Parsons and Percival, 2005; Zahir et al., 2005; Holmes et al., 2009).

c. Skeletal implications

Diagenetic processes must be considered when investigating bone mercury concentrations in archaeological individuals. These processes include the absorption of heavy metals in bone and other bone changes occurring in the post-mortem environment, such as collagen destruction, microbial attack, ion, and mineral matrix alteration (Hedges, 2002). A study by Rasmussen et al. (2015) found that soil samples from the graves of skeletal individuals with elevated mercurial bone content had no correlation to eachother, demonstrating that diagenesis was not at play (Rasmussen et al., 2013a; 2015). Research by Yamada et al. (1995), Zuckerman (2017a), Emslie et al. (2019) and López-Costas (2020) also reported no evidence of soil derived diagenesis in bone. Mercury accrues in the bone hydroxyapatite through the replacement of calcium and by forming bonds with carbonates ante-mortem (Lee et al., 2005; Ávila et al., 2014). Thus, elevated mercurial bone concentrations are strong indicators of *in vivo* exposure (Schwarz et al., 2013; Rasmussen et al., 2015). High concentrations of mercury, indicating its use as a medicine, have been found in the remains of members of royalty as well as the more general population from Russia, Denmark, England, Poland, and the Iberian Peninsula (Shorter, 2006; Kępa et al., 2012; Charlier et al., 2014; Rasmussen et al., 2015; Zuckerman, 2016, 2017a; Emslie et al., 2019; López-Costas et al., 2020). In soft tissue, the half-life is just 60 days – eliminated via respiration primarily – but the matrix of bone is a long-term elemental reservoir (>2 years) that retains metals like mercury until resorption or remodeling begin (Miculescu et al., 2011). In non-skeletal tissues mercury has a half-life of about 60 days (Boyd et al., 2000; Ozuah, 2000) and is eliminated primarily through human excreta and secondarily through exhalation, saliva and sweat (Holmes et al., 2009). Mercury exposure does not appear to normally cause diagnostically useful skeletal changes in adults and is believed to remain relatively stable within bone postmortem (Tucker, 2007; Rasmussen et al., 2013; Ávila et al., 2014). However, if mercurial medicine is provided during childhood it may cause enamel defects, which are unlike those associated with congenital syphilis (Ioannou et al., 2016). While dental changes are known to occur within a wide percentage (circa 10%-65%) of individuals affected by congenital syphilis (Ioannou et al., 2016), the percentage of dental changes resulting from mercury exposure in children appears to be entirely unknown. Mercury alters calcium homeostasis, which can result in hypercalcemia, aside from directly altering the function of bone cells (Suzuki et al., 2004). Studies conducted on modern animal bones have shown that aside from decreasing calcemia, mercury poisoning also decreases estrogen receptor expression while increasing metallothionein synthesis. Furthermore, it has notable, negative effects on fetal development, decreasing long bone length and delaying ossification (Rodríguez and Mandalunis, 2018).

In a recent study, López-Costas (2020) found no correlation between collagen preservation and bone mineral constituents (e.g., Sr, P, Ca) and noted that there was an absence of mercury in geological sources around the site the analysed individuals were excavated from. Previous studies of archaeological populations have suggested that mercury concentrations in bone range 0.08 (cortical) to 0.3 ppm (trabecular) (see Rasmussen et al., 2015). However, due to volcanically released mercury (e.g., Mt. Hekla) (see Coderre and Steinthorsson, 1977; Thordarson and Larsen, 2007), Iceland has a higher atmospheric mercury concentration than the areas that these background values were derived from. It is therefore likely that the normal threshold is higher in Iceland, therefore concentrations were not deemed to be elevated unless they exceed 0.3 ppm even though trabecular bone was not used in this study. In ancient and modern dental enamel from human teeth, Rasmussen (1974) found the range of mercury concentrations to be ca. ≤0.001-1.88 ppm.

2.3.2 Fluoride (F)

a. History

Fluorite, or fluorspar, is a mineral form of calcium fluoride (CaF_2). Fluorite crystals were highly prized gemstones in ancient Greece and Rome and the material was used to create costly vessels known as *vasae murrinae*, such as the famous Roman Barber and Crawford cups (AD 50-100) (Tressaud and Vickers, 2007). However, the first known historical descriptions of the use of fluorine compounds do not appear until the 16th century in books written by George Agricola (AD 1494-1555), where he describes its use in smelting to lower the melting point of various ores (Langley and Welch, 1983; Wisniak, 2002). Although the uses of various compounds of fluorine were well known, it was not isolated as an element until AD 1886 (Langley and Welch, 1983). In the 18th and 19th centuries, experiments with fluorine chemistry resulted in the production of hydrofluoric acid, which became an important scientific and chemical commodity (Wisniak, 2002). Due to the extreme dangers of working with fluorine, large-scale production was not performed until around the time of World War II when it became an essential component for the development of nuclear devices (Wisniak, 2002). In the past, the most common exposure to fluoride, the ionic form of fluorite, likely occurred due to volcanic emissions from eruptions or passive degassing. In one study of skeletal remains from Herculaneum (decimated during the AD 79 Vesuvius eruption), the researchers identified numerous cases of osteoscleroses, entheseopathies, and exceedingly high fluorine concentrations. These skeletal changes were indicative of fluorosis and suggested that it was endemic in the population (Petrone et al., 2011, 2013).

Fluoride's use as an anticaries supplement in water resulted from observations of populations with low caries rates in areas of high concentrations of fluoride in drinking water (Marquis et al., 2003; Browne et al., 2005). The first experimental use of fluoride in water for caries control began in 1945 in the US and 1946 in Canada in four trial cities. After the beneficial effects were recognized in North America, experiments with water fluoridation were initiated in the United Kingdom and then in Ireland (Browne et al., 2005). Fluoridated water thereby became one of the most important methods for reducing the prevalence of carious lesions (Ishiguro et al., 1993; Browne et al., 2005). The World Health Organisation has set the maximum fluoride level in drinking water at 1.5 ppm (Yoshimura et al., 2006). While some countries have begun to limit this to lower than 1 ppm, numerous government agencies still maintain a maximum fluoride allowance of 4 mg/L (4 ppm), despite the adverse health effects often described even with concentrations lower than 2 mg/L (2 ppm) (Littleton, 1999; Ayoob and Gupta, 2006; Yoshimura et al., 2006). Fluorosis is a major public health problem in some regions of the world that can result in physical impairment and disability in individuals suffering from it (Seeley, 2001a, 2001b). Around 317 million people throughout the world are currently ingesting fluoride from naturally and artificially fluoridated water (Browne et al., 2005). Tens of millions of people suffer from endemic fluorosis in over 25 countries, many of which display substantial volcanic activity (D'Alessandro, 2006). Today, Icelandic dentists occasionally discover fluorosed dental enamel in the modern population (Árnadóttir et al., 2005), but archaeological cases of human osteofluorosis have not yet been found Iceland.

b. Mechanisms

A variety of forms, doses, and lengths of exposure to fluoride can result in chemical and biological changes to cells and tissues. There is currently no way of determining the exact cutoff point between safe and dangerous levels of fluoride because so many biological, environmental, dietary, and cultural factors are at play (Arnala et al., 1985; Littleton, 1999; Everett, 2011; Petrone et al., 2013; Nelson et al., 2016). Low fluoride supplementation in drinking water should not cause fluorosis, but some studies have shown that under certain circumstances skeletal and dental changes associated with fluorosis can occur even with very low concentrations (Browne et al. 2005). People conducting physical labor, particularly in the heat, tend to be more susceptible as they require more water to rehydrate thereby also ingesting higher concentrations of fluoride. Although fluorosis is more commonly diagnosed in men, it tends to occur more rapidly in women (Alhava et al., 1980; Ebel et al., 1992; EFSA, 2010). In humans, skeletal fluorosis normally occurs with water fluoride concentrations over 4 ppm, but severe cases of fluorosis have been identified with concentrations as low as 1.35 ppm due to biocultural and environmental predisposing factors (e.g., malnutrition, water storage methods, climate) (Littleton, 1999; Savas et al., 2001). For example, endemic fluorosis has been seen in regions with water fluoride concentrations of 2-3 ppm. Dental fluorosis has been observed at water concentrations of 0.5 ppm, skeletal fluorosis at 0.7 ppm and crippling skeletal fluorosis concentrations as low as 2.8 ppm (Ayoob and Gupta, 2006; Petrone et al., 2013).

Fluoride exposure normally occurs through diet and the consumption of water and although it does not absorb well through skin, it can cause severe skin burns. Neither does it systemically absorb particularly well via inhalation, but exposure to it increases the risk of developing chronic lung conditions, such as asthma (D'Alessandro 2006). Fluoride toxicity tends to be cumulative and gradual (Den Besten, 1999a, 1999b; Nelson et al., 2016) although acute toxicity can occur in extreme cases (Littleton, 1999; Everett, 2011). The circulatory system distributes it, and it can easily accrue in bone, especially during development or growth. As a result, its concentration increases with age as it is permanently retained in bones and teeth (Arnala et al., 1985). If an individual is suffering from calcium deficiency, up to 80% of ingested fluoride can be absorbed through the gastrointestinal tract and retained in the bone, compared to around 50% under normal circumstances. The remainder is excreted primarily through urine (Whyte et al. 2008). The skeletal system contains approximately 99% of one's total fluoride burden – fluoride (F-) substitutes for hydroxide (OH-) within the bone hydroxyapatite. This substitution forms hydroxyfluoroapatite (partial substitution) as well as fluoroapatite (full substitution), which are more compact and less soluble than unaltered hydroxyapatite (Whyte et al. 2008).

Excess fluoride ingestion occurring during the development of the teeth can cause porotic, hypomineralised enamel (i.e., dental fluorosis) (Browne et al., 2005) as it disrupts crystal development, thus permanently altering the mineralizing matrix through (Den Besten, 1999a). Hydroxyfluoroapatite and fluoroapatite can reduce caries risk, especially combined with the antimicrobial properties of fluoride, but it can also cause changes in the stability and appearance of the teeth. Many researchers reduce these changes to merely aesthetic or cosmetic defects, while others consider the important biological and psychological impacts that poorly mineralized, discolored, cracked, and pitted teeth can cause (D'Alessandro, 2006; Susheela, 2007; Santa-Rosa et al., 2014; Rajeswari et al., 2016). For example, a study of red deer dentition showed that fluorotic teeth had reduced enamel hardness, which resulted in increased periapical lesions, periodontal disease and dental attrition and made the teeth fracture-prone (Schultz et al., 1998).

Skeletal fluorosis causes the densification and thickening of bone (with increased fracture risk), anemia, ossification of soft tissues (decreases mobility), chronic pain, hormone imbalances and numerous other signs and symptoms (Baxter, 2009). In the early phases, the effects of fluorosis poisoning usually include pain, stiffness, nausea, constipation, and discomfort (ca. two weeks of chronic exposure) (Jolly et al., 1969; Littleton, 1999; EFSA, 2010; Petrone et al., 2013; Nelson et al., 2016;). Lower back pain may be a consequence of the early stages of fluorosis even when other symptoms are not present (Namkaew and Wiwatanadate, 2012). As the condition progresses, neuropathic, reproductive, and vascular complications arise, in addition to the development of thyroid dysfunction, early sexual maturation, diabetes and the restriction or stiffness of the muscles (Nelson et al., 2016). Soft tissues undergo calcification, resulting in limited joint mobility (such as inability to make a fist). The skeletal system begins to exhibit increased bone density and production and the teeth exhibit enamel staining, mottling and other enamel defects. During the later stages of the condition, difficulty in walking and other crippling effects may ensue as osteosclerosis and ankylosis occur throughout the skeleton, especially in the spine and chest (Littleton, 1999; Savas et al., 2001; Petrone et al., 2013). Fluoride may also play a role in the promotion of cancer, particularly bone cancers such as osteosarcoma. For example, one study identified a seven-fold increase in bone cancer risk in children exposed to fluorosilicates used in water treatments (Bassin et al., 2006).

Genes play a role in fluoride absorption, particularly regarding amelogenesis and bone homeostasis. Fluoride increases osteoblast proliferation at low concentrations, causes cellular death at high concentrations, alters regulating genes, and disrupts the formation and modeling of the intracellular matrix of skeletal and dental tissues (Mousny et al., 2006, 2008; Barbier et al., 2010; Everett, 2011; Kobayashi et al., 2014; Nelson et al., 2016). One study showed that several proteins are targeted for varying fluoride responses in skeletal tissues (Kobayashi et al. 2014). In genetic research on murinae and nematoda it was discovered that some fluoride resistance genes decrease biological responses to toxic levels of fluoride (see Mousny et al., 2006; Mousny et al., 2008), indicating that genetics involvement in fluorosis susceptibility (Everett, 2011; Kobayashi et al., 2014; Nelson et al. 2016).

c. Skeletal implications

Essentially, fluoride alters bone mineral metabolism (i.e., bone turnover) through changes to the accretion and resorption rates of skeletal tissues (Savas et al., 2001; Mousny et al., 2006). Following bone proliferation, soft tissue ossification, joint ankyloses, and fusion occur, severely limiting mobility (Arnala et al., 1985; Littleton, 1999; Browne et al., 2005; D'Alessandro, 2006; Mousny et al., 2008; Baxter, 2009;

EFSA, 2010; Petrone et al., 2013; Kobayashi et al., 2014; Nelson et al., 2016). These biochemical processes occur most rapidly in trabecular bone (Alhava et al., 1980; Ebel et al., 1992; Littleton, 1999; EFSA, 2010). Fluoride causes irregular bone proliferaration, increasing bone mass (increased osteoid volume), density and ossification at soft tissue attachment sites (Whyte et al., 2008). It has actually been used medicinally in low concentrations to increase bone density in the lumbar spine of osteoporotic patients (Arnala et al., 1985; Ishiguro et al., 1993; Mousny et al., 2006), although some studies show that both high and low concentrations increase fracture rates (EFSA, 2010; Namkaew and Wiwatanadate, 2012).

Although fluoride can enter plants through the roots or via foliar absorption, research has indicated that fruits and vegetables are generally an unlikely source of fluoride poisoning (EFSA, 2010) possibly because of the cleaning, processing, and controlled sourcing of food (D'Alessandro, 2006). However, certain plants (e.g., tea) that are prone to fluoride accumulation can cause toxicity with habitual consumption (Whyte et al., 2008). Even if fluoride is unlikely to accumulate substantially within the tissues of most foliage, ash and other volcanic particulate matter and emissions are highly likely to deposit upon grazing fields. Thus, grazing terrestrial mammals are generally first and most severely affected (D'Alessandro, 2006; EFSA, 2010). In numerous historic and modern-day instances, fluoride poisoning has caused death to livestock in Iceland and New Zealand (Grattan, 2006). For example, in the 1970 eruption of Hekla, sheep died of acute fluorosis within a week of bone concentrations increasing to 30-60 ppm. Fluoride poisoning in animals generally depends upon the duration of ingestion, diet, nutrition, individual biological response, body size, overall health and the type, solubility and amount of fluoride ingested (Shupe et al., 1963). Essentially, these principles apply to humans as well. Chronic fluorosis has been diagnosed in lambs at ca. 698 ppm (normally 116), one-year old sheep at ca. 1683 ppm (normally 560) and adult sheep at ca. 1329 ppm (normal 830), showing that younger and smaller animals may accumulate fluoride more rapidly or in larger concentrations. Some animals tolerate higher concentrations better than others regardless of size or age, such as whales, which have shown concentrations between 8605-12700 ppm (Stefánsdóttir, 2016). As fluoride undergoes mobility in bone, not all individual's skeletal elements will contain the same fluoride levels and there can also be disparity between bones and bone tissues (cortical or trabecular) (Littleton, 1999; Hedges, 2002). Recent research found that fluoride tends to be more elevated in trabecular bone than cortical bone (Lanocha-Arendarczyk et al., 2015b). Arnala et al. (1985) analysed trabecular bone samples from the cadavers of individuals that resided in two regions (high-fluoridated and low-fluoride) in Finland, demonstrating that concentrations above ca. 1500 ppm may be considered elevated. Additionally, the individuals with impaired renal functions generally had higher fluorine concentrations (2090 ± 1010 ppm) (Arnala et al., 1985). People that are exposed to just 1 ppm of fluoride daily generally exhibit normal bone fluoride concentrations (<500-1500 ppm).

Since fluoride accrues with increasing age, fluorosis especially affects older individuals (Arnala et al., 1985; Barbier et al., 2010; Petrone et al., 2011). As a result, elderly people often have elevated fluoride bone concentrations in even when they have no signs/ symptoms of osteofluorosis (Richards et al., 1994). In both palaeopathological (e.g., Jolly et al., 1969; Frohlich et al., 1989; Grimaldo et al., 1995; Littleton, 1999; Yoshimura et al., 2006; Petrone et al., 2011, 2013) and modern research (e.g., Ayoob and Gupta, 2006), categorize fluoride concentrations in bone into poorly defined ranges related to subjectively progressive changes. Depending upon geographic provenance and overall age, normal baselines for fluoride in unaffected human skeletal tissues ranges from <500 to 3000 ppm (Ebel et al., 1992; Sastri et al., 2001; Petrone et al., 2013). Advanced bone chages often occur with concentrations between 6000 and 9000 ppm (clinical phase 1-2), even though osteosclerosis and impairment of the renal system may begin with concentrations just exceeding the normal threshold. Bone concentrations exceeding 9000 ppm are associated with severe, crippling skeletal fluorosis (clinical phase 3) (USDHHS, 1991; Ayoob and Gupta, 2006).

2.3.3 Lead (Pb)

a. History

The mining and use of lead in human history started at least 6000-7000 years ago, but the history of lead poisoning can only be traced to at least 2500 year ago (Lessler, 1988; Hernberg, 2000). Lead is a highly toxic heavy metal that easily bioaccumulates through inhalation, dermal contact, or ingestion due to natural or anthropogenic exposure (Bower et al., 2005; Rasmussen et al. 2014; Evans et al., 2018). Natural sources of lead exposure include geology (i.e., rock, volcanic particulate matter and associated plants and groundwater) (Åberg et al., 1998), while anthropogenic sources include its use as a material for buildings, pipes, gutters, roofs, windows, statues, pigments/colorants, kitchenware, pottery, ceramics and coins and through activities, such as mining, ship building, plumbing, food preparation, wine making, bronze or brass production, glass making, smelting and deliberate use in food (Nriagu, 1983; Hernberg, 2000; Rasmussen et al., 2015; Evans et al., 2018).

Lead poisoning, in fact, is one of the earliest examples of disease caused both by environmental sources and occupational activities, with cases described since antiquity (Nriagu, 1983; Riva et al., 2012). For example, in the 2nd century BC Nicander of Colophon diagnosed lead poisoning based upon its acute presentation of saturnine colic and paralysis (Riva et al., 2012). Hippocrates (460-377 BC) noted lead poisoning symptoms, including colic, appetite and weight loss, irritability, nerve spasms and pallor, which are many of the same symptoms observed today (Lessler, 1988). Typically, the people normally exposed to lead prior to the Roman Empire were artisans or individuals from the lower ranks of society (Riva et al., 2012). Several populations, including the Egyptians, Greeks and Romans documented the effects of lead poisoning from mining activities, generally amongs slaves or lower-class workers (Lessler, 1988). The Roman Empire was responsible for increasing the availability of lead by locating lead-rich ores for various uses, such as the construction of water and sewage systems. It was believed that lead salts released into drinking water flowing through these lead pipes may have been a primary cause in the downfall of the Roman Empire as the ruling class began to struggle with health problems and infertility due to lead poisoning (Riva et al., 2012). However, more recent research suggests that the raw mountain water within these pipes was rich in calcium carbonate, thereby coating them with a protective layer that would have prevented the release of lead salts into them. Thus, lead poisoning amongst upper-class Roman society was likely more associated with the use of lead acetate ("lead sugar"), produced from slowly cooking "sapa" (i.e., a preparation of must: freshly crushed fruit juice used for winemaking), to preserve and sweeten wine (Riva et al., 2012). Cerussa (white lead or 'lead sugar') was also used in cosmetic preparations, especially by women, to whiten the face, and other poisonous substances including cinnabar (mercury sulphide) and minium (red lead) were components of other cosmetics (Olson, 2009).

In the medieval period, the use of lead widely increased, and lead poisoning became associated particularly with artists using lead-based paints and metallurgists, the latter being instructed to cover their mouths with rags and keep their windows open during metal work (Lessler, 1988; Riva et al., 2012). Alchemists and physicians quickly became aware of the extent of the toxic attributes of lead, identifying various occupations that were at high risk such as painters, potters, tinsmiths, and miners (Riva et al., 2012). However, the use of "sapa" prepared or stored in lead containers also persisted into the 17th century, resulting in outbreaks of lead poisoning characterized by saturnine colic (Riva et al., 2012). In Iceland, lead-glazed kitchenware increased in availability around the 17th century, although some examples have been found at medieval sites (Þorgeirsdóttir, 2010). The use of lead exponentially increased during the Industrial Revolution as it became a necessity for metal products and other fabrications (Lessler, 1988). It was not until the 18th-19th centuries that scientists began to clearly link outbreaks of lead poisoning to water and dietary intake (Lessler, 1988; Riva et al., 2012). The analysis of the Greenland ice cores corroborates historical evidence that the mining and exploitation of lead caused worldwide pollution. Although lead pollution was pronounced in the Medieval and Rennaisance periods and during the Industrial Revolution, it was remarkably high during the times of the ancient Greek and Roman empires (Hong et al., 1994).

b. Mechanisms

Upon exposure, lead is distributed throughout the soft tissues, especially to the liver and kidneys. Most (80-90%) of the absorbed lead is excreted, while the rest is retained in bone (Lessler, 1988). Lead exposure can happen during skin contact with lead objects/structures, lead dust or paint inhalation, or the consumption of food, soil, or other materials (Hernberg, 2000; Vahter, 2007; Riva et al., 2012). Additionally, the accumulation/retention and susceptibility to the health effects of toxic metal exposure differ between males and females (Vahter et al., 2007). Females generally accumulate lead in the skeletal system more rapidly than males, mostly due to biological changes that alter the production of estrogen (e.g., menopause, pregnancy) (Vahter et al., 2007). Due to outdoor play and hand-to-mouth behavior, children are often environmentally exposed to it and absorb it easily (see Wittmers et al., 2002; Jacobs and Nevin, 2006).

c. Skeletal Implications

Lead exposure may result in notable skeletal pathologies including osteopenia, osteoporosis and increased fracture rates as its accumulation causes bone turnover malfunctions and reduction of bone density (Brito et al., 2014). Furthermore, vitamin D alters and stimulates the absorption of essential minerals, toxic metals and radioactive isotopes and may contribute to the severity of pathological conditions caused by exposure to toxic metals by increasing the rate of absorption and retention. Thus, lead exposure interferes with the normal vitamin D metabolism (Moon, 2013). Over 90% of the body's lead burden is contained in bone and it becomes mobilized during periods of increased rates of bone turnover, such as during menopause, pregnancy, lactation, or pathological conditions altering bone metabolism (Vahter et al., 2007). Some studies have shown that bone remodelling occurs predominately (55%-80%) in the trabecular/cancellous bone as it is more metabolically active (Brito et al., 2014). Lead also acts to alter the balance of bone turnover, possibly by disrupting bone architecture and resorption or reduction of bone mass, thereby leading to conditions such as osteopenia and osteoporosis (Brito et al., 2014). Concentrations up to 5 ppm are normal in modern cortical bone and 7 ppm in normal modern trabecular bone (Rasmussen et al., 2015). Diagenesis, however, is a major complication in bone lead studies. Long bones have been shown to contain as much as double the lead concentration of flat bones (e.g., rib) (Barry and Mossman, 1970). It is well established that lead concentrations increase in bone according to age, particularly if an individual is periodically exposed to lead throughout life (Barry and Mossman, 1970; Hisanaga et al., 1989; Rasmussen et al., 2015). Dental enamel samples of prehistoric individuals buried in England show concentrations of 0.04 to ca. 0.4 ppm, while individuals from the Romano-British and Medieval period had concentrations of up to 40 ppm. This marked increase is associated with anthropogenic exposure to lead during life from industrial pollution. Lead concentrations in dental enamel that are associated with geological sources through the diet, rather than from technological exposure, generally range from ca. 0.5-1.0 ppm regardless of time period (including modern individuals) (Budd et al., 2004).

2.3.4 Cadmium (Cd)

a. History

As a distinct metal, cadmium was only discovered in AD 1817, despite its ubiquitous presence as an impurity of various metal ores that were extensively exploited in the ancient past, such as lead, zinc and copper (Hong et al., 1997). Aside from steel making and the incineration of wastes, volcanic eruptions and zinc production are the most significant sources of cadmium emissions in the environment (Hutton, 1983). During periods of major volcanic events, cadmium emissions were likely notably elevated, but according to Greenland ice core studies, cadmium concentrations did not increase beyond current natural background levels during the Greek, Roman or Medieval periods (Hong et al., 1997).

Cadmium toxicity appears to primarily affect contemporary populations and those living in the recent past because intentional production has only been extensively performed in modernity for use in batteries, paint pigments, electroplating, polyvinyl chloride plastic and other industry (Rahimzadeh et al., 2017). Significant increases in cadmium concentrations in human liver and kidney samples from 1980 were observed when compared with samples from 1897-1937, further indicating that increased cadmium exposure relates to modern industry (Ericson et al., 1991). Historical exposure could have occurred through the consumption of contaminated fish or shellfish, use in pigments, cooking with earthenware (Hisanaga et al., 1989) or perhaps through mining, smelting and amalgamation with other metals, such as lead. An association between osteomalacia and serious cadmium exposure was first demonstrated in a Japanese population residing in the Jinzu river basin region during the 1950s (Johri et al., 2010). The people residing there developed Itai-itai disease, characterized by severe skeletal pain, long bone malformation, increased fracture rate and other typical signs and symptoms associated with rickets or osteomalacia (Johri et al., 2010). It was later determined that cadmium pollution entered the river because of industrial waste discharge from an upstream zinc mine (Vahter et al., 2007; Johri et al., 2010).

Cadmium is low in Icelandic seafood today (Traustadóttir, 2016). Thus, it was unlikely to be a notable source of exposure in the past. While it is also generally very low in the environment in Iceland, it can increase in the atmosphere and environment following eruptions. For example, researchers found that over 25 tons of poisonous cadmium were degassed during the 2014 eruption of Holuhraun (Bárðarbunga-Veiðivötn volcanic system) in southern Iceland, making it a serious atmospheric and environmental pollutant (Jónsdóttir and Smáradóttir, 2015; Gauthier et al., 2016). As previously mentioned, an AD 1477 eruption occurring in the same system causing catastrophic damage in eastern Iceland, not far from the monastery Skriðuklaustur (Larsen, 1988; Rafnsson, 1990; Thordarson and Larsen, 2007, Global Volcanism Program, 2013).

b. Mechanisms

Cadmium exposure occurs easily through the inhalation of tobacco smoke or industrially polluted air and through ingestion of contaminated food, water, or soil (Loganathan et al., 2003; Vahter et al., 2007; Rahimzadeh et al., 2017). Exposure may also occur from volcanic activity, mining for silver and lead and from objects, such as pottery, made with these materials (González-Reimers et al., 2003). Cadmium causes toxicity by altering the proliferation, differentiation, and apoptosis of cells, binding with mitochondria and by interfering with DNA repair mechanisms, which can result in the breakage of DNA strands, chromosomal deletions, and other mutations (Rahimzadeh et al., 2017). Low-level cadmium exposure has been shown to result in increased risk for the development of osteoporosis and kidney damage (Kazantzis, 2004). Kazantzis (2004) describes studies that found that older inviduals (>60 years old) who were exposed to cadmium had a much higher bone fracture risk associated with decreased bone mineral density. Cadmium exposure promotes skeletal demineralization as it can directly interact with bone cells and inhibit collagen production (Rahimzadeh et al., 2017). Elevated cadmium concentrations in bodily tissues are a contributing factor to various conditions, including osteoporosis, increased fracture risk, osteomalacia, osteopenia, cancer (e.g., prostate, bladder, liver, stomach) and renal diseases (Black, 1988; Johri et al., 2010; Brodziak-Dopierala et al., 2015; Nordberg et al., 2015; Chen et al., 2019). Cadmium accumulates predominately in the liver and kidneys, although it is also retained by the placenta, bone, and other bodily tissues, and most frequently causes renal damage, calcium loss, proteinuria, and nephrotoxicity (Rahimzadeh et al., 2017). The signs and symptoms of acute cadmium toxicity via ingestion include increased salivation, abdominal pain, choking, vomiting, vertigo and loss of consciousness while toxicity via inhalation includes a dry throat and cough, choking, vomiting, chest pains, pulmonary edema or fibrosis, bronchospasms, leg pains, muscle weakness, pneumonitis, bronchitis, emphysema, muscular weakness, headache, and flu-like symptoms (Johri et al., 2010; Chen et al., 2019). It has also been associated with reproductive system damage, heart disorders and neurotoxicity (Suzuki et al., 2004; Vahter et al., 2007; Rahimzadeh et al., 2017)

Cadmium toxicity in skeletal tissues (Itai-itai disease) is characterized by an imbalance in bone resorption and formation, critical mechanisms important for maintaining biomechanical properties of bone (Johri et al., 2010; Chen et al., 2019). Its skeletal effects include painful osteoporosis, increased fracture and pseudofracture rate, severe skeletal decalcification, femoral and lower back pain, osteomalacia and physical impairment resulting in decreased quality of life (Rahimzadeh et al., 2017). The effects of cadmium on bone include loss of trabecular bone, decreased number of osteocytes, inhibited osteoblastic activity, increased osteoclast formation and cortical bone thinning (Johri et al., 2010; Rahimzadeh et al., 2017). Cadmium exposure disrupts the production of vitamin D within the kidneys thereby decreasing intestinal calcium absorption, but it also has direct effects on bone (Vahter et al., 2007; Johri et al., 2010; Chen et al., 2019). The removal of cadmium accumulation from bodily tissues occurs very slowly with a biological half-life of 10-30 years (Longanathan et al., 2003; Rahimzadeh et al., 2017), thus low quantity long-term exposure to cadmium leads to toxic concentrations (Black, 1988; Loganathan et al., 2003), altering bone metabolism (e.g., increased bone resorption, hypercalcemia) and integrity of bone structure and impeding cellular activity, which reduces the density of bone matrix and other mineral elements (Suzuki et al., 2004; Vahter et al., 2007; Brodziak-Dopierala et al., 2015). Sex and age are significantly associated with the extent of bone damage resulting from cadmium exposure, which most often occurs in elderly women (Vahter et al., 2007; Chen et al., 2019). Additionally, women tend to have higher concentrations of cadmium in the body due to increased absorption through the gastrointestinal system (Brodziak-Dopierala et al., 2015). Diet and nutrition also seem to play a major role in an individual's susceptibility to cadmium toxicity (Chen et al., 2019). For example, a diet high in zinc, magnesium, iron, copper, fibre and calcium can reverse cadmium-induced toxicity and alter the gastrointestinal absorption of cadmium. However, a diet high in rice could increase cadmium exposure as rice tends to easily absorb cadmium in polluted areas (Vahter et al., 2007; Brodziak-Dopierala et al., 2015; Rahimzadeh et al., 2017; Chen et al., 2019). Zinc appears to be particularly antagonistic to the effects of cadmium on bone tissue (Brodziak-Dopierala et al., 2015).

c. Skeletal Implications

Compartable to mercury toxicity, the non-specific nature of cadmium's effect on bone makes it implausible to use osteological methods alone for the diagnosis of cadmium toxicity. However, Itai-itai disease is characterized by osteoporosis, osteomalacia, kidney damage and multiple, spontenous skeletal fractures (Vahter et al., 2007). In an archaeological context, González-Reimers et al. (2003, 2005) analysed prehistoric individuals from the Canary Islands, but the results did not indicate cadmium toxicity. The results rather demonstrated a notable increase in bone cadmium content in modern populations in comparison to the prehistoric skeletal material (González-Reimers et al., 2003, 2005). Another study conducted on prehistoric Native American individuals, demonstrated a cadmium concentration range of 0.008 to 0.36 ppm (Ericson et al., 1991). Research by Martinez-García et al. (2005) found that cadmium concentrations were typically higher in ribs than in other skeletal tissues. Amongst all Medieval samples (8-13th centuries) the cadmium concentration was 0.01 to 19.0 ppm and the mean value was 1.5 ppm (Martinez-García et al., 2005). Brodziak-Dopierala et al. (2015) analysed cadmium concentrations from modern hip joint samples, resulting in a mean of 0.52 ppm in cortical bone and noted a correlation (Mann-Whitney U test, $p<0.025$) between elevated cadmium concentrations, increased degenerative changes (average 0.64 ppm) and fracture (average 0.85 ppm) rates. González-Reimers et al. (2003) demonstrated that modern individuals' bone cadmium averages 0.517 ± 0.352 ppm and lead averages 30.53 ± 14.62 ppm, while the mean concentrations in ancient individuals from Gran Canaria (dated 1405 ± 60 to 1213 ± 60 bp) was 0.085 ± 0.129 ppm in Cd and 4.06 ± 4.63 ppm in Pb. One study observed cadmium concentrations in children's teeth in the range of 0.007-0.610 ppm (Bayo et al., 2001).

2.3.5 Arsenic (As)

a. History

Arsenic, known as the *King of Poisons* or the *Poison of Kings*, has been used therapeutically, for the treatment of skin lesions (e.g., ulcers, abscesses) and other ailments, as far back as 2000 BC (Jolliffe, 1993; Hughes et al., 2011). While most forms are highly unlikely to be essential nutrients, some studies have suggested that trace amounts of inorganic arsenic might be (Hughes et al., 2011). Arsenicals typically used in traditional medicine throughout history include orpiment, realgar, arsenolite or arsenic trioxide (Liu et al., 2008). Produced from the smelting of copper, arsenic trioxide is the form that has been used since at least 2000 BC as both a poison and medicine (Jolliffe, 1993). Early physicians, such as Hippocrates, Aristotle, Paracelsus, Galen and Pliny the Elder all described its extensive medical uses (Jolliffe, 1993; Hughes et al., 2011). Hippocrates used orpiment and realgar to treat necrotic or dead tissues, while Paracelsus frequently used elemental arsenic (Jolliffe, 1993). Realgar, an arsenic sulphide (AsS) mineral also known as ruby of arsenic or ruby sulphur, has likewise been used in traditional Indian and Chinese medicine since at least 200 BC, as an elixir for treating disease or to induce perpetual life (Liu et al., 2008), and even as a poison (Bolt, 2012). The poisonous effects of arsenic and other heavy metals have been noted since ancient times (Liu et al., 2008; Hughes et al., 2011).

In the Medieval period, arsenic was commonly used medicinally, but also as a means of committing homicide and suicide. For example, the Medici and Borgia families used arsenic trioxide to assassinate their adversaries (Hughes et al., 2011). Arsenic trioxide ("white arsenic") was a popular choice for poison because it was inexpensive, odorless, inconspicuous in appearance, widely available and tasteless unless burned (Jolliffe, 1993; Hughes et al., 2011). Arsenic preparations became common treatments for skin conditions (e.g., eczema, psoriasis, skin cancer), sexually transmitted diseases (e.g., syphilis and chlamydia) and other diseases, such as chorea, malaria, breast cancer and asthma (Hughes et al., 2011). They were also used to prevent diseases believed to be caused by miasma and treat lesions associated with plague (Tosetti, 2014; Legan, 2015; Hughes et al., 2011). By the 18th century, arsenic trioxide was espoused as the cure for remittent fevers, periodic headaches, and agues (Jolliffe, 1993). In medicinal preparations, arsenic was often used alongside other ingredients, such as sulphur, mercury, antimony, and numerous herbs (Jolliffe, 1993; Tosetti, 2014; Legan, 2015). At least 60 different types of arsenic preparations were used throughout history before they were replaced with less toxic and more effective medicines, yet hundreds of traditional Chinese medications containing arsenicals remain in use today (Liu et al., 2008).

Arsenic is a component trace element of basaltic rocks in Iceland, and it can become concentrated in hydrothermal fluids (ca. 0.15 ppm As) following water-rock reactions occurring in geothermal or volcanic systems. During these interactions, it can contaminate surface waters and it becomes a health concern if elevated in drinking water above 0.01 ppm (Olsen et al., 2010). Relevant to this study is the possibility that in the past, drinking water sources merging with water from geothermally or volcanically active areas could have been toxically contaminated. However, it would have been unlikely that people would collect water for consumption directly from geothermal sources, which may be foul tasting and smell strongly of sulphur.

b. Mechanisms

Arsenic exposure can occur naturally through soil, food, water and air or anthropogenically as a result of metallurgy or the use of arsenic containing chemical compounds in folk medicine and as preservatives, pesticides, fungicides or for industrial waste disposal (Hughes et al., 2011; Swift et al., 2015). Arsenic accumulation and incorporation occurs in the body especially through the consumption of contaminated drinking water and via inhalation (Kabata-Pendias and Mukherjee, 2007). In the past, arsenic exposure in Iceland may have occurred through locally produced food and drinking water, particularly following volcanic eruptions. While some volcano types (e.g., arc, hotspot) contain greater quantities of particular metals, Icelandic volcanic emissions have been shown to contain notable amounts of toxic heavy metals relevant to this study, such as arsenic, cadmium and lead (Edmonds et al., 2018). Additionally, geothermal waters, which are ubiquitous in Iceland, are commonly high in arsenic (Weaver et al., 2019). The prolonged consumption of arsenic concentrations above 0.8 mg/L in drinking water can result in arsenic toxicity (Aras and Ataman, 2006). Seafood (e.g., fish, marine mammals, molluscs, bivalves) and seaweed, both of which were frequently consumed in historical Iceland (Walser III et al., 2020a), contain far higher arsenic concentrations than terrestrial foods and may have been significant sources of arsenic exposure (Aras and Ataman, 2006; Swift et al., 2015). Furthermore, cooking causes the solubilisation of arsenic, increasing its concentration in food according to factors such as temperature, cooking time, volume of water and level of contamination in water (Swift et al., 2015). Other exposures include tobacco smoking, drinking water pollution, close proximity to industrial emissions (e.g., metals, coal), gardening or agriculture, its use as a pigment and metallurgic practices (e.g., copper ore, metal, slag and smelting furnaces) (Brodziak-Dopierala et al., 2011; Hughes et al., 2011; Bolt, 2012; Swift et al., 2015). Arsenic often coexists with sulphur in the natural environment (Fisher et al., 2008) and it interacts with sulphur to form stable As-S complexes (Hughes et al., 2011). Sulphur is found in the Icelandic environment, which made it viable as an important trade commodity in the past, and has been recovered from Medieval archaeological sites, such as Skriðuklaustur (Kristjánsdóttir, 2012; Mehler, 2015). Thus, exposure to arsenic may have occurred amongst individuals responsible for metallurgic productions or sulphur processing and those administering or receiving arsenical and sulphur-based medicinal treatments.

Exposure to arsenic is a serious health risk, both acutely (immediate toxicity) and chronically (e.g. as a highly potent carcinogen (Vahter et al., 2007; Swift et al., 2015). It also causes genotoxicity and is capable of interfering with fetal development transplacentally (Hughes et al., 2011; Swift et al., 2015). Arsenic exposure easily occurs through drinking water and certain foods, particularly rice and vegetables (Vahter et al., 2007). Co-exposure of arsenic with other toxic elements or compounds, such as fluoride, is a major concern in countries with groundwater or river pollution and where coal is combusted indoors (Bolt, 2012). The combined action of co-exposure to arsenic and fluoride, as well as arsenic and barium, have been demonstrated to result in cell death (Bolt, 2012). However, the correlation between strontium and arsenic in bone has been shown to be antagonistic (Brodziak-Dopierala et al., 2011). Symptoms of acute arsenic toxicity include liver, myocardial, vascular toxicity, kidney and gastrointestinal disease, and in extreme or high-dose cases, brain stem failure (Liu et al., 2008; Swift et al., 2015). Chronic arsenic exposure can cause cardiovascular disease, skin lesions, neuropathies, respiratory dysfunction, diabetes, goitres and numerous cancers (e.g. skin, bladder, liver, lung, kidney) (Liu et al., 2008; Swift et al., 2015). Illness can occur decades after limited arsenic exposure (Smith et al., 2006). However, it is important to note that some forms of arsenic have poor solubility in water and are poorly absorbed into the body (e.g., orpiment, realgar), thereby being quickly eliminated through urine and excrement (Liu et al., 2008). Other forms (e.g., arsenolite, arsenic oxide) are highly bioavailable with arsenic trioxide being extremely toxic (Liu et al., 2008; Hu et al., 2012). Some arsenic preparations used in traditional medicine have been demonstrated to be effective and have beneficial properties (e.g., killing leukemia cells), but their toxic risks and carcinogenic nature complicate the justification for their use (Liu et al., 2008). Nonetheless, arsenic trioxide is prescribed in specific situations, particularly for the treatment of acute promyelocytic leukemia (Bolt, 2012).

c. Skeletal implications

Arsenic replaces phosphorus, thereby localizing in bone where it can be retained for years and contribute significantly to bone marrow abnormality and other skeletal disorders (Wu et al., 2014). Arsenic alters bone metabolism, inhibiting osteoblast differentiation and may result in osteopenia (Wu et al., 2014). The earliest evidence of arsenic poisoning in archaeological remains due to environmental exposure was found in “Ötzi”, the Tyrolean Neolithic mummy that lived and died sometime between 3359-3105 BC (Bolt, 2012). Swift et al. (2015) found toxic elevations of arsenic in the skeletal remains of a pre-Columbian population from northern Chile, which were likely associated with the volcanic setting and local geology of the Andes.

The concentration of arsenic in skeletal tissues increases with age (Brodziak-Dopierala et al., 2011). Arsenic poisoning has been noted in modern cases with bone concentrations ranging from 0.5-1.0 ppm, but toxicity normally occurs with concentrations above 1 ppm (Swift et al., 2015). On the other hand, dental enamel concentrations of arsenic have been reported in ancient and contemporary human teeth as ranging between <0.001-0.406 ppm (Rasmussen, 1974).

Arsenic exposure increases the risks of bone disorders but does not cause characteristic changes useful for the osteological diagnosis of arsenic toxicity (Wu et al., 2014). A notable skeletal effect of arsenic exposure in modern populations is bone cancer (Tsai et al., 1999). The results of Akbal et al. (2014) demonstrated a relationship between arsenic exposure and the functionality of bone metabolism as indicated by increased rates of osteopenia. Essentially, arsenic exposure interferes with bone remodeling and may result in the inihibition of osteoblast differentiation of bone marrow stromal cells (Hu et al., 2012; Wu et al., 2014). While some of these skeletal disorders or bone changes could indicate cadmium or arsenic toxicity, the same changes may also indicate toxicity with other heavy metals, such as mercury or lead, in addition to a vast array of other pathogeneses unrelated to exposure to toxic substances. It is thus vital to assess the concentration of multiple metal species in bone if any relationship or correlation between bone changes and toxic metal exposure may be indicated.

2.3.6 Strontium (Sr)

a. History

Strontium, an alkaline earth metal, was first discovered in 1790 in a Scottish mine, but was not isolated until 1808. Strontium does not occur free in nature and instead it occurs as a result of metallic strontium oxidizing to form the yellow-colored strontium oxide. It is present in water and soil because it is a component of the earth's crust and humans primarily ingest it through drinking water and the consumption of vegetables and cereals (Nielsen, 2004). Most forms of low dose strontium are only minimally toxic or non-toxic, however, it can result in hypocalcemia as it increases renal calcium excretion (Cabrera et al., 1999; Nielsen, 2004). Experiments on animals have demonstrated that the amount of consumed strontium can vary significantly without the appearance of toxic effects. However, one study showed that when pigs were fed with 6700 ppm strontium and just 0.16% calcium, they experienced a lack of coordination, posterior paralysis, and weakness (Nielsen, 2004). Additionally, older research suggested that a diet high in strontium produced insoluble strontium phosphates, which may result in rickets or phosphorous deficiency (Jones, 1938; Nielsen, 2004). Human and animal research has since shown that harmful bone changes, such as rachitic lesions, occur with simultaneously increased strontium intake (e.g., high Sr in soil, vegetables, or water) and decreased vitamin D and calcium intake (e.g., lack of sunlight or dietary deficiency) (Nielsen, 2004).

b. Mechanisms

Strontium is generally poorly absorbed through the intestines, particularly in the presence of calcium, although vitamin D has been shown to promote absorption within the intestinal tract. Meanwhile, carbohydrates and lactose increase the absorption of both calcium and strontium. The typical diet consumed in most western countries results in neglible strontium exposure, particularly compared to calcium. Although strontium is a non-essential trace element, it is used in various medical treatments (e.g., radiation therapy for cancer; drug therapy with strontium ranelate for osteoporosis and has been shown to provide some preventative mechanism for the development of carious lesions. While the total amount of calcium within the skeleton is far greater than the amount of strontium (i.e., amount of Sr making up just 0.035 of the Ca content), strontium deposits almost entirely within skeletal tissues (Nielsen, 2004). Strontium substitutes for calcium within the hydroxyapatite of skeletal tissues but the amount absorbed is less than the dietary amount due to biopurification processes (Burton et al., 2003). Non-absorbed strontium is predominately excreted through the renal system. Strontium behaves very similarly to calcium, both of which seek out bone, but competes with calcium for absorption within the intestinal tract and kidneys for example (Nielsen, 2004). Approximately 90% of the strontium stored within the body is retained in bone (Ezzo, 1994). Since Iceland's landmass is composed primarily of basaltic rocks (~90%), most of which contain high concentrations of strontium (100-1000 ppm), all life forms residing in Iceland ingest basalt dietarily (Davidson, 1999; Snæbjörnsdóttir et al., 2014).

c. Skeletal Implications

Using ICP-MS, Liu et al. (2013) found that dental enamel from modern samples of teeth (Taiwan) show a mean strontium concentration of 108.31 ± 35.71 ppm. Strontium has been shown to engage in cariostatic activity (Dogan, 2018). Some studies have noted that strontium concentrations in dental enamel are lower in carious teeth (79.7-85.8 ppm) than in healthy teeth (128-156.8 ppm), regardless of age or sex (e.g., Li et al., 2013; Dogan, 2018). Therefore, dental enamel samples subjected to trace element analyses should be selected from non-carious, healthy teeth with well-preserved dental enamel to acquire data useful for archaeological interpretations about diet and geographic provenance. Regarding dietary reconstruction using strontium concentrations, completely vegan diets do not produce Ba/Ca and Sr/Ca ratios that are significantly different from the diet of organisms consuming a 60-70% carnivorous diet because calcium in both vegan and predominately carnivorous diets is derived from plant sources (Burton et al., 2003). High barium and strontium concentrations resulting from this biopurification at the herbivorous level overpowers the limited alkaline earth contribution derived from meat (Burton et al., 2003). Considering this, little variance in diet amongst group members residing in the same area is shown through these biomarkers (Burton et al., 2003). Strontium isotope ratios ($^{87}Sr/^{86}Sr$) are, however, one of the most useful and effective isotopic ratios for examining mobility and geographic provenance in ancient animal and human populations, particularly if paired with other isotope analyses (e.g., oxygen, carbon, lead, sulphur). As previously discussed, strontium isotopes enter the human skeleton through the consumption of food and water; the geochemical signatures of ingested strontium isotopes are recorded in the skeleton and can be mapped to past geological regions within the environment of an individual's residence. Strontium ratios derived from bone, which provides a record connected to the time of death, are wrought with issues of diagenesis. However, dental enamel is highly resistant to diagenesis, but reflects signatures recorded during the development or formation of the enamel (i.e., *in utero* and during childhood) (Bentley, 2006).

2.3.7 Zinc (Zn)

a. History

Zinc was extensively produced during Antiquity and the Medieval period as an important material used to produce brass and copper-zinc alloys, which were likely produced as early as ca. 3000-4000 years ago (Craddock et al., 1987; Hong et al., 1997). The form known as zinc oxide was used by early physicians (e.g., Galen) in medical treatments, especially in combination with lead for the treatment of cancers (Karpozilos et al., 2004). Zinc was heavily mined and used for brass making in India and China and historical and archaeological evidence indicates that substantial technology transfer occurred between India and the western world (Biswas, 2006). For example, zinc distillation technology spread and increased in use significantly in the 13th century AD although it was likely in use as early as the 4th century BC (Biswas, 2006). While it has been found at ancient Greek archaeological sites, it was likely mined in Germany, India or China or even brought as a souvenir from India (Biswas, 2006). Ores containing zinc normally also contain lead and often silver, cadmium and copper as well, most of which were exploited significantly throughout Antiquity and the Medieval period (Craddock et al., 1987; Hong et al., 1997). Despite the extensive exploitation of zinc in the past, no significant changes to zinc concentrations are represented in Greenland ice cores during the Greek, Roman and Medieval period. Unlike lead, it appears that zinc mining and smelting was not intense enough to cause emissions that would leave notable signals in Greenland ice beyond the normal background concentrations. Except during periods of severe volcanic eruptions, volcanic emissions were likely an insignificant source of atmospheric zinc emissions according to the results of Greenland ice studies (Hong et al., 1997). Nonetheless, zinc pollution did occur in the past from metallurgic and mining activities, as it continues to occur today along with additional pollution contributed by automobile traffic, waste incineration, galvanic industry and the production of rubber, paint, and plastic (Tvinnereim et al., 1999). In Iceland, zinc is released during volcanic eruptions, with large amounts being released during the eruption of Laki (AD 1783-1784) for example (Hong et al., 1997). Otherwise, zinc appears to be low in the environment and one study even indicated that grazing animals in Iceland (i.e., cattle and sheep) sometimes exhibit parakeratosis associated with zinc deficiency (Jóhannesson et al., 2007).

b. Mechanisms

Zinc is an essential dietary element, important for proper metabolic and other human body functions (Tvinnereim et al., 1999; Brodziak-Dopierala et al., 2015). Zinc is derived predominately from high protein food sources, such as meat, dairy, poultry, seafood, and some whole grain products (Tvinnereim et al., 1999). Its functions include the stimulation of metallothionein synthesis and proliferation of osteoblasts, regulation of vitamin D activity, regulation of hormones and cell growth, protection against free radicals and prevention of parathyroid hormone stimulated bone resorption (Tvinnereim et al., 1999; Brodziak-Dopierala et al., 2015). It also plays a role in protein synthesis and acts as a co-factor for enzymes responsible for the regulation of gene transcription (Tvinnereim et al., 1999).

Zinc deficiency can contribute to osteoporosis, with studies demonstrating that low-zinc diets lead to reduced bone size, length and stability thereby increasing fracture risk and diminishing trabecular bone content (Brodziak-Dopierala et al., 2015). It is also important for the proper mineralization of bones, acting to enhance and stimulate the synthesis of growth factors (e.g., IGF-1) on skeletal tissues (Brodziak-Dopierala et al., 2015). Severe zinc deficiency can even cause dwarfism (Ezzo, 1994). It reduces bone tissue growth and induces bone resorption, resulting in thin, brittle bone prone to fracture (Ezzo, 1994; Brodziak-Dopierala et al., 2015). Signs and symptoms include poor skeletal development, poor appetite, reduced wound healing, impaired immune response, and skin changes (Tvinnereim et al., 1999). On the other hand, zinc toxicity, which is rare, can cause immune system abnormalities, as well as nausea, lethargy, fatigue, vomiting and epigastric pain (Fosmire, 1990; Tvinnereim et al., 1999). Excess zinc can also induce copper deficiency, adding additional signs of neutropenia and anemia (Fosmire, 1990). Zinc and cadmium have an antagonistic relationship, with cadmium inhibiting zinc absorption within the intestinal tract. Furthermore, cadmium acts to block zinc stimulated growth factor (IGF) production, which acts to reduce osteoblastic bone matrix protein production, particularly in populations of older cells, potentially resulting in toxic bone tissue effects as age increases (Brodziak-Dopierala et al., 2015).

c. Skeletal implications

Zinc absorption occurs predominately within the intestinal tract, with the absorption rate generally correlated inversely with the quantity of zinc consumed in the diet (Ezzo, 1994). The skeletal system retains approximately 28% of the body’s zinc store (Ezzo, 1994). As zinc is a trace element under homeostatic regulation, the amount of dietary intake does not necessarily correlate with the total zinc within the body unless zinc deficiency is indicated (Ezzo, 1994). Bone and dentine are prone to the diagenetic uptake of zinc from the burial environment, correlating with the zinc content of the soil; zinc is best assessed in dental enamel, which is far less susceptible to diagenesis (Ezzo, 1994). Lappaleinen et al. (1981) showed an increase in zinc concentration in dental enamel according to increasing age. A combination of studies reviewed by Ezzo (1994), suggest ancient and modern bone concentrations of zinc as ranging from 10-1550 ppm and in enamel from 58-2100 ppm. Enamel concentrations may relate to zinc absorption, with research results showing a positive correlation between extent of industrialisation/urbanisation and zinc/lead concentrations (Tvinnereim et al., 1999). Clinical research has shown an association between low enamel zinc concentrations and malnutrition (Fosse and Justesen, 1978; Tvinnereim et al., 1999; Brown et al., 2004). Norwegian research on deciduous dental enamel showed that zinc concentrations of less than 90 ppm might indicate poor dietary zinc intake in childhood (e.g., Fosse and Justesen, 1978; Tvinnereim et al., 1999). Furthermore, essential trace element deficiencies (e.g., calcium) may cause the rapid and irregular absorption of toxic heavy metals, especially in malnourished children (Talpur et al., 2018). Nonetheless, since zinc is subject to homeostatic control, zinc measured in enamel may not correlate with palaeodiet (Ezzo, 1994; Dolphin and Goodman, 2009).

2.3.8 Barium (Ba)

a. History

In the past, blue and purple pigments were produced by the ancient Egyptians (i.e., Egyptian Blue) and Chinese (i.e., Han Blue, Han Purple) from barium copper silicates (an artificially produced, pigmented mineral). Egyptian Blue was used throughout Egypt, the Mediterranean, Western Asia and parts of central Europe until the fall of the Roman Empire (Ma et al., 2006). The Romans acquired the recipe for Egyptian Blue and thereby began to produce vestorianum (Pompeii Blue). The archaic Greeks and Persians also used Egyptian Blue on architectural structures and statues or figurines (Thieme, 2001). Han Blue and Purple, on the other hand, were important pigments used on bronze objects and pottery in China, particularly during the Han Dynasty (206 BC-AD 220) (Thieme, 2001; Ma et al., 2006). These were also used to adorn jewelry, the Terracotta Army and other objects (Thieme, 2001; Ma et al., 2006). Notably, almost all samples of artifacts adorned with Han Blue or Purple also contain substantial amounts of lead (Ma et al., 2006), adding additional toxicity to objects with barium copper silicate-based pigments. Today, toxic soluble barium salts are found in rodenticides, fireworks, fertilizer, insecticides, and depilatory substances and are also used during textile dyeing, welding, semiconductor production, glass manufacturing and in the steel and petroleum industry (Talwar et al., 2007; Naimy, 2008; Kravchenko et al., 2014). Various barium compounds are also used in the manufacturing of electronics, ceramics, pharmaceuticals, rubbers, plastics, bricks, cosmetics, and paper (Kravchenko et al., 2014). One study showed barium concentrations in welders were elevated up to 60 times the concentration found in the average population (Zschiesche et al., 1992; Kravchenko et al., 2014). Metallurgic activity in the past may therefore be considered a potential source of barium toxicity, as previously noted with mercury, lead, arsenic and cadmium. Cases of toxic natural exposure due to the combination of high barium levels in the local environment, cultural practices and diet have also occurred. For example, in Szechuan province (Pa Ping and Kiating areas) of China, where the local geology includes purple shales high in barium, phosphorous and calcium, subacute barium poisoning was endemic due to contaminated table salt and flour (Kravchenko et al., 2014; Oskarsson, 2015). Today, elevated barium concentrations are also found where industrial waste is disposed (Llugany et al., 2000).

b. Mechanisms

Barium is an alkaline earth metal that is present in substantial quantities in the natural environment and is concentrated in the earth’s crust (Llugany et al., 2000; Kravchenko et al., 2014). Of interest in this thesis, is that geographic regions with volcanic activity are also usually high in barium content (Kravchenko et al., 2014). Past eruptions in Iceland show that barium concentrations increase with silica concentrations (Alnethary, 2018). Increased barium may thus occur with certain eruptions, such as the 2010 eruption of Eyjafjallajökull where 58% of the mass of ash was composed of silica (Gislason et al., 2011). Barium is absorbed via inhalation through the lungs or through the intestinal tract via ingestion (ca. 70-80% from food, ca. 20% from water) and predominately accumulated within the skeletal system (Kravchenko et al., 2014; Oskarsson, 2015). Its concentration in soil is low to moderate (up to 1200 ppm; ca. 75 in Icelandic bedrock) but marginal in seawater (ca. 0.013 ppm) and freshwater (0.0026 ppm) (Llugany et al., 2000; Naimy, 2008). Due to the high content of barium in soil and geological features, plants tend to have the highest amount of barium, followed by terrestrial herbivores, while carnivores have the lowest concentrations (Burton et al., 2003). Similarly, sea fish and marine mammals have substantially lower barium concentrations than plants, terrestrial animals, and freshwater fish (Llugany et al., 2000; Burton et al., 2003; Naimy, 2008; Szostek et al., 2009). In theory, bone barium concentrations should be lower in humans that consume more meat than plants (Burton et al., 2003). Freshwater barium concentrations range between 0.01-302 ppb (average 3.6 ppb) in Iceland (Naimy, 2008).

Some compounds of barium are toxic dependent upon their solubility and mechanism of absorption, while others are used medicinally (e.g., barium sulfate) and do not necessarily absorb at all (Kravchenko et al., 2014; Oskarsson, 2015). Most barium found in the environment has low solubility, making the risk of exposure to toxic barium compounds low (Llugany et al., 2000). However, cases of barium poisoning resulting in death have occurred when individuals ingest, accidentally or suicidally, compounds containing soluble barium (Naimy, 2008; Kravchenko et al., 2014; Oskarsson, 2015). Toxicity may occur with the ingestion of as little as 200 mg of soluble barium salts, but a lethal dose can occur with the ingestion of 1-30 g of various soluble barium salts depending upon the degree of absorption, diet, and gastric pH (Naimy, 2008). It is a common pollutant in urban air as particulate matter produced through combustion of diesel or coal and the incineration of waste materials. Major sources of ingested barium include nuts, milk, freshwater fish, seafood, and legumes. Lactose in milk increases the overall absorption of barium (Kravchenko et al., 2014), which may be a relevant factor in the examination of barium concentrations in Iceland due to the high dietary dependency on dairy products in the past. If inhaled, it can cause benign pneumoconiosis (Oskarsson, 2015).

Animal experiments and cases of human exposure have demonstrated that toxic barium salts cause cardiac malfunction, renal and liver toxicity or failure, reproductive system alterations, instestinal hemorrhage, hypokalemia (i.e., low potassium), cardiac arrythmias, pulmonary edema, hearing loss and hypertension (Talwar et al., 2007; Kravchenko et al., 2014; Oskarsson, 2015). Signs and symptoms of barium poisoning include nausea, vomiting, tremors, headache, confusion, seizures, diarrhea, abdominal pain, respiratory paralysis and quadripilegia, all of which may begin within just hours of exposure (Talwar et al., 2007; Kravchenko et al., 2014). These experiments also showed that the kidney is a sensitive target organ in animals ingesting drinking water that contains barium chloride (Oskarsson, 2015). Barium poisoning blocks potassium channels in cell membranes, redistributing it, thereby causing severe hypokalemia that may lead to paralysis (Talwar et al., 2007; Oskarsson, 2015). Soluble forms of barium (e.g., Ba^{2+} ion) act as a muscle poison ultimately leading to paralysis (Oskarsson, 2015).

c. Skeletal implications

Barium enters the body through the diet, such as via the consumption of plants or terrestrial animal tissues (Szostek et al., 2009). It is a non-essential element that is not under homeostatic control in the human body, has no biological function and it varies according to the consumer's trophic level (Szostek et al., 2009). Dependent upon the chemical form of the ingested barium, only about 5-30% of it is absorbed within the intestinal tract, with anywhere between 9-98% of it being excreted through urine and excrement (Kravchenko et al., 2014). Barium absorption may be inhibited by the intestinal concentrations of antagonistic compounds, such as zinc, phosphorous and calcium (Kravchenko et al., 2014). Barium replaces calcium within the hydroxyapatite, or mineral component, of bone where 90-99% of the total barium amount is retained (Szostek et al., 2009). The total amount of barium retained in bone appears to also be partly determined by the amount of casein (i.e., milk proteins) present in the human diet, potentially reflecting the inclusion of dietary components of an animal origin. A similar effect has been noted with the elevation of zinc concentrations (Szostek et al., 2009). One study showed barium concentrations in dental enamel to be 6.4 ppm in dry weight (Manea-Krichten et al., 1991). Another study found that deciduous teeth contain 6.41 ppm while permanent teeth contain 4.4 ppm (Brown et al., 2002, 2004; Liu et al., 2013). Liu et al. (2013) found a lower average of barium (1.96 ± 1.01 ppm) in dental enamel. In the case of lead exposure, barium concentrations in dental enamel remain lower than lead concentrations (Manea-Krichten et al., 1991; Liu et al., 2013).

Barium analysis can be applied in archaeological studies to examine diet and geographic provenance. However, as calcium in either vegan or predominately carnivorous diets is derived from plant sources, completely vegan diets do not produce Ba/Ca and Sr/Ca ratios that are significantly different than the diet of organisms consuming a 60-70% carnivorous diet. This biopurification at the herbivorous level, resulting in high barium and strontium, overpowers the limited alkaline earth contribution that is derived from meat. Considering this, these biomarkers show little variance in diet amongst group members residing in the same area (Burton et al., 2003). As a result, barium analysis can corroborate the findings of other isotope analyses that suggest whether any non-local individuals (from childhood) were members of a local group (or if all members were local).

2.3.9 Antimony (Sb)

a. History

Antimony (Sb) has been used since ancient times in cosmetics, alchemical experiments (e.g., transmutation into gold), medicine and amalgamation (Hansell, 2015). The ancient Egyptians, Greeks and Assyrians all made use of antimony for various ailments, such as for conditions of the urinary system, and the Eberus papyrus (ca. 1550 BC) discusses its use as a medicinal remedy (McCallum, 1999). As previously discussed (section 2.3.3), scholars formerly believed that the fall of the Roman Empire may have been partly caused by chronic lead poisoning among the population, but this theory has been dismissed as the calcite deposits in the pipes would have prevented the transfer of lead into running water (Riva et al., 2012). Recent research by Charlier et al. (2017) instead suggests that the Romans living at Pompeii may have been poisoned by antimony leaching out of their lead pipes. In Iceland, antimony is released during volcanic eruptions, such as during the 2014-2015 eruption of Holuhraun (Bárðarbunga-Veiðivötn volcanic system) when the air became enriched with antimony and other trace metals (Jónsdóttir and Smáradóttir, 2015).

Antimony is rarely found naturally in a metallic state and is usually found in an ore called stibnite (Sb_2S_3) (Hansell, 2015). In antiquity, stibnite was frequently used as an eye makeup due to its deep, black color, but it was also used to treat skin conditions (Hansell, 2015). The widespread use of antimony in Europe appeared in the Medieval period, only gaining extensive attention from ca. 1300s, particularly in medicine (McCallum, 1999). It was also frequently used in glass production and to produce domestic vessels (e.g., cooking vessels) (Dungworth and Nicholas, 2004). However, despite the knowledge of its poisonous nature, it played an important role in humouric medicine as an alternative to bloodletting as it was believed to balance the humours by inducing sweating, vomiting, or having laxative effects (McCallum, 1999; Hansell, 2015). It was taken as a pill or pellet, which was later collected for re-use probably because it was expensive (Hansell, 2015). By the 1600s antimony pellets were banned and new, alternative treatments came into play such as drinking wine that was left in an antimony cup overnight (Hansell, 2015). In fact, the use of antimony was banned in 1566 and in 1615 in Paris due to its poisonous properties but the bans were then overturned in 1666, with antimony thus becoming officially approved as a medicine. King Louis XIV of France used a medicinal preparation containing antimony with purportedly successful results, thereby diminishing concerns over its use and effectiveness (Cooper and Harrison, 2009). However, it has also been suggested that antimony poisoning may have contributed to Mozart's young age-at-death (35 years old) (McCallum, 1999; Hansell, 2015).

Although numerous metals and herbs were used in medical preparations, antimony stirred the most conflict and controversy, with the period of ca. 1560 to ca. 1660 being called the "Antimony War" in Western Europe, particularly in France and Germany (McCallum, 1999). Antimony (and arsenic) was often a component found in *speiss*, a residue of silver smelting that often problematically absorbed some of the silver and presented an inhalational health hazard to metallurgical practitioners. This residue was often discarded in antiquity, but metallurgists and alchemists later learned to extract the silver from it (Williams, 2009). It was common to alloy antimony with other metals to improve its strength and hardness for use in various products. For example, in the printing press invented by Johannes Gutenburg in the 1400s, lead-tin-antimony alloys were used for the metallic type blocks (Hansell, 2015). The medical use of antimony declined substantially in the 18th century and after the 19th century its use shifted primarily to industry (McCallum, 1999). In the 20th century, anthropologists described the use of mineral antimony for staining the teeth black by some cultures in the Philippines to beautify and strengthen the teeth (Atienza, 2014). Antimony is still used today as a flame retardant in plastics, fabrics and brake linings and it is also used as a component in batteries, rubber compounding, electronics, sheet and pipe metal, solder, ammunition, pewter, bearings, type metal, castings, paint pigments and some alloys and ceramics (Cooper and Harrison, 2009; Hansell, 2015). Some pentavalent compounds of antimony remain in medical use today, predominately for treating tropical diseases such as the parasitic infection leishmaniasis (Al Jaser et al., 1995; McCallum, 1999; Cooper and Harrison, 2009; Hansell, 2015).

b. Mechanisms

Antimony is a toxic substance and non-essential element that has also been found to be carcinogenic (Hansell, 2015; Tylenda et al., 2015). It enters the environment predominately through anthropogenic means, such as mining, smelting, amalgamation, alloying and the processing of its ores in general (Cooper and Harrison, 2009). Exposure occurs through ingestion via water, food or soil contact or inhalation via airborne dust (Cooper and Harrison, 2009; Hansell, 2015). Antimony exposure may likewise occur through milk consumption, but exposure through food or diet appears to be insignificant. Exposure to airborne antimony results in eye, lung, and skin irritation, with chronic exposure resulting in antimoniosis (a form of pneumoconiosis) (Cooper and Harrison, 2009). Another source of exposure to antimony in the past and the present is through glass making (Jackson, 2005; Cooper and Harrison, 2009). For example, the Romans used it extensively to create colourless glass as antimony acts as a decolorizor and fining agent resulting in bright, clear glass (Jackson, 2005). In addition to its oncogenic and mutagenic potential, other effects of chronic exposure include heart, gastrointestinal, lung conditions (e.g., cancer), chromosome damage, and reproductive disorders (Cooper and Harrison, 2009). Antimony exposure has also occurred with cases of lead toxicity because antimony ores are frequently associated with lead and arsenic, causing symptoms such as constipation, loss of appetite, colic, mouth ulcers, weight loss, glycosuria, albuminuria, dizziness, abdominal pain, and headaches (Cooper and Harrison, 2009). Like most toxic metals, the risk of antimony toxicity relates to age, sex, nutritional condition, lifestyle, family traits, overall health and the dose, route, and duration of exposure. Contemporaneous exposure to other toxic metals, such as lead, increases the toxicity risk of antimony. Unlike many other toxic elements (such as lead), antimony is not known to accumulate in aquatic organisms and does not leach through drinking water pipes (Cooper and Harrison, 2009). Additionally, antimony does not systemically distribute to all the same tissues that other toxic metals do, according to post-mortem examinations that found insignificant concentrations of it in liver and kidney tissues (Cooper and Harrison, 2009). It is primarily excreted through urine and feces (Tylenda et al., 2015). Antimony is generally very low in soil (<1 ppm) with high concentrations (109-2550 ppm) being found today predominately at industrial or hazardous waste sites (Harrison and Cooper, 2009).

c. Skeletal implications

Antimony is poorly absorbed by the body, but the antimony that is absorbed is retained long-term and has a long biological half-life, particularly in lung tissues. Other tissues that may exhibit high concentrations are the thyroid and adrenal glands, liver, and kidneys (Tylenda et al., 2015). As the mechanisms of antimony absorption remain poorly understood (Hansell, 2015), the skeletal implications of exposure to it are unclear. Some studies have examined its function in the liver, spleen, and bone marrow (Al Jaser et al., 1995). Retief et al. (1970) found a mean antimony concentration of 0.96 ppm, Underwood (1977) found a range of 0.005-0.67 ppm and Jones (2014) found a range of 0.00-1.01 ppm with a mean of 0.05 ± 0.15 ppm in modern dental enamel samples. Rasmussen (1974) found a range of <0.001-1.59 ppm in human dental enamel from a combination of archaeological and modern individuals.

2.4 Diagenesis in bone

Numerous factors, involving burial conditions, contribute to the diagenetic alteration of bone, such as the presence of groundwater and soil contamination via anthropogenic or environmental means. While dental enamel can incur diagenetic enrichment if poorly preserved, this is far less likely than in bone (Hollund et al., 2015). Therefore, the following section will address diagenesis in bone, the far more susceptible skeletal tissue, with respects to the elements discussed above and analysed in this thesis. The presence of groundwater is an important factor as elements such as arsenic are mobilized within it, thereby causing surface contact between the element in the water and the buried skeletal material (Swift et al., 2015). Various scholars recommend comparing the soil concentrations of the burial environment with the concentrations found in bone samples to examine the likelihood of diagenetic enrichment (e.g., González-Reimers et al., 2003, 2005; Rasmussen et al., 2008, 2013a, 2013b, 2015, 2017; Ávila et al., 2014; Swift et al., 2015). Another way to examine possible diagenetic change is to investigate the bone element concentrations in non-adults, especially infants, whose bones are less mineralized and more porous and therefore more prone to diagenetic uptake (Swift et al., 2015).

European soil background concentrations of arsenic are reported to 11.6 ppm (range 0.32-282 ppm), of mercury to 0.061 ppm (range 0.005-1.35 ppm), of cadmium to 0.284 ppm (range 0.145-14.1 ppm) and of lead to 32.6 ppm (range 5.32-970 ppm) (Salmien et al., 2001). Total fluoride concentration in most soil types can range anywhere from from 20-1000 ppm (Edmunds and Smedley, 2005; Ozsvath, 2008; WHO, 2002). Strontium, zinc, barium, and antimony are not discussed below because they were only measured in dental enamel, which is far less likely to undergo diagenetic enrichment than bone (Kendall et al., 2018). Additionally, the elements only measured in dental enamel have little to no comparative data for bone measurements and their diagenetic behaviors in bone have yet to be deeply investigated.

Mercury

Rasmussen et al. (2013a, 2015) measured soil samples associated with skeletons that had high mercury concentrations and these showed no correlation with soil mercury levels, thus indicating that diagenesis was not a factor. Yamada et al. (1995) and Zuckerman (2017a) likewise found no evidence for diagenetic transfer of mercury between bone and soil. Mercury is rare in nature and humans are not prone to the post-mortem uptake of it into bone, unlike with lead (Swanston et al., 2012) mercury is uncommon in the natural environment and humans are not prone to post-mortem uptake (Ávila et al., 2014; Rasmussen et al., 2015). Bone hydroxyapatite retains mercury by replacing calcium and bonding with carbonates during life (Lee et al., 2005; Ávila et al., 2014). Therefore, high concentrations of mercury in skeletal remains are robust indicators of *in vivo* exposure (Schwarz et al., 2013; Rasmussen et al., 2015). As noted earlier (section 2.3.1), the matrix of bone behaves as a long-term (>2 years) heavy metal reservoir prior to remodelling, but mercury has a half-life of just 60 days in non-skeletal tissues (Boyd et al., 2000; Ozuah, 2000; Miculescu et al., 2011).

Fluoride

Fluoride has a high affinity for water and as a result it is most concentrated in areas of the natural environment where water flows. Accordingly, the diagenesis of fluoride into bone tends to occur predominately in permanently waterlogged burial environments. For example, Petrone et al. (2011) examined the extent of fluoride diagenesis in bone samples from Vesuvius, where the ash deposit was rich in fluoride and permanently saturated with groundwater. The researchers measured fluoride concentrations in the surrounding environment, which proved to be fluoride-laden, and investigated structural change in bone through histology, which showed no changes correlated with microbial activity. Despite the fluoride contamination in the burial environment and surrounding area, they suggest that the anoxic nature of the waterlogged burial environment likely reduced the extent of diagenesis as these types of conditions inhibit microbial activities and other related diagenetic process. A few of the samples were found to have non-biogenic fluoride, such as the infants that likely showed diagenetic bone fluoride concentrations due to the porosity of the bones and their propensity for diagenetic uptake in general and were thereby excluded from further analysis (Petrone et al., 2011). Soil fluoride concentration in Iceland may increase following eruptions, such as the Hekla eruption of 1947-1948, when fluoride concentrations temporarily increased in fresh water sources from a low, normal maximum of 0.57 ppm to 9.5 ppm in some samples. Aside from ashfall deposit, such increases in stream water may contribute to temporarily increased fluoride concentrations in surrounding soil, which normally has a very low concentration in Iceland (Stefánsson and Sigurjónsson, 1957; Sigurðsson and Pálsson, 1957; Stewart et al., 2006; Gunnarsdóttir et al., 2016). Essentially diagenesis of fluoride in bone is primarily a concern if the surrounding environment contains a rich fluoride deposit in combination with significant groundwater saturation or water flow. Total fluoride concentration in most soil types worldwide generally range between 30-500 ppm (Edmunds and Smedley, 2005; Ozsvath, 2008), but can technically range anywhere between 20-1000 ppm (WHO, 2002). Volcanic soils defluoridate naturally, but slowly, over time through adsorption via contact with various forms of aluminum and clay minerals (D'Alessandro, 2006), both of which are common components of Icelandic geology.

Lead

The diagenesis of lead into skeletal remains has been thoroughly addressed in the literature and a correlation between soil and bone lead concentrations has been widely acknowledged (e.g., Waldron et al., 1979; Waldron, 1983; Vuorinen et al., 1990; Skytte and Rasmussen, 2013; Rasmussen et al., 2008, 2013a, 2013b, 2015). Trabecular tissue has been demonstrated to undergo far more diagenetic change than cortical bone, which can be mechanically cleaned to significantly reduce or even eliminate non-biogenic lead contamination from the surface of bone samples (Rasmussen et al., 2015). Some sites may be more prone to lead diagenesis due to factors such as pH (scale of acidity or basicity) and groundwater flux (inflows and outflows). Furthermore, earlier studies reporting diagenesis of lead in bone samples have often not reported what bone tissue was used for analysis or the methods employed for cleaning, decontaminating, and preparing the samples (e.g., Waldron, 1983; Vuorinen et al., 1990), perhaps because whole bone samples (cortical plus trabecular) were used and minimal or no sample decontamination was performed (Rasmussen et al., 2015).

Cadmium

Research conducted across Iceland showed average cadmium concentrations of 0.63 ppm (Panek and Kepinska, 2002). While the propensity for bone to undergo diagenetic change with natural background levels of cadmium in the burial environment have not been directly investigated, González-Reimers et al. (2003, 2005) did find a correlation between lead and cadmium concentrations in bone samples from the Canary Islands. Following that correlation and the lack of apparent lead diagenesis in bone, the research implied that diagenesis of cadmium in the bone samples did not play a significant role (González-Reimers et al., 2003).

Arsenic

While arsenic incorporation in bone is likely to occur biogenically via inhalation or the consumptions of contaminated water (Kabata-Pendias and Mukherjee, 2007), it may also be integrated into bone diagenetically via infiltration into bone through soil pore water (Mahoney et al., 2005). Unfortunately, arsenic appears to be an element which is quite susceptible to diagenetic enrichment, particularly if the burial environment contains groundwater and clay minerals (e.g., iron, hydrous manganese, and aluminum oxides) and can therefore accumulate in bone even if surrounding arsenic concentrations are low (Pike and Richards, 2002). European soil background concentrations of arsenic are reported as 11.6 ppm (range 0.32-282 ppm) (Salmien et al., 2001), yet one Icelandic survey of soil arsenic concentrations found extremely low concentrations of just 0.10 to 0.50 ppm (with the concentrations closer to 0.50 ppm being found in the closest vicinity to smelters and industrial plants (Magnússon and Thomas, 2007).

3 Origins and Diet

3.1 The origins of the Icelandic population

Isotope (enamel $^{87}Sr/^{86}Sr$, $\delta^{18}O$; collagen $\delta^{34}S$), trace element (e.g., Pb, Sb, Ba, Sr, Zn), osteological and ancient (aDNA) analyses are generally used to obtain mobility information from archaeological skeletal remains (Walser III et al., 2020a). Ancient DNA analyses can also be used to examine the ancestry of a population by assessing changes or differences in genetic variation and drift (Ebenesersdóttir et al., 2018). These methods are particularly powerful when used in conjunction with historical documents, ethnographic research, and the literary record – an approach that was taken in this study.

3.1.1 Icelandic ancestry according to mtDNA and ancient DNA analysis

Despite its degraded nature, ancient DNA from human skeletal remains can be used to help explore the genealogical relationships between individuals and populations, as well as the burden of infectious disease (Roberts and Manchester, 2010; Anastasiou and Mitchell, 2013; Ebenesersdóttir et al., 2018). Earlier DNA studies indicated that the ancestry of Icelanders was primarily (between 60-90%) Norse and 10-30% Gaelic and that the population was, to a degree, genetically homogenous (e.g., Helgason et al., 2000a, 2000b, 2001; Goodacre et al., 2005). Research also showed that most of the matrilineal ancestry (62%) originated from Britain and Ireland while Y-chromosome microsatellite variations suggests that most of the patrilineal ancestry (75%) is of Scandinavian origin (Helgason et al., 2000b, 2009). Recently, genomic analyses based on Next Generation Sequencing (NGS) from 27 ancient Icelanders demonstrated that the founding population was not only composed of Norse and Gaelic individuals, but also of admixed individuals. A *D*-statistic test *D(YRI,X; Gaelic, Norse)* further revealed a greater affinity between Norse and contemporary Icelanders than between Norse and ancient Icelanders, indicating that settlers of Norse ancestry may have had greater reproductive success than the Gaelic settlers. This was possibly a result of social constraints (e.g., slavery of Gaels, language inequality, social marginalisation) and later immigration from Denmark, which controlled Iceland between the 14th and 20th centuries. The study also showed that the ancient individuals were notably more similar to their source populations than modern day Icelanders after a millennium of genetic drift (see Ebenesersdóttir et al., 2018). Other research has shown that certain pathological conditions have a high degree of heritability (e.g., inflammatory bowel disease, ankylosing spondylitis) (Thjodleifsson et al., 2007). The small founding population and founding events may have thus been relevant to the overall risk of developing certain diseases in historical Iceland.

3.1.2 Ancestry according to osteological analyses of non-metric traits

Previous non-metric studies, such as Hallgrimsson et al. (2004), have made suggestions about the origin of the Icelanders. Mandibular (*torus mandibularis)* and palatine tori (*torus palatinus)* are extremely common non-metric traits in both historical and present Icelandic populations, exhibiting one of the world's highest prevalence rates of oral tori (see Figure 3.1) (Hooton, 1918; Axelsson and Hedegaard, 1981, 1985; Halffman et al., 1992; Richter and Eliasson, 2008). Oral tori are bony growths extending from the mandible or maxilla towards the tongue. These traits are also frequently seen in other Scandinavian populations and the indigenous Greenlanders (e.g., the Inuit) and it is believed that there may be additional factors specific to the arctic region's environment, such as masticatory stress-related to tough food (e.g., harðfiskur) and a primarily meat-based diet (Hooton, 1918; Suzuki and Sakai, 1960; Halffman et al., 1992). Hooton (1918) found that amongst the Icelandic individuals from Alftanes and Haffjarðarey he analysed, 68% presented with mandibular torus and 71% presented with palatine torus. Richter and Eliasson (2008) found that mandibular tori were present in 50% of the individuals excavated from Skeljastaðir, while palatine tori were present in 40% of them.

Scott et al. (2016) noted that ancient Icelanders and the Greenlandic Norse showed a very high frequency (65-97%) of mandibular torus, while Norwegians showed a somewhat lower frequency (48%) and Danish Vikings showed a very low frequency (9%). Because of the small population in Medieval Iceland, a very small gene pool may have existed allowing for greater admixture of these potentially heritable skeletal expressions (Hallgrimsson et al., 2004). However, the study also indicated that the expression of mandibular tori was far more likely to result from environmental pressures or local stresses than from genetic inheritance or dental attrition although they suggest that cold, bruxism and high-protein diets may play roles in torus formation (Scott et al., 2016). Nonetheless, some clinical research suggests that genetic factors are dominant in the expression of oral tori (e.g., Auškalnis et al., 2015) yet others suggest environmental factors, abnormal continuous growth, masticatory hyperfunction, mechanical stress, trauma, mandibular morphology, or a combination of these variables, demonstrating that a single aetiology for torus expression cannot yet be validated (Cortes et al., 2014; Smitha and Smitha, 2015). Irritation to the oral mucosa may also be correlated with torus formation, which is relevant to this study of Icelandic skeletal remains in light of the irritation to eyes, connective tissue and mucous membranes that volcanic emissions are known to cause. Torus mandibularis is found situated between the periosteum and the mucous membrane and the exostosis itself is composed of lamellar bony tissues (Hooton, 1918; Suzuki and Sakai 1960; Halffman et al., 1992; Gorsky et al., 1998; Hallgrimsson et al., 2004).

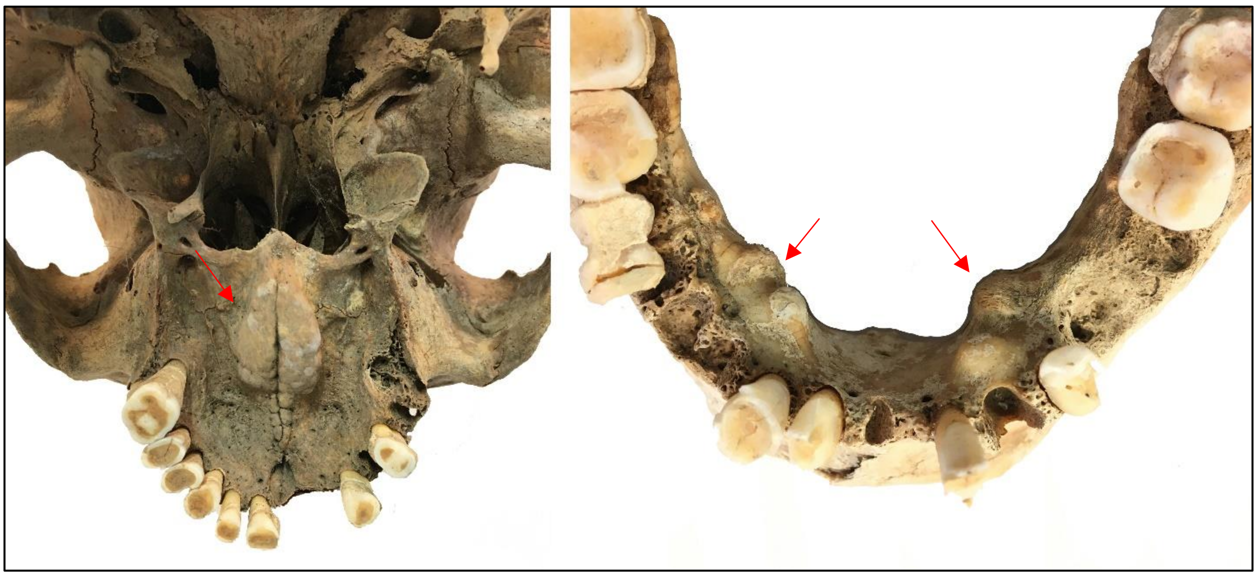

Figure 3.1 Left, palatine torus (SKR 169) – inferior view. Right, mandibular torus (ÞSK 55) – superior view

3.1.3 Principles: using $\delta^{18}O$, $^{87}Sr/^{86}Sr$ and trace element analyses (e.g., Sr, Pb, Zn, Ba) to evaluate geographic provenance

Mobility or geographic residence reconstructions are normally performed by determining isotopic ratios of strontium ($^{87}Sr/^{86}Sr$), oxygen ($\delta^{18}O$) (Åberg et al., 1995; Katzenberg, 2012) and trace elements (Kamenov and Gulson, 2014; Åberg et al., 1998). For the geographic provenancing of archaeological individuals, $\delta^{18}O$, $^{87}Sr/^{86}Sr$ and trace element analyses (e.g., Sr, Pb, Zn, Ba) may be conducted on dental enamel samples. Dental enamel is composed of a calcium phosphate lattice and is very resistant to diagenesis. Trace element ions can accumulate in enamel when they substitute for calcium during dental development (Åberg et al., 1998; Montgomery, 2002; Neil et al., 2017). Enamel does not remodel once it is mineralised and $^{87}Sr/^{86}Sr$ values do not significantly vary by trophic levels, so the ratio reflects the sources of strontium that people are exposed to during dental formation (beginning *in utero*, persisting through childhood and adolescence) (Montgomery, 2002; Bentley, 2006; Neil et al., 2017). The identification of the geographic provenance of archaeological individuals using strontium isotopes is possible because humans dietarily acquire strontium through biosphere sources; as geological weathering occurs, strontium is released from rocks and enters groundwater and soil where it becomes bioavailable, entering the food chain thereafter (Capo et al., 1998; Neil et al., 2017). While these levels vary between biota and specific tissues for several reasons, strontium isotope ratios do not undergo biologically induced fractionation due to the small differences in mass between strontium isotopes (Price et al. 2002). Isotopic studies have noted that strontium concentrations generally tend to be higher amongst individuals excavated from coastal or island sites (Neil et al., 2017).

Oxygen isotopes determined from skeletal remains primarily represent the isotopic composition of the drinking water people consume, however, they can also reflect variation resulting from culturally mediated behaviors (Brettell et al., 2012a, 2012b). Some variables that can alter oxygen isotope ratios include chemically bound oxygen in food, respiration, consuming fluids that undergo fractionations (e.g., cow's milk), food preparation methods (e.g., slow cooking, stewing), climate, ale drinking and breastfeeding/weaning (Brettel et al., 2012a; Neil et al., 2017). For example, the regular consumption of brewed drinks or slow cooked food can raise $\delta^{18}O$ values significantly (Brettell et al. 2012a). Furthermore, oxygen isotope ratios ($\delta^{18}O$) decrease according to increasing latitude, altitude, and the distance from sources of atmospheric water vapor (Laffoon et al., 2012). Since oxygen concentrations in human bioapatite are primarily derived from fluid ingestion, oxygen isotope ratios reflect drinking water and are thereby useful for estimating provenance (Laffoon et al., 2012; Price et al., 2015; Neil et al., 2017). Modern phosphate oxygen isotope ratios in precipitation were originally estimated on average to be around -7.0‰ in Iceland (Lecolle, 1985; Fricke et al., 1995). Modern bottled water from Reykjavík averaged $\delta^{18}O$ -8.7 (source) and -8.8 (purchase) (Bowen et al., 2005). A 2013 study reported the results of isotopic analyses for $\delta^{18}O$ in 11 groundwater samples from Iceland as ranging between -8.8 and -8.2 (Friedrich and Schlosser, 2013).

The mechanisms of uptake of strontium and lead differ and therefore respond to migration differently (Budd et al., 2004; Montgomery 2002). Lead concentrations in dental enamel generally range between 0.5-0.7 ppm, therefore a Pb value of more than 0.7 ppm in dental enamel potentially indicates exposure beyond the normal environment and individual outliers may represent migrants amongst a sample population (Evans et al., 2018). Lastly, other trace elements in dental enamel can also be used to provide details about diet and provenance. Zinc (Zn) values are correlated with meat consumption, environmental sources and anthropogenic pollution and are reported to range between 9.9 and 1550 ppm in dental enamel (Jaouen et al., 2016, 2017; Guede et al., 2017). Barium (Ba) and strontium (Sr) reflect local geology and the plant and water sources consumed (Liu et al., 2013; Guede et al., 2017). One study demonstrated that Pb values tend to be higher than Ba values when Pb exposure beyond the normal environment occurs (see Liu et al., 2013).

3.1.4 Geographic provenance as revealed by previous isotope and trace element analyses

Price and Gestsdóttir (2006) identified 32 non-Icelandic migrants using strontium isotope ($^{87}Sr/^{86}Sr$) ratio analysis on enamel from pre-Christian individuals (n=83) excavated from around the country. Individuals were identified as non-local if their $^{87}Sr/^{86}Sr$ values were greater than ca. 0.7092, which is the value for seawater and rain and the upper $^{87}Sr/^{86}Sr$ end-member for Iceland's basaltic biosphere. Icelandic geologic and bioavailable strontium isotope baselines ($^{87}Sr/^{86}Sr$) are presented in Table 3.1. This study likewise indicated $^{87}Sr/^{86}Sr$ variations may relate to differing diets between people residing inland (values closer to 0.703) and coastally (values closer to 0.7092) (Gestsdóttir and Price, 2006; Price and Gestsdóttir, 2006). Icelandic bioavailable strontium isotope ratios are greater than those derived from whole rock (ca. 0.703) due to the seasplash and spray occurring all over the country. A small subset (n=10) of the same Viking Age samples also underwent oxygen isotope analyses, though $\delta^{18}O$ values were only published for five of them (see Gestsdóttir and Price, 2006; Price et al., 2015). The combination of the $^{87}Sr/^{86}Sr$ and $\delta^{18}O$ values determined for the sampled individuals imply non-local geographic origins. Lastly, tooth and bone samples of one early Settlement Period female migrant, *Bláklædda Konan* (LKS 1), underwent $^{87}Sr/^{86}Sr$, $\delta^{18}O$, $\delta^{15}N$, $\delta^{13}C$ isotope analyses, in addition to the trace element analyses of strontium and lead (Montgomery and Jakob, 2015). The results of these two studies provide a limited, but interesting pool of comparative data.

Table 3.1 Strontium ratios ($^{87}Sr/^{86}Sr$) determined in bedrock, grass grown on volcanic soil, modern barley, and seawater and from modern and archaeological faunal dental enamel. The sheep samples were collected from inland sheep that did not graze upon seaweed, which may raise strontium isotope ratios (Price et al., 2015)

Enamel sample	87Sr/86Sr	n	Source	Material	87Sr/86Sr	Source
Archaeological Cattle	0.7042	2	Price et al. (2015)	Bedrock	0.7030-0.7037	Sigmarsson et al., (1992); Price et al. (2015)
Archaeological Pig	0.7042	1	Price et al. (2015)	Grass (volcanic soil)	0.7030-0.7040	Åberg (1995)
Modern Redshank Bird	0.7057	5	Evans and Bullman (2009)	Barley	0.7068	Price et al. (2015)
Modern Sheep	0.7059-0.7069	5	Price and Gestsdóttir (2006)	Seawater	0.7092	Åberg (1995)
Modern Reindeer	0.7060	1	Åberg (1995)	Rainwater	0.7090	Åberg (1995)

Intra- and inter-population differences in geographic provenance can be observed by assessing trace element variability between individuals. These applications are particularly strong with non-essential elements that are not under homestatic control (Jaouen and Pons, 2017). Prior to this study, apart from the single individual (LKS 1), trace element analyses (i.e., Zn, Pb, Ba, Sr) have not been performed on Icelandic human dental enamel samples. It is possible that such analytical data could improve geographic provenance determination for Icelandic archaeological populations (Burton et al., 2003; Montgomery et al., 2014). However, these elements were previously measured in Icelandic geology (rock, soil, plants, and groundwater) (see Table 3.2). The results of Panek and Kepinska (2002) showed that there was very little anthropogenic lead input soil and foliage from Iceland, especially when compared with the concentrations seen in Poland and Sweden.

Table 3.2 Zinc, lead, and barium concentrations (ppm) found in two species of moss (Racomitrium sp. and Drepanocladus sp.), topsoils (andosols, regosols, leptosols, organic soil), bedrock (basalt) and groundwater in Iceland

Material	Zinc	Lead	Barium	Source
Racomitrium sp.	46.1	5.5	-	Panek and Kepinska (2002)
Drepanocladus sp.	54.1	5.9	-	Panek and Kepinska (2002)
Topsoil	83	5.8	-	Panek and Kepinska (2002)
Bedrock	63	4.7	75	Panek and Kepinska (2002); Naimy (2008)
Groundwater	-	-	0.0036	Naimy (2008)

3.2 The diet of past Icelandic populations

Dietary reconstructions from skeletal remains are usually performed by determining the isotope ratios of carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$) (Katzenberg, 2008) and more recently, sulphur ($\delta^{34}S$) isotopes (Nehlich, 2015; Sayle et al., 2016). The water and food sources (plants or animals) humans ingest are recorded within the consumers' tissues and can be revealed through isotope analyses (Sealy, 2001) providing powerful information, especially when used in combination with historical documents and archaeozoology.

3.2.1 Principles: using carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$) and ($\delta^{34}S$) isotope analysis and trace elements (Pb, Zn, Ba, Sr) for palaeodietary reconstructions

Carbon and nitrogen stable isotope analysis on human bone collagen is the most common method of reconstructing the diets of past populations. Carbon isotope ratios are dependent upon diet, based upon differences in photosynthesic pathways between C_3, C_4 and CAM (crassulacean acid metabolism) plants (Katzenberg, 2012). C_3 plants (e.g., wheat, oats, rye) are the most abundant (around ca. 85% of earth plant species) and are best adapted to cool environments, while C_4 plants (e.g., corn, millet) are rather adapted to warm and often arid environments (Yamori et al., 2014). Lastly, CAM plants (e.g., pineapple, aloe, cacti) are adapted to hot, dry environments (Katzenberg, 2012). An individual consuming primarily C_3 plants (average value around -26.5‰) will usually exhibit $\delta^{13}C$ values of around -21.5‰ in bone collagen. In bone collagen this value rises by about +1‰ in the consumer relative to the individual's dietary protein source (e.g., $\delta^{13}C$ for exclusively terrestrial protein consumers = ca. -20.5‰, $\delta^{13}C$ for exclusively marine protein consumers = ca. -12‰) (Smith and Epstein, 1971; DeNiro and Epstein, 1978; Schoeninger et al., 1983). Additionally, $\delta^{13}C$ values can vary significantly between or even within species (Katzenberg, 2012). $\delta^{13}C$ values measured in enamel carbonate can also impart dietary information that is closely correlated with $\delta^{13}C$ values measured in bone collagen (Loftus and Sealy, 2012). While $\delta^{13}C$ and $\delta^{15}N$ values determined in bone collagen predominately reflect the protein component and the consumption of C_3 and C_4 plants in a person's adult diet, $\delta^{13}C_{carbonate}$ values determined from bioapatite (i.e., dental enamel) reflect whole diet during the time the sampled tooth was forming. Individuals primarily consuming C_3 food sources generally exhibit $\delta^{13}C_{carbonate}$ values between -17.0 and -14.0 ‰ (Froehle et al., 2012; Neil et al., 2017).

Nitrogen isotope ratios ($\delta^{15}N$) are used to examine an organism's trophic level, which reflects the number of steps it is removed from the starting point of a food chain. Determining $\delta^{15}N$ can thus be very helpful during dietary reconstruction if an individual's diet contains more heavily enriched carbon isotopes, such as marine foods and C_4 plants. The $\delta^{15}N$ offset in human bone collagen is estimated to be around +5.5 ± 0.5‰ indicating a trophic shift value of around +4.5‰ (e.g., terrestrial herbivores range +2.5‰ to +6.5‰, terrestrial carnivores range +7‰ to +11‰) (Fernandes, 2015). Marine food webs are more complex, resulting in $\delta^{15}N$ values reaching between 15‰-20‰ in exclusively marine protein consumers (predators) (DeNiro and Epstein, 1981; Schoeninger et al.,1983; Schoeninger and DeNiro, 1984; Sayle et al., 2016). More recently, sulphur isotope analysis has been conducted, enabling deeper differentiation between protein sources in palaeodietary studies. Sulphur isotope ratio ($\delta^{34}S$) values measured in consumers reflect the local environment as characterised by geological processes. Plants absorb sulphur through their roots in the form of sulfate and oxidized sulphides, which leach into earth and water via bedrock weathering. Sulphur isotopes undergo little to no fractionation and therefore $\delta^{34}S$ values can provide information about the geographic provenance of an organism (Trust and Fry, 1992; Richards et al., 2001). However, sulphur ($\delta^{34}S$) ratios are most useful for identifying the differences between or the combination of marine, freshwater, and terrestrial dietary resources in archaeological remains (Sayle et al., 2016). Marine plants and algae exhibit a small range of $\delta^{34}S$ values around 17‰ to 21‰, while according to the local geology, terrestrial and freshwater plants exhibit a vast range of $\delta^{34}S$ values from -22‰ to 22‰ (Peterson and Fry, 1987). $\delta^{34}S$ values determined in bone collagen show an increase of about 1‰ from the dietary protein source (Peterson and Howarth, 1987). Researchers have also attempted to use trace element analysis (e.g., Pb, Sr, Ba, Zn) for palaeodietary reconstruction, as the variations seen in the results of such analyses may help supplement other data that can discriminate between populations or groups with differing diets (Safont et al., 1998).

3.2.2 Past diet according to archaeological research and historical and literary records

Despite the precarious sub-polar climate of Iceland, people have supplied themselves with a diverse selection of foodstuff since the start of the Settlement period (AD 871±2) (Karlsson, 2000). Though barley was cultivated, there was short, challenging, and limited growing season (Mehler, 2011; Svanberg and Ægisson, 2012; Mooney and Guðmundsdóttir, 2020). The spread of epidemics along with volcanic and climatic events occurring during the Little Ice Age (ca. 13th to 19th century AD) further complicated local cultivation (Dugmore and Véststeinsson, 2012; McGovern et al., 2014). Though it is often thought that plants were only a small component of the diet (Mehler, 2011; McGovern et al., 2014), subsistence gardening was practiced at some sites, such as at Skriðuklaustur (Kristjánsdóttir et al., 2014) (see Table 3.3). A list of the most common edible plants found in Iceland can be seen in Table 3.3.

Table 3.3 Examples of some of the most consumed edible wild plants in historical Iceland (see Gísladóttir, 1999; Mehler, 2011; Svanberg and Ægisson, 2012)

Common name	**Icelandic name**	**Scientific name**	**Plant part**	**Purpose**
Common silverweed	Tágamura	*Potentilla anserine*	Root	Subsistence
Common horsetail	Klóelfting	*Equisetum arvense*	Root	Subsistence
Garden angelica	Ætihvönn	*Angelica archangelica*	Root, leaves	Subsistence, medicinal
Scurvy grass	Skarfakál	*Cochlearia officinalis*	Root, leaves	Subsistence, medicinal
Common sorrel	Túnsúra	*Rumex acetosa*	Leaves	Subsistence
Iceland moss	Fjallagrös	*Cetraria islandica*	Moss	Subsistence, medicinal
Bilberry	Aðalbláberjalyng	*Vaccinium myrtillus*	Berries	Subsistence, seasoning
Bog bilberry	Bláberjalyng	*Vaccinium uliginosum*	Berries	Subsistence, seasoning
Crowberry	Krækilyng	*Empetrum nigrum*	Berries	Subsistence, seasoning
Dulse	Söl	*Palmaria palmata*	Seaweed	Subsistence
Carrageen moss	Fjörugrös	*Chondrus crispus*	Seaweed	Subsistence
Wild thyme	Blóðberg	*Thymus praecox*	Leaves, flowers	Tea, seasoning
Caraway	Kúmen	*Carum carvi*	Seeds	Seasoning
Common juniper	Einir	*Juniperus communis*	Berries	Seasoning
Common butterwort	Lyfjagras	*Pinguicula vulgaris*	Leaves	Seasoning, medicinal
Mountain avens	Holtasóley	*Dryas octopetala*	Leaves, flowers	Tea

The staples of the diet were diary, sheep meat and fish, much of which was often salt-preserved, fermented, smoked, or acidified/soured (Mehler, 2011). Fish and meats were also exchanged for various goods with foreign traders despite ample local production (Mehler, 2011). Archaeozoological research indicates that the subsistence economy in Iceland during the Settlement period was likely much healthier and far more stable than in Greenland where almost all available fat was exploited from all livestock bones except the ribs, indicating a less healthy diet compared to Iceland, where fat from bone marrow was not utilized to the same extent. This evidence of subsistence stress is probably the result of dwindling seasonal supplies due to differing climate conditions, soil quality and fishing behaviour between Norse Iceland and Greenland (Outram, 2003).

3.2.3 Past diet according to previous isotope research

The first palaeodietary isotopic study relevant to Icelandic populations was conducted in 1999 on the Greenland Norse, the Icelandic migrants that resided in Greenland (ca. AD 1000-1450). The results determined that these individuals underwent significant dietary change between the initial settlement, moving from a primarily terrestrial diet to a largely marine protein diet by the end of their occupation in Greenland (Arneborg et al. 1999). Nelson et al. (2012) found that the dietary economy of Norse settlements in Greenland was focused on a combination of domestic livestock and hunting. In 2010, Sveinbjörnsdóttir et al. (2010) isotopically analysed 83 skeletal samples (79 humans, two horses and two dogs) for the dietary reconstruction of early Icelandic settlers and to make comparisons with the study conducted on the Greenland Norse. The sampled individuals all lived between AD 900-1250: those from Christian burials (n=45) showed an average $\delta^{13}C$ value of -19.39 ± 0.46‰ and those from "pagan" burials (n=30) had an average $\delta^{13}C$ value of -18.73 ± 1.05‰. The results showed a dietary variation between people, likely due to geographic residence (i.e., proximity to the sea) and possibly non-local migration and social status (see Sveinbjörnsdóttir et al., 2010). The consumption of fish and seaweed can raise $\delta^{13}C$ values, resulting in a dietary signature that appears to be more marine based (Schulting et al., 2017). The consumption of fish and seaweed in addition to the heavy dietary reliance on seaweed- and fishmeal-eating sheep in historical Iceland (Hallsson, 1964; Sigurðsson, 1988) could potentially alter isotope values (Schulting et al., 2017; Balasse et al., 2019).

Other isotopic studies noted that $\delta^{15}N$ values in human bone collagen samples contain dietary-derived freshwater carbon resulting in a complicated reservoir effect and have described several problems with palaeodietary reconstructions based solely upon $\delta^{13}C$ and $\delta^{15}N$ in Iceland (Ascough et al. 2012, 2014). Thus in 2013, 129 archaeological animal bones from the Settlement period (*landnám*, ca. AD 874-930) site Skútustaðir in north-eastern Iceland were isotopically analysed ($\delta^{13}C$, $\delta^{15}N$ and $\delta^{34}S$) (Sayle et al., 2013). With the addition of sulphur isotope analysis, the researchers were able to differentiate between terrestrial, freshwater- and marine-based diets amongst animals as well as explore aspects of animal husbandry and the trade of livestock (Sayle et al., 2013). Recent isotope research ($\delta^{13}C$, $\delta^{15}N$ and $\delta^{34}S$) on 46 human and 39 animal bone samples showed a wide range of isotopic values, indicating that the individuals living at the Late Viking Age (ca. AD 940-1070) site Hofstaðir í Mývatnsveit were consuming a varied diet, with outliers suggesting migrants to the area (Sayle et al. 2016). Volcanic activity causes fluctuations in sulphur concentrations found in water sources and foliage: one study demonstrated that sulphur concentrations in water in the Hekla region were elevated even 15 years after the last eruption due to magmatic degassing (Holm et al., 2010; Sayle et al., 2013). For the study presented here, it is therefore important to consider the effect that volcanic activity may have had on populations residing close to volcanic systems, such as those living at Skeljastaðir (near Mt. Hekla) and Skriðuklaustur (near the Veiðivötn-Bárðarbunga volcanic system).

4 Materials and Methods

From the seven sites that were included in this analysis osteological and palaeopathological analysis was undertaken on a total of 186 adult (>17 years) individuals. A total of 50 individuals from two sites (Skriðuklaustur and Skeljastaðir) were sampled for stable isotopes of $\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$ and bone element concentrations of Hg, Pb, Cd. F and As. While most of the samples were selected from the adult individuals that were osteologically analysed, six non-adults were also sampled. All the individuals that were analysed for elemental or isotope data underwent osteological analyses, but the results and statistical findings presented in Chapter 5 only include adult individuals (the osteological data of the non-adults that were sampled for elemental and isotope analyses was not included in those tabulations). Trace element analysis was undertaken on dental enamel for Hg, Pb, Cd, As, Sb, Sr, Zn and Ba on a total of 31 of the same individuals. Soil samples (n=22) from Skriðuklaustur and Skeljastaðir were analysed alongside human bone samples to examine natural background levels of various elements in the local environment and to control for concerns over diagenetic enrichment. Animal bones (n=25) were examined for this purpose as well, but also to investigate differences in exposure between livestock, wild animals, and the human population. The animal bone samples were also used to provide isotope baselines for the palaeodietary reconstruction. Details of the archaeological sites and methods (e.g., ion-selective electrode, ISE; isotope ratio mass spectrometry, IRMS; inductively coupled plasma mass spectrometry, ICP-MS; osteological analyses) are included in this chapter.

4.1 Materials: archaeological sites, population background and volcanic history

4.1.1 Sites from the 10th to 12th centuries

From the initial Settlement (late 9th century), the Icelandic economy and subsistence was based upon the import and farming of domestic stock. Foraged foods, hunting (e.g., fish, birds, seals) and likely small-scale cultivation of cereals (e.g., barley) also contributed to the subsistence economy (Harrison and Snæsdóttir, 2012; Riddell et al., 2018). People resided on farms that practiced inter-regional exchange, but the country was entirely devoid of urban centers (McGovern et al., 2006, 2007; Harrison, 2009; Harrison and Snæsdóttir, 2012). During this period, the population began observing Christian regulations and were influenced by Christian habits (Kristjánsdóttir, 2017: 18-26).

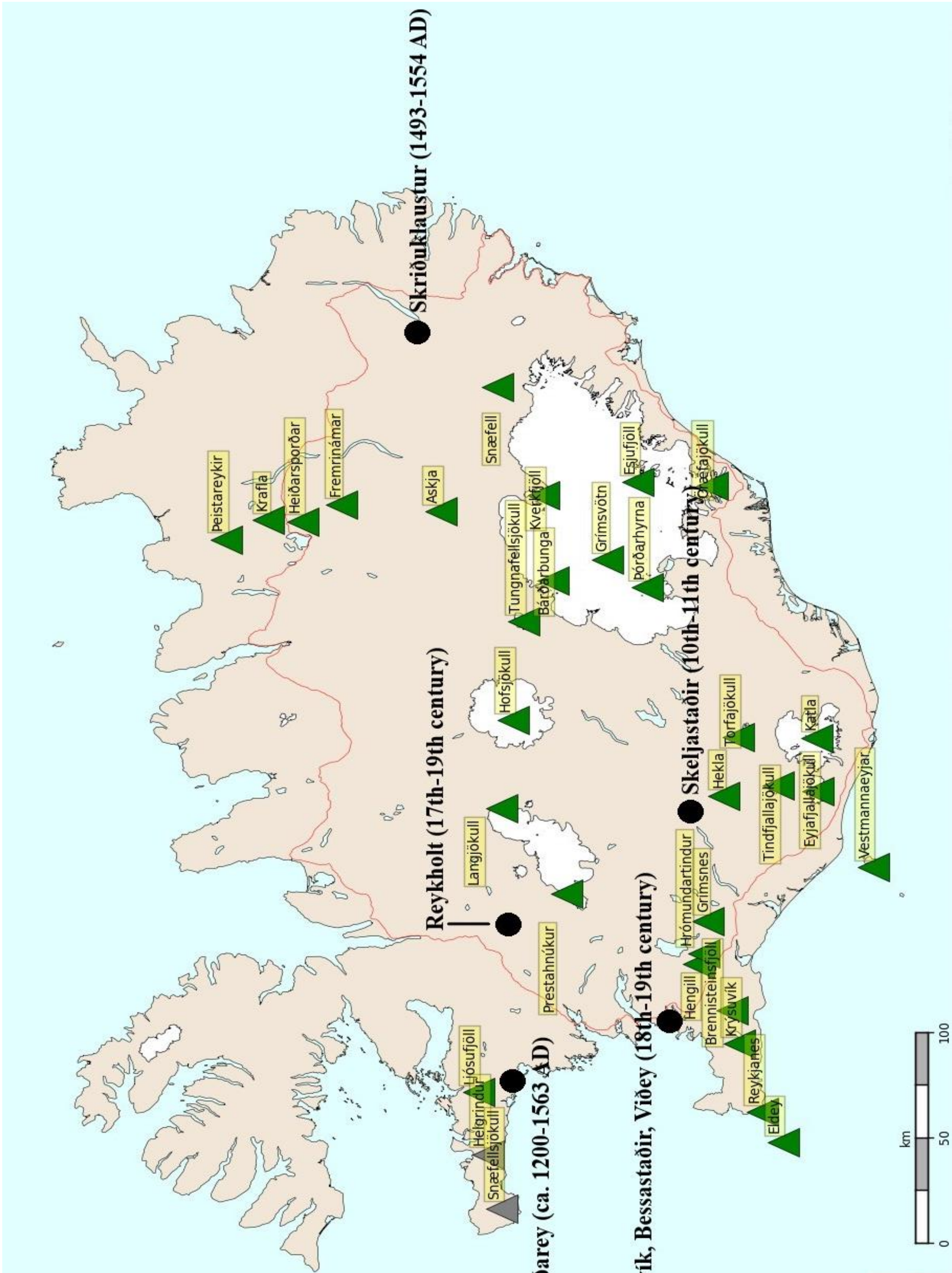

Figure 4.1. Map depicting the volcanoes of Iceland (green triangles) and the archaeological sites (black dots) analysed in this research. The dates correspond to the skeletal assemblages rather than the entirety of the sites use. The map is presented in a modified form from the Volcanic Hazards map issued by the Icelandic Meteorological Office (Veðurstofa Íslands), http://www.vedur.is/skjalftar-og-eldgos/eldgos, where a larger, clearer version of the map and names of volcanoes may be seen

Skeljastaðir (ÞSK). Skeljastaðir is in the Þjórsárdalur valley by Skeljafell in southern Iceland, and the cemetery was used up until the severe AD 1104 eruption of Mt. Hekla, a volcano located nearby (Steffensen, 1943; Þórðarson, 1943; Dugmore et al., 2007). It is believed that the cemetery was used from AD 1000-1104 (Gestsdóttir, 2014). During the excavation (1939) 63 individuals were recovered from the cemetery (Steffensen, 1943; Þórðarson, 1943). Only 56 of these individuals are currently available for study at the National Museum of Iceland due to the commingling of remains following the excavation. Previous isotopic research on 33 individuals from Skeljastaðir demonstrated that only one individual (ÞSK 44) was of non-local origin (Price and Gestsdóttir, 2006). Radiocarbon dating conducted on seven skeletons resulted in a range of AD 890-1220 (68.2% probability) (Sveinbjörnsdóttir et al., 2010), indicating that some of the individuals were possibly buried following the eruption.

4.1.2 Sites from 13th to 16th centuries

By the 13th century, power struggles between the elite families in Iceland were amplified and Iceland fell under Norwegian rule. The zenith of the Catholic church's influence occurred during this period, with noticeable changes in subsistence habits, medical care, manuscript production (from vellum to parchment) and the import of fine metallic church objects, such as chalices (Kristjánsdóttir, 2017). During this period there were still no urban centers, thus international trade occurred seasonally at certain coastal trading posts on an increasingly larger scale due to the favourable weather conditions (Barrett et al., 2004; Mehler, 2007, 2015; Perdikaris and McGovern, 2009; Véstseinsson et al., 2010; Harrison and Snæsdóttir, 2012; Barrett, 2016; Hoffman, 2018). The climatic conditions during the Medieval Warm Period (9th to 13th centuries) also increased the population density of certain fish species, such as cod, which was essential to the diet and herring, which was an important commodity. These favorable conditions further contributed to the increasing human population density and the presence of foreign fishermen and traders in the North Atlantic, as the demand for fish grew exponentially due to religious fasting rules (Barrett et al., 2004; Perdikaris and McGovern, 2009; Barrett, 2016; Hoffman, 2018). During the 14th and 15th centuries, Iceland came under Danish rule and trade with England and Germany significantly increased, particularly in the exchange of refined sulphur, woollen items and fish for various goods, such as alcohol and grain (Mehler, 2007, 2015). Considering the monastic functions of Skriðuklaustur, and its role as a center of commerce, the use of materials such as lead for structures and objects was likely far more common than at fishing or farm sites, such as Skeljastaðir or Haffjarðarey.

Haffjarðarey (HFE). The Catholic church and cemetery on the island Haffjarðarey (HFE) in Haffjörður, off the Snæfellsnes Peninsula, are believed to have been in use from approximately AD 1200-1563 (Þorkelsson, 1888; Steffensen, 1946). The cemetery ceased to be used in 1563, likely because of severe coastal erosion and the Reformation (Steffensen, 1946; Hoffman, 2018). Though none erupted during the time the site was used, the region has three volcanic systems (i.e., Snæfellsjökull, Lýsuskarð, Ljósufjöll) (Harðarson, 1993). Two excavations were carried out, one beginning in 1905 and the other in 1945. A total of 24 individuals were excavated along with the remains of 34 more individuals from disturbed burials. Fifty-four of these individuals are curated in the National Museum today. The people residing at Haffjarðarey likely represented the general population, made up of laborers and farmers (Gestsdóttir, 2004). Strontium isotope analysis conducted on 11 of the excavated individuals showed only local geographic provenance (Price and Gestsdóttir, 2006).

Skriðuklaustur (SKR). Skriðuklaustur (SKR) in Fljótsdalur was an Augustinian monastery (AD 1493-1554) and hospital located inland in eastern Iceland, in the Vatnajökull region. It began operating just 16 years after a severe eruption of the Bárðarbunga-Veiðvötn volcanic system (AD 1477) (Thordarson and Larsen 2007, Kristjánsdóttir 2012), that resulted in a massive tephra fall and place abandonment in the Hrafnkelsdalur valley. It devastated the crops and landscape, as well as the livestock and human populations (Larsen, 1988; Rafnsson, 1990; Thordarson and Larsen, 2007; Global Volcanism Program, 2013). A total of 295 individuals, around one-third of which are nonadults (<17 years), were excavated from the site and around half show notable pathological conditions. The discovery of surgical instruments, healing plants and numerous cases of infectious disease (e.g., treponematosis, tuberculosis, hydatidosis) and other pathological conditions (e.g., Paget's disease, cleft palate, traumatic injuries) demonstrated its role as a monastic hospital (Kristjánsdóttir, 2012; Walser III et al., 2019). According to zooarchaeological, isotopic and historical data, trade, particularly for fish, and small-scale gardening were the basis of their subsistence economy (Pálsdóttir, 2006; Hamilton-Dyer, 2010; Kristjánsdóttir, 2012; Kristjánsdóttir et al., 2014; Mehler, 2015; Walser III et al., 2020a). The monastery and the hospital were closed shortly after the Lutheran Reformation (mid-16th century) in Iceland (Kristjánsdóttir, 2011, 2012, 2015).

4.1.3 Sites from the 17th to 19th centuries

During the Protestant era, printing and textile production became more industrialised (Arnórsdóttir, 2013). Between AD 1602-1787 the Kingdom of Denmark maintained a trade monopoly in Iceland, enacted through designated trading centers throughout the country. Regardless, unauthorized trade amongst fishermen and merchants from England, Germany, Holland and elsewhere persisted (Karlsson, 2000; Þórhallsson and Joensen, 2014). During this period, the Icelandic population was still dispersed across small, rural villages and farms throughout the country, with no towns or urban centers (Harrison and Snæsdóttir, 2012). In the mid-18th century, Reykjavík underwent a rapid transition from a predominately rural to a densely populated urban society over just a few generations, shifting occupational dependency from farming and seasonal fishing to specialized production (Jónsson and Þorsteinsson, 1991; Björnsson, 1998; Harrison and Snæsdóttir, 2012). The importance of trade goods also increased, marking a major shift in occupational roles and the subsistence economy, which were previously centered on food production (Jónsson, 1998).

Reykjavík was an excellent location for fishing and gathering of marine resources, but the area was less suited for farming. Until the end of the 18th century, fishermen operated seasonally, and most were therefore also farmers. The Danish trade monopoly restricted socio-economic development and prevented the necessary technological advancements that later lead to the significant increase in fisheries and the fishing economy. In the last decades of the trade monopoly, the company Innréttingar was established by high-status Icelanders, marking the beginning of urbanization in Reykjavík. The company developed specialist factories or workshops, thereby also forming the first real streets in the city, and focused on improving the fishing, manufacturing, and farming industries in Iceland. The trade monopoly was finally abolished in AD 1787 (Karlsson, 2000; Óskarsson, 2002; Pálsdóttir, 2008; Harrison and Snæsdóttir, 2012). The 19th century saw the increased import of sugar, coffee, and tobacco as well as the replacement of whole rye and barley products with wheat flour, which is much lower in nutritional value. Grains and vegetables began to replace animal proteins and sugar consumption increased from AD 1870. Nonetheless, the increased availability of grain and its decreasing price led to population growth as the society became less dependent upon livestock supply. Despite the increased consumption of grain, nutrition seems to have improved dramatically towards the mid-19th century, except among the poor who suffered from malnutrition and disease (Jónsson, 1998). However, unlike during the late 18th to early 19th centuries in Britain, the Industrial Revolution did not begin in Reykjavík until the 20th century (Karlsson, 2000). None of the volcanic systems in the region of Reykjanes Peninsula (Viðey, Reykjavík or Bessastaðir) erupted during the times that the sites were occupied (17th to 19th centuries) (Figure 4.1) (Global Volcanism Program, 2013). However, the Laki eruption of AD 1783-1784 was devastating and caused a volcanic winter that led to famine and climatic changes, as well as mass mortality to the human (20-25%) and livestock (75%) populations (Jónsson, 1994; Thordarson et al., 1996; Thordarson and Larsen, 2007; Guðmundsson et al., 2008; Halldórsson, 2013).

Reykholt (RKH). The Reykholt farm site, located in Borgafjörður in western Iceland, was occupied soon after the Settlement of Iceland, but a church is not mentioned in the historical records until AD 1185 (DI. I., 279-280; Sveinbjarnardóttir, 2012). Archaeological remains were found at Reykholt during construction work in 1930, followed by numerous research expeditions over the years (Sveinbjarnardóttir and Jónsson, 1998). Since the cemetery is still in use, and the area was not under development, the archaeologists chose to excavate as few graves as possible. Eighteen individuals were excavated from the site, ten of whom are adults and eight non-adults. The burials date to between the mid-16th and the late 19th centuries and include some high-status, known individuals (e.g., Rev. Þorleifur Bjarnason) (Sveinbjarnardóttir, 2016). Prestahnúkur (Langjökull system), the only volcano closeby, last erupted in 3550 BC and thus did not affect the people living at Reykholt (Global Volcanism Program, 2013). However, the Book of Settlements describes that the geothermal pool located at Reykholt was used by an individual that resided there and by the neighboring settlement farm since at least the 11th century (Íslenzk Fornrit I, 1968: 78, 192).

Bessastaðir (BES). Bessastaðir in Álftanes, in the Reykjavík area, is the current presidential residence, but historical records suggest that the area has been used since about AD 1000. It became the residence of the Danish King's highest-ranking officers as early as the 13th century. The Bessastaðir church and cemetery are still in use today. In 1987, a total of 18 individuals dating between the 18th and 19th centuries were excavated from the cemetery due to road development. The burials likely represent a mixture of the local populace and high-status individuals associated with the governor's household (Amarosi et al., 1992; Gestsdóttir 2004; personal correspondence: Guðmundur Ólafsson, Archaeologist, 2019).

Viðey (VEY). On the island of Viðey, located less than 1 km off the coast of Reykjavík in southern Iceland, 36 out of a total of 91 burials were excavated between 1987-1988 (Hallgrímsdóttir, 1989, 1993). Since before the monastic period a church and cemetery were located there. Although an Augustinian monastery operated there from AD 1226-1539, the excavated burials rather date to the 18th and 19th centuries as they are associated with the church constructed in AD 1774 (Kristjánsdóttir, 1995a, 1995b, 1996). Hallgrímsdóttir, 1989, 1991, 1993; Gestsdóttir, 2012). After, a farm and leprosy hospital were established. During the time that Skúli Magnússon, an 18th century representative of the Danish King in Iceland, lived on Viðey, the island hosted various establishments (e.g., a printing office). Analyses of faunal and pollen remains indicate that the individuals were of a high social status (Amarosi, 1996; Riddell et al., 2018). The island of Viðey itself has no volcanic or geothermal activity, but the area is subject to the previously described Reykjanes peninsula volcanic systems.

Reykjavík (RVK). In Reykjavík, the first church charter dates to AD 1397, however an earlier church is mentioned by the bishop Páll Jónsson from Skálholt in AD 1200. The cemetery in Reykjavík was used until 1838 when it was replaced by the nearby, modern cemetery known as Hólavallagarður (Óla, 1963; Gestsdóttir, 2009). The site was first excavated in 1940 and again in 1967 (Gestsdóttir, 2012). The skeletal assemblage contains numerous disarticulated remains in addition to 17 articulated individuals (Gestsdóttir, 2009). Although another 38 individuals were excavated from the site in 2016 (Zoëga, 2018), they were not available for research until recently and are therefore not included in this study. The individuals included in the Reykjavík skeletal assemblage likely represented members of the general public and have been dated to the 18th and 19th centuries based upon artefact identifications (e.g., buttons) (Gestsdóttir, 2012). No volcanic systems are found in the greater Reykjavík area, but the area is subject to the previously described Reykjanes peninsula volcanic systems.

4.2 Methods: skeletal analysis

4.2.1 Anthropological descriptions and osteological analyses

The osteological analysis was conducted on a total of 186 adult skeletal individuals from the previously described archaeological sites, dating between the 10th-19th centuries (see Figure 4.1, Table 4.1, and Section 4.1). Though six non-adults were included in the elemental bone analyses, they were not included in the osteological results.

Table 4.1 Table indicating the numbers of individuals analysed osteologically from each site

Osteological analysis						
Period	**Site**	**n**	**Male**	**Female**	**YA**	**OA**
10th - 12th centuries	Skeljastaðir	50	24	26	20	30
Total	10th - 12th centuries	50	24	26	20	30
13th - 16th centuries	Haffjarðarey	16	7	9	6	10
	Skriðuklaustur	66	24	42	45	21
Total	13th - 16th centuries	82	31	51	51	31
17th - 19th centuries	Reykjavík	13	7	6	6	7
	Viðey	21	12	9	11	10
	Bessastaðir	10	5	5	4	6
	Reykholt	10	7	3	6	4
Total	17th - 19th centuries	54	31	23	27	27
Sum Total	10th - 19th centuries	186	86	100	98	88

The aim of the osteological analysis was predominately to investigate overall health and the possible ante-mortem exposure to fluoride, which is known to cause characteristic, but diffuse skeletal changes. While the other elements measured in bone in this study, including lead (Pb), cadmium (Cd), arsenic (As) and mercury (Hg), all interact with the skeleton, and none of them regularly cause bone changes that can be directly correlated with toxic exposure. Standard anthropological methods as outlined by Buikstra and Ubelaker (1994) and Brickley and McKinley (2004) were used to estimate age, sex, stature, and pathological conditions. Due to the multitude of skeletal markers necessary for an accurate investigation into skeletal fluorosis and other conditions relevant to this research, only adult individuals with more than 50% completeness and good preservation (grade 0-1; see Brickley and McKinley, 2004, Chapter 5, Section 5.3) were analysed from the seven sites used in the present study. Sex determination was performed using metrical estimations, cranial morphology and os coxae morphology (Buikstra and Ubelaker, 1994; Brickley and McKinley, 2004, Chapter 8) and distal humerus morphology (Rogers et al., 1999; Falys et al., 2005; Vance et al., 2011). Age was estimated according to phase changes on the morphology of the auricular surface (Lovejoy et al., 1985) and the pubic symphysis (Brooks and Suchey, 1990), epiphyseal fusion (Buikstra and Ubelaker, 1994; Scheuer and Black, 2000) and dental eruption and development (AlQahtani, 2010). Secondary methods used to estimate age include degree of dental attrition (Brothwell, 1981), cranial suture closure (İşcan et al., 1984, 1985; Meindl and Lovejoy, 1985) and acetabular degeneration (Calce, 2012). Age categories were limited to younger adult (YA; 17-36 years) and older adult (OA; 36+ years) because dental attrition-based age estimation methods appear to overestimate the age of skeletal individuals from Iceland. These overestimations are caused by the high dental wear rate and acid erosion occurring from the consumption of certain acidic dietary staples (see Lanigan and Bartlett, 2013; Richter and Eliasson, 2017). Differential diagnoses for pathological conditions were considered according to the descriptions in reference material by Ortner (2003), Roberts and Manchester (2010) and Aufderheide and Rodriguez-Martin (2011) in addition to recent journal articles (see methods of Articles I, II, III and supplementary information of Article III). Osteological data was first documented on skeletal recording sheets prior to registration in a bespoke electronic database (*Kupa*, National Museum of Iceland). Skeletal conditions were presented as crude prevalence rates (number of individuals with a condition/number of observable individuals) and dental conditions were presented as true prevalence rates (number of teeth or alveolar positions with conditions/number of observable teeth or alveolar positions). T-tests were performed using Excel and chi-squared tests were conducted using *PAST (PAleontological STatistics)* software, with p-values of ≤ 0.05 being considered statistically significant.

4.2.2 Diagnosing heavy metal toxicity (Hg, Pb, Cd, As)

The identification of heavy metal toxicity from skeletal remains is highly dependent upon measurements conducted on skeletal and dental tissues. Bone changes associated with heavy metal toxicity are not pathognomonic and predominately appear as osteopenia of trabecular bone and increased fracture risk, with only a few exceptions (see Table 4.2; Rodríguez and Mandalunis, 2018). For example, cadmium can cause yellow staining on dental enamel, while mercury can cause grey staining in addition to enamel mottling and malformation if exposure occurs during childhood (Mortazavi et al., 2014). However, these bone or dental changes cannot be systematically evaluated as evidence of toxic exposure and may only be described as potentially correlated if bone element concentrations exceed normal background levels.

Mercury exposure does not normally cause diagnostically useful skeletal changes in adults (Tucker, 2007; Rasmussen et al., 2013; Ávila et al., 2014). However, in children, treatment with mercury can cause enamel abnormalities, which are different from those caused by congenital syphilis (Ioannou et al., 2016). While dental changes are known to occur within a wide percentage (ca. 10%-65%) of individuals affected by congenital syphilis (Ioannou et al., 2016), the percentage of dental changes resulting from mercury exposure in children appears to be entirely unknown. Mercury alters calcium homeostasis, which can result in hypercalcemia, and directly alters the function of bone cells (Suzuki et al., 2004). Therefore, mercury exposure can only be macroscopically assessed to some degree in non-adult skeletal remains, while adult skeletal remains must be sampled for elemental analysis because of the lack of bone changes diagnostic or indicative of mercury toxicity. Some studies draw correlations between mercury exposure and osteopenia and osteoporosis, but exposure to other toxic elements (e.g., Pb, Cd, F, As) may also be indicated. These conditions can also result from numerous other etiologies and are therefore not useful for estimating exposure to toxic elements. Some of the skeletal changes associated with fluorosis can also result from heavy metal poisoning (see Tables 4.2 and 4.3; Whyte et al., 2008). For example, cadmium toxicity can result in osteomalacia, osteoporosis and increased fracture rate (Lanocha-Arendarczyk et al., 2015a), while lead can increase the severity of fluorosis (Leite et al., 2011). Common volcanic pollutants that can result in human toxicity were therefore analysed in this research.

Table 4.2 Table showing the primary in vivo effects of metals on bone analysed in this study on bone, adapted from Rodríguez and Mandalunis (2018)

***In vivo* effects of metal on bone**	**Stimulation (increase)**	**Inhibition (decrease)**	**Alteration (either)**
Bone Formation	Arsenic (As), Lead (Pb)	Cadmium (Cd)	-
Osteoblast Differentiation	-	Cadmium (Cd)	-
Bone Resorption	Cadmium (Cd), Lead (Pb)	-	Arsenic (As)
Osteoclast Differetiation	-	Arsenic (As)	-
Mineralisation	-	Cadmium (Cd)	Lead (Pb)
Endochondral Ossification	-	Arsenic (As), Lead (Pb)	-

4.2.3 Diagnosing fluoride (F) toxicity and skeletal fluorosis

Due to the cumulative and progressive nature of skeletal fluorosis, a large range of skeletal changes must be considered in combination with measured fluoride concentrations in bone (see Table 4.5; Littleton, 1999; Petrone et al., 2013; Nelson et al., 2016). Digital radiography was used to distinguish fluorosis from other conditions causing similar bone changes (e.g., Paget's disease). Light microscopy was used to analyse enamel defects (e.g., mottling, hypomineralisation, chipping) as well as enhance the observation of skeletal changes when necessary. Indicators of dental and skeletal fluorosis were recorded following clinical and palaeopathological descriptions (e.g., Dean, 1936; Littleton and Frohlich, 1993; Littleton, 1999; Hillson, 2008; Petrone et al., 2013, 2016; Nelson et al., 2016). Each bone was assessed for changes potentially related to skeletal fluorosis and scored according to presence or absence, ossification of soft tissues (e.g. glands, ligamentum flavum, atlanto-occipital membrane) (Steinbock, 1989; Binder et al., 2016; Geber and

Hammer, 2018), calcification of soft tissue attachment sites (Mariotti et al., 2007; Petrone et al., 2013), ankyloses (Rogers et al., 1985; Petrone et al., 2013; Ventades et al., 2018), vitamin D deficiency (Brickley et al., 2005, 2010; Ives and Brickley 2014) and enamel defects (Dean, 1936; Hillson, 2008). Detailed descriptions of the methodologies used in this study to assess skeletal fluorosis are included in the Supplementary materials of Article III.

Table 4.3 Skeletal changes associated with fluorosis investigated in this study. Additional skeletal changes and differential diagnoses can be seen in Table 4.4

Skeletal change	*Description*
Osteomalacia and residual rickets	Morphological deformities, fractures, others
Ossification of ligamentum flavum	Cranial or caudal ossifications on vertebral arch attachments
Atlanto-occipital membrane	Increased rugosity or mineralisation
Entheseal & interosseous calcification	Increased rugosity or mineralisation
Ankyloses	Joints or bones otherwise fused to one another
Fractures	Healed or unhealed perimortem or antemortem fractures
Enamel defects	Linear enamel hypoplasia; hypomineralisation (opacities); mottling; chipping
Calculus	Presence of mineralised plaque formation on dentition
Caries	Carious lesions on any aspect of tooth roots or crowns
Other ossifications	Ossification or calcification of glands (e.g. thyroid), cartilage or organs

The bone and dental changes observed in sheep (*gaddur)* following the Hekla eruption of 1970 appear to have inspired the diagnosis of skeletal fluorosis in an archaic human (*Homo erectus*) from present-day Java, Indonesia that was found in geological strata composed primarily of volcanic ash. The research suggested that the individual acquired skeletal fluorosis due to the consumption of plants or fruits that had been contaminated with volcanogenic fluoride. The diagnosis was based upon an analogous soft tissue ossification noted in the archaic human and an autopsy patient with skeletal fluorosis who died of hepatic cirrhosis from the daily consumption of fluoridated table wine (Soriano, 1970). Since then, this bone formation has been re-diagnosed multiple times as evidence of anything from an injury or myositis ossificans to an infection (Ruff et al., 2015). This case illustrates some of the complications associated with the retrospective diagnosis of skeletal lesions, perhaps fluorosis especially, as bone displays limited variation in reactive changes to a wide range of substantially different pathological conditions. Some other limitations in retrospective diagnosis include the “Osteological Paradox” (see Wood et al., 1992), inadequate translations of historical texts, the presence of multiple or concurrent conditions, the disappearance of some disease and the emergence of others (Mitchell, 2011).

Skeletal fluorosis is an important differential diagnosis for Paget’s disease of bone (see section 2.2.4), of which one case is included in this study. Some modern literary analyses have attempted to identify what condition may have caused the reportedly striking appearance of the poet-Viking Egill Skallagrimsson of Egil’s saga. Egill has been described as ugly, with misshapen bones, and as having the characteristic Pagetic whitening of the cranium with a soft, pumice-like outer table when he was reportedly struck with an axe. Some scholars suggest that the description of Egil’s condition may be attributable to Paget’s disease, thereby retrospectively diagnosing him with this condition (Byock, 1993). However, the skeletal changes could also be attributable to skeletal fluorosis (Weinstein, 2005) as well as other conditions (e.g., thyroid disorders, osteopetrosis, myositis ossifcans) and – as previously discussed – retrospective diagnosis based upon historical or literary descriptions is problematic and often sensationalized (see Mitchell, 2011).

Fluorosis can cause different forms of osteoarthritis including seronegative and inflammatory arthritis, for example (Shukla, 2016). Its symptoms and bone changes often resemble osteoarthritis, particularly in early stages. Partly due to this resemblance, it is often misdiagnosed (Connett, 2012; Namkaew and Wiwatanadate, 2012; Petrone et al., 2013). Similarly, osteoarthritis itself can actually be induced by chronic fluorosis. Osteoarthritis occurs prior to detectable osteosclerosis of the spine, thereby rendering early diagnostic differentiation difficult when based upon skeletal changes alone. For example, in a study conducted in China it was demonstrated that osteoarthritis and fracture risk was significantly higher in one known fluorosis region than in its control group or even in the rest of the country (Liang et al., 1997; EFSA, 2010). Another study conducted on individuals from an area of endemic fluorosis in Turkey found that 66.1% of individuals with fluorosis experienced symptoms of knee pain primarily due to osteophyte formation. Despite a small sample size (n=96: 56 patients and 40 age- and sex-matched controls), the study suggests that the severity of knee osteoarthritis increases with endemic fluorosis. It also found infrequent radiological evidence of the hallmark skeletal changes associated with fluorosis, such as osteosclerosis and soft tissue calcifications (Savas et al., 2001). These results indicate that regardless of toxicity, skeletal manifestations of fluorosis are differential, not always observable, and likely underrepresented diagnostically. Common differential diagnoses and skeletal changes associated with skeletal fluorosis are shown in Table 4.4.

Table 4.4 Skeletal and dental changes associated with skeletal fluorosis. Compiled from the criteria described by Dean (1936), Shupe et al. (1963), Den Besten (1999a, 1999b), Littleton (1999), Pendrys et al. (1999), Savas et al. (2001), Brown et al. (2005), Ayoob and Gupta (2006), Yoshimura et al. (2006), Hillson (2008), Whyte et al. (2008), Alvarez et al. (2009), EFSA (2010), Blinkhorn and Mekertichian (2013), Petrone et al. (2013), Faccia et al. (2015), Nelson et al. (2016). Differential diagnoses as described by Littleton (1999), Yoshimura et al. (2006), Whyte et al. (2008), Faccia et al. (2015) and Nelson et al. (2016). Common causes of increased bone mass as described by Whyte et al. (2008)

Skeletal and dental changes associated with skeletal fluorosis		
Bone formation or changes	*Vertebral changes*	*Dental changes*
dense periosteal deposition	widened vertebral appearance	discoloration
extensive new bone production	disc space narrowing	enamel pitting
joint disease or ostoearthritis	osteophytic vertebral fusion	mottling
osteosclerosis	ossification of spinal ligaments	hypoplasia
periosteal hyperostosis	thoracic kyphosis	brown staining
hypertrophic bony exostoses	degenerative joint disease	white opacities
Ossification or calcification	*Microstructure*	*Other*
f. magnum ligaments	thickened cranial diploe	increased fracture rate
tendons	osteopenia	osteomalacia
ligaments	osteoporosis	diaphyseal widening
interosseous membranes	osteophytosis	genu varum
costo-vertebral & -sternal joints	coarse trabecular pattern	genu valgum
intercostal calcification	intermittent growth lines	flexion deformations
Differential diagnoses and other common causes of increased bone mass		
Differential diagnoses	*Common causes of increased bone mass*	
DISH	Craniodyaphyseal dysplasia	Lymphoma
Ankylosing spondylitis	Craniometaphyseal dysplasia	Hypervitaminosis A
Hematogenous osteomyelitis	Endosteal hyperostosis	Hypervitaminosis D
Hyper- or hypoparathyroidism	Melorheostosis	Renal osteodystrophy
Paget's disease	Myelofibrosis	Fibrogenesis imperfecta ossium
Myositis ossificans	Sarcoidosis	Skeletal metastases
Osteopetrosis	Heavy metal toxicty	Engelmann disease

Diagenetic and taphonomic factors must always be considered when conducting elemental analysis on archaeological bones for the purpose of diagnosis because skeletal remains tend to accumulate certain elements from the burial environment, particularly in areas that are rich in organic matter and moisture (Krajcarz, 2017). Due to diagenetic processes, skeletal remains may exhibit elevated concentrations from elements in the soil rather than from antemortem bioaccumulation (Hedges, 2002; Nelson et al., 2016; Krajcarz, 2017). Fluoride has a natural affinity for water, thus environmental fluoride concentrations are usually elevated in regions with significant groundwater water movement and saturation (King et al., 2011). It is therefore important to measure the elemental concentrations of soil from the burial environment as well (Littleton, 1999; Petrone et al., 2013; Nelson et al., 2016). Unfortunately, soil concentrations may not always reflect the exact concentration of an element present at the temporal period under investigation due to its persistent exposure to rainwater and groundwater over the years (EFSA, 2010). Generally, the range for fluoride concentrations in soil is between 30 and 500 ppm (Edmunds and Smedley, 2005; Ozvath, 2008). Volcanic eruptions can substantially elevate this concentration (Pyle and Mather, 2009), however, it normalizes over time as it rinses out of soil, ash, and flora. For example, volcanic ash from the Hekla eruption of 1970 presented with fluoride concentrations up to 2000 ppm and some vegetation measured up to 4000 ppm (Thorarinsson and Sigvaldason, 1972). Such high values only occur due to extreme conditions and are almost always attributable to volcanic activity (D'Alessandro, 2006). Human bones generally exhibit fluoride concentrations ranging from 300-7000 ppm according to overall lifetime exposure. Individuals exposed to large amounts of fluoride during life tend to exhibit bone fluoride concentrations that are 2 to 3 times higher than normal (Aras and Ataman, 2006).

4.3 Methods: elemental analyses of cortical bone samples

Elemental analyses (ICP-MS and ISE) were used to investigate the concentrations of mercury, lead, cadmium, arsenic and fluorine in bone samples. Bone samples (n=50) were selected from Skeljastaðir (10^{th}-12^{th} centuries) and Skriðuklaustur (15^{th}-16^{th} centuries). Samples were not selected from the 17^{th}-19^{th} centuries sites due to funding limitations as well as difficulties with controlling industry-related variables that notably increased during this period. For example, mercury was used in the production of hats and other textiles, tobacco use increased thereby increasing cadmium exposure and lead or lead-glazed goods became widely available. Also, a pilot study examining fluoride exposure in 17^{th}-19^{th} centuries skeletal remains was conducted by Gestsdóttir et al. (2006), thereby providing some comparative material.

Mercury concentations in cortical bone were considered elevated if they exceeded 0.3 ppm (see section 2.3.1). While osteofluorosis may occur at relatively low concentrations in bone, this research defines concentrations greater than 3500 ppm (pre-clinical phase) in cortical bone as the cutoff for elevated fluoride concentrations that might indicate skeletal fluorosis (see section 2.3.2; see Franke et al., 1975; Smith and Hodge, 1979; USDHHS, 1991). Lead concentrations in bone were considered elevated if they exceed 7 ppm even though trabecular bone was not used in this study (see section 2.3.3). Cadmium concentrations in bone were only considered elevated if they exceeded 1 ppm in this study (see section 2.3.4). Lastly, arsenic concentrations below 1 ppm are considered normal in this study (see section 2.3.5).

4.3.1 Inductively coupled plasma mass spectrometry (ICP-MS) analysis

Inductively coupled plasma mass spectrometry was used to assess elemental concentrations, including mercury, lead, cadmium and arsenic, in bone samples. A total of 50 rib samples were selected according to preservation, pathology, sex, age, and completeness (>50%) following the methods described above (section 4.2.1). Cortical bone samples were selected primarily from non-pathologically altered ribs. In a few cases, the samples were selected from long or cranial bones due to the absence of suitable rib samples. Ribs were selected for conservation and ethical reasons, as they are often fragmentary, do not contain much trabecular bone – which was discarded in this study – and could mostly be selected without further destruction. Thirty-six samples in total were selected from individuals buried in the cemetery at Skriðuklaustur. Five of these samples were selected from individuals with no bone changes related to infectious disease. From the Skeljastaðir cemetery, 14 rib samples were selected, but only six of them displayed skeletal markers suggestive of infectious disease.

The method of sample preparation was adapted from Skytte and Rasmussen (2013). The human bone samples were cut from complete, well-preserved ribs. They were photographed, cleaned, and then placed in sterile, labeled containers. A dental bur was used to abrade the cortical surfaces and then the samples were cleaned with a synthetic brush and ultrapure water. The trabecular bone was removed mechanically using a scalpel. The samples of cortical bone were each pulverized with a basic analytical mill. Only cortical bone from the rib samples was used because it is much less susceptible to post-mortem contamination from the burial environment than trabecular bone (see Rasmussen et al., 2015). Rib samples provide average values that are skewed towards the end of an individual's life because the bone turnover rate is faster for ribs than most other bones (Fahy et al., 2017). High metal concentrations in rib samples can indicate a period of exposure within a few years prior to death. Previous research by Rasmussen et al. (2013b), found that intra-skeletal differences in cortical bone metal concentrations are marginal, while significant differences were noted in trabecular bone. The study also showed that concentrations are higher in trabecular bone found in the thoracic cavity, probably because of close proximity to the organs (i.e. kidneys, lungs, liver) that absorb and retain the majority of ingested or inhaled heavy metals found in the body.

Elemental concentrations in the bone and soil samples were determined by ICP-MS (inductively coupled plasma mass spectrometry) after mineralisation with closed vessel acid digestion. Portions (up to 200 mg weighed to 0.1 mg) of pulverised samples together with 3 ml HNO_3 (nitric acid) were transferred to 50 ml digestion vessels. They were then digested in a Milestone Ultrawave Acid Digestion System (Milestone Inc.), according to method SV-25-02-SN in the Matís Quality Manual. The digested sample solutions were quantitatively transferred to 50 ml polypropylene tubes and diluted to 30 ml with Milli-Q water. The mercury concentrations in these digests were determined by ICP-MS (Agilent 7500ce, Waldbronn, Germany). ^{115}Indium was used as an internal standard. A detailed description of the analyses of inorganic contaminants is presented in method SV-22-02-SN-1 in Matís Quality manual. Certified reference materials are routinely treated and analysed in the same manner as the samples to assure the quality of metal analysis. All samples, standard and wash solutions contain 200 ppb Au, which reduces the memory effect of Hg (see Thermo Electron Corp., 2003). All samples were run in triplicates and all blanks were carefully monitored.

To control for diagenetic factors and evaluate environmental baselines for mercury, lead, cadmium and arsenic, animal bones and soil samples were also analysed. The samples were primarily selected from ribs and from long bone fragments when ribs were not available. Animal bones (n=23) from Skriðuklaustur, representing dog and fox (*Canidae* sp.), cattle (*Bos taurus)*, fish, seals (*Phocidae* sp.), sheep (*Ovis aries)* or goat (*Capra hircus)*, horses (*Equus* sp.), and swan *(Cygnus sp.)* were measured. Soil samples (n=14) from outside the site and from several locations around it were also measured. Soil samples (n=9) from within the cemetery at Skeljastaðir were analysed, but no animal bones were preserved or available for study.

4.3.2 Ion-selective electrode (ISE) analysis

Ion-selective electrode (ISE) was used to assess fluorine bone concentrations. A total of 50 well-preserved bone elements were sampled for fluoride analysis: 36 from Skriðuklaustur (n=295 individuals) and 14 from Skeljastaðir (n=56 individuals). All the samples were taken from ribs except six, two of which were from long bones, one from a temporal bone, one from an os coxa, and two from parietal bones. First, the samples were cleaned with a synthetic brush and distilled water. The trabecular bone was mechanically removed with a scalpel, and to remove surface contamination the cortical bone surfaces were abraded using a dental bur. Trabecular bone was discarded as it is far more susceptible to post-mortem diagenesis than cortical bone (see Rasmussen et al., 2015). Preservation, age, sex, and pathological markers informed sample selection. Soil samples were collected from the Skeljastaðir (n=2) and Skriðuklaustur (n=2) cemeteries.

Fifty archaeological human bone samples and four soil samples were analysed for cumulative bone fluoride concentrations using ion-selective electrode (ISE). Prior to analysis, NaOh (9 grams) was melted at 500ºC in a nickel-crucible and then cooled down to room temperature. Portions (1 gram) of ground sample material, blanks (same procedure without sample) and control samples (containing 10 mg CaF2 instead of a bone sample) were digested at 500ºC for 60 minutes. After cooling, they were dissolved in 100 ml of water. Aliquots of the samples were neutralized and TISAB-solution (NaCl, Titriplex and acetic acid in water, pH 5.5) was added. The samples, blanks and controls were then measured with ISE for fluoride. The ISE device is calibrated daily to ensure the accuracy of the instrument.

4.4 Methods: isotope analyses of bone collagen and dental enamel

For the purpose of this research, bone samples from 50 humans (36 from Skriðuklaustur and 14 from Skeljastaðir) and 25 animals (from Skriðuklaustur) underwent isotope analysis for carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$) and sulphur ($\delta^{34}S$). The sample selection was informed by the overall skeletal completeness, state of preservation, sex, age and pathologies. The samples were taken from skeletal elements that did not have pathological bone changes. The human bone samples were primarily selected from ribs, which provide a dietary record of the last few years of life. A previous study performed dietary isotope analysis of 13 individuals from Skeljastaðir (Sveinbjörnsdóttir et al., 2010). Three of these 13 individuals were re-analysed here to control for inter-laboratory differences. No animal bones were available for sampling from Skeljastaðir. From Skriðuklaustur, bone samples representing several animal species were selected, including *Cygnus cygnus* (swan), *Capra hircus* sp. or *Ovis aries* sp. (sheep or goat), *Bos taurus* sp. (cattle), *Equus* sp. (horse), *Phocidae* sp. (seal), *Canidae* sp. (dog or fox), and marine fish, considering their differences in dietary resources and animal-human interactions. A modified Long method was used for bone collagen extraction (Longin, 1971; O'Connell & Hedges, 1999) and the $\delta^{13}C$ and $\delta^{15}N$ stable isotope analyses were conducted using a Costech Elemental Analyser (ECS 4010) connected to a Thermo Delta V Advantage isotope ratio mass spectrometer. Finally, $\delta^{34}S$ stable isotope analysis was performed with a Costech Elemental Analyser (ECS 4010) attached to a Thermo Scientific Delta V Plus isotope ratio mass spectrometer (see Article II and Supplementary Materials, Article II).

Dental enamel samples from 31 of the same individuals sampled from Skriðuklaustur also underwent isotope analysis for oxygen ($\delta^{18}O$), strontium ($^{87}Sr/^{86}Sr$), and trace elements, including lead (Pb), zinc (Zn), strontium (Sr), and barium (Ba). The enamel samples were primarily selected from premolars, the enamel of which mineralises within about three years between the ages of 2.5 to 8.5 years (AlQahtani et al. 2010). Only the 3rd molars were available for sampling in three individuals, the enamel of which mineralises within about four years between 7.5 to 16.6 years of age (AlQahtani et al., 2010). Dental analysis from Skeljastaðir was not performed here because results from a previous study by Price and Gestsdóttir (2006) were available.

All isotope and trace element analyses were performed in the Department of Earth Sciences, Durham University. Oxygen ($\delta^{18}O$) and Carbon ($\delta^{13}C$) isotope ratios were measured in the carbonate (CO_3) component of tooth enamel by Thermo Fisher Scientific MAT 253 gas source mass spectrometer for isotope analysis. The $^{87}Sr/^{86}Sr$ ratios were acquired using a Neptune Multi-Collector Inductively Coupled Plasma Mass Spectrometer (MC-ICP-MS). Finally, enamel samples were analysed for Sr, Ba, Zn and Pb by ICP-MS (Thermo Scientific XSeries2) and the final enamel concentrations were then determined according to sample weights and total dilution volumes. Detailed analytical methods are provided below.

4.4.1 Bone collagen carbon and nitrogen isotope analysis

Bone collagen was extracted using the modified Longin method (see Brown et al., 1988). Total organic carbon, total nitrogen content and stable isotope analysis of the samples were performed using a Costech

Elemental Analyser (ECS 4010) connected to a Thermo Finnigan Delta V Advantage isotope ratio mass spectrometer. Carbon isotope ratios were corrected for ^{17}O contribution and reported in standard delta (δ) notation in per mil (‰) relative to Vienna Pee Dee Belemnite (VPDB). Isotopic accuracy was monitored through routine analyses of in-house standards, which were stringently calibrated against international standards (e.g., USGS 40, USGS 24, IAEA 600, IAEA N1, IAEA N2): this provided a linear range in $\delta^{13}C$ between –46 ‰ and +3 ‰ and in $\delta^{15}N$ between –4.5 ‰ and +20.4 ‰. Analytical uncertainty in carbon and nitrogen isotope analysis was typically ±0.1 ‰ for replicate analyses of the international standards and typically <0.2 ‰ on replicate sample analysis. Total organic carbon and nitrogen data was obtained as part of the isotopic analysis using an internal standard (Glutamic Acid, 40.82 % C, 9.52 % N).

4.4.2 Bone collagen sulphur isotope analysis

Sulphur isotopic analysis of collagen samples were performed using a Costech Elemental Analyser (ECS 4010) connected to a Thermo Scientific Delta V Plus isotope ratio mass spectrometer. Collagen was weighed out into 10x10mm tin capsules (between 4 and 6 mg) and approximately the same weight of vanadium pentoxide (V_2O_5) was added to aid in the combustion process to release sulphur. Isotopic accuracy was monitored using the following international sulphur standards: IAEA-S-2, IAEA-S-3, IAEA-S-4, IAEA-SO-5, and NBS 127. Analytical uncertainty in sulphur isotope analysis was typically <0.2 ‰ for replicate analyses of the international standards. Total sulphur was obtained as part of the isotopic analysis using the international standards listed above.

4.4.3 Enamel carbon and oxygen isotope analysis

Carbon ($\delta^{13}C$) and oxygen ($\delta^{18}O$) isotope ratios were measured in the carbonate (CO_3) component of tooth enamel. For each tooth, approximately 2mg of powdered sample was weighed and transferred into an individual exetainer vial. Vials were flushed with helium (grade 4.5) then CO_2 was liberated by reaction with 99% ortho-phosphoric acid for 2 hours at 70ºC. The resultant gas mix of helium and CO_2 was transferred through a Thermo Fisher Scientific Gasbench II in which a gas chromatographic column separated the CO_2 from the gas mixture then passed into a Thermo Fisher Scientific MAT 253 gas source mass spectrometer for isotopic analysis.

The following international reference materials were analysed with each batch of samples: NBS 18 (calcite, *n*=3), IAEA-CO-1 (marble, *n*=3) and LSVEC (Lithium Carbonate, *n*=3). In addition, two internal standards: DCS01 (calcium carbonate, *n*=6) and Dobbins (horse tooth, *n*=2) were also analysed. Repeated analysis of both international and internal standards yielded an analytical precision of 0.20‰ (2 s.d.) for $\delta^{13}C$ and 0.24‰ (2 s.d.) for $\delta^{18}O$. Duplicate analyses of the same sample reproduced within or better than 0.06‰ and corrections were made using IAEA-CO-1 and LSVEC, with all $\delta^{13}C$ and $\delta^{18}O$ values reported relative to the Vienna PeeDee Belemnite (VPDB) standard. $\delta^{18}O$ was additionally reported relative to the Vienna Standard Mean Ocean Water (VSMOW) standard for comparison purposes.

4.4.4 Enamel strontium isotope analysis

The enamel samples were collected following the procedure given in Montgomery (2002). For each molar, a single chip of enamel weighing approximately 20mg was collected using a diamond-tipped rotary dental saw. All surfaces of the enamel samples were cleaned and polished with a diamond-tipped dental burr to a depth of >100 μm to remove traces of contaminants such as soil and dentine. Cleaned enamel samples were analysed in the Arthur Holmes Isotope Geology laboratory, Department of Earth Sciences, Durham University using column chemistry methods outlined in Font et al. (2008). Samples were first dissolved in 3M HNO_3 and heated overnight on a hot plate. The samples were loaded onto cleansed and preconditioned columns containing 60μl of strontium-specific resin. 2x250 μl 3M HNO_3 was passed through to elute the waste, then 2x200 μl MQ H_2O was passed through to elute the strontium, which was collected. Seventeen μl of ca. 15.5M HNO3 was added to the Sr fraction to make the solution 3% HNO_3. Following preparation, the size of the ^{86}Sr beam was tested for each sample to assess the strontium concentrations. From this analysis, a dilution factor could be calculated for each sample and each was diluted to yield a beam size of approximately 20V ^{88}Sr, where possible, to match the beam size of the isotope reference material, NBS987. The strontium samples were analysed by Multi-Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICP-MS) using a Neptune MC-ICP-MS. Samples were introduced into this using an ESI PFA50 nebuliser and a glass expansion cinnabar micro-cyclonic spray chamber. Instrumental mass bias was corrected for using an $^{88}Sr/^{86}Sr$ ratio of 8.3752 (the reciprocal of the accepted $^{86}Sr/^{88}Sr$ ratio of 0.1194) and an exponential mass fractionation law. Corrections were also applied for Kr (krypton) interferences on ^{84}Sr and ^{86}Sr, derived from Ar (argon) gas supply, and the Rb (rubidium) interference on ^{87}Sr, derived from the sample, by monitoring masses ^{82}Kr, ^{83}Kr and ^{85}Rb respectively. The average ^{83}Kr intensity throughout the analytical session was ca. 0.22mV, which is insignificant considering the Sr beam size (^{88}Sr between 4.6 and 27V, average of 14.8V). The average ^{85}Rb intensity was slightly greater at ca. 0.61mV (range: 0.20-1.1 mV) but again, given the range in Sr beam size, the Rb correction on the $^{87}Sr/^{86}Sr$, was very small (<0.00001) and is accurate at that magnitude. The samples were analysed in two analytical sessions during which the average $^{87}Sr/^{86}Sr$ value and reproducibility for the international isotope reference material NBS987 was 0.710258±0.000013 (2SD; n=12) and 0.710266±0.000009 (2SD; n=6). Data are renormalized to an accepted value for NBS 987 of 0.71024. Total procedural Sr blanks run during the period of study were <100 pg, which is insignificant relative to the average sample size of 143 ng (0.07% blank) and even the minimum sample size of 26ng (0.38% blank) and do not require application of a blank correction.

4.4.5 Enamel trace element analysis (ICP-MS*)*

In a 1.5ml plastic vial, the pre-weighed sample (5-10mg) had 1ml of 3N HNO3 added and was left overnight to dissolve. Subsequently, 0.5 ml was then transferred to a 15ml autosampler vial and diluted to 10ml. Samples were analysed by ICP-MS (Thermo Scientific XSeries2) previously optimised for low oxide and double charge interferences and calibrated for Sr, Ba, Zn and Pb. Calibration standards and blanks were analysed throughout the sample sequence to monitor and correct for any instrumental drift. Final enamel concentration was then determined based on sample weight and total dilution volume. Re-analysing the same sample from the 15 ml vial is reproducible ±2%.

5 Results and Discussion

The results of all analyses, and the discussion of the results, are summarized below. Further detailed discussions about the bone mercury (Article I), isotope (Article II) and fluoride (Article III) findings are contained with the respective articles.

5.1 Summary of Results

5.1.1 Soil samples

European soil background concentrations of arsenic are reported to 11.6 ppm (range 0.32-282 ppm), of mercury to 0.061 ppm (range 0.005-1.35 ppm), of cadmium to 0.284 ppm (range 0.145-14.1 ppm) and of lead to 32.6 ppm (range 5.32-970 ppm) (Salmien et al., 2001). Research conducted on modern samples from across Iceland showed average lead concentrations of 5.8 ppm in topsoil (4.7 pp in bedrock) and cadmium concentrations of 0.63 ppm (Panek and Kepinska, 2002). Generally, the background range of fluoride in soil worldwide is 30-500 ppm (Edmunds and Smedley, 2005; Ozsvath, 2008), however, it can temporarily increase after volcanic eruptions (Pyle and Mather, 2009). Topsoil leaching can result in inaccurate measurements or interpretation of historic fluoride concentrations in the environment, but soil concentrations eventually return to normal over time, as previously discussed at the end of section 4.2.3 (Thorarinsson and Sigvaldason, 1972; EFSA, 2010).

Elemental concentrations (Pb, Hg, As, Cd) were measured in 22 soil samples using ICP-MS and four samples were measured for fluoride. The results are presented in Table 5.1. The soil samples were measured for trace elements from the Skriðuklaustur and Skeljastaðir cemeteries. Lead concentrations in soil were low, ranging from 0.2 to 4.4 ppm for lead. Archaeological bone is prone to the diagenetic uptake of easily mobilised elements such as lead (see section 2.3.10; see Rasmussen et al., 2015). These concentrations suggest an absence of environmental lead pollution in the Icelandic environment, including the present day (see Panek and Kapinska, 2002), thereby reducing the likelihood of diagenetic enrichment. Mercury concentrations were also low, ranging from 0.06 to 0.10 ppm (all were ≤0.06 ppm except a single sample from just outside Skriðuklaustur that showed 0.10 ppm). The cadmium concentrations ranged from 0.03 to 0.13 ppm (all were ≤0.03 ppm except a single sample from just outside Skriðuklaustur that showed 0.13 ppm) for cadmium. The arsenic concentrations were lower than those found in most European nations, ranging from 0.237 to 1.913 ppm. However, due to the likelihood of diagenetic enrinchment of arsenic in bone, interpretations of such data should be considered irresolute. Lastly, fluoride concentrations in soil (n=4) were very low at both sites (<68 ppm), again indicating that diagenesis was not likely to have been at play. Overall, these results indicate that diagenesis was not a significant confounding variable on elemental data derived from human bone samples.

All the soil samples showed negligible amounts of mercury and cadmium, with most of the concentrations being below the level of detection. In other words, none of the cadmium, mercury or lead concentrations in soil exceeded the normal background concentrations at either site. However, despite being within the normal range, it is worth noting that four out of 22 samples (all from Skriðuklaustur) exceeded arsenic concentrations of 0.50 ppm because concentrations above this figure increase risk of toxicity. Considering the lack of permanent sources of groundwater at the analysed sites and the overall results of the soil and non-adult (n=6, see section 5.1.3) bone samples analysed here, diagenesis does not appear to have played a discernible role in this research. However, it is not possible to entirely dismiss the potential for post-depositional (diagenetic) uptake to have occurred. The additional elements (Sb, Zn, Sr, Ba) that were only measured in dental enamel were not analysed in soil samples because well-preserved enamel is highly resistant to diagenesis.

Table 5.1 Heavy metal concentrations (ppm in soil samples from Skriðuklaustur (SKR) and Skeljastaðir (ÞSK). All fluoride concentrations determined in soil (n=4) were below <68 ppm

Sample	As	Cd	Hg	Pb	Sample	As	Cd	Hg	Pb
SKR soil 1	1.913	0.130	0.059	4.368	ÞSK soil 1	0.304	<0.03	<0.06	0.176
SKR soil 2	0.439	<0.03	<0.06	1.297	ÞSK soil 2	0.311	<0.03	<0.06	0.254
SKR soil 3	0.475	<0.03	<0.06	1.111	ÞSK soil 3	0.337	<0.03	<0.06	0.673
SKR soil 4	0.401	<0.03	<0.06	0.409	ÞSK soil 4	0.334	<0.03	<0.06	0.540
SKR soil 5	0.447	<0.03	<0.06	0.812	ÞSK soil 5	0.348	<0.03	<0.06	0.239
SKR Soil 6	0.615	<0.03	0.097	2.254	ÞSK soil 6	0.355	<0.03	<0.06	0.199
SKR soil 7	0.364	<0.03	<0.06	0.584	ÞSK soil 7	0.342	<0.03	<0.06	0.234
SKR soil 8	0.508	<0.03	<0.06	1.373	ÞSK soil 8	0.313	<0.03	<0.06	0.200
SKR soil 9	0.411	<0.03	<0.06	0.344	ÞSK soil 9	0.237	<0.03	<0.06	0.452
SKR soil 10	0.416	<0.03	<0.06	0.375					
SKR soil 11	0.545	<0.03	<0.06	1.689					
SKR soil 12	0.274	<0.03	<0.06	0.218					
SKR soil 13	0.311	<0.03	<0.06	0.203					

5.1.2 Animal bone samples

All the animal bone samples were selected from Skriðuklaustur as no animal remains were available for sampling from Skeljastaðir. None of the animal bone samples (n=23) had elevated concentrations of cadmium (all <1 ppm), mercury (all <0.3 ppm) or lead (all <7 ppm), however, one cow had a notably elevated arsenic concentration of 3 ppm (Table 5.2). In the Nordic countries, during the Medieval Period, arsenic or other mineral salts (e.g., copper) were often mixed with antiparasitic botanicals and other substances to deworm livestock (Waller et al., 2001). It is therefore possible that the cow with elevated heavy metal concentrations could have been receiving antiparasitic or other treatment, although these elevations may have also occurred simply from consuming contaminated food or water, if diagenetic enrichment could be ruled out. In any case, the low concentrations of these heavy metals in animal bone (and the soil samples previously described) further indicate that the trace element concentrations determined in the human bone samples are biogenic rather than diagenetic.

Table 5.2 Trace element (As, Pb, Cd, Hg) concentrations (ppm) determined in animal bone samples from Skriðuklaustur

Sample	As	Cd	Hg	Pb	Sample	As	Cd	Hg	Pb
Seal 1	<0,04	<0,03	<0,03	0.122	Cow 1	<0,04	<0,03	<0,03	<0,06
Seal 2	<0,04	<0,03	<0,03	0.091	Cow 2	0.081	0.086	<0,03	0.936
Seal 3	0.173	0.031	0.03	2.200	Cow 3	<0,04	0.074	<0,03	<0,06
Seal 4	<0,04	<0,03	<0,03	0.145	Cow 4	<0,04	<0,03	<0,03	<0,06
Sheep 1	0.182	<0,03	<0,03	<0,06	Cow 5	0.083	0.067	<0,03	0.122
Sheep 2	<0,04	0.169	<0,03	<0,06	Cow 6	3.000	0.051	<0,03	0.065
Sheep 3	0.131	0.355	<0,03	0.182	Fish 1	0.181	0.184	<0,03	0.315
Sheep 4	0.207	0.221	0.037	0.132	Fish 2	0.201	0.217	<0,03	0.262
Sheep 5	<0,04	0.409	<0,03	0.095	Fish 3	0.184	0.249	<0,03	0.163
Swan 1	0.07	0.055	<0,03	0.763	Fish 4	0.078	0.062	<0,03	<0,06
Horse 1	<0,04	0.045	<0,03	<0,06	Dog 1	0.057	0.21	0.038	<0,06
Horse 2	0.22	0.076	0.059	0.07					

5.1.3 Human bone analyses

The tables presented in this section (5.1.3) are discussed throughout Chapter 5. Cumulative bone element concentrations (Hg, Pb, Cd, As) were determined in the 50 human bone samples from Skriðuklaustur and Skeljastaðir as previously described. The results are presented in Table 5.3 and summary statistics in Tables 5.4 and 5.5. The results of each of these elemental analyses are discussed in detail in this chapter. The fluoride bone concentration (n=50) results are presented in Supplementary Table I of Article III and summary statistics in Table 7 of Article III. Isotope and trace element analysis results – dental enamel (n=31) and bone collagen (n=50) – are presented in Table 5.6 below and the findings are discussed in detail throughout this chapter. The results of the osteological analyses (n=186) are presented in Tables 3-5 from Article III and Supplementary Table 1 from Article III and are also discussed in section 5.3.1.

Table 5.3 Trace element (As, Pb, Cd, Hg) concentrations (ppm) determined in human bone samples from Skriðuklaustur (SKR) and Skeljastaðir (ÞSK)

Sample	Sex	Age	Bone	As	Cd	Hg	Pb		Sample	Sex	Age	Bone	As	Cd	Hg	Pb
SKR 4	M	OA	Rib	0.066	0.325	0.252	5.0		SKR 169	F	OA	Rib	0.208	0.592	0.112	9.5
SKR 10	F	OA	Rib	0.036	0.295	0.111	0.9		SKR 172	M	OA	Rib	0.278	1.948	0.397	22.2
SKR 14	NA	NA	Rib	0.069	0.716	0.494	3.6		SKR 174	M	OA	Femur	0.041	0.030	0.030	13.5
SKR 22	NA	NA	Rib	0.085	0.446	0.139	7.1		SKR 181	F	OA	Temporal	0.331	0.797	0.192	77.2
SKR 23	F	YA	Rib	0.003	0.188	0.069	7.7		SKR 189	F	YA	Rib	0.275	1.078	0.295	2.9
SKR 29	F	YA	Rib	0.284	1.004	0.092	11.9		SKR 195	F	YA	Rib	0.271	2.679	0.513	11.4
SKR 30	F	OA	Rib	0.045	5.293	1.854	1.2		SKR 201	F	YA	Rib	0.225	2.273	1.823	93.8
SKR 33	F	OA	Rib	0.112	0.927	0.278	1.7		SKR 221	NA	NA	Rib	0.129	0.438	0.062	1.2
SKR 46	NA	NA	Rib	0.062	0.612	0.251	9.4		SKR 236	F	OA	Rib	0.057	1.931	0.157	0.6
SKR 65	F	YA	Rib	0.782	1.330	0.476	37.3		SKR 241	F	OA	Rib	0.165	0.849	0.087	0.9
SKR 81	F	YA	Rib	0.269	1.464	0.066	1.7									
SKR 91	M	YA	Rib	0.073	0.329	0.193	4.0		ÞSK 3	F	OA	Rib	0.057	0.326	0.572	0.5
SKR 100	M	YA	Rib	0.291	1.139	0.107	2.5		ÞSK 4	F	YA	Rib	0.115	3.654	10.134	2.4
SKR 115	M	OA	Rib	0.369	1.022	0.178	5.8		ÞSK 16	F	OA	Rib	0.054	0.361	0.496	1.3
SKR 122	F	YA	Parietal	0.151	0.729	0.195	9.6		ÞSK 17	F	OA	Rib	0.035	0.172	1.585	0.7
SKR 126	F	OA	Rib	0.169	0.548	0.107	0.7		ÞSK 29	M	OA	Rib	0.079	1.447	12.860	1.2
SKR 128	F	OA	Rib	0.058	1.055	0.163	60.7		ÞSK 32	M	YA	Rib	0.033	0.090	1.431	0.8
SKR 130	M	OA	Rib	0.037	1.737	0.598	4.3		ÞSK 34	M	YA	Rib	0.064	0.463	1.738	0.6
SKR 135	M	YA	Clavicle	0.340	0.610	0.121	5.3		ÞSK 37	M	OA	Rib	0.169	4.059	13.059	1.2
SKR 144	F	OA	Rib	0.947	1.834	0.283	1.9		ÞSK 41b	M	OA	Rib	0.038	0.340	0.490	0.9
SKR 146	NA	NA	Rib	0.157	0.254	3.429	2.0		ÞSK 42	M	OA	Rib	0.074	0.165	0.523	1.6
SKR 150	M	YA	Rib	0.408	7.138	1.782	18.5		ÞSK 44	M	OA	Os coxa	0.172	0.485	11.327	7.0
SKR 152	M	YA	Rib	0.256	1.108	0.289	3.2		ÞSK 48	M	OA	Parietal	0.099	0.156	0.478	1.4
SKR 155	M	OA	Rib	0.127	0.482	0.435	5.4		ÞSK 51	F	OA	Rib	0.049	0.144	0.438	0.6
SKR 163	NA	NA	Rib	0.093	0.368	0.933	5.8		ÞSK 54	M	YA	Rib	0.087	0.444	3.340	0.6
SKR 167	M	YA	Rib	0.212	0.818	0.343	3.4									

Key: SKR= Skriðuklaustur, ÞSK = Skeljastaðir; NA = Non-adult (<18), YA = Younger adult (18-35), OA = Older adult (35+), M = Male, F = Female

Table 5.4 Average (mean) concentrations and standard deviations of cadmium (Cd), arsenic (As), lead (Pb) and mercury (Hg) in ppm among samples from Skriðuklaustur and Skeljastaðir

	n	Cd	SD	As	SD	Pb	SD	Hg	SD
SKR	36	1.233	1.391	0.208	0.196	12.6	21.4	0.470	0.696
ÞSK	14	0.879	1.307	0.080	0.045	1.5	1.7	4.176	5.136
SKR and ÞSK	50	1.134	1.364	0.172	0.177	9.5	18.8	1.507	3.189
SKR Male	12	1.390	1.898	0.208	0.198	7.8	6.6	0.394	0.465
SKR Female	18	1.381	1.184	0.244	0.248	18.4	29.0	0.382	0.545
ÞSK Male	9	0.850	1.269	0.091	0.050	1.7	2.0	5.027	5.631
ÞSK Female	5	0.932	1.525	0.062	0.031	1.1	0.8	2.645	4.213
All Males	21	1.159	1.643	0.158	0.121	5.2	5.9	2.380	4.281
All Females	23	1.284	1.242	0.204	0.231	14.7	26.5	0.874	2.090
SKR Non-Adults	6	0.472	0.167	0.099	0.037	4.9	3.1	0.885	1.285
SKR Younger Adults	14	1.563	1.741	0.274	0.179	15.2	24.5	0.455	0.589
SKR Older Adults	16	1.229	1.244	0.190	0.229	13.2	22.7	0.327	0.433
ÞSK Younger Adults	4	1.163	1.670	0.075	0.035	1.1	0.9	4.161	4.069
ÞSK Older Adults	10	0.765	1.220	0.083	0.050	1.6	1.9	4.183	5.708
All Younger Adults	18	1.474	1.685	0.230	0.179	12.1	22.2	1.278	2.388
All Older Adults	26	1.051	1.232	0.149	0.187	8.8	18.5	1.810	3.937

Table 5.5 Table with p-values (t-test) detailing statistically significant differences between Skriðuklaustur and Skeljastaðir across sites, ages and sex categories

	As	Cd	Pb	Hg
SKR vs ÞSK	**0.0207**	0.4157	0.0597	**0.0002**
SKR Males vs Females	0.6539	0.9874	0.2222	0.9511
SKR Younger vs Older	0.2765	0.5456	0.8174	0.5008
SKR Adults vs Nonadults	0.1396	0.1448	0.3392	0.1105
ÞSK Males vs Females	0.2716	0.9159	0.5410	0.4275
ÞSK Younger vs Older	0.7844	0.7022	0.6086	0.9945
SKR Males vs ÞSK Males	**0.0228**	0.4697	**0.0156**	**0.0100**
SKR Females vs ÞSK Females	0.1226	0.4863	0.2029	**0.0285**
SKR Younger vs ÞSK Younger	**0.0452**	0.6880	0.2760	**0.0027**
SKR Older vs ÞSK Older	0.1585	0.3609	0.1235	**0.0119**

Table 5.6 All isotope and trace element data from the Skriðuklaustur and Skeljastaðir samples. Samples labeled with italics indicate $\delta^{13}C$ and $\delta^{15}N$ results reported by Sveinbjörnsdóttir et al. (2010). All strontium concentrations from Skeljastaðir (ÞSK), conducted on 1^{st} molars unless unavailable, indicate results reported by Gestsdóttir & Price (2003, 2006) and Price & Gestsdóttir (2006). Enamel samples from SKR 30, 130, 144, 146 and 163 were unavailable or not preserved. $\delta^{34}S$ is not reported for SKR 174 due to insufficient collagen for analysis

Sample	Tooth	Bone	Sex	Age	$^{87}Sr/^{86}Sr$	$\delta18O_p$ ‰	$\delta18O_{dw}$ ‰	$\delta13C_{carb}$ ‰	$\delta13C_{co}$ ‰	$\delta15N_{co}$ ‰	$\delta34S_{co}$ ‰	Sr ppm	Pb ppm	Zn ppm	Ba ppm	Cd ppm	Hg ppm	As ppm	Sb ppm
SKR 4	LPM2 max	Rib	M	OA	0.70706	13.9	-12.3	-15.4	-19.6	11.9	9.5	47.84	9.60	115.94	0.27	0.007	0.191	0.067	0.033
SKR 10	RM3 max	Rib	F	OA	0.70874	14.4	-11.5	-13.2	-19.2	11.0	8.26	156.68	0.30	145.83	0.51	0.015	nd	0.038	0.016
SKR 14	RPM1 mand	Rib	-	NA	0.70887	15.6	-9.7	-12.9	-17.9	15.7	10.72	140.41	0.58	119.40	0.25	0.010	0.090	0.079	0.022
SKR 22	LPM1 mand	Rib	-	NA	0.70746	15.7	-9.5	-15.3	-21.1	8.6	7.10	46.04	2.65	112.90	0.11	nd	0.045	0.078	0.022
SKR 23	RPM2 mand	Rib	F	YA	0.70798	15.8	-9.4	-15.3	-19.2	11.1	10.43	72.39	4.08	90.91	0.80	0.044	0.012	0.080	0.043
SKR 29	RPM1 mand	Rib	F	YA	0.70753	14.7	-11.1	-15.5	-19.2	12.5	8.63	40.80	2.28	73.51	0.41	nd	nd	0.020	0.023
SKR 30	-	Rib	F	OA	-	-	-	-	-19.5	12.3	11.23	-	-	-	-	-	-	-	-
SKR 33	LPM2 mand	Rib	F	OA	0.70859	15.9	-9.3	-14.7	-18.2	14.2	9.64	97.45	1.69	90.85	0.43	nd	nd	0.062	0.007
SKR 46	LPM2 mand	Rib	-	NA	0.70845	15.9	-9.3	-14.8	-19.1	12.3	9.38	88.18	2.73	101.84	0.17	0.009	nd	0.070	0.016
SKR 65	LPM2 max	Rib	F	YA	0.70742	13.8	-12.5	-16.4	-19.1	13.3	9.75	44.14	3.51	66.99	0.81	0.008	nd	0.124	0.012
SKR 81	LPM2 mand	Rib	F	YA	0.70757	16.0	-9.1	-15.7	-19.4	11.9	6.28	45.15	0.73	70.79	0.69	nd	nd	0.052	0.010
SKR 91	LPM2 mand	Rib	M	YA	0.70790	15.6	-9.7	-15.1	-19.1	12.3	9.26	52.90	0.53	91.57	0.17	0.013	nd	0.039	0.009
SKR 100	LPM2 mand	Rib	M	YA	0.70834	15.2	-10.3	-14.5	-18.9	11.9	11.58	93.70	2.68	104.60	0.40	0.004	0.400	0.017	0.003
SKR 115	LPM1 mand	Rib	M	OA	0.70686	15.9	-9.3	-15.7	-18.2	13.2	8.11	71.20	0.35	95.30	0.60	0.012	0.600	0.054	0.002
SKR 122	LPM2 max	Parietal	F	YA	0.70747	14.4	-11.6	-15.5	-19.3	13.5	9.49	44.40	1.38	134.59	0.62	0.010	nd	0.058	0.181
SKR 126	LPM2 max	Rib	F	OA	0.70781	15.6	-9.7	-15.2	-18.6	13.0	8.67	67.25	0.51	126.08	0.63	nd	nd	0.008	nd
SKR 128	LPM2 max	Rib	F	OA	0.70813	14.9	-10.8	-16.0	-17.5	13.9	8.61	64.13	0.51	87.03	0.29	0.016	0.098	0.077	0.103
SKR 130	-	Rib	M	OA	-	-	-	-	-17.5	14.5	8.52	-	-	-	-	-	-	-	-
SKR 135	LM3 max	Rib	M	YA	0.70727	16.1	-8.9	-15.2	-18.3	13.8	12.86	45.83	0.64	73.10	0.72	0.009	nd	0.068	0.043
SKR 144	-	Rib	F	YA	-	-	-	-	-18.5	13.0	8.01	-	-	-	-	-	-	-	-
SKR 146	-	Rib	-	NA	-	-	-	-	-18.3	14.5	8.70	-	-	-	-	-	-	-	-
SKR 150	RPM2 mand	Rib	M	YA	0.70726	14.4	-11.5	-16.1	-18.4	14.2	8.54	43.69	0.39	47.30	0.46	0.071	nd	0.060	0.023
SKR 152	LPM2 mand	Rib	M	YA	0.70832	15.6	-9.7	-14.5	-16.9	14.6	11.06	53.13	0.23	112.00	0.27	nd	nd	0.042	0.013
SKR 155	LM3 max	Rib	M	OA	0.70840	15.4	-10.0	-14.5	-18.5	12.5	9.05	73.54	0.21	123.89	0.29	nd	nd	0.037	0.008
SKR 163	-	Rib	-	NA	-	-	-	-	-18.3	13.9	7.95	-	-	-	-	-	-	-	-
SKR 167	RPM1 mand	Rib	M	YA	0.70723	14.4	-11.5	-16.6	-19.5	10.4	7.08	18.31	0.26	67.54	0.17	nd	nd	0.093	0.008
SKR 169	RPM1 max	Rib	F	OA	0.70809	15.5	-9.9	-14.9	-19.0	11.2	10.57	82.28	0.33	70.56	0.70	nd	nd	0.074	0.011
SKR 172	RPM1 max	Rib	M	OA	0.70684	15.2	-10.4	-15.1	-17.7	13.8	5.10	43.18	0.34	81.92	0.13	nd	nd	0.064	0.013
SKR 174	RC max	Femur	M	OA	0.70602	16.0	-9.1	-15.5	-18.9	12.7	-	74.97	1.46	104.97	0.61	0.016	nd	0.044	0.013
SKR 181	LPM2 max	Temporal	F	OA	0.70857	16.0	-9.1	-14.5	-19.4	12.4	6.81	79.55	0.41	116.02	0.18	0.009	nd	0.048	0.009
SKR 189	LPM1 mand	Rib	F	YA	0.70700	15.1	-10.4	-15.8	-19.6	12.3	9.47	43.90	9.40	145.50	0.40	0.002	0.400	0.005	0.012
SKR 195	LPM2 mand	Rib	F	YA	0.70874	15.6	-9.7	-14.0	-17.9	14.1	9.28	111.26	0.45	43.76	0.51	nd	nd	0.065	0.011
SKR 201	LPM1 max	Rib	F	YA	0.70816	15.1	-10.4	-16.3	-18.2	13.4	8.54	80.90	1.36	141.54	0.23	0.007	nd	0.035	0.013
SKR 221	RPM2 mand	Rib	-	NA	0.70767	15.5	-9.8	-15.3	-19.5	11.8	7.84	25.79	0.13	98.61	0.22	0.008	0.126	0.060	0.123
SKR 226	RPM1 mand	Rib	F	OA	0.70778	15.4	-10.1	-15.1	-19.5	11.1	6.48	87.33	0.61	75.60	0.46	0.138	nd	0.043	0.021
SKR 241	LPM1 mand	Rib	F	OA	0.70800	15.3	-10.1	-15.4	-17.8	12.9	11.43	50.39	3.12	88.33	0.30	nd	nd	0.021	0.038

Key: SKR = Skriðuklaustur, ÞSK = Skeljastaðir; LPM1 = left 1st premolar, LPM2 = left 2nd premolar, LM3 = left 3rd molar, RPM1 = right 1st premolar, RPM2 = right 2nd premolar, RC = right canine, RM3 = right 3rd molar, max = maxilla, mand = mandible; M = Male, F = Female; NA = Nonadult (<17), YA = Young adult (17-36), OA = Older adult (36+); $\delta18O_p = \delta18O_{phosphate\ VSMOW}$ (‰), $\delta18O_{dw} = \delta18O_{drinkingwater\ VSMOW}$ (‰), $\delta13C_{carb} = \delta13C_{carbonate\ VPDB}$ (‰), $\delta13C_{co} = \delta13C_{collagen}$ (‰), $\delta15N_{co} = \delta15N_{collagen}$ (‰), $\delta34S_{co} = \delta34S_{collagen}$ (‰), Sr/Pb/Zn/Ba ppm = Strontium/Lead/Zinc/Barium parts per million, nd = not detected

Sample	Tooth	Bone	Sex	Age	$^{87}Sr/^{86}Sr$	$\delta18O_p$ ‰	$\delta18O_{dw}$ ‰	$\delta13C_{carb}$ ‰	$\delta13C_{co}$ ‰	$\delta15N_{co}$ ‰	$\delta34S_{co}$ ‰	Sr ppm	Pb ppm	Zn ppm	Ba ppm	Cd ppm	Hg ppm	As ppm	Sb ppm
ÞSK 2	?	?	F	OA	0.70905	-	-	-	-19.7	7.5	-	-	-	-	-	-	-	-	-
ÞSK 3	-	Rib	F	OA	-	-	-	-	-21.0	6.6	8.5	-	-	-	-	-	-	-	-
ÞSK 4	-	Rib	F	YA	-	-	-	-	-20.7	8.8	7.7	-	-	-	-	-	-	-	-
ÞSK 5	?	?	F	YA	0.70591	-	-	-	-19.8	7.2	-	-	-	-	-	-	-	-	-
ÞSK 12	?	?	F	OA	0.70562	-	-	-	-19.2	7.6	-	-	-	-	-	-	-	-	-
ÞSK 15	?	?	F	OA	0.70615	-	-	-	-19.6	7.6	-	-	-	-	-	-	-	-	-
ÞSK 16	?	Rib	F	OA	0.70754	-	-	-	-21.0	6.7	11.5	-	-	-	-	-	-	-	-
ÞSK 17	?	Rib	F	OA	0.70662	-	-	-	-21.2	8.7	9.6	-	-	-	-	-	-	-	-
ÞSK 26	?	?	M	OA	0.70610	-	-	-	-19.7	6.8	-	-	-	-	-	-	-	-	-
ÞSK 29	-	Rib	M	OA	-	-	-	-	-21.5	7.1	9.5	-	-	-	-	-	-	-	-
ÞSK 32	?	Rib	M	YA	0.70685	-	-	-	-21.4	8.8	8.1	-	-	-	-	-	-	-	-
ÞSK 34	-	Rib	M	YA	-	-	-	-	-20.3	9.1	8.8	-	-	-	-	-	-	-	-
ÞSK 37	?	Rib	M	OA	0.70900	-	-	-	-20.6	7.9	9.1	-	-	-	-	-	-	-	-
ÞSK 38	?	?	M	OA	0.70608	-	-	-	-19.5	7.0	-	-	-	-	-	-	-	-	-
ÞSK 39	?	?	F	OA	0.70972	-	-	-	-19.9	10.5	-	-	-	-	-	-	-	-	-
ÞSK 41a	?	?	M	YA	0.70704	-	-	-	-19.7	7.8	-	-	-	-	-	-	-	-	-
ÞSK 41b	-	Rib	M	OA	-	-	-	-	-21.3	7.9	8.2	-	-	-	-	-	-	-	-
ÞSK 42	?	Rib	M	OA	0.70653	-	-	-	-21.4	8.0	8.6	-	-	-	-	-	-	-	-
ÞSK 44	-	Fibula	M	OA	-	-	-	-	-21.6	8.1	13.7	-	-	-	-	-	-	-	-
ÞSK 47	?	?	M	YA	0.70682	-	-	-	-19.8	6.9	-	-	-	-	-	-	-	-	-
ÞSK 48	?	Rib	M	OA	0.70614	-	-	-	-21.4	8.0	9.2	-	-	-	-	-	-	-	-
ÞSK 51	-	Rib	F	OA	-	-	-	-	-21.1	7.3	11.4	-	-	-	-	-	-	-	-
ÞSK 54	-	Rib	F	YA	-	-	-	-	-21.3	7.0	7.7	-	-	-	-	-	-	-	-
ÞSK 60	?	?	M	OA	0.70718	-	-	-	-19.4	7.4	-	-	-	-	-	-	-	-	-

Key: SKR = Skriðuklaustur, ÞSK = Skeljastaðir; LPM1 = left 1st premolar, LPM2 = left 2nd premolar, LM3 = left 3rd molar, RPM1 = right 1st premolar, RPM2 = right 2nd premolar, RC = right canine, RM3 = right 3rd molar, max = maxilla, mand = mandible; M = Male, F = Female; NA = Nonadult (<17), YA = Young adult (17-36), OA = Older adult (36+); $\delta18O_p = \delta18O_{phosphate\ VSMOW}$ (‰), $\delta18O_{dw} = \delta18O_{drinkingwater\ VSMOW}$ (‰), $\delta13C_{carb} = \delta13C_{carbonate\ VPDB}$ (‰), $\delta13C_{co} = \delta13C_{collagen}$ (‰), $\delta15N_{co} = \delta15N_{collagen}$ (‰), $\delta34S_{co} = \delta34S_{collagen}$ (‰), Sr/Pb/Zn/Ba ppm = Strontium/Lead/Zinc/Barium parts per million, nd = not detected

Table 5.7 Means and ranges of isotope ratios determined in the human dental enamel samples from Skriðuklaustur. Samples SKR 100, 115 and 189 were run during the second analytical session which gave the average $^{87}Sr/^{86}Sr$ value and reproducibility for the international isotope reference material NBS987 as 0.710266±0.000009 (2SD; n=6) while all others were run in the first analytical session which gave 0.710258±0.000013 (2SD; n=12)

Samples	n	$^{87}Sr/^{86}Sr$ ± 2SD	δ18O$_{phosphate}$ ‰ (VSMOW)	δ18O$_{drinkingwater}$ ‰ (VSMOW)	δ13C$_{carbonate}$ ‰ (VPDB)
Male	11	0.70741 ± 0.00144	15.2 ± 0.7	-10.2 ± 1.1	-15.3 ± 0.6
Female	16	0.70797 ± 0.00099	15.2 ± 0.6	-10.3 ± 1.0	-15.2 ± 0.8
Non-Adult	4	0.70811 ± 0.00114	15.7 ± 0.1	-9.6 ± 0.2	-14.6 ± 1.0
All	31	0.70779 ± 0.00132	15.3 ± 0.6	-10.2 ± 1.0	-15.2 ± 0.8
Ranges (All)	31	0.70602 to 0.70887	13.8 to 16.1	-12.3 to -8.3	-16.6 to -12.9

Table 5.8 Medians, standard deviation, and ranges (highest and lowest values) of trace elements determined in the human dental enamel samples from Skriðuklaustur. Concentrations from the samples that were not detectable are not included. Lowest concentration of the range represents the sample with the lowest detectable concentration

Samples	n	Lead (Pb) ppm	Barium (Ba) ppm	Zinc (Zn) ppm	Strontium (Sr) ppm
Male	11	0.39 ± 0.71	0.29 ± 0.19	95.30 ± 22.12	52.90 ± 19.64
Female	16	1.05 ± 2.26	0.49 ± 0.19	89.59 ± 31.33	69.82 ± 29.99
Non-Adult	4	1.62 ± 1.18	0.20 ± 0.05	107.37 ± 8.36	67.11± 43.91
All	31	0.61 ± 1.82	0.40 ± 0.21	95.30 ± 26.70	64.13 ± 30.30
Ranges (All)	31	0.13 - 9.40	0.11 - 0.81	43.76 - 145.83	25.79 - 156.68
Samples	n	Cadmium (Cd) ppm	Mercury (Hg) ppm	Arsenic (As) ppm	Antimony (Sb) ppm
Male	11	0.012 ± 0.022	0.400 ± 0.167	0.054 ± 0.019	0.013 ± 0.012
Female	16	0.010 ± 0.041	0.098 ± 0.166	0.050 ± 0.030	0.013 ± 0.046
Non-Adult	4	0.009 ± 0.001	0.090 ± 0.033	0.074 ± 0.008	0.022 ± 0.045
All	31	0.010 ± 0.032	0.126 ± 0.190	0.058 ± 0.025	0.013 ± 0.039
Ranges (All)	31	0.002 - 0.138	0.012 - 0.600	0.017 - 0.124	0.002 - 0.181

5.2 Health and cultural implications of mercury (Hg) in bone and dental enamel

5.2.1 Mercury concentrations in bone

Cumulative bone mercury concentrations were determined in 22 soil samples (Table 5.1), 23 faunal bones (Table 5.2) and the 50 human bone samples (Table 5.3). This section addresses Research Questions 1.2.3, 1.2.4, 1.2.6. The human bone mercury concentrations, along with palaeopathological descriptions and differential diagnoses, are presented in Table 1 of Article I. Increased mercury concentrations (>0.3 ppm) were found in eleven (n=11) individuals from Skriðuklaustur. Among females (n=18), the mean concentration was 0.382 ppm, while it was it was 0.394 ppm among males (n=12). When one male outlier (SKR 174; the only sample with <0.03 ppm) is excluded, the mean for males (n=11) was 0.427 ppm. Even if access to medical care was different between social classes or biological sexes, the mercury concentrations reported in this study did not show a statistically significant difference. For older adults (>36 years) (n=16) the mean was 0.327 ppm. Though younger adults (17-35 years) (n=14) had a higher mean of

0.455 ppm, it was not statistically significant (see Tables 5.4 and 5.5 above and Figure 3 and 4 from Article I). Zuckerman (2016) found a similar trend, with older adults having lower concentrations than younger individuals. In this study, at least nine of the analysed individuals had bone markers associated with treponemal disease (e.g., venereal syphilis). The 11 individuals that had increased mercury concentrations all showed bone markers indicative of either hydatid, treponemal, or non-specific infectious disease. Therapy with mercurial medicines was normally initiated following the onset of skin lesions (Zuckerman, 2017a), which usually appear in the first (primary) and secondary phases of venereal syphilis (Baughn and Musher, 2005; Nyatsanza and Tipple, 2016). A latent period sometimes follows and can potentially last for 30+ years (Nyatsanza and Tipple, 2016). Thus, if older individuals were in the latent or tertiary stages of the disease, it is possible that they may have stopped taking mercurial medicine prior to death. Meanwhile, a young adult female (SKR 201) had the highest concentration (1.823 ppm) (excluding non-adults) and exhibited destructive bone activity that caused multiple perforations to the palate. It must be considered that mercury toxicity itself can also lead to a younger age-at-death, especially in light of the the non-standardised dosage given medicinally during the 16th century (see Ioannou et al., 2016; Zuckerman, 2016). Two neonates had increased mercury in bone, potentially originating during gestation via transplacental transfer, or directly through mercurial therapy following birth. Dental enamel defects were absent on the observable teeth; however, enamel changes only appear in an unknown number (circa 10%-65%) of affected individuals (Ioannou et al., 2016). Six individuals (SKR 10, 65, 115, 144, 174 and 221) had normal concenrations ranging from <0.03 to 0.283 ppm and no skeletal markers indicative of infectious disease.

Overall, the mercury concentrations determined from Skeljastaðir were all elevated and are statistically significantly higher than those from Skriðuklaustur, not only between sites (*t-test, p=0.002)*, but also across age and sex categories (Tables 5.4 and 5.5 above and Figure 7 and 8 from Article I). The mean concentration for those without pathological conditions (n= 8) was 4.945 ppm. Individuals with skeletal pathologies (n=6) had a mean of 3.151 ppm. These means are markedly greater than they were at Skriðuklaustur, predominately due to the individuals (n=5) with severely elevated concentrations. Four individuals (ÞSK 17, 32, 34 and 54) had elevate dconcentrations that ranged from 1.585 ppm to 3.340 ppm, but another four (ÞSK 4, 29, 37 and 44) had exceedingly high concentrations that ranged between 10.134 ppm and 13.059 ppm. Since some individuals sampled from Skeljastaðir date to after the Mt. Hekla eruption of AD 1104 (see Sveinbjörnsdóttir et al., 2010), it is possible that acute mercury exposure occurred during and following it. It is worth nothing that mercury alone can cause bone and dental pathologies, such as periodontitis, dental enamel defects, brittle teeth, dental attrition, and higher rates of ante-mortem tooth loss, in addition to maxillary and mandibular new bone formation (Zuckerman, 2016). One individual (ÞSK 29) had ante-mortem tooth loss of all mandibular teeth (maxilla unobservable), lingual wear, enamel defects on the anterior teeth (mottling), advanced alveolar resorption, and a bone mercury concentration of 12.860 ppm. These bony alterations could have been caused by exposure to mercury, fluoride or by numerous other pathological conditions.

Elevated concentrations of mercury found in the skeletal population from Skriðuklaustur could have arisen from several points of possible exposure, such as from medical use, through pigments featured in scholarly writing (e.g., vermilion) or from the dietary importance of marine resources (see Parsons and Percival, 2005; Mehler, 2015). While mercury was used (e.g., treatment for skin conditions) during the time period that Skeljastaðir was inhabited, it was not commonplace until around the time that syphilis plagued western continental Europe (late 15th century) (Parsons and Percival, 2005; Swiderski, 2008). Marine mercury contamination rose five-fold in the 1800s and ten-fold during the 1900s (Hylander and Meili, 2003; Parsons and Percival, 2005). Therefore, prior to the 19th century, marine animals had far lower levels of mercury pollution than in the present. Thus, diet was not likely a major source of exposure to mercury in Medieval Iceland. Additionally, isotopic results showed that marine protein was unimportant to the diet at Skeljastaðir, whereas the opposite was the case at Skriðuklaustur (Sveinbjörnsdóttir et al., 2010; Walser III et al., 2020a). As volcanoes almost constantly release emissions (passive degassing) and eruption events may persist for months or even years (Simkin and Siebert, 1994; D'Alessandro, 2006), exposure could have happened during eruptions or via residential proximity so close to Mt. Hekla. Rib samples showing notably elevated concentrations imply exposure occurring within weeks to a few years of the sampled individual's time-of-death. Though mercury can be high in the atmosphere due to degassing or eruptions (Coderre and Steinthorsson, 1977), cinnabar ore does not occur naturally in Iceland (personal correspondence: Kristján Jónasson, Geologist, Icelandic Institute of Natural History, 31.01.2016). Therefore, any forms of mercury (e.g., ore, elemental) that were used in would have been purchased from abroad for medicinal purposes.

5.2.2 Mercury concentrations in enamel

In ancient and modern dental enamel from human teeth, Rasmussen (1974) found the range of mercury concentrations to be ca. <0.001-1.88 ppm. In this study, only six out of 31 individuals presented with detectable concentrations of mercury in their tooth enamel (Table 5.7). The six samples with detectable mercury concentrations ranged between 0.01-0.60 ppm, indicating that none of them were exposed to mercury during childhood (Table 5.8).

5.3 Health and culturual implications of fluoride (F) in bone and associated skeletal and dental changes

5.3.1 Results of osteological analysis

The results of all the osteological analyses are provided in Tables 3-5 from Article III and Supplementary Table 1 from Article III. This section addresses Research Questions 1.2.1, 1.2.4, 1.2.6, 1.2.7 and 1.2.8. Overall, the observed skeletal markers increased with age. This was to be expected, considering that many of the recorded pathologies are directly associated with increasing age. There were low rates of vertebral ankyloses (6%) and other joint ankyloses (8%), with no observed significant differences between demographic categories. Nearly half (42%) of the individuals displayed at least one ante-mortem fracture, with males and individuals aged 36+ years most affected across time. Periosteal new bone formation was seen in 57% of the analysed individuals. Fractures, entheseal and interosseous calcifications were higher (with some cases statistically significantly higher) in all time periods among males and individuals from the 36+ category, perhaps because differences in behaviour and biology are strongly correlated with increased rates of calcifications (see e.g., Meyer et al., 2011; Shuler et al., 2012; Milella et al., 2012; Henderson and Cardoso, 2013; Santana-Cabrera et al., 2015). Therefore, entheseal changes have a highly multifactorial aetiological origin (Villotte and Knüsel, 2013). Since the accumulation of fluoride is age related, long-term, low-dose fluoride exposure may instead be indicated (Arnala et al., 1985; Barbier et al., 2010; Petrone et al., 2011). Comparative data from Herculaneum (see Petrone et al., 2013) is presented in Table 5.9. Unfortunately, the demographic data provided in the Petrone et al. (2013) publication is limited and thus detailed demographic differences between Iceland and Herculaneum cannot be directly compared. Categories requiring more detailed discussion are presented below and in the text of the Article (III).

Table 5.9 Comparison of results of skeletal analyses performed on individuals from Herculaneum (Petrone et al., 2013) and Iceland (this study). Petrone et al. (2013) only presented the results as percentages and not as frequencies

	Herculaneum	Iceland
Fractures	32%	42%
Spinal Ankylosis	20%	6%
Interosseous/Entheseal	92%	41%
Osteomalacia	8%	3%
DJD Appendicular	47%	46%
DJD Spinal	28%	72%
Fluoride Concentrations	mean: 6672 ppm	mean: 2056 ppm

Fractures

In this research, 42% of individuals had a minimum of one ante-mortem post-cranial fracture (e.g., Figure 5.1, 5.2 and 5.3 and Table 3 from Article III), providing a higher prevalence rate than what was seen at Herculaneum (32%), where osteofluorosis was endemic and severe (Petrone et al., 2013). Aside from fluorosis, the higher fracture rate seen in the Icelandic assemblages might also be related to the local ecology, human behaviour, or geography (e.g., mountains, lava fields, glaciers).

Figure 5.1 Photograph and radiograph a healing oblique fracture to distal shaft of the right tibia of an adult female (ÞSK 16) – lateral view

Figure 5.2 Radiograph and photograph of healed oblique fractures on the left tibia and fibula of an adult male (HFE 18) – anterior view

Figure 5.3 Lumbar vertebra (L5) with spondylolisis, a fracture that can occur from heavy lifting, in an adult male (BES 5) – superior (body) and posterior (neural arch) view

Ankylosis

Ankyloses of spinal (6%) and non-spinal joints (8%) had low prevalence rates and do not suggest skeletal fluorosis (e.g., Figures 5.4, 5.5 and 5.6 and see Table 3 in Article III). At Herculaneum, for comparison, Petrone et al. (2013) observed ankylosis on a minimum of one skeletal element in 39% of individuals: 20% spinal, 28% foot distal interphalangeal joints and 22% manubriosternal joint.

Figure 5.4 Ankylosis of two thoracic vertebrae in an adult individual (RKH 7) – posterior view

Figure 5.5 Ankylosis of proximal and distal ends of right tibia and fibula of an older adult male (ÞSK 29) – anterolateral view. The individual also had ossified cartilage on the manubrium, sternal body, xiphoid process, and sternal rib ends. However, these bone changes may rather be correlated with older age and diffuse idiopathic skeletal hyperostosis, which may be seen throughout the spine

Figure 5.6 Diffuse idiopathic skeletal hyperostosis in an older adult male (ÞSK 29). Some of the lower spine has fused completely – lateral view

Degenerative joint disease

The highest prevalence of skeletal changes occurred in the form of joint disease, with 72% individuals showing evidence of vertebral joint degeneration and 46% of individuals exhibited non-spinal joint degeneration of at least one joint surface (e.g., Figures 5.7, 5.8). Spinal joint disease and/or osteoarthritis of at least one joint surface was noted in ca. 63% of younger adults (17-35 years) and ca. 81% of older adults (36+ years) and non-spinal joint disease and/or osteoarthritis was observed in ca. 25% of younger adults and ca. 65% of older adults. In this study, non-spinal degenerative joint disease was statistically highest (*chi-squared test, p=0.00003*) across all time periods amongst individuals aged 36+ from Skeljastaðir. Non-spinal joint disease was also significantly higher in the 17^{th}-19^{th} centuries assemblages than in the assemblage from the 10^{th}-12^{th} centuries. Non-spinal joint disease was significantly higher in older individuals than in younger individuals in the 10^{th}-12^{th} centuries and in the assemblages from the 13^{th}-16^{th} centuries. While the prevalence of non-spinal joint disease was also higher amongst older adults compared to younger adults in the assemblages from the 17^{th}-19^{th} centuries, it was not statistically significant (*chi-squared test, p=0.057*).

Previous research noted that ca. 32% of individuals at Skeljastaðir had evidence of osteoarthritis and that there was a high prevalence of hip osteoarthritis compared with English, Swedish and Danish populations (Gestsdóttir, 2006, 2014; Gestsdóttir et al., 2006b). When compared with the skeletal assemblage representing the Greenland Norse, a similar pattern of osteoarthritis was noted, likely because these individuals were practicing similar subsistence strategies and physical activities and had the same genetic background as those residing in Iceland at the same time (Gestsdóttir, 2006). Furthermore, research has shown a correlation between hip osteoarthritis and farming activities, such as those practiced at Skeljastaðir (Gestsdóttir, 2014). At a contemporaneous site (10^{th}-12^{th} centuries) in northern Iceland, Keldudalur, 44%

of the population showed evidence of degenerative joint disease, consistent with a strenuous farming and animal husbandry lifestyle (Zoëga and Murphy, 2016). For a non-local comparison by Rogers et al. (1981), a large multiperiod study (n=400) on skeletons from England, found that 50% of individuals exhibited spinal joint disease and 40% of individuals showed evidence of non-spinal joint disease. Therefore, the high prevalence of spinal (39 out of 45; 86%) and non-spinal (28 out of 50; 56%) degenerative joint disease affecting at least one joint surface at Skeljastaðir is likely predominately correlated with age, underlying genetic factors and subsistence strategies centered around farming. At Herculaneum spinal degenerative joint disease was noted in 28% of individuals, while it was present in 72% of individuals analysed from Iceland. Meanwhile, the prevalence of appendicular degenerative joint at Herculaneum was 47%, similar to the 46% prevalence noted from Iceland. Petrone et al. (2013) suggest an age-related explanation for degenerative joint disease at Herculaneum rather than an association with osteofluorosis.

Figure 5.7 Degenerative joint disease in a lumbar vertabra of an adult female (BES 20) – posterior view

Figure 5.8 Left femur of an adult male (HFE 18) with osteoarthritis (eburnation, porosity, osteophytes, contour/morphological change) – anterior view

Infectious disease

Periosteal new bone formation was observed in 57% of individuals (e.g., Figures 5.9 and 5.10). At Skriðuklaustur, individuals showing evidence of infectious diseases were common, possibly inflated by the medical activities performed there; osteitis/osteomyelitis (e.g., Figure 5.9), several cases of tuberculosis, the highest prevalence of hydatid disease (e.g., Figure 2.4) and the only confidently diagnosed cases of treponemal disease across all skeletal assemblages in Iceland (e.g., Figure 2.3; Kristjánsdóttir and Collins, 2011; Kristjánsdóttir, 2012; Walser III et al., 2019). Haffjarðarey, unlike Skriðuklaustur, was a coastal site that likely substantially depended upon fishing and the gathering of other marine resources (Gestsdóttir, 2014). An intensive fishing economy was in place on the Snæfellnes peninsula, with people fishing year around in addition to farming (Guðmundsson et al., 1988; Valdimarsson and Bjarnason, 1997), indicating that those residing at Haffjarðarey were likely subject to more rigorous activities and occupational behaviors that may have mechanically contributed to periosteal changes, particularly in the lower leg bones (see Lassus, 2002). The population at Haffjarðarey likely had a less varied diet than was consumed at inland sites and probably depended upon seafood, especially *harðfiskur* (dried fish) (Hoffman, 2018). As fresh or dried sea food can contain high fluoride concentrations (Marya, 2011; Ganta et al., 2015), the importance of fish and other marine resources at both these sites, unlike the 10^{th}-12^{th}-century site (ÞSK), may also indicate an additional source of fluoride exposure beyond volcanic emissions.

Figure 5.9 Photograph and radiograph of the fibulae, tibiae, and left femur of a non-adult individual (SKR 46) with periosteal change, osteitis, and osteomyelitis. On the radiograph, destructive lesions that are not apparent in the photograph can be seen below the cortical surfaces of the bones, particularly in the tibiae – anterior view

Figure 5.10 Primary (woven) new bone formation on the left distal fibula of an older adult male (RVK-C-6) – medial view

Ossification of the ligamentum flavum, atlanto-occipital membrane and other soft tissues

Ligamentum flavum ossification (e.g., Figure 5.11) was seen in 26% of individuals. Females dating from the 17th to 19th centuries (RKH, BES, RVK and VEY) had the highest prevalence; see Table 3 in Article III). The distribution of ligamentum flavum ossification was even across sex and age categories when all assemblages were pooled together. Bone changes at the attachment site of the atlanto-occipital membrane (e.g., Figure 5.12) (foramen magnum, posterior margin), were analysed in this study because of the anatomical correspondence it has to the ligamentum flavum (Cramer and Darby, 2017; Gonzales and Iwanaga, 2018). Antlanto-occipital membrane attachment site ossification was seen in 37% of individuals and no differences were observed across sex or age categories, aside from in the 10th-12th century assemblage (ÞSK) which had a less frequent rate in younger adults (24%) than in older adults (44%) (Table 3 in Article III). Atlanto-occipital membrane or attachment site changes are sometimes discussed in clinical studies after traumatic accidents (e.g., vehicular accidents causing whiplash). In some cases, underlying congenital or developmental conditions act as predisposing factors (Vangilder and Menezes, 1983; Lustrin et al., 2003; Riascos et al., 2015). Occupation, such as the heavy lifting performed during farm work, could potentially explain the higher rate observed among older adults from the 10^{th-}12th century assemblage.

The pathogenesis of ligamentum flavum ossification is related to genetics and diet, but various additional factors might contribute as well (e.g., Mobbs and Dvorak, 2007; Shepard et al., 2015; Geber and Hammer, 2018). The 17th-19th-century assemblages (43%) had a higher prevalence than the 13th-16th-century (22%) and 10th-12th-century (16%) assemblages possibly due to to a mixture of factors such as the diverse diet available to the upper class (see Jónsson, 1998), genetic admixture (see Ebenesersdóttir et al., 2018), changes in occupation connected to urbanization (see Harrison and Snæsdóttir 2013) and environmental fluoride pollution following the eruption of Laki in AD 1783-1784 (see Steingrímsson, 1998; Halldórsson, 2013). Ligamentum flavum ossification was seen in 15.6% of female and in 23.6% of male thoracic vertebrae in a Medieval Polish sample (AD 900-1000), for comparison (Swedborg, 1974). In another example, research on the *Mary Rose* skeletal assemblage from England showed that 46.3% of individuals had ligamentum flavum ossification throughout the thoracic spine, which was interpreted to have been caused by rigorous activity or occupation (Stirland and Waldron, 1997).

Other soft tissue ossifications (e.g., Figure 5.13), such as glands or cartilage, were observed in 20% (36 out of 184) of individuals. They were found to be more common in older individuals (25%; 22 out of 87), as expected, than in younger individuals (14%; 14 out of 97), likely related to increasing age. Other soft tissue ossifications were associated with increasing age, aside from the 17th-19th-century assemblages, which had similar frequencies between younger (15%) and older adults (11%) (Table 3 in Article III).

Figure 5.11 Ossification of the ligamentum flavum (blue arrow) in a thoracic vertebra of an adult male (ÞSK 26) – posterior view

Figure 5.12 Inferior view of older adult male cranium. Slight changes to the attachment site of the atlanto-occipital membrane at the posterior aspect of the foramen magnum are indicated by the blue arrows

Figure 5.13 Ossification of cartilage on manubrium and sternum of an adult male (ÞSK 29)

Mineralization of interosseous and entheseal attachment sites

The results of entheseal changes (e.g., Figure 5.14, 5.15, 5.16 and 5.17) analysis are presented in Table 4a in Article III and interosseous mineralization (e.g., Figure 5.18 and 5.19) in Table 4b in Article III. Entheseal changes had a non-significantly higher prevalence in the 10^{th}-12^{th}-century assemblage than was observed in the 13^{th}-16^{th} century assemblages yet statistically lower prevalence than those seen from the 17^{th}-19^{th} centuries. Interosseous mineralization was lowest in the 10^{th}-12^{th}-century assemblage, while the 17-19^{th} century assemblages had statistically significantly higher prevalence rates (*chi-square test*, $p<0.02$ for all long bones except fibulae) than both the 10^{th}-12^{th} and 13^{th}-16^{th}-century assemblages. Though no pattern was observed according to sex, the older adults representing the 17^{th}-19^{th} century assemblages had the highest frequencies – sometimes significantly higher – of calcifications on interosseous and entheseal attachment sites. Calcification of the interosseous crest of the ulnae, radii, fibulae, and tibiae was seen on 12-24% of each bone in younger adults and on 26-46% of each bone in older adults. Entheseal calcification was recorded in 6-27% of each long bone or os coxa in younger adults and in 30-57% of older adults. In line with the findings of previous studies of entheseal changes, the severity and prevalence of entheseal changes increased among older adults (the 36+ OA category) and were higher in males than in females. From Skeljastaðir, at least 14 individuals had severe entheseophyte formation (Gestsdóttir, 2008; this study), suggesting entheseal change beyond normal increased rugosity.

Figure 5.14 Right and left humerus with entheseal changes to the deltoid tuberosity (blue arrows) – anterior view. This cortical defect, also known as a chronic avulsive injury, is a benign reactive lesion secondary to high force movements at the pectoralis major insertion on the proximal humerus shaft (see Fulton et al., 1979; Villotte et al., 2016)

Figure 5.15 Posterior aspect of right femur, of an older adult female (SKR 169), with slight entheseal changes to the linea aspera

Figure 5.16 Posterior aspect of the right and left femur, of an older adult male (ÞSK 30), with moderate entheseal changes to linea aspera, spiral line and gluteal tuberosity

Figure 5.17 Os coxa from an older adult male (ÞSK 30) with entheseal changes to iliac crest – posterior view

Across all sites and all ages, under 41% of individuals showed indicators, on at least one bone, of mineralization of entheseal or interosseous attachment sites. Meanwhile, at Herculaneum, 92% of individuals of all ages had clear evidence of calcificying/ossifying changes on at least one bone (see Petrone et al., 2013). Similar to the rates presented here, previous studies have all shown a strong correlation to increasing age instead of activity patterns or pathologies (e.g., Campanacho and Santos, 2013; Henderson and Cardoso, 2013; Henderson et al., 2013). Rates of calcification on the entheseal attachment sites and interosseous crests were only slightly lower in the older assemblages than in the 17th-19th-century assemblages. As fluoride burden increases according to age, it is possible that these rates of entheseal and interosseous calcifications are related to elevated environmental fluoride contamination originating from Iceland's most catastrophic historic eruption – the Skaftáreldar fires (Laki) of AD 1783-1784. The population dealt with dietary malnourishment, aside from fluoride pollution, as metabolic deficiency conditions became endemic (e.g., rickets, scurvy, osteomalacia) and diseases, like smallpox, spread throughout the country (Pétursson et al. 1984; Halldórsson, 2013).

Figure 5.18 Slight mineralization of the interosseous crest of the right tibia of a young adult female (RVK-C-4) – medial view

Figure 5.19 Ossification of the interosseous crest of the left tibia of an older adult female (SKR 169) – lateral view. Corresponding changes can be seen on the left fibula, which is not pictured here

The results of this research demonstrated a higher prevalence of changes to the interosseous crest of the lower leg in females (*chi-squared test*, $p<0.009$), while males had a higher prevalence of interosseous crest changes on the lower arm (*chi-squared test, $p<0.04$*). While these results may have implications for gendered activities, Halldórsdóttir (2018) notes that women were not just farmer's wives but were often farmers themselves and, in some cases, managed the farm alone. Farm work performed by women particularly increased in the winter, especially with tasks related to the preparation of wool for textile production (Róbertsdóttir, 2014). Women also worked in the wool and fish factories, as housekeepers and even as fisherwomen (Róbertsdóttir, 1998, 2001, 2008; Willson, 2016; Frangoudes and Gerrard, 2018). This implies that even if any distinct gendered social roles were in place at the time, women still participated in equally strenuous work. Though it is known that heavy or repetitive physical activities exacerbate skeletal changes resulting from fluorosis, particularly at entheses (Maheshwari, 2006; Nelson et al., 2016), increased rugosity and other degenerative changes may occur just as extensively without fluoride exposure. Furthermore, other pathological changes that can occur at the attachment site, such as those resulting from neoplastic disease, may occasionally be misidentified or difficult to differentiate from non-specific entheseal change (e.g., Figure 5.20). The repetitive strain causes minor trauma, making bone respond with remodeling (Littleton, 1991, 1993; Maheshwari, 2006; Nelson et al., 2016). However, Meyer et al. (2011) noted that entheseal change studies often interpret sexually dimorphic differences as the effect of gendered divisions of labor, but the results may instead just reflect intrinsic sexual dimorphism rather than cultural or population-related differences. Henderson et al. (2016), for example, found that textural changes at the biceps brachii insertion are common and relate rather to normal biology than directly to bodily strain or activity. In light of this, changes to the radial tuberosity and similar attachment sites, although recorded, were not included in this research. It is also important to note that skeletal activity marker research often lacks consistency between methods and is notorious for issues with inter-observer differences, inter-sample comparison and reproducibility (Henderson, 2013). Also, the impact of age and its connection with entheseal features has not been evaluated deeply enough, thus substantially reducing the application of their study to the examination of specific activity patterns (Henderson et al., 2016). For a more nuanced perspective on the possible gendered differences seen through skeletal activity markers, a wider study using the Coimbra method (see Henderson et al., 2016) could be undertaken in the future, particularly as Palmer et al. (2018) showed that data produced using the Coimbra method is broadly comparable with the Mariotti method (Mariotti et al., 2004, 2007), the method used in this study in a modified form.

Figure 5.20 Posterior aspect of right femur, of a young adult female (ÞSK 5), with possible traumatic soft tissue changes to the greater trochanter, such as from entheseal or muscular trauma. However, a strong possible differential diagnosis is a benign neoplasm (e.g., osteochondroma exostosis)

Vitamin D deficiency

According to historical records, folklore, and supposition due to latitude, it is believed that rickets (and residual rickets) and osteomalacia (vitamin D deficiency) were prevalent in Icelandic history (e.g., Figure 5.21; Jónsson, 1998; Sigurðardóttir, 2017). However, vitamin D defieciency diseases are not often identified in Icelandic skeletal assemblages (Steffensen, 1939; Gestsdóttir, 1991; Sundman, 2011; Zoëga and Murphy, 2016). The results of the research described here also provided low rates for these conditions. Osteomalacia was observed in in six individuals (3%) and residual rickets in 13 individuals (7%); vitamin D deficiency overall was thus noted in just 10% of the sample set (Table 3 in Article III). About 32% of vitamin D deficiency cases were from Skeljastaðir and 42% were from Skriðuklaustur. There were five individuals with vitamin D deficiency from the 17th-19th century assemblages, or 26% of the total observed cases. The vitamin D deficiency frequency is thereby comparable with results of studies on adult skeletons from the British Isles dated between the 18th and 19th centuries AD: there was a prevalence of 14% (20 out of 135) for residual rickets (Brickley et al., 2010) and 1.43% (19 out of 1,323) for osteomalacia (Ives and Brickley, 2014). However, Newman and Gowland et al. (2018) found a far higher rate of rickets at some Northern sites in the British Isles. The prevalence rates from Iceland described here might be lower than expected because of the long-period of sunlight during the summer months and the dietary importance of fish and other vitamin D rich foodstuff (e.g. foraged mushrooms and Iceland moss, *Cetraria islandica*).

Osteomalacia is a notable predisposing condition for osteofluorosis (Gupta et al., 1996; Khandare et al., 2005) but diagnosis can be difficult (Ives and Brickley, 2014). Though vitamin D deficiency was equal between females (n=3) and males (n=3) at Skeljastaðir (10th-12th centuries), it varied insignificantly across females from the 13th-16th-century (n=6) and 17th-19th-century (n=4) assemblages and males (n=2 and n=1, respectively). While this may result from the small sample sizes, gendered social roles could also be indicated: women and children of the past may have spent more time indoors than men, especially in the later periods (see Norrman, 2008; Hayeur-Smith, 2014; Veselka et al., 2018). Aproximately 3% of individuals had osteomalacia, while at Herculaneum around 8% of the individuals that had fluorosis also had osteomalacia (see Petrone et al., 2013).

Figure 5.21 Femoral bowing, possibly due to residual rickets, in a young adult male (ÞSK 34) – anterior view. However, the right femur was fused to the acetabulum (detached post-mortem), causing notable angulation and immobility. Thus, it is possible that the bowing and atrophy may be due to mobility impairment rather than vitamin D deficiency

Dental Changes

Dental chipping, which may relate to poor enamel mineralisation (e.g., from dental fluorosis) (Thylstrup and Fejerskov, 1978), was present in 7% of teeth, though in this research it is likely associated primarily with diet instead of enamel mineralization. Linear enamel hypoplasia (e.g., Figure 2.9, 2.10) may occur due to childhood fluoride exposure (Thylstrup and Fejerskov, 1978; Petrone et al., 2011, 2013; Marklein et al., 2016;) and was the most common (16%) enamel defect observed in this research (mottling, 3%; hypomineralisation/opacities, 8%) (see Table 5 in Article III). Enamel hypomineralisation (opacities) (e.g., Figure 5.22) was only just higher than normally reported in healthy, modern teeth (ca. 5%) (Hillson, 1996). Hypercementosis prevalence (e.g., Figures 5.23 and 5.24) was very low (mandible, <1%; maxilla, 4%). For comparison, a very high prevalence (85% of 104) of hypercementosis was reported in 17th-19th-century slaves from Barbados by Corruccini et al. (1987). Hypercementosis is often idiopathic, but it can result from a wide range of systemic and local factors (Pinheiro et al., 2008; García-González et al., 2019) aside from dental fluorosis (Littleton, 1999). None of the individuals seen in this study with elevated bone fluoride concentrations presented with evidence of hypercementosis, but linear enamel hypoplasia, enamel hypomineralisation, dental chipping, and mottling were observed among those with pre-clinical concentrations.

Figure 5.22 Mandible of a young adult female (BES 11) with hypomineralisation (dental enamel opacities, e.g., blue arrow)

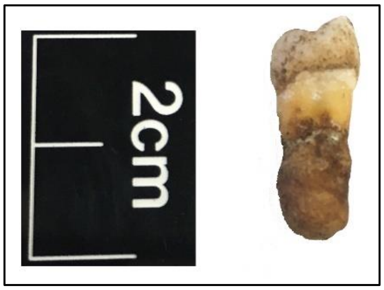

Figure 5.23 Premolar with hypercementosis of the root from a female adult (SKR 96) – lateral view

Figure 5.24 Photograph and radiograph of an adult male (HFE 18) with hypercementosis: the tooth could not be removed from the maxillary alveolar bone due to the pathologically enlarged morphology of the tooth root – lateral view

Calculus (60% maxillary and 68% of mandibular teeth; see e.g., Figures 2.9 and 2.10) and periodontal changes/alveolar resorption (ca. 72% of individuals) had high prevalence rates. However, the prevalence of carious lesions (ca. 1%) was very low. There were 286 periapical lesions (ca. 5% of all alveolar sockets) and 540 teeth lost ante-mortem (ca. 8% of all teeth) (see Table 5.10).

Table 5.10 Results of dental analyses for presence, post-mortem loss, ante-mortem loss, and periapical lesions. See Table 5 in Article III for remaining dental analyses

Maxilla						***Mandible***				
Tooth	***Present***	***Unobservable***	***AM loss***	***Periapical***		***Tooth***	***Present***	***Unobservable***	***AM loss***	***Periapical***
RM3	84/186 (45)	78/186 (42)	24/186 (13)	11/186 (6)		RM3	109/186 (59)	63/186 (34)	14/186 (8)	12/186 (6)
RM2	129/186 (69)	24/186 (13)	23/186 (12)	24/186 (13)		RM2	147/186 (79)	24/186 (13)	15/186 (8)	12/186 (6)
RM1	135/186 (73)	35/186 (19)	16/186 (9)	26/186 (14)		RM1	142/186 (76)	25/186 (13)	19/186 (10)	24/186 (13)
RPM2	134/186 (72)	36/186 (19)	16/186 (9)	8/186 (4)		RPM2	141/186 (76)	26/186 (14)	19/186 (10)	7/186 (4)
RPM1	139/186 (75)	31/186 (17)	16/186 (9)	11/186 (6)		RPM1	145/186 (78)	27/186 (15)	14/186 (8)	9/186 (5)
RC	144/186 (77)	32/186 (17)	10/186 (5)	6/186 (3)		RC	145/186 (78)	33/186 (18)	8/186 (4)	6/186 (3)
RLI	130/186 (70)	38/186 (20)	18/186 (10)	2/186 (1)		RLI	142/186 (76)	29/186 (16)	15/186 (8)	2/186 (1)
RCI	127/186 (68)	37/186 (20)	22/186 (12)	2/186 (1)		RCI	136/186 (73)	28/186 (15)	22/186 (12)	1/186 (<1)
LCI	129/186 (69)	34/186 (18)	33/186 (18)	3/186 (2)		LCI	133/186 (72)	32/186 (17)	21/186 (11)	2/186 (1)
LLI	127/186 (68)	32/186 (17)	27/186 (15)	2/186 (1)		LLI	142/186 (76)	32/186 (17)	12/186 (6)	1/186 (<1)
LC	141/186 (76)	36/186 (19)	9/186 (5)	5/186 (3)		LC	148/186 (80)	29/186 (16)	9/186 (5)	2/186 (1)
LPM1	138/186 (74)	34/186 (18)	14/186 (8)	4/186 (2)		LPM1	145/186 (78)	30/186 (16)	11/186 (6)	3/186 (2)
LPM2	139/186 (75)	36/186 (19)	11/186 (6)	3/186 (2)		LPM2	144/186 (77)	30/186 (16)	12/186 (6)	7/186 (4)
LM1	137/186 (74)	36/186 (19)	13/186 (7)	15/186 (8)		LM1	135/186 (73)	32/186 (17)	19/186 (10)	25/186 (13)
LM2	120/186 (65)	39/186 (21)	27/186 (15)	13/186 (7)		LM2	140/186 (75)	27/186 (15)	19/186 (10)	17/186 (9)
LM3	88/186 (47)	80/186 (43)	19/186 (10)	11/186 (6)		LM3	108/186 (58)	65/186 (35)	13/186 (7)	10/186 (5)
All	2041/2976 (69)	648/2976 (68)	298/2976 (8)	146/2976 (5)		All	2202/2976 (74)	532/2976 (18)	242/2976 (8)	140/2976 (5)

Key: Presence/Absence (%) pooled across all sites/periods; Present (presence of tooth); Unobservable (post-mortem loss, unerupted, other); AM loss (ante-mortem loss); Mottling (dental enamel mottling or pitting); Periapical (periapical lesions)

Respiratory disease

Sinusitis is an important indicator of respiratory health in past populations (Merrett and Pfeiffer, 2000; Sande and Gwaltney, 2004; Hedayati et al., 2007). According to the study presented here (n=186), 70% of individuals with observable sinuses (n=130) displayed evidence of either lamellar or woven new bone formation in the maxillary sinuses, indicating a high rate of sinusitis (e.g., Figure 5.25). Meanwhile, up to 60%, dependent on bone and side, of individuals showed evidence of lower respiratory disease in the form of woven or lamellar new bone deposition on the visceral aspects of the ribs. This analysis was conducted to provide additional information about the impact of volcanic particulate matter on respiratory health, but due to the widespread and diverse etiology of sinusitis it is not included as evidence of outdoor environment-related illness in this study and showed no discernible pattern by age or sex. A recent investigation concluded that the high rate of otitis media and sinusitis observed in ancient Icelandic skeletal remains is likely to be correlated with infectious diseases, especially tuberculosis, rather than from poor air quality (Collins, 2019).

Figure 5.25 Small area of remodelling (lamellar), spiculated new bone formation (within blue circle) in the right maxillary sinus of a younger adult female (BES 20) – superior view

5.3.2 Fluoride concentrations in bone

Bone fluoride concentrations are presented alongside osteological findings in Supplementary Table 1 of Article III and summary statistics for bone fluoride concentrations are presented in Table 7 and 8 of Article III. This section addresses Research Questions 1.2.7 and 1.2.8. Cumulative bone fluoride concentrations were determined in the human bone samples (n=50) and four soil samples. The fluoride concentrations of the bone samples (n=50) ranged from 223 to 4370 ppm and had a mean of 2056 ± 1112 ppm. The bone fluoride concentrations at Skriðuklaustur (n=36) ranged from 223 to 4370 ppm and had an overall mean of 2324 ± 1067 ppm. The bone fluoride concentrations at Skeljastaðir (n=14) ranged from 223 to 3030 ppm and had an overall mean of 1366 ± 937 ppm. Inter-site differences in fluoride concentrations were statistically significant (*t-test*, $p<0.002$ excluding non-adults; *t-test*, $p<0.005$ including non-adults*;* Table 8 of Article III), but intra-site differences according to sex and age categories were not. At Skriðuklaustur, bone fluoride concentrations were significantly higher than at Skeljastaðir, even across sex and age categories. The majority (24 out of 36; 67%) of individuals from Skriðuklaustur had fluoride concentrations falling within the normal range (<3000 ppm), though the remainder (12 out of 36; 33%) were elevated. While no concentrations were indicative of clinical fluorosis, some were consistent with the pre-clinical phase (>3500 ppm) were noted in 5 out of 36 (14%) of the individuals (Table 6 of Article III). The mean for non-adults from Skriðuklaustur was 1638 ± 1018 ppm. The mean for non-adults was not significantly lower (p = 0.084) than it was for the adults (2461 ± 1039 ppm). Lastly, only 2 out of 14 (14%) individulals from Skeljastaðir presented with elevated concentrations.

At Herculaneum, for example, the fluoride bone concentrations clearly indicated widespread fluorosis. The concentrations ranged between 2042 and 11342 ppm and had a mean of 6672 ppm. In contrast with the results from Iceland, the fluoride concentrations at Herculaneum were strongly correlated with increasing age (Petrone et al., 2013). In this study, all except two individuals with elevated concentrations were categorized as younger adults (YA), possibly suggesting the occurrence of increased exposure (e.g., from volcanic emissions) beyond normal age-related fluoride accumulation from consuming uncontaminated drinking water. One older female (SKR 128) had multiple vertebral fractures as well as reduced bone density observable under radiography and microscopy, indicating osteoporosis. Clinical studies have demonstrated that bone fluoride concentrations do indeed steadily increase with age (Ishiguro et al., 1993).

Women over 55 years of age had the most noticeable elevations, probably because of the concurrence of senile osteoporosis and menopause. Rather than from toxic exposure, the elevated concentration (3190 ppm) in SKR 128 likely relates to osteoporosis and age-related accumulation.

The individuals with elevated fluoride concentrations had higher than average rates of linear enamel hypoplasia (83%, 10 out of 12), enamel hypomineralisation or opacities (50%, 6 out of 12) and mottling and ante-mortem chipping (23%, 3 out of 12). The rates of interosseous and/or entheseal calcifications (50%, 6 out of 12), fractures (33%, 4 out of 12), ankyloses (16%, 2 out of 12), joint disease (25%, 3 out of 12), and atlanto-occipital membrane attachment site changes (8%; 1 out of 12), however, were not markedly different between those with elevated fluoride concentrations and the rest of the population sample when controlled for sex and age. None had ossification of the ligamentum flavum, though one individual (SKR 150) had Eagle's syndrome, displaying an elongated, ossified stylohyoid chain measuring 56.3 mm. On average, the styloid process usually measures about 20-30 mm. The symptoms (e.g., dysphagia, cervicofacial pain, cerebral ischemia) of Eagle's syndrome are clinically observed in people with elongated styloid processes longer than 40 mm on average (see Balcioglu et al., 2009; Salega and Farba, 2018). The generally low fluoride concentrations seen in the non-adults (mean 1638 ± 1018) provides further evidence that diagenesis was not at play because porous, less mineralized non-adult bones are generally particularly prone to it. Despite bone fluoride concentrations increasing with increasing age, they should remain relatively low if toxic exposure is not indicated. An older male adult (SKR 174) with Paget's disease of bone had a fluoride concentration of 2120 ppm, suggesting that osteofluorosis is an unlikely differential diagnosis (see Figures 2.7 and 2.8). The low-level uptake of fluoride among people buried at Skriðuklaustur may have resulted from volcanic emissions, normal age-related accumulation and possibly from the dietary importance of marine resources.

The absorption of fluoride into the bone hydroxyapatite can be altered or decreased by the simultaneous absorption of lead, calcium, and other trace elements. Clinical studies have shown that concurrent exposure to lead and fluoride increases lead concentrations in calcified tissues, without increasing fluoride (Whyte et al., 2008; Sawan et al., 2010; Leite et al., 2011). The individuals with treponemal disease, who also had been exposed to lead during childhood (>0.7 ppm in enamel), also had the lowest fluoride concentrations (<525 ppm F): SKR 22 (2.7 ppm Pb) and SKR 23 (4.1 ppm Pb) (see Walser III et al., 2020a). The low concentrations might partly relate to concurrent lead exposure – if it continued into adulthood – or other poisonous elements that interact with bone hydroxyapatite. Icelandic aquifers (range 1.3-52.8 ppm; median 5.5 ppm) have 3-5 times more calcium than the aquifers of other Nordic countries (Gunnarsdóttir et al., 2016) and important historic dietary staples (e.g. bone marrow, dairy) are also high in calcium. Since the uptake of fluoride can be reduced by the uptake of calcium (Whyte et al., 2008), it could be that these individuals received nourishing diets as patients of the monastery.

At Skeljastaðir, the mean bone fluoride concentration was actually lower than first expected, especially considering the site is to Mt. Hekla – a known heavy fluoride (and mercury) emitter (Thorarinsson and Sigvaldason 1972; Coderre and Steinthorsson, 1977; Thordarson and Larsen, 2007). The highest fluoride concentrations were predominately observed in those (ÞSK 4, 44) who also had the highest mercury concentrations. It may be that the individuals found to have elevated elemental bone concentrations had exposure to toxic emissions during the volcanic eruption, especially since some of the individuals post-date the eruption (AD 1104). Meanwhile, the others were perhaps only exposed to passive emissions from the surrounding area. One older adult male (ÞSK 44), for example, had a fluoride concentration of 3010 ppm in addition to interosseous and entheseal calcifications upon the long bones and osteochondritis dissecans on the right ulna (proximal end). Moreover, 14 individuals had robust entheseal changes which could potentially be correlated with mild osteofluorosis (Gestsdóttir 1998, 2009; this study).

5.4 Health and cultural implications of lead (Pb) and barium (Ba) in dental enamel and lead (Pb) in bone

5.4.1 Lead in bone

Cumulative bone lead concentrations were determined for 22 soil samples (Table 5.1), 23 faunal bones (Table 5.2), and 50 human bone samples (see Table 5.3). This section addresses Research Questions 1.2.2, 1.2.3 and 1.2.4. Across all human samples (n=50) the lead concentrations ranged from 0.52 to 93.81 ppm and had a mean of 9.49 ppm (Tables 5.4 and 5.5). At Skriðuklaustur (n=36), they ranged from 0.65 to 93.81 ppm and the overall mean was 12.6 ± 21.4 ppm. At Skeljastaðir (n=14), they ranged from 0.52 to 6.98 ppm and had an overall mean of 1.5 ± 1.7 ppm (Table 5.4). The samples from Skriðuklaustur had higher bone lead concentrations than at Skeljastaðir, but this difference was not statistically significant (Table 5.5). However, lead concentrations among males from Skriðuklaustur were significantly higher (*t-test, p=0.02)* than concentrations in males from Skeljastaðir. From Skriðuklaustur, 14 out of 36 (39%) of individuals exhibited elevated lead concentrations (>7 ppm) while the remainder (22 out of 36; 61%) had normal concentrations. Furthermore, a little over half (8 out of 14; 57%) of the Skriðuklaustur indviduals with elevated bone lead concentrations also represent 72% (8 out of 11) of the individuals that had elevated lead concentrations in their dental enamel (Table 5.6). This implies that these individuals were likely exposed to lead not only in childhood but also as adults. Meanwhile, at Skeljastaðir, only one (1 out of 14) individual (ÞSK 44, 7 ppm) had an elevated lead concentration. Considering his migrant status (see Price and Gestsdóttir, 2006), it is possible that the lead exposure did not actually occur at Skeljastaðir, but elsewhere prior to his migration to the farm site.

According to Rasmussen et al. (2015) bone lead concentrations may be considered elevated if they exceed >7 ppm in trabecular bone and >5ppm in cortical bone and post-mortem diagenesis is not indicated. In this study, bone lead concentrations were only considered elevated if they exceeded 7 ppm, although trabecular bone was not analysed. From Skriðuklaustur, 39% (14 out of 36) of individuals had bone lead concentrations greater than 7 ppm; however, some parts of the cemetery appear to have been contaminated by lead objects (e.g., tools) and infrastructure (e.g., window frames) potentially indicating that diagenesis may have altered the bone lead concentrations post-mortem. Nevertheless, it is interesting to note that 57% (8 out of 14) of those individuals with elevated bone lead concentrations also exhibit elevated dental enamel concentrations, which may imply that these individuals were exposed to lead both during childhood and throughout life or close to the time of death, if diagenetic uptake did not occur post-mortem within the burial environment. Exposure to lead at Skeljastaðir was probably lower because less trade/commerce occurred there (or during that time period), compared with Skriðuklaustur, which was occupied later. At Skriðuklaustur, due to the monastic functions and its role as a center of commerce, the use of materials such as lead for structures and objects was probably far more common and accessible than at the earlier farm site Skeljastaðir.

5.4.2 Lead and barium in dental enamel

This section addresses Research Questions 1.2.2, 1.2.3 and 1.2.4. The dental enamel trace element analyses showed low barium concentrations, likely because seawater and Icelandic groundwater and basalt are low in barium content (see Naimy, 2008) (Table 5.6). The low values, little variation, and small range in barium concentrations also further indicate that the people of Skriðuklaustur were of local origin. The means for lead concentrations are higher than they were for barium, which may suggest lead exposure (see Liu et al., 2013) from anthropogenic sources at Skriðuklaustur (Supplementary Figure S1 of Article II). No individuals presented with elevated or outlying barium concentrations. However, one individual happened to have both the highest strontium and barium concentrations in dental enamel. Twelve sampled individuals had a lead content higher than ca. 0.7 ppm (Tables 5.6 and 5.8), the concentration that is usually thought to be the threshold for lead exposure in archaeological human dental enamel (Montgomery et al., 2010; Millard et al., 2014).

All the samples presenting with anthropogenically elevated lead (e.g., from inhaled lead dust/paint, skin contact, or ingestion of contamined food or soil) were from non-adults or individuals that are biologically female with the exception of the older adult male with Paget's disease of bone (SKR 174). These findings may indicate differing routes of exposure due to the life course in general or gendered socio-cultural roles. Women may have spent more time inside, as it is thought that they were especially responsible for housekeeping, preparing food, and the production of woven textiles, which were important both as a currency (*vaðmál*) and for clothing and other goods. Textile production was highly standardised because it was essential for paying taxes, tithes, and other economic and legal transactions. It was an important occupation and form of female agency in historic Iceland that was potentially regulated by law to some degree (Norrman, 2008; Hayeur-Smith, 2014). On the other hand, biological differences between men and women (e.g., menopause or pregnancy) also play an important role in the retention and susceptibility to negative health impacts resulting from exposure to heavy metals (Vahter et al., 2007).

A young adult female (SKR 189) with cystic echinococcosis (hydatid disease) had a lead concentration of 9.40 ppm in bone, which was significantly higher than the rest of sample set. She also had the highest lead concentration in enamel, indicating childhood lead exposure occurred, which may have then continued into adulthood. The second highest lead concentration (4.1 ppm) was from a young adult (ca. 17-25) female (SKR 23) displaying skeletal evidence of treponemal disease, consistent with probable venereal syphilis (see Hackett, 1976; Ortner, 2003). From the 17th century onwards lead-glazed goods became much more common and accessible in Iceland, though they remained generally associated with high-status sites (Þorgeirsdóttir, 2010). For context, Rasmussen et al. (2015) measured lead concentrations in archaeological assemblages from monastic, rural, and urban sites in northern Germany and Denmark. The results showed that people with high social status were more likely to reside close to or within lead buildings as well as be able to afford lead or lead-glazed goods. One of the few individuals (SKR 65) buried inside the church, which may suggest that she held some special status there (e.g., as a benefactor), had a lead concentration of 3.51 ppm (Kristjánsdóttir, 2010; Walser III et al., 2019). Thus, some anthropogenic exposure likely occurred on site, particularly among those residing there in childhood. However, with just a few exceptions, the lead enamel concentrations are mostly under ca. 0.7 ppm, implying that the samples from Skriðuklaustur represent people that were raised in a generally unpolluted environment, such as Iceland (see Montgomery et al., 2014; Walser et al., 2020a).

5.5 Health implications of cadmium (Cd) and arsenic (As) concentrations in bone and dental enamel and antimony (Sb) in dental enamel

Overall, the low concentrations of cadmium and arsenic in bone and dental enamel and antimony in dental enamel indicate that these individuals lived within an environment and culture that did not heavily rely on these elements for medicine, production, or trade. If these elements were used in society, this was likely limited to artisans or individuals working within specialized centers such as schools or monasteries. Furthermore, the low concentrations and little variability in concentrations of each element seen between individuals corroborates the interpretation that the sampled individuals represent a local group of people that were born and grew up in Iceland.

5.5.1 Cadmium in bone

Cumulative cadmium concentrations were analysed in 22 soil samples (Table 5.1), 23 faunal bones (Table 5.2), and 50 human bone samples (Table 5.3). This section addresses Research Question 1.2.3 and 1.2.4.

The cadmium concentrations of all human samples (n=50) ranged from <0.03-7.14 ppm with a mean of 1.13 ppm. At Skriðuklaustur (n=36), the bone cadmium concentrations ranged between <0.03-7.14 ppm and the overall mean was 1.23 ± 1.39 ppm. At Skeljastaðir (n=14), the bone cadmium concentrations ranged between 0.09-4.06 ppm and the overall mean was 0.88 ± 1.31 ppm (Tables 5.3 and 5.4). Overall, the bone cadmium concentrations were slightly higher at Skriðuklaustur than at Skeljastaðir, but inter-site differences and differences between sex and age groups in cadmium concentrations were not statistically significant (Table 5.5).

According to the results of this research, some individuals may have been exposed to cadmium close to the time of their death. Assuming no diagenetic processs were at play, 16 out of 36 (44%) of individuals from Skriðuklaustur exhibited elevated cadmium concentrations (>1 ppm) while the remainder (20 out of 36; 55%) had normal concentrations. At Skeljastaðir, two out of 14 individuals had elevated concentrations, while the rest had normal cadmium levels. Research by Martinez-García et al. (2005) on Medieval skeletal remains from Spain found that cadmium concentrations were typically higher in ribs than in other skeletal tissues. This factor further demonstrates that the values determined in this research are relatively low, particularly considering that some of their results were as high as 19.0 ppm (Martinez-García et al., 2005). However, cadmium and lead concentrations have been observed to be significantly correlated in other bioarchaeological studies (e.g., González-Reimers et al., 2003). It is therefore possible that the elevated bone cadmium concentrations observed at Skriðuklaustur in this study could be correlated with lead pollution at the monastery, particularly considering the low prevalence of elevated cadmium concentrations at Skeljastaðir where lead pollution was not indicated. Although elevated concentrations could reflect exposure to volcanogenic cadmium, other sources such as smoke inhalation, tobacco smoking and working with objects containing cadmium are more likely to be the cause when considered in combination with the other bone element concentrations.

5.5.2 Arsenic in bone

Cumulative bone arsenic concentrations were determined in 22 soil samples (Table 5.1), animal bones (Table 5.2) and all the human bone samples (Table 5.3). This section addresses Research Question 1.2.3 and 1.2.4. The arsenic concentrations of all samples (n=50) ranged between <0.003-0.947 ppm with a mean of 0.17 ± 0.18 ppm. At Skriðuklaustur (n=36), the bone arsenic concentrations ranged between 0.003-0.947 ppm and the overall mean was 0.21 ± 0.20 ppm. At Skeljastaðir (n=14), the bone arsenic concentrations ranged between 0.033-0.172 ppm and the overall mean was 0.08 ppm ± 0.05 (Table 5.4). Overall, the bone arsenic concentrations were significantly higher (*t-test, p=0.02)* at Skriðuklaustur than at Skeljastaðir. The males at Skriðuklaustur had significantly higher arsenic concentrations (*t-test, p=0.02)* than the males at Skeljastaðir and this was the same for younger individuals (*t-test, p=0.05)*. No other inter-site differences or differences between sex and age groups were statistically significant (Table 5.5). None of the individuals analysed in this research presented with concentrations indicative of toxicity (>1 ppm), although toxicity has been noted in modern individuals within the range of 0.5-1.0 ppm. If this range is used as an indication of potentially elevated arsenic levels, then two individuals (SKR 144, 0.947 ppm; SKR 65, 0.782 ppm) from Skriðuklaustur may have been exposed to arsenic. However, considering the propensity of arsenic to infiltrate bone diagenetically, such interpretations must be made cautiously regardless of the extremely low background levels found in the natural environment in Iceland.

Since arsenic toxicity normally occurs with concentrations greater than 1 ppm (Swift et al., 2015), the results of this study indicate that none of the analysed individuals suffered from arsenic toxicity. However, seven individuals from Skriðuklaustur had notably higher concentrations than the others, including one older adult female (SKR 144) that presented with a bone arsenic concentration of 0.947 ppm, which is just under the threshold of potential toxic exposure. Additionally, a younger adult female (SKR 65) also presented with a slightly elevated bone arsenic concentration (0.782 ppm). This individual, who was one of just seven people buried within the church itself, also had elevated cadmium (1.330 ppm), mercury (0.476

ppm) and lead (37.3 ppm) bone concentrations, all of which may be correlated with a special status as a benefactor, layperson, or medical practitioner within the monastery. Furthermore, arsenic is a byproduct of tin and copper mining and it is also commonly found in sulphur and other metals. Since refined sulphur was found at Skriðuklaustur (Walser III et al., 2019) and was an important commodity sourced in Iceland (Mehler, 2016), it is possible that elevated arsenic concentrations in the people buried at Skriðuklaustur are correlated with medicinal uses of sulphur or with occupational exposure through sulphur mining and processing. On the other hand, at Skeljastaðir, none of the individuals showed increases in bone arsenic concentrations, which is worth noting considering its proximity to use and during a volcanic eruption. This may indicate that anthropogenic activities (e.g., medical preparations, sulphur mining or refining) were a more likely source of arsenic exposure than environmental emissions in Medieval Iceland. However, despite extremely low background levels of arsenic in the natural environment in Iceland, arsenic is known to infiltrate bone diagenetically and thus such interpretations must be made cautiously.

5.5.3 Arsenic, cadmium, and antimony in dental enamel

This section addresses Research Questions 1.2.2, 1.2.3 and 1.2.4. Dental enamel concentrations of arsenic have been reported in ancient and contemporary human teeth as ranging between <0.001-0.406 ppm (Rasmussen, 1974). In this study, all individuals presented with detectable concentrations of arsenic in enamel that ranged between 0.005-0.124 ppm, indicating that none of them were substantially exposed to arsenic during childhood (Table 5.6 and 5.8). Similar to the cadmium concentrations found in dental enamel, the concentrations determined in this research are well below the baselines determined in ancient and modern human teeth, thereby indicating that the sampled individuals were not substantially exposed to arsenic during childhood dental development.

In this study, 19 out of 31 individuals presented with detectable concentrations of cadmium in enamel. The 19 samples with detectable cadmium concentrations ranged between 0.002-0.138 ppm (Tables 5.6 and 5.8). Few studies have assessed cadmium concentrations in dental enamel, but in modern non-adult dental enamel Bayo et al. (2001) found cadmium concentrations to range from 0.007-0.610 ppm and Tvinnereim et al. (2000) found a mean of 0.113 ± 392 ppm. It is unlikely that substantial exposure to cadmium occurred during childhood in any of the individuals sampled in this study considering the results fall within a range that is lower than the baseline concentrations determined in modern individuals. This indicates that cadmium exposure among the individuals analysed here did not exceed normal background limits.

Dental enamel concentrations of antimony have been reported in ancient and contemporary human teeth as ranging between <0.001-1.59 ppm (Rasmussen, 1974). In this study, 30 out of 31 individuals presented with detectable concentrations of antimony in enamel that ranged between 0.002-0.181 ppm, indicating that none of them were substantially exposed to antimony during childhood (Tables 5.6 and 5.8). The dental enamel antimony concentrations determined in this study are far lower than the upper limits of the baselines reported by Rasmussen (1974).

5.6 Origins and elements

5.6.1 Health and cultural implications of mobility or migration according to bone collagen isotope ($\delta^{34}S$) and dental enamel isotope ($^{87}Sr/^{86}Sr$, $\delta^{18}O$) analyses

This section addresses Research Questions 1.2.1, 1.2.2 and 1.2.4. The $\delta^{18}O$, $\delta^{13}C$ and $^{87}Sr/^{86}Sr$ (n=31) from enamel samples can be seen in Table 5.6 and isotope ratio mean averages and ranges in Table 5.7. The human samples from Skriðuklaustur range from 0.7060 to 0.7088 and all fall within the lower geological end-member of basalt and the upper end-member of the Icelandic biosphere of rain and seawater, i.e. 0.7030 to 0.7092. The animals and humans were all consistent with geographic provenance in Iceland or other areas of marine or basaltic limestones, the only two rock types that produce biosphere values under 0.7092 (Figure 5.26). The human $\delta^{18}O_{carbonate}$ values range from 22.8 to 25.0 ‰, mean = 24.2 ± 0.6‰. Using the equations provided in Chenery et al. (2012) ($\delta^{18}O_{phosphate} = 1.0322 \times \delta^{18}O_{carbonate} - 9.6849$ and $\delta^{18}O_{drinkingwater} = 1.590 \times \delta^{18}O_{carbonate\ (VSMOW)} - 48.634$), this equates to a $\delta^{18}O_{phosphate}$ range of 13.9 to 16.1‰ and mean of 15.3 ± 0.6‰. The $\delta^{18}O_{drinkingwater}$ values range from -12.3 to -8.9‰. Converting enamel $\delta^{18}O$ to precipitation significantly raises the uncertainty of individual measurements, which is ±1‰ (2SD) according to Chenery et al. (2012). Nevertheless, the range for human dental enamel falls into the annual Icelandic $\delta^{18}O$ range for modern precipitation (-13‰ to -8‰) (see Price et al., 2015: Fig. 20; Bowen, 2018). It is also close to the values seen in groundwater in modern contexts (range -8.2 to -8.8; n=11) (Friedrich and Schlosser, 2013). Water sources in the North Atlantic region may have been affected by climate changes in the past, such as those that occurred during the Little Ice Age (Fricke et al., 1995; Daux et al., 2005, 2008), but these results provide a $\delta^{18}O_{dw}$ range exceeding 3‰ at -12.3 to -8.9 ± 1‰ (2SD) for Medieval humans of apparent Icelandic geographic provenance based upon their $^{87}Sr/^{86}Sr$ ratios (Article II).

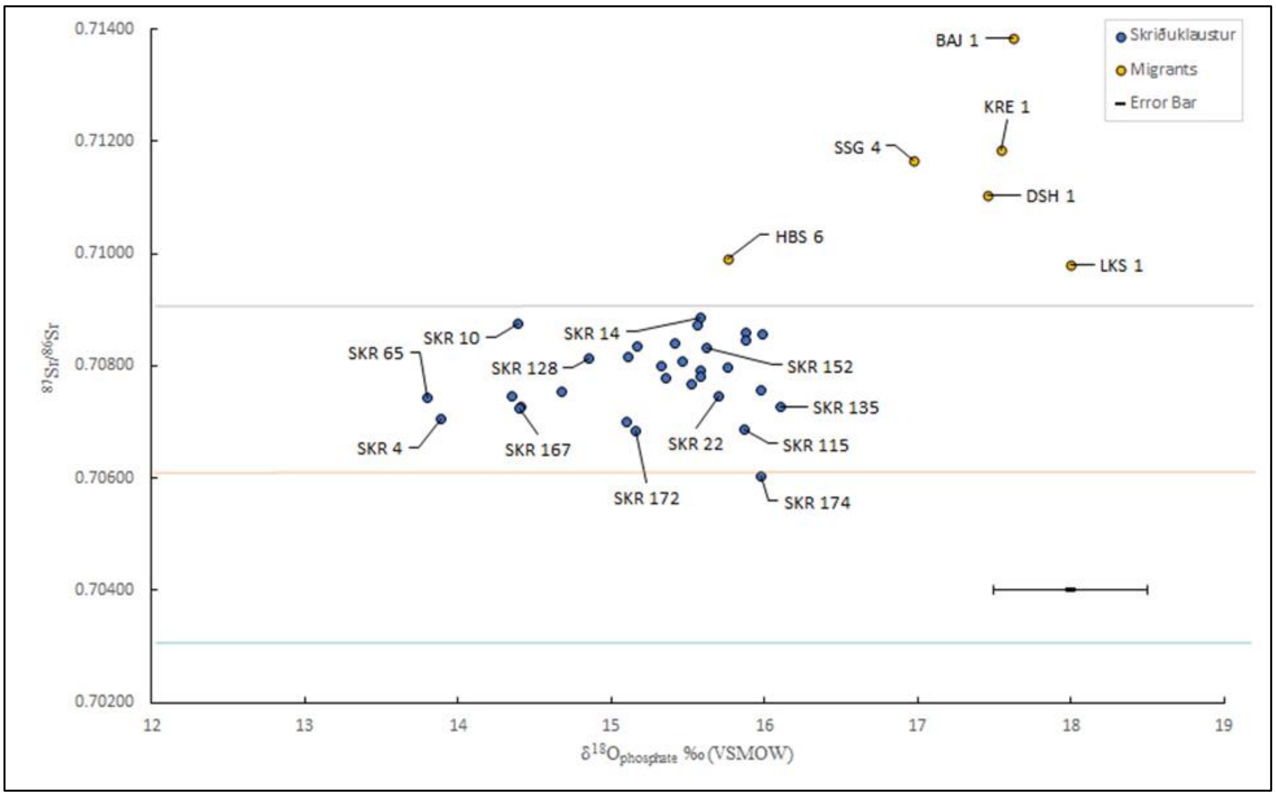

Figure 5.26 A plot of $\delta^{18}O_{phosphate}$ and $^{87}Sr/^{86}Sr$ values for the enamel samples from individuals buried at Skriðuklaustur (SKR; dark blue). For comparison, Settlement period migrants are indicated by the light-yellow markers (Gestsdóttir and Price, 2006 except LKS 1). LKS 1 indicates the settlement period migrant known as Bláklædda konan (Montgomery and Jakob, 2015). Icelandic bedrock (0.703), sheep (0.706) and seawater (0.7092) are indicated by the horizontal lines (also see Table 3.1). Error bar for standard deviation for $\delta^{18}O$ at ±0.5‰ and $^{87}Sr/^{86}Sr$ at 0.002

The geographic provenance of the people buried in the temporally constrained cemetery at Skriðuklaustur (AD 1493-1554) was unclear prior to this research (Article II), but it was thought they might have been a mixture of locals, foreign traders, patients, and pilgrims seeking treatment, hospice and commerce, particularly in light of the site's hospital status and international network (Kristjánsdóttir, 2012, 2017). The oxygen and strontium isotope findings indicated that the sampled individuals were all of local provenance (no samples exceeded the $^{87}Sr/^{86}Sr$ value of rain and seawater, 0.7092). However, it is still possible that some of them first resided on chalk, which has a range of 0.708-0.709 (e.g., Southern Britain and Denmark) (see Evans et al., 2010; Evans et al., 2012; Montgomery et al. 2014). Nevertheless, the $\delta^{18}O_{phosphate}$ values are not consistent with a geographic provenance in Ireland or Britain, where $\delta^{18}O_{phosphate}$ would then fall into the range of 16.3‰ to 19.1‰ (see Montgomery et al., 2014). Additionally, the $\delta^{18}O_{phosphate}$ range is ca. 2‰, the standard range of a single temporally contemporaneous population. When $^{87}Sr/^{86}Sr$ approaches the value of sea and/or rainwater, $\delta^{13}C_{carbonate}$ values move away from a terrestrial value thus indicating greater marine-derived carbon and strontium input in the diet (Figure 5 of Article II). It must be remembered though that $\delta^{13}C_{carbonate}$ reflects whole diet (e.g., protein, carbohydrates, fat) instead of just the protein portion of it. The $\delta^{13}C_{carbonate}$ and $^{87}Sr/^{86}Sr$ variability might suggest though that some individuals (e.g., SKR 167) subsisted on a wholly terrestrial diet while also residing further away from the coast (lower $\delta^{13}C_{carbonate}$, more basaltic-derived $^{87}Sr/^{86}Sr$ values) in childhood. Meanwhile, those with higher $\delta^{13}C_{carbonate}$ and more marine-derived $^{87}Sr/^{86}Sr$ values (e.g., SKR 10 and 14) seem to have subsisted on seafood and resided closer to the coast where seaspray and splash were more prevalent. This positive correlation is reinforced by the Sr concentrations, that are likewise positively correlated with $\delta^{13}C_{carbonate}$ and $^{87}Sr/^{86}Sr$: enamel Sr content increases while Sr isotope ratios move towards the seawater value (see Figure 5.26). This implies that the higher values are associated with people that grew up in areas close to the coast where the consumption of seaweed and seaweed-grazing fauna and marine seasplash and spray affected the foodweb. On the other hand, individuals with lower ratios and concentrations lived further from the coast in inland areas where the food chain was dominated by basalt. Though there is a logical correlation between these three parameters, it is only rarely seen this clearly in human populations. These findings imply that isotope analyses could even be used to identify geographic provenance within Iceland.

The older adult male (SKR 174) that had bone changes associated with Paget's disease of bone (e.g., endocranial "cotton wool" appearance, cranial diploë expansion, diffuse and abnormal new bone formation on the long bones and cranium) (see Ortner, 2003) had the lowest $^{87}Sr/^{86}Sr$ value, which suggests that he was originally from an inland area (see Figure 5.26 and Figure 5 of Article II). Clinical research shows that patients with Paget's disease of bone involving the femur, tibia, or acetabular region of the ilium experience statistically and clinically significant mobility and functional impairments (Lyles et al., 1995). In addition to mobility limitations, it frequently leads to muscular atrophy and sensory and psychological (e.g., deafness, dementia) impairments (Monsell, 2004; Kimonis et al., 2008) which can increase the risk of social disability (see Roberts, 2000). They may have relocated to the monastery from an inland area to seek medical assistance, possibly aided by members of his local community. Medieval Icelandic monasteries, according to the written record, were each responsible for designated areas of the country. Thus, people went to the monastery within their region when they required their services (Kristjánsdóttir, 2016; 2017). Sometimes corpses were transported to the designated monastery from the place of death to receive funerary services and proper burial rites (Kristjánsdóttir, 2017: 134, 248-249). Though Icelandic monasteries did not all function as hospitals, they were principally required to deliver aid to those seeking it, especially travelers and the poor or homeless (Kristjánsdóttir, 2016; 2017). Since Skriðuklaustur was responsible for the south-eastern quarter of Iceland (Kristjánsdóttir, 2016), at least some of the samples from there are likely to represent individuals coming from somewhere within this district. Meanwhile, from Skeljastaðir, only one sampled individual (ÞSK 39) (n=33) was of foreign geographic provenance (Price and Gestsdóttir, 2006). They also had a substantially higher $\delta^{15}N$ value than the rest of the sample set, both in the results presented here and in those presented by Sveinbjörnsdóttir et al. (2010) (Figures 5.27 and 5.28). This dietary difference could thereby be related to their non-local geographic origin and the diet they ate before they relocated and resided at Skeljastaðir.

5.6.2 Other indicators of ancestry, provenance, and environmental influences

According to the study presented here (n=186), 63% (95% confidence interval, 55% to 70%) of the analysed skeletal individuals dated between the 10th-19th centuries exhibited mandibular torus, while 40% (95% condence interval, 32% to 49%) exhibited palatine torus. In addition, 27% (95% confidence interval, 20% to 34%) of these individuals also exhibited maxillary tori. The decreased prevalence in oral tori from the ancient to modern Icelanders is unlikely to be associated with ancestry-linked genetic components, rather, it is more likely to be due to a decrease in environmental stressors and significant changes in subsistence. Scott et al. (2016) found that archaeological Greenlandic Norse and Icelanders had a high frequency of morphologically pronounced (65-97%) mandibular torus, while comparative Medieval Norwegians had a significantly lower frequency (48%) and Danish Viking skeletons had a very low rate (9%). Axelsson and Hedegaard (1981) found that modern Icelanders (n=976) had a mandibular torus prevalence of 13% in one region (n= 213; North Þingeyjarsýsla) and 30% in another (n=763; South Þingeyjarsýsla), notably lower than prevalence rates found in archaeological individuals from Settlement Period Iceland. Since the prevalence of mandibular torus has decreased over time and considering that Norse ancestry increased from the Settlement period to the present (see Ebenesersdóttir et al., 2018), any heredity association with this trait would have to be positively linked with Gaelic ancestry. Although earlier family studies (e.g., Moorrees et al., 1952; Suzuki and Sakai, 1960) suggested a hereditary origin of mandibular torus (autosomal dominant mode of inheritance), they did not consider diet, socio-economic status and other variables that may be shared within or between families in each population. Recent evidence rather indicates the strong environmental, dietary, and mechanical components of variance associated with the expression of mandibular (Scott et al., 2016; Baumann et al., 2017) and palatine torus (Halffman and Irish, 2004).

5.7 Dietary health implications according to bone collagen isotope ($\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$) and dental enamel isotope ($\delta^{13}C_{carbonate}$) analysis, dental enamel trace element (Zn) analysis and osteological analyses

5.7.1 Diet implications of bone collagen ($\delta^{13}C$, $\delta^{15}N$, $\delta^{34}S$) and enamel ($\delta^{13}C_{carbonate}$) analysis

This section addresses Research Questions 1.2.5 and 1.2.6. C:N atomic ratios (SKR C:N atomic mean of 3.2, ÞSK C:N atomic mean of 3.3) were between 3.0 and 3.4 for all human and animal bone samples, which provided well preserved bone collagen (see Ambrose, 1990; DeNiro, 1985). While no significant differences in $\delta^{13}C$ or $\delta^{15}N$ values were found at either site across age groups or between males and females, one male from Skeljastaðir (ÞSK 44, higher) and two males from Skriðuklaustur (SKR 135, lower, and SKR 172, higher) showed outlying $\delta^{34}S$ values. Descriptive statistics are presented in Table 5 of Article II. Following the values indicated by Arneborg et al. (1999), which are derived from research performed on humans from Canada, Norway, Sweden, and western Greenland (see Sveinbjörnsdóttir et al., 2010), the Skeljastaðir assemblage (n=14) showed a terrestrial dietary signal (closer to the terrestrial dietary end-member of -21‰) while the Skriðuklaustur assemblage (n=36) presented with a range and overall $\delta^{13}C$ mean indicative of a marine diet (closer to the marine dietary end-member of -12.5‰). The $\delta^{13}C$ and $\delta^{15}N$ results found by Sveinbjörnsdóttir et al. (2010) from individuals analysed from Skeljastaðir are included in the means, thus increasing the total number of sampled individuals to n=24 (excluding $\delta^{34}S$ values). Despite the nearly identical $\delta^{15}N$ values Sveinbjörnsdóttir et al. (2010) reports, their $\delta^{13}C$ sample values are 1-2‰ higher than the values found in this research. A 1-2‰ offset of $\delta^{13}C$ was even observed among the three samples which were re-analysed during this study (ÞSK 16, 34 and 48). Thus, when the two sets of data are examined together it becomes clear that the overall $\delta^{13}C$ mean at Skeljastaðir is elevated by ca. 1‰. The offset is likely the result of inter-laboratory differences.

The nitrogen and carbon isotope ratios (bone collagen) from adult individuals sampled from Skriðuklaustur indicated a diet dominated by mixed C_3 terrestrial and marine protein. The significantly different diet consumed at Skeljastaðir, on the other hand, was C_3 terrestrial protein dominated. The $\delta^{13}C$, $\delta^{15}N$ and $\delta^{34}S$ mean values derived from faunal bones (n=25) from Skriðuklaustur can be seen in Table 6 of Article II and plotted in Figures 5.27 and 5.29. As no faunal bones from Skeljastaðir were available for sampling, the human isotope results from there are only comparable with animal baselines from Skriðuklaustur (Figure 5.27 and Table 6 of Article II) and other Icelandic sites (Table 2 of Article II). With *Canidae* species, a single sample showed a notably low $\delta^{15}N$ (1.6‰) value, yet the other showed a substantially higher $\delta^{15}N$ (9.1‰). This difference could imply that these two samples are from different species (e.g., wild arctic fox and domestic dog) or that they could be from the same species with different diets. Also, two domestic goat or sheep samples presented with notably higher carbon and nitrogen isotope ratios ($\delta^{13}C$ ca. -13.8, $\delta^{15}N$ ca. 14.3, $\delta^{34}S$ ca. 13.4) than the others (n=5) (mean $\delta^{13}C$ -21.6 ± 0.5‰, $\delta^{15}N$ 2.5 ± 1.3‰ and $\delta^{34}S$ 4.1 ± 1.6‰), implying that they heavily consumed marine resources (see Schulting et al., 2017). Sayle et al. (2013). Sayle et al. (2013), for comparison, published sheep or goat samples with means of $\delta^{13}C$ -21.2 ± 0.4‰, $\delta^{15}N$ 2.5 ± 1.1‰ and $\delta^{34}S$ 6.7 ± 1.9‰ but no $\delta^{15}N$ outliers in were observed. These findings indicate that, for domestic animals, different feeding strategies were practiced.

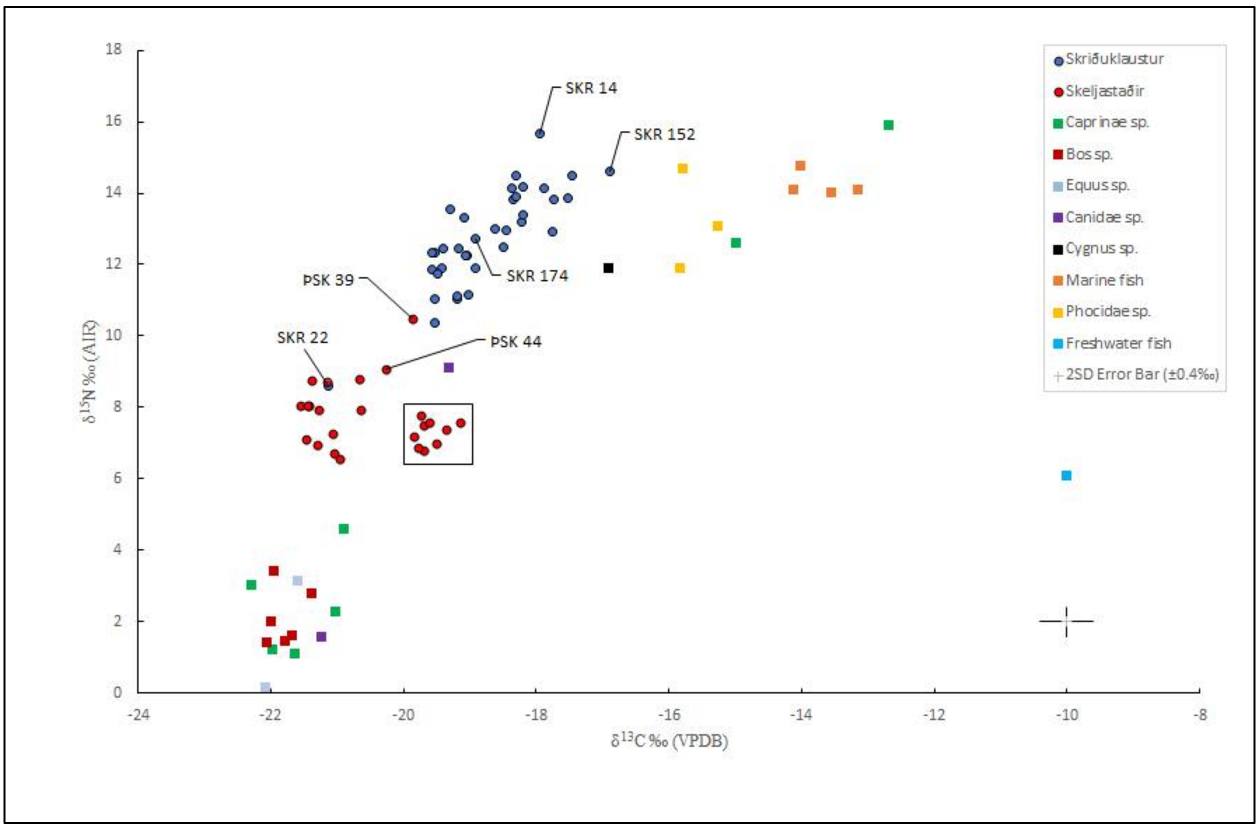

Figure 5.27 Plot of $\delta^{13}C$ versus $\delta^{15}N$ values for sampled individuals from Skriðuklaustur and Skeljastaðir. Also plotted are the $\delta^{13}C$ versus $\delta^{15}N$ values of the archaeological animal bone samples from Skriðuklaustur, except for freshwater fish, which were reported in Sayle et al. (2013). The data points within the black square and ÞSK 39 represent individuals from Skeljastaðir as reported in Sveinbjörnsdóttir et al. (2010). 2SD error ±0.4‰

The $\delta^{13}C_{carbonate}$ values, representing childhood whole diet, have a mean of -15.2 ± 0.8‰ and range from -16.6 to -12.9‰. Aside from SKR 10 and SKR 14, all the individuals were in the expected range of a diet predominately acquired from C_3 terrestrial resources (-17.0 to -14.0 ‰; see Kellner and Schoeninger, 2007; Froehle et al., 2012; Neil et al., 2017). When the $\delta^{13}C_{carbonate}$ values are plotted against the $\delta^{13}C_{collagen}$ it may be seen that all sampled individuals fell between the C_4/marine protein line and the C_3 protein line, further demonstrating the mixed marine and terrestrial subsistence strategy held at Skriðuklaustur (Figure 5.28).

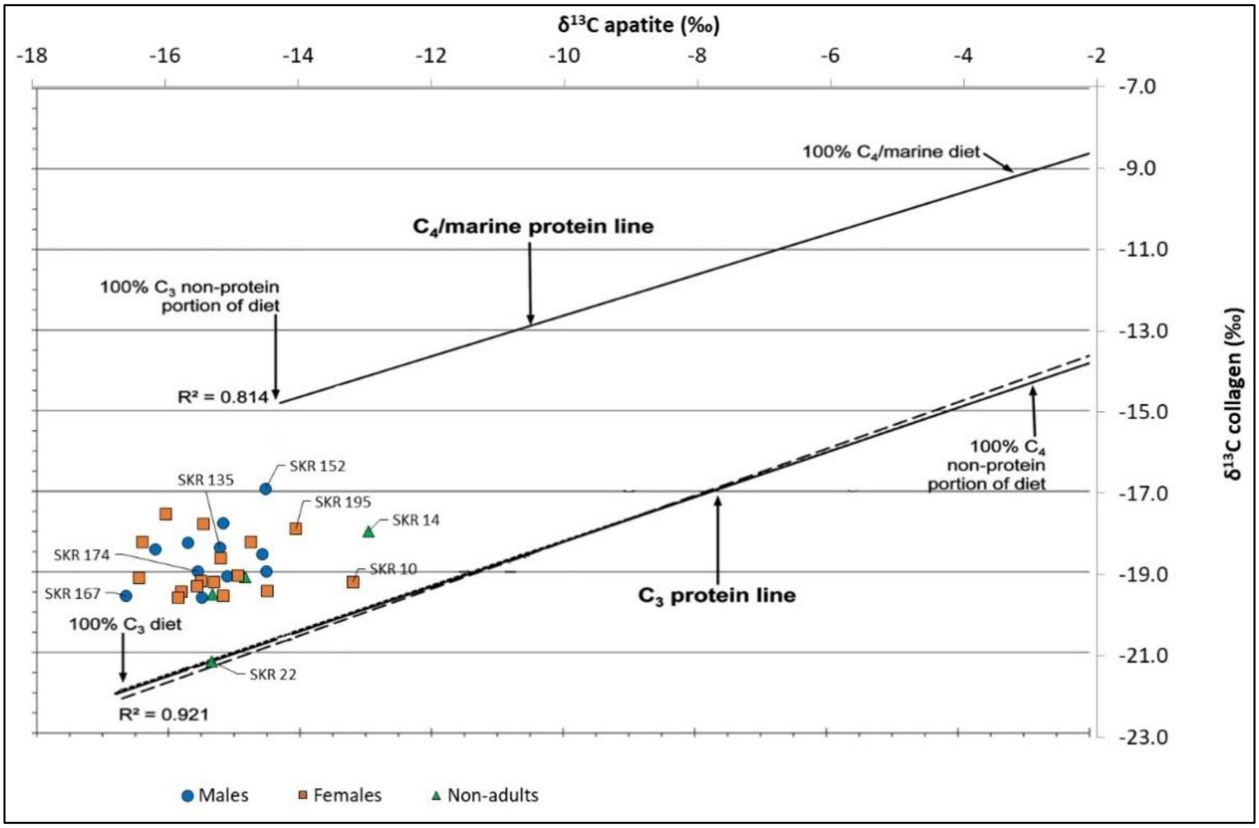

Figure 5.28 $\delta^{13}C_{carbonate}$ vs. $\delta^{13}C_{collagen}$ plot of individuals from Skriðuklaustur

As previously shown by Sveinbjörnsdóttir et al. (2010), subsistence at Skeljastaðir was predominately based on terrestrial protein resources. During the Medieval Warm Period, within just a few decades of AD 1000, North Atlantic sea fishing increased substantially (Barrett et al., 2004). At Skeljastaðir there was a low input of marine resources, likely because subsistence strategies focused on sea-fishing were less common at the time of the site's occupation (AD 1000-1104). It is however possible that this just relates to the long geographical distance (ca. 60 km) between Skeljastaðir and the coast. Similarly, $\delta^{13}C$ and $\delta^{15}N$ isotope analyses conducted on skeletal remains from the United Kingdom by Müldner and Richards (2007) showed that High Medieval groups subsisted on significantly more marine resources than earlier populations and that church-imposed regulations regarding dietary fasting were likely responsible. While no documentary evidence about Skeljastaðir is available, archaeological data demonstrates that as many as 15 other farms existed in the valley before the Hekla eruption of AD 1104. The valley is a verdant grazing land and has numerous freshwater sources containing fish (Steffensen, 1943; Þórðarson, 1943; Dugmore et al., 2007; Gestsdóttir, 2014). The sulphur isotopes ($\delta^{34}S$), when compared with bone collagen derived $\delta^{13}C$ and $\delta^{15}N$ values, indicate that some individuals from Skeljastaðir consumed freshwater protein in a greater quantity than others, while individuals from Skriðuklaustur subsisted more heavily on a mixture of fresh and saltwater resources (see Figure 5.29).

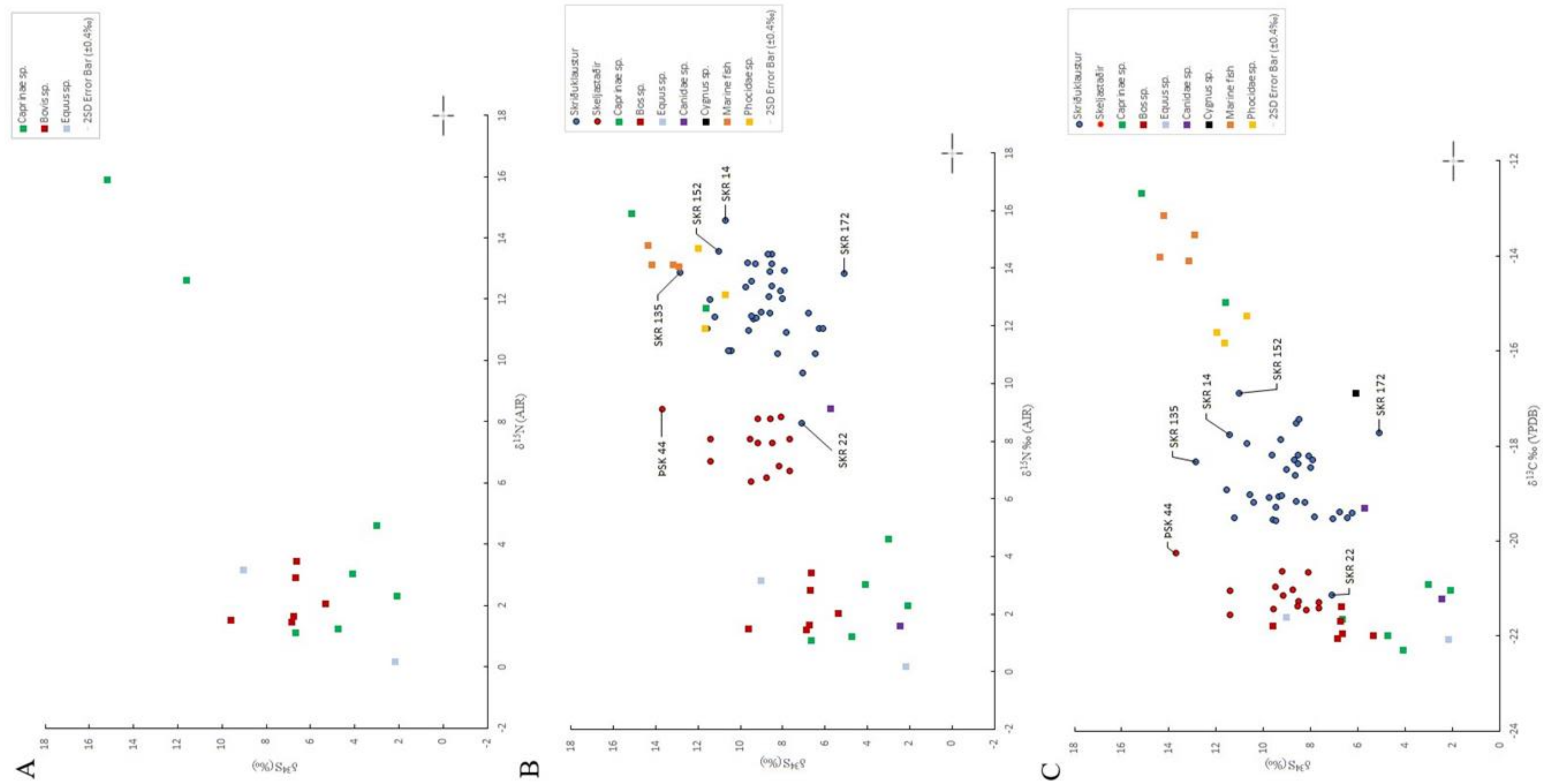

Figure 5.29 A. Plot of $\delta^{15}N$ versus $\delta^{34}S$ values determined from the sampled herbivores from Skriðuklaustur. B. Plot of $\delta^{15}N$ versus $\delta^{34}S$ values determined from the archaeological human and animal bone samples from both sites. C. Plot of $\delta^{13}C$ versus $\delta^{34}S$ values determined from the archaeological human and animal bone samples from both sites. Freshwater fish (n=12; not shown on plots) exhibit mean $\delta^{13}C$ values of -9.8 ± 0.6, $\delta^{15}N$ values of 5.9 ± 0.6 and $\delta^{34}S$ values of -2.7 ± 1.4, according to Sayle et al. (2013). There was not enough collagen to measure $\delta^{34}S$ for SKR 174

The carbon and nitrogen results derived from human bone collagen from Skriðuklaustur line up with zooarchaeological findings and historical evidence, showing a subsistence strategy high in marine and possibly freshwater fish protein. No sex differences were observed in dietary intake. Following a trophic shift of +1‰ for carbon and 5.5 ± 0.5‰ for nitrogen (see Fernandes, 2015; Sayle et al. 2016), which results in a $\delta^{13}C$ value of -20.7‰ and a $\delta^{15}N$ value of 7.7 ± 0.5‰, none of the humans sampled from Skriðuklaustur consumed a completely terrestrial protein diet during adulthood. If the two fishmeal or seaweed grazing sheep or goats (see Figure 5.27) are included, however, then a +5.5 ± 0.5‰ trophic shift provides a $\delta^{15}N$ value of 9.2 ± 0.5‰. In that case, a single individual (SKR 22; 16.5-18.5 years old) with a significantly lower $\delta^{15}N$ value (8.6‰) subsisted on a solely terrestrial diet. As the individual has a cleft lip (cleft premaxilla) and palate (cleft maxilla) (Barnes, 2012) (Figure 2.5) as well as evidence of treponemal disease marked by gummatous tibial and cranial lesions (see Hackett, 1976; Ortner, 2003; Aufderheide and Rodríguez-Martín, 2011), dietary restrictions and poor general health may be indicated. As described in section 2.2.3, palatal clefts or perforations have several origins, such as from congenital birth defects or late-stage venereal syphilis, and frequently result in notable problems with talking, drinking, and eating (Patil, 2016; Ilczuk-Rypula et al., 2017). Overall, the isotope results suggest that the people living at Skriðuklaustur ate a varied diet of marine and terrestrial resources. Considering the reliable access that the monastery had to both imported and local foodstuff, it is suggested that the people living there acquired a fair supply of nutritious food.

5.7.2 Dietary implications of palaeodental analyses

This section addresses Research Questions 1.2.1, 1.2.5 and 1.2.6. Earlier studies have indicated a notably high dental wear rate, probably resulting from parafunctional behaviours (e.g., weaving *vaðmál*, a woollen fabric) and the local past diet of tough/gritty foods, acidic drinks, and dairy products (Scott and Jolie, 2008; Lanigan and Bartlett, 2013; Richter and Eliasson, 2017; Hoffman, 2018). As previously mentioned (section 3.2.2), the staples of the diet were dried or cured meat, dried fish, stone-ground grain, cheese, milk, and fermented milk products (i.e., *súr mysa,*) (Gísladóttir, 1999; Mehler, 2011; Svanberg and Ægisson, 2012).

Carious lesions (<1%) had a low prevalence, which is probably related to the high rate of calculus (>60%) (see Green et al., 2005) as well as the past diet, which was high in protein and low in flour, grains, and sugar until the 20th century (see Jónsson, 1998; Gísladóttir, 1999; Sigurðsson, 2010; Mehler, 2011; Bjarnadóttir, 2016). In combination with the low fluoride content found in Icelandic aquifers and drinking water, fluoride bound within large calculus deposits, which were common in the skeletal assemblages, might have likewise aided in cariostasis (see Tatevossian, 1990; Aspiras et al., 2010; Gunnarsdóttir et al., 2016). The childhood disease burden of dental fluorosis was low overall overall. At Herculaneum, for comparison, 47% of teeth had linear enamel hypoplasia, 55% had enamel mottling, 18% had hypomineralisation and 20% of teeth had dental caries. However, hypercementosis was only observed in a few individuals (Table 5.11; Petrone et al., 2013).

Table 5.11 Comparison of results of dental analyses performed on teeth from Herculaneum (Petrone et al., 2013) and Iceland (this study). The percentages are derived from the total number of teeth analysed from each site. Percentage for hypercementosis was not reported in Petrone et al. (2013). The percentages of dental changes indicative of fluorosis are much higher among individuals from Herculaneum than Iceland

	Herculaneum	Iceland
Linear enamel hypoplasia	47%	16%
Enamel mottling	55%	3%
Hypomineralisation	18%	8%
Dental caries	20%	<1%
Hypercementosis	2 teeth	8%

Metabolic deficiencies (e.g., vitamin D, rickets and osteomalacia; vitamin C, scurvy) are important predisposing factors for skeletal fluorosis (Gupta et al., 1996; Khandare et al., 2005). However, as previously mentioned (see section 2.2.5), both have only rarely been observed in Icelandic osteological studies. Additionally, scurvy can contribute to periodontal disease (Timmerman et al., 2007; Roberts and Manchester, 2010; Zoëga and Murphy, 2016), which was observed in ca. 72% of the individuals analysed in this study. Dental calculus and periodontal disease were also common in historical Iceland and more prevalent than in neighbouring countries (Sigurðardóttir, 2017). For example, at the early Christian cemetery of Keldudalur in northern Iceland, all the individuals (n=21) presented with calculus (Zoëga and Murphy, 2016). As high protein diets increase oral alkalinity (Moynihan, 2000; Hillson, 2008; Roberts and Manchester, 2010), the high prevalence of calculus deposition is likely correlated with the historical staple diet that was predominately composed of meat, fish, eggs and dairy (Gísladóttir, 1999; Mehler, 2011). Diets high in dairy or milk proteins can further contribute to the hardening of dental plaque, thereby increasing calculus deposition; the calcium, phosphate and casein found in milk and cheese are cariostatic and therefore provide some prevention from caries, particularly in the absence of sugar (Moynihan, 2000; Johansson, 2002). Grains, flour, and sugar were uncommon and were not found in large quantities until the 20th century (Gísladóttir, 1999; Sigurðsson, 2010; Mehler, 2011; Bjarnadóttir, 2016). Other factors that contribute to calculus formation include genetic variation, dental hygiene, local pH, and salivary flow (Hardy et al., 2009) and these must also be considered. For example, both cheese consumption and heavy mastication increase salivary flow, which also raises oral pH/alkalinity (Moynihan, 2000). Furthermore, calculus (a mineralisation process) acts as a shield from the bacterial acids that cause carious lesions (a demineralisation process), despite being partly composed of the same microorganisms responsible for caries. Essentially, the increase of calculus leads to an increase in periodontal disease and a decrease in carious lesions (Greene et al., 2005).

As calculus itself is a significant contributor to periodontal disease, the high rate of calculus (60% maxillary and 68% of mandibular teeth affected) noted in this research likely contributed to the high rate of periodontal changes (i.e., alveolar resorption in ca. 72% of individuals) as it displaces gingival epithelial tissues enabling bacteria and non-calcified plaque to reach the alveolar process (Riethe, 1974; Albandar and Kingman 1999; Kinane, 2002). The high calculus prevalence likely also contributed to the prevention of carious lesions, which had a very low prevalence (ca. 1%) as previously mentioned (see Table 5 in Article III). While there may be some correlation with dental calculus fluoride content and cariostasis (see Tatevossian, 1990; Aspiras et al., 2010), the generally low fluoride concentrations in Icelandic aquifers (see Gunnarsdóttir et al., 2016) imply that low fluoridation in natural drinking water was not an important contributor to caries prevention in the historical population. Therefore, it is far more likely that dietary factors are responsible for the low prevalence of carious lesions. Since carious lesions were rare, the 146 maxillary and 140 mandibular periapical lesions (ca. 5% of all alveolar sockets) and the 298 maxillary and 242 mandibular teeth lost ante-mortem (ca. 8% of all teeth) observed in this research are likely correlated with non-carious pulp exposure and mechanical oral stress (see Clarke and Hirsche, 1991; Molnar, 2008) as was noted in a previous study of the Skeljastaðir assemblage (Richter and Eliasson, 2017). Other factors, such as vitamin D deficiency and hormonal changes that alter bone density could have also contributed to the rates of periodontal disease and ante-mortem tooth loss (Burnett, 2016; Hoffman, 2018). As previously noted, dental hygiene was limited and although toothpicks were historically common, toothbrushes were not until well into the 20th century (Sigurðardóttir, 2017). These observations indicate that subsistence significantly contributed to the notable dental wear, which was likely an important factor in overall dental health in historical Iceland. Indeed, other osteological studies conducted on Icelandic assemblages have shown extremely low caries rates combined with extremely high rates of calculus, dental attrition, periodontal disease and other oral or dental pathologies (see Gestsdóttir, 2004, 2008, 2009; Kristjánsdóttir, 2012; Lanigan and Bartlett, 2013; Zoëga, 2016; Richter and Eliasson, 2017).

5.7.3 Dietary implication of zinc concentrations determined in dental enamel

This section addresses Research Question 1.2.6. Clinical research has found a connection between malnutrition and lower enamel zinc concentrations (Brown et al., 2004). It has also shown that enamel zinc concentrations of more than 90 ppm might indicate poor zinc uptake in childhood (e.g., Tvinnereim et al., 1999). Other research demonstrates that essential trace element deficiencies, like calcium, can lead to the rapid and abnormal absorption of poisonous metals (e.g., lead), especially in malnourished non-adults (Talpur et al., 2018). Zinc is a homeostatically-controlled essential trace element. Nevertheless, its concentrations can be altered by multiple complex interactions occurring with disease, diet, digestion, absorption, and individual variation. Thus, zinc concentrations derived from dental enamel might not actually relate to either diet or health (Ezzo, 1994; Dolphin and Goodman, 2009). The lowest zinc concentrations seen in this research were found in an adult male (SKR 150) (47.3 ppm) and an adult female (SKR 195) (43.8 ppm), potentially suggesting they received a limited supply of zinc as children (see Supplementary Figure S2 in Article II). They also had dental enamel hypoplasia, a skeletal marker associated with periods of metabolic or general health stress occuring childhood (see Ortner, 2003). The highest zinc concentration (145.83; SKR 10) was still well within the lower side of the scope of expected dental enamel zinc concentrations (9.9-1550 ppm) (see Jaouen et al., 2017).

5.8 Hidden dangers?

This section addresses all the research questions, but especially addresses Research Questions 1.2.1 and 1.2.7. Environmental conditions, changes and events have partly shaped how space and human settlements were used by people and their animals and vice-versa in a reciprocal process (Black, 1981; McKinzey et al., 2005). Humans are known to react to both the immediate and anticipated dangers they face (e.g., environmental, interpersonal/intergroup conflict) (Lowe et al., 2002). Therefore, it is worth considering whether assessments of volcanic risks held a secondary role in choosing locations for villages or farms in the past. For example, ethnographic data and oral history from the Maori of New Zealand discuss taboos around visiting regions subject to substantial volcanic activity (Lowe et al., 2002). Historical texts also imply that people were aware of the devastating effects of volcanism and subsequent fallout (Creighton, 1965) and likely acted to minimize their exposure risk perhaps by considering evacuation pathways, resource supply and geographic proximity of residence to volatile areas (Black, 1981; Grattan, 2006). However, records indicate that this may not have been the case in historical Iceland (Karlsson, 2000). In some cases, entire regions or settlements were destroyed leading to the displacement of the population and cultural discontinuity with known allies, neighbouring villages, or traders/merchants. The psycho-social impacts of such displacements, in addition to the severe alteration of weather patterns and light conditions, can be as debilitating in terms of health and survival as the direct physical impacts (Black 1981; Noji, 1997; Grattan, 2006; Dugmore and Véststeinsson, 2012). Additionally, challenging geographic topography (e.g., Icelandic lava fields and glaciers) often imposes mobility limitations for individuals with physical impairments (Institute of Medicine, 1997), such as skeletal fluorosis, that may lead to social disability (Seeley, 2001a, 2001b). The ethnographic record has also demonstrated that tectonic and volcanic activities have resulted in the abandonment or fleeing of settlements due to fear and panic (e.g., 19th century Aleutian villages abandoned following earthquakes and volcanic eruptions) (see Black, 1981). Regardless, these abandonments should likely be perceived as short-term responses: evidence shows that these abandoned places are often later reinhabited or are reestablished in nearby locations (Black, 1981). While Skeljastaðir and other Þjórsárdalur farms were abandoned during the Hekla eruption of AD 1104, for example, Dugmore et al. (2007) report evidence of human activity and animal grazing persisting at least until a subsequent eruption in AD 1300. Regardless, few historic Icelandic eruptions have directly resulted in individually known fatalities, although mass fatalities certainly occurred during volcanic fallout due to famine, disease, and extreme environmental change (D'Alessandro, 2006; Gestsdóttir et al., 2006; Grattan, 2006).

The disease burden of skeletal fluorosis, even when considering possible predisposing factors, appears to be lower in historical Iceland than originally hypothesised. The arid climate in the Arabian Gulf has contributed significantly to the prevalence of fluorosis, which remains common in that region today. Meanwhile, despite the high volcanic activity, Iceland has a subarctic climate, which was likely a key factor in the low prevalence of skeletal fluorosis. The results suggests that the dental and skeletal changes reported in this research appear to be more connected to population dynamics, culturally mediated behaviors (e.g., gendered social roles, marine food sourcing), environment, and increasing urbanization than widespread fluoride pollution. These findings agree with the results of a pilot study (n=3) by Gestsdóttir et al. (2006), which used palaeopathology, radiography, and ICP-MS to investigate osteofluorosis in skeletal individuals from two burial grounds close to the Laki fissure, that were used around the time of the AD 1783-1784 eruption. The results did not show any bone changes suggestive of skeletal fluorosis and the concentrations did not exceed normal baselines (Gestsdóttir et al., 2006).

Volcanogenic mercury emissions on the other hand seem to have affected all the analysed individuals from Skeljastaðir, either through chronic exposure via passive volcanic degassing or due to heavy subacute exposure during the eruption of Mt. Hekla (AD 1104). At Skriðuklaustur, in contrast, ca. 31% of the analysed individuals exhibited elevated mercury concentrations consistent with the long-term, low-dose exposure that patients undergoing mercurial medicinal treatments received. The dietary isotope analyses demonstrated the importance of marine dietary resources in Iceland (e.g., Skriðuklaustur), but also showed that freshwater fish were consumed in areas far from the sea (e.g., Skeljastaðir). It does not appear that the consumption of fish or marine mammals (or freshwater fish) resulted in toxic exposure to biologically cycled mercury (methylmercury) or fluoride. Marine contamination with mercury and fluoride is believed to have increased dramatically only after the Industrial Revolution, which may also explain the apparent lack of elevated bone element concentrations in the fish and marine mammal bones. The results of bone cadmium and arsenic concentrations were not notably elevated and any low-level exposure to these elements indicated in this research is more likely to correlate with anthropogenic activities and cultural behaviors than volcanogenic exposure. Most of the analysed individuals with elevated bone lead concentrations from Skriðuklaustur – who were all of local geographic provenance according to the multi-isotope and trace element analyses presented here – also had elevated concentrations in dental enamel, indicating that people were likely exposed to lead during childhood and into adulthood, possibly from the lead infrastructure or objects found at the site. Meanwhile, Skeljastaðir only had one individual with an elevated bone lead concentration, which is likely correlated with their non-local provenance.

Epidemiological and environmental conditions were probably major catalysts for migration or travel to Skriðuklaustur as the monasteries could provide aid in times of need (Figure 5.30). Medieval monasteries in Iceland were usually found on important travelling paths or close to settlements on the coast where much of the population lived (Figure 5.31). While Skriðuklaustur seems to be in an isolated inland valley, at the time it was occupied it was actually a central place on an essential path connecting the northern and southern halves of eastern Iceland (Kristjánsdóttir, 2012: 296; 2016). Up until the 17th century – at which time the travelling route fell out of use because of climate change – pilgrims, patients, merchants, and others regularly traversed the Vatnajökull glacier to reach the Fljótsdalur valley (Kristjánsdóttir, 2016) (Figure 5.32); the area was heavily traveled and thus diseases spread by travellers and patients seeking healing at the monasteryr. The Black Death, or the Plague, came to Iceland for the first time in the beginning of the 15th century, resulting in the death of over half of the population (Karlsson, 2000: 114-117; Kristjánsdóttir, 2016; Júlíusson, 2018). In 1495-1496, just after Skriðuklaustur was established, the second wave of the Black Death appeared in Iceland (Kristjánsdóttir, 2016). Moreover, cases of treponemal disease in Iceland have only been confidently diagnosed in individuals excavated from Skriðuklaustur (Kristjánsdóttir, 2012; Walser III et al., 2019). This suggests that immediately after the Black Death, cases of treponemal disease broke out in Iceland simultaneously with the late 15th century epidemic occurring in mainland Europe (see Walker et al., 2014). Various plagues, including the Black Death, likewise coincided with changes in the climate (e.g., cooling weather and sustained summer rains), leading to crop and grass failure, shortage of food, and increased disease burden and the population of homeless people (Kristjánsdóttir, 2016).

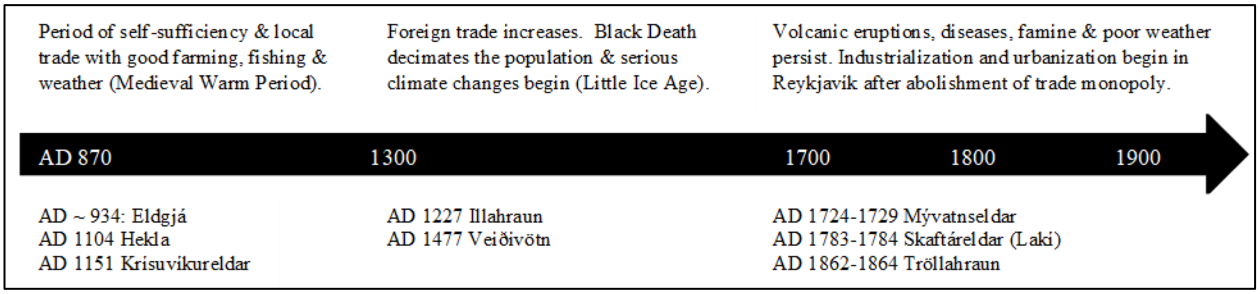

Figure 5.30 Changes in the environment, subsistence economy and public health in Iceland over time. Adapted from Mehler (2011). The listed volcanic eruptions were historically recorded as "fires" and in some cases one event represents numerous consecutive eruptions (e.g., 10-11 eruptions occurred during the AD 1783-1784 Skaftáreldar (Laki) fires) (Thordarsen and Larsen, 2007)

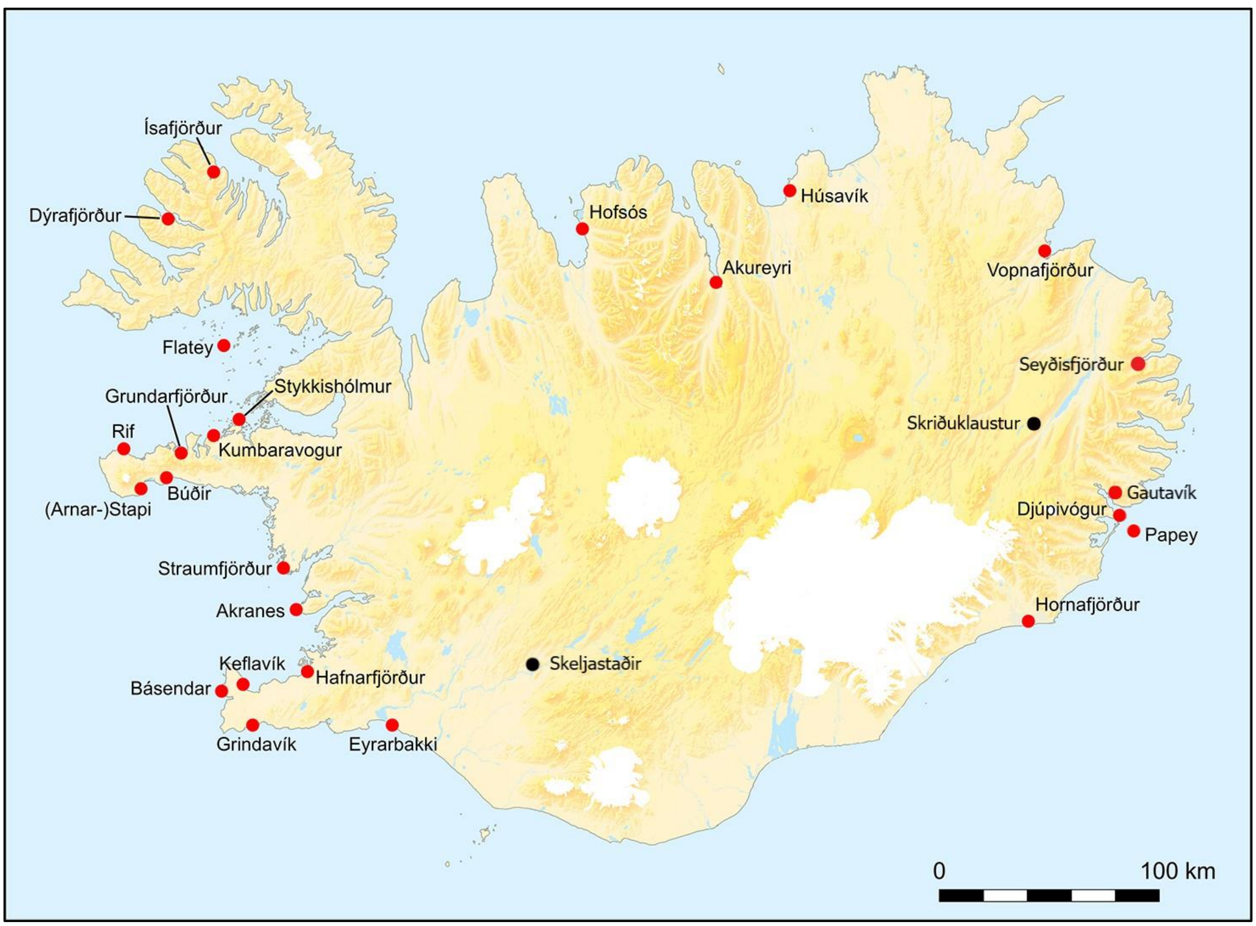

Figure 5.31 Map of Iceland depicting the locations of Skriðuklaustur (east), Skeljastaðir (south) and the trading ports active during the 16th century. Presented here as a courtesy of Natascha Mehler (© Natascha Mehler), with additions

The brethren living at the Skriðuklaustur monastery bought coastal farms so they would have stable access to the important resources (e.g., driftwood, fish, seals, and whales) that the sea offered. These coastal properties were clearly essential to their dietary subsistence, especially for religious fasting, but they were also important for the trade of fish to other countries and monasteries across northwestern Europe. For example, refined sulphur, which was an important trade commodity at the time, was discvovered there (Mehler, 2011; Kristjánsdóttir, 2012), potentially for medicinal uses (Leslie et al., 2004) or to produce vermilion (Mehler, 2015). Other imports of note include a monastic trumpet, an effigy of Saint Barbara, and uncommon ceramics imported to Iceland from France (Kristjánsdóttir, 2012; Mehler et al., 2018). Monetary sources included payment for medical services, donations from local benefactors, local and international trade, community charity, and education (Steinsson, 1965: 108, 1966; Kristjánsdóttir, 2016). Foreign commerce was also a vital part of the subsistence strategy followed at the monastery.

Numerous fish bones, especially from ling *(Molva molva)*, cod *(Gadus morhua)*, haddock *(Melanogranmus aeglefinus)*, rays and were recovered from Skriðuklaustur, again suggesting that fish were a vital part of the monastic subsistence strategy. Smaller fish (60-80 cm in length) are normally found around the eastern and northern coasts of Iceland and in the Greenland Sea. Meanwhile, larger fish (often over 100 cm), which were characteristic at the monastery, are usually fished in the vicinity of the western and southern coasts (Kristjánsdóttir, 2016). Relevant to this study is that, compared to smaller fish, larger fish tend to have elevated isotope values and higher trophic levels, which could potentially impact consumer's isotope ratios (Schoeninger and DeNiro, 1984; Häberle et al., 2016). Fish were culturally and dietarily essential to the monastic subsistence strategy, where dietary abstinence or fasting for religious reasons was observed (Kristjánsdóttir, 2017). Though uncommon at inland sites, where mostly dried fish are found, archaeozoological analysis indicated that fresh fish were often eaten there (Pálsdóttir, 2006; Hamilton-Dyer, 2010). This marked difference was presumably related to religious fasting (Pálsdóttir, 2006). Nevertheless, in some Augustinian and other monastic orders, seals, and bipedal animals (e.g., poultry) could be eaten during periods of the fast as well (Kristjánsdóttir, 2017).

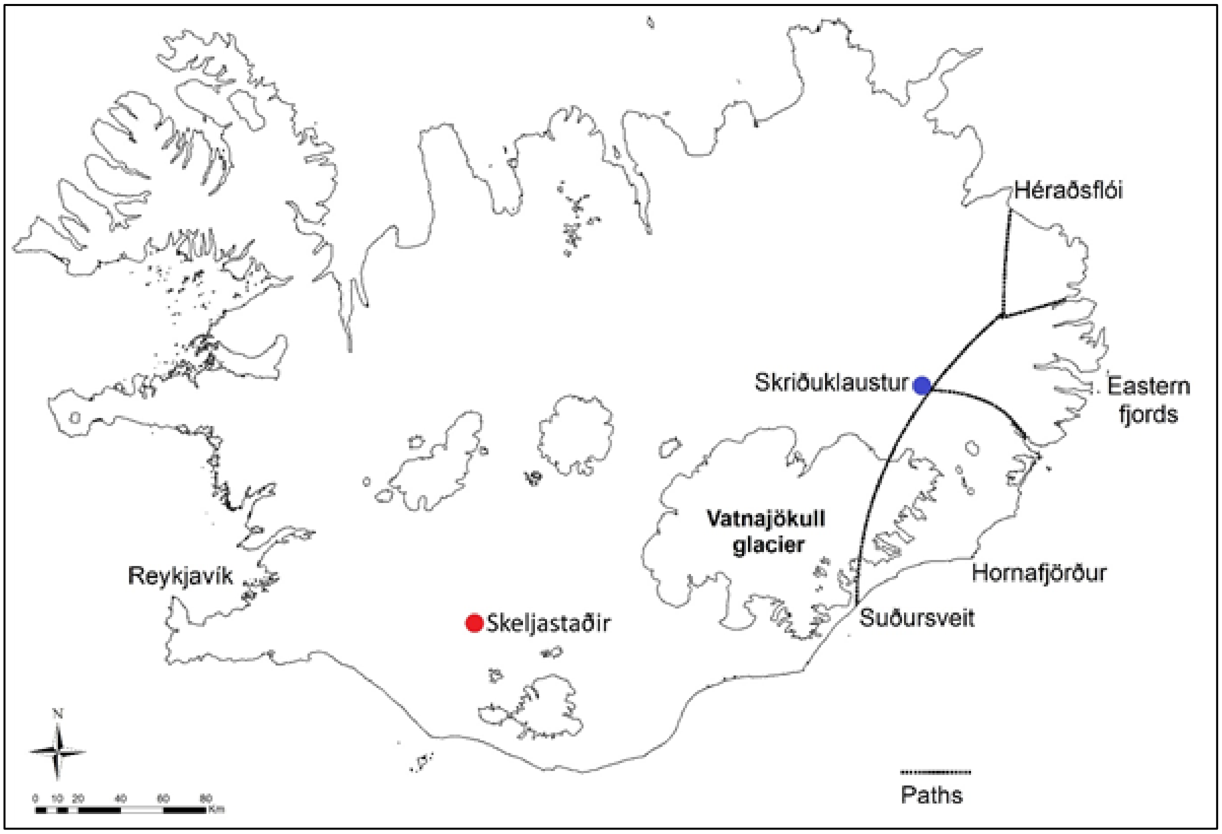

Figure 5.32 A map of Iceland depicting the locations of Skriðuklaustur and Skeljastaðir. The major routes of trade and travel to and from Skriðuklaustur are also shown (© S. Kristjánsdóttir & V. Gunnarsdóttir)

From 1600 to the late 19th century, weather conditions in Iceland were variable, unpredictable, and characterized by cold periods, particularly when sea ice floating on ocean currents landed on the coasts, thereby preventing fishing, and even reducing the growth of grass and temperature on land (Ogilvie and Jónsson, 2001; Ogilvie, 2005, 2010). During the Laki fissure eruption of AD 1783-1784, a highly concentrated toxic haze of sulphur, fluorine and chlorine was released into the air causing environmental changes, darkness, cooler weather and severe damage and death to people, animals, and vegetation (Gunnlaugsson and Rafnsson, 1984; Rafnsson, 1984). The eruption caused one of four major famines that occurred during the 17th-18th centuries (Júlíusson et al., 2017). An estimated 8700 people (20-25%) of Iceland's human population (Guðmundsson et al., 2008) and tens of thousands of sheep, cattle and horses died (up to 75% of Iceland's livestock at the time) (Gunnlaugsson and Rafnsson, 1984; Jónsson, 1994; Halldórsson, 2013). Reverend Jón Steingrímsson (1728-1791) provided a detailed report of disease following the eruption, describing symptoms associated with fluorosis in both dead and living animals and humans, describing that people survived by eating cooked skin or hide, hay mixed with water and meal to make porridge and boiled fish bones crushed into milk (Steingrímsson, 1998; Halldórsson, 2013). He also recounts that people who depended upon locally sourced water and food – contaminated with fluoride among other pollutants – developed the same bone and tooth changes associated with fluorosis as the livestock. The health conditions were likely compounded by poor nutrition following food shortages during the long-lasting volcanic eruption and subsequent fallout (D'Alessandro, 2006). Descriptions of human health in 1783 in Iceland, the Netherlands and the UK are consistent between regions, specifically mentioning farmers, some of whom were incapable of work due to severe respiratory distress. Accordingly, asthmatics suffered more than usual due to an overall decrease in lung function, and people were plagued with headaches, eye conditions and a permeating sulphurous stench (Thorarinsson, 1981; Grattan, 2006;

Grattan et al., 2012; Moffat and Wilson, 2012). A haze followed the eruption causing difficulties in visibility and navigation, particularly for fishermen (Rafnsson, 1984). The bishop Hannes Finnsson from Skálholt, which was an important cultural, intellectual, and spiritual center and seat of governmental power containing an episcopal see, school, cathedral, and cemetery, described the haze-famine which followed the Laki eruption in a work entitled *Um mannfækkun af hallærum á Íslandi (Decimation of the population in Iceland due to famine)* (Gjerset, 1925; Rafnsson, 1984; Karlsson, 2000; Lucas, 2010). He described that "hunger and starvation marked people's appearance with all the diseases which thereof spread; particularly dysentery, scurvy and mumps. The hunger was so common that it could even be seen on many priests and well-off farmers" (Finnsson, 1970: 117-118; translation by Halldórsson, 2013: 32). These records suggest that the population faced serious malnourishment, deficiency diseases (e.g., scurvy) and plagues, such as smallpox (Pétursson, 1984; Halldórsson, 2013). However, these conditions were certainly temporary and were likely alleviated shortly after the volcanic eruption and subsequent fallout ended. For example, the import of grain temporarily increased considerably immediately following the eruption (Beck, 2020). Earthquakes also occurred throughout Iceland, one of which caused severe destruction to Skálholt in AD 1784, forcing the bishopric to relocate to Reykjavík (Hambrecht, 2011). At the time, Skálholt had one of the largest populations (>100 individuals), making it the place that most resembled a town in Iceland until the growth of Reykjavík occurred in the late 18th century (Lucas, 2010). The famine, pestilence and climatic changes that occurred following the eruption threatened the very survival of the country, reconfiguring Iceland both physically and politically and provided additional impetus for the abolishment of the trade monopoly just three years later, in AD 1787 (Streeter et al., 2012; Clark and Jones, 2016).

Ideology is an important component of state formation and landscapes were focal points for national identity and political consciousness during this period (Hastrup, 2008), so it may be argued that landscape change and habitability contributed to shifting ideological principles, which are likewise vital to state formation (Mann, 1986; Clark and Jones, 2016). Thus, in addition to the constraints of the Danish trade monopoly, the natural disasters, diseases, and famine occurring at the time likely also contributed to the dispersal of the population and the eventual urbanization of Reykjavík. The aftermath of historic volcanic eruptions has often been characterized by innovation, development, and social agency throughout the world (Grattan, 2006), yet some scholars argue that volcanic impacts stifled innovation and even promoted conservatism in Iceland (see Karlsson, 2000; Dugmore and Vésteinsson, 2012). However, this point of view may be correlated with hegemonic or nationalist tendencies that posit historical Icelanders as highly strategic survivors against all odds, particularly in the earliest centuries of Iceland's settlement. Some scholars (e.g., Ebel, 1977) have even argued that Icelanders could not have survived without foreign trade due to a lack of raw materials, but archaeological investigations have proved this argument to be entirely invalid (Gardiner and Mehler, 2007). Nonetheless, 18th century Iceland simultaneously underwent the worst natural and epidemiological disasters that led up to a positive shift in cultural core values (Þórhallsson and Joensen, 2014). In effect, these natural disasters, and periods of pestilence, in combination with the abolishment of the Danish trade monopoly (1787), may have contributed significantly to the multi-factorial changes in population dynamics and the steady economic and urban growth occurring in the 18th century. For example, in AD 1801 the population census of Reykjavík recorded just 307 inhabitants but grew steadily and rapidly in population density and urban development, thereby reaching a population of nearly 6700 individuals by AD 1900 (Malmström, 1958). Iceland was rural, comprised primarily of farmers, and, as in most places throughout history, poverty and poor hygiene likely contributed to periods of poor health in the past (McGovern et al., 2007; Loftsdóttir, 2008). Infectious diseases (e.g., leprosy, tuberculosis) spread easily as people greeted one another with a kiss (Sigurðardóttir, 2017). Sites indicating archaeological and documentary evidence for a trade economy between Europe and Iceland are indicative of significant subsistence upon fish and terrestrial animal meats that were traded for Icelandic goods and resources such as sulphur and walrus ivory (Harrison et al., 2008).

Modern ethnographic studies illustrate commonly held criteria for avoiding contaminated water, such as by evaluating its smell, taste, and color (Scherzer et al., 2010). Unfortunately, clear, and scentless water does not necessarily indicate safety, the same way that foul smelling or discolored water does not always indicate that it is unsafe (Napier and Kodner, 2008). However, the salty and bitter taste of fluoride-contaminated water has prevented cases of lethal exposure in some instances: two-thirds of 34 children that drank water highly contaminated with fluoride recovered within a 24-hour period, for example (Hoffman et al., 1980). In some areas, the consumption of locally sourced water carries stigma, thereby leading to collective beliefs about what should or should not be consumed and what sources are appropriate to draw from. People's beliefs and experiences are important factors in their following behaviors and practices (Scherzer et al., 2010). A study conducted in India, where water is often contaminated with fluoride and arsenic, demonstrated that when options for sourcing water are limited, people adapt to what is available and attempt to mitigate the problems associated with it. However, when multiple options are presented, people begin to differentiate and prioritise water quality (Linneck, 2016). With regards to this study, people in the past likely implemented strategies for acquiring uncontaminated food and water and likely managed to evacuate areas affected by volcanic eruptions at least to some degree (Black, 1981; Grattan, 2006). Overall, a social group's preparedness to manage environmental stresses is vital for mitigating the impacts of natural disasters (Black, 1981; Noji, 1997).

6 Conclusions

Despite Iceland being a rural country well into the modern period, the general health and living conditions do not appear to have been vastly different from any of the neighboring countries (Júlíusson, 2018; Turchin et al., 2018). Human groups probably fled from certain regions that were especially affected by volcanic fallout and unlike the livestock, they probably managed to avoid contaminated drinking water when possible. Therefore, volcanic dangers were not well hidden, especially when it was possible to move away from the ashfall and visible fires that spread across the country. These dangers were probably only concealed during passive periods, when natural degassing still posed health risks to those living within close proximity to active volcanic regions. Looking towards the future, the preparedness and stability of geo-political and socio-economic structures of regions prone to volcanic activity should be especially considered rather than focusing solely upon the abrupt, short-term consequences that eruptions cause.

In this concluding chapter, the original research questions presented in section 1.2 are reiterated and summaries of the results and findings of the articles (see Articles I, II, and II) and additional results published in Chapter 5 of this thesis summary are provided below to address the research questions.

Research Question 1.2.1 How did environmental conditions impact subsistence, landscapes, human health, and culture in past Icelandic populations?

Overall, this research (see Articles I, II, II and Thesis Summary) aimed to investigate diet, geographic provenance, and exposure to volcanogenic or anthropogenic contaminants and their connection with the environment and overall health among the historic population of Iceland. The research suggests that Medieval Icelanders were aware of the volcanic impact on their health, environment, livestock, and surroundings, perhaps even informing the selection of settlement places and land use. While volcanic eruptions cause disturbances in individuals residing in certain areas and the community's forms of subsistence, a culture's ability to cope with environmental stresses plays a major role in mitigating the effects of detrimental impact (Black, 1981; Noji, 1997; Grattan, 2006). Because of Iceland's small population, which in the Middle Ages was only a fraction (ca. 1/6th) of today's population, close genetic and familial structure, and the struggle for survival during volcanic fallout, Icelanders may have emphasized reciprocity and altruism during difficult times (Grattan, 2006).

Research Question 1.2.2 What were the geographic origins of the people residing at Skeljastaðir and Skriðuklaustur and does their provenance relate to pathological conditions or exposure to toxic elements?

Previous research showed that Skeljastaðir was inhabited by locals aside from a single migrant (Price and Gestsdóttir, 2006). It was similarly important to determine the geographic origins of the population at Skriðuklaustur, as such information provides indications about where people might have lived, what they may have been exposed to earlier in life and whether changes in environment or locale were correlated with pathological changes. It was hypothesized that the people buried at Skriðuklaustur would be both local and foreign, such as traders, considering the high rate of infectious disease cases found at the site. For example, individuals with long-standing, probable venereal syphilis infections were found at Skriðuklaustur, demonstrating that the disease arrived in Iceland close to the time that it reached epidemic levels on mainland Europe. However, the research ultimately found that none of the analysed individuals were foreigners and that the cemetery population is likely predominately representative of a local population of people born in Iceland that resided both at inland and coastal sites within and around the southwestern quarter of the country. People from throughout the region traveled or relocated to Skriðuklaustur for medical treatment of a wide range of conditions, hospice, trade, religious reasons and perhaps because of homelessness. One older adult male from Skriðuklaustur with advanced, potentially disabling, Paget's disease resided further inland as a child (see Article II). This individual also had very low trace element concentrations in bone, probably due to the irregular bone remodeling that occurs with Paget's disease (see

Article I; Chapter 5 of Thesis Summary). Trace elements used as indicators of provenance (Sr, Pb, Ba, Zn) analysed in this study were low in variation and when considered together with the strontium and oxygen isotope ratios determined in dental enamel, further indicate a local population that was born in Iceland (see Article II).

Research Question 1.2.3 Did volcanogenic emissions of mercury (e.g., from the Hekla eruption of AD 1104) or other elements affect the health of the local population of Skeljastaðir? Were any other toxic substances used medicinally at the monastery?

Considering the volcanogenic emission of mercury in Iceland and the known Medieval use of the chemical element as a medicine at monastaries across the present-day European continent, the research sought to investigate whether the individuals buried at two sites, Skriðuklaustur and Skeljastaðir, were exposed to it during life. Elevated mercury bone concentrations were found at both sites: the results from Skeljastaðir suggest high-dose and possibly acute volcanogenic exposure, while the results from Skriðuklaustur are consistent with long-term, low-dose exposure to mercury, probably associated with medical treatment. Soil samples and animal bones were also analysed, all of which showed around or under the normal background threshold of mercury, indicating that diet and diagenesis were not significant contributors to the elevated ante-mortem mercury values found among the analysed individuals (see Article I). None of the human bone samples indicated arsenic exposure, despite the widespread use of arsenic in medical preparations in the past. Only one animal bone sample had an elevated arsenic concentration, but none of the other animal bones showed any elevated concentrations of the elements analysed here. It is possible that the cow with elevated heavy metal concentrations could have been receiving an antiparasitic treatment, which often contained arsenic and minerals salts (e.g., copper) at the time in the Nordic countries, although the elevation could have also occurred from contaminated food or water as well. Low-level cadmium exposure was indicated at Skriðuklaustur, while only a couple of cases presented at Skeljastaðir. These elevated concentrations could reflect exposure to volcanogenic cadmium, but other sources such as smoke inhalation, tobacco smoking and working with objects containing cadmium are more likely to be the cause when considered in combination with the other bone element concentrations (see Chapter 5 of Thesis Summary). Elevated bone lead concentrations were noted from Skriðuklaustur, particularly in individuals that were also exposed to lead during childhood, possibly from the lead infrastructure or objects found at the site. Only one individual from Skeljastaðir had an elevated bone lead concentration, probably related to their non-local provenance (see Article II; Chapter 5 of Thesis Summary).

Research Question 1.2.4 Regarding exposure to toxic elements, were there any identifiable differences (e.g., behavioral, or occupational exposure) between men, women, children or individuals of different social status or age groups?

There were no statistical differences between men and women in mercury exposure in bone samples from Skriðuklaustur even though access to medical treatment could have differed between the sexes and social classes in the past. Meanwhile, younger adults had higher mercury concentrations than older adults, possibly because they had entered the latent or tertiary stages (e.g., the latent stage lasts >30 years) and had stopped taking mercurial medicines prior to death. Younger age-at-death can also occur from mercury poisoning, especially in light of the non-standardised dosage provided for medical treatment during the 16th century (see Article I). Trace element analysis of dental enamel showed that children and women were often exposed to lead: in children this was likely due to hand-to-mouth activities, while in women this was probably because of occupational activities that may have involved more time spent indoors around lead objects and structures containing lead (see Article II).

Research Question 1.2.5 Were socio-cultural or environmental conditions responsible for dietary shifts between the populations living during the Medieval Warm Period (Skeljastaðir assemblage) and the Little Ice Age (Skriðuklaustur assemblage)?

Archaeological evidence, historical records and isotope analyses demonstrate that the diet was predominately based upon mixed marine (i.e., fish, seals) and terrestrial resources (i.e., foraged edibles, gardening, imported goods and livestock). Freshwater fish were likely important to the subsistence strategy of inland farm sites, especially towards the beginning of the Settlement Period: stable isotope analyses showed that the protein portion of the diet at Skeljastaðir was mainly terrestrial with some freshwater fish consumption, while at Skriðuklaustur there was highly marine diet (mixed marine and terrestrial) with the addition of some freshwater fish input. Despite being an inland site, historical records and zooarchaeological studies conducted at Skriðuklaustur are in line with the findings of this research, showing that fish was important to the diet, probably because of religious fasting. The steady exploitation of marine resources began during the beginning of the Settlement of Iceland but increased significantly over time. While the early migrants to Iceland may have grown crops such as barley, the cooling weather leading up to the Little Ice Age impacted the growth, practice and yield of grain farming. Trade networks were important from the earliest period, but the importance and frequency of visiting traders increased steadily and significantly leading up to the Little Ice Age, resulting in a stable and varied diet that included various imported goods such as fruit (see Article II). Historical records from the more recent past (17^{th}-19^{th} centuries) indicate that the import of flour, sugar and other refined and processed goods became important, contemporaneously with the urbanization of Reykjavík – the only real urban center established during the country's history (see Article III).

Research Question 1.2.6 Does osteological evidence of metabolic or nutritional distress show any relationship with pathological conditions, toxic element exposure or diet?

Diet is important to overall health, but it is also a primary source of exposure to certain toxic elements, such as lead, mercury or fluoride. Marine fish and mammals are particularly known to accrue high amounts of the highly toxic, biologically cycled methylmercury. Likewise, marine fish and mammals are also prone to fluoride uptake. Therefore, it was important to examine the subsistence of the analysed populations to control for diet-derived exposure. The lack of elevated bone element concentrations in the fish and marine mammal bones indicates that the consumption of fish or marine mammals was unlikely to have been a significant source of toxic mercury exposure. This is probably because marine mercury contamination only increased dramatically after the Industrial Revolution (see Article I). A few cases of possible metabolic or health stress in childhood were suggested by dental enamel hypoplasia in combination with apparent zinc deficiency, although being under homeostatic control, it remains uncertain whether zinc concentrations in dental enamel actually reflect palaeodiet or not. Additionally, only one individual from Skriðuklaustur consumed a completely terrestrial diet, probably correlated with dietary restrictions caused by a congenital case of cleft maxilla and premaxilla (palate and lip) (see Article II). The high rate of calculus and the low rate of caries was likely correlated with the historical diet, which was high in protein derived from dairy products, fish, and meat and low in sugar, flour, and other processed foods prior to the urbanization of Reykjavík. Dental calculus may have provided some protection from carious lesions, but likely played a part in the high rate of periodontal disease seen in the archaeological population (see Article III).

Research Question 1.2.7 Did historic eruptions in Iceland result in mass human mortality such as occurred among livestock?

The results show that the human fluorosis burden was very low, which suggests that people were aware of the toxic effects of volcanic emissions and likely knew not to consume water, livestock or foliage immediately or directly affected by volcanic fallout. Similarly, fluoride can significantly bitter the taste of water, which was probably a clear indicator to people that it was not safe to drink as has occurred in some modern cases of drinking water contaminated with fluoride. The bone changes reported here are probably related rather to environmental conditions, population dynamics, culturally mediated behaviors and increasing urbanization and population density than to serious fluoride exposure (see Article III).

Research Question 1.2.8 What osseous pathologies present in the human skeletal assemblages and are they associated with toxic heavy metal or fluoride exposure, as revealed by ICP-MS and ISE?

The human health impact of fluoride emissions in historical Iceland from volcanogenic sources was investigated by examining bone fluoride concentrations and skeletal changes potentially associated with skeletal fluorosis, a disease caused by fluoride toxicity. Considering historical reports and modern findings of the devastating impact volcanogenic fluoride has had on past and present livestock, it was hypothesized that people residing at sites situated nearby and occupied during or following volcanic eruptions would have been subject to enough fluoride emissions to cause toxicity. Unless relocated, livestock tend to continue drinking the water and eating the foliage within their local vicinity even during volcanic fallout. Overall, the skeletal and dental changes likely reflect occupational activities (e.g., fishing and farming), culturally mediated behaviours (e.g., gendered occupations), increasing urbanization, population dynamics, and diet rather than exposure to volcanic pollution, such as from fluoride (see Article III).

Concluding remarks

Overall, this research indicates that volcanic emissions predominately only affected the people residing near actual eruption events. Although these events dramatically impacted the living conditions and the habitability of zones subject to volcanic fallout, sometimes resulting in site abandonment, the actual health burden of skeletal fluorosis and other toxic elements of volcanogenic origin appears to be minimal. Instead, anthropogenic sources of toxic substances seem to have been much more commonplace, such as the use of mercury as a medicine and lead exposure resulting from regular contact with lead objects or infrastructure. Relevant to the present day, this research suggests that it may be even more vital to reinforce socioeconomic circumstances, geopolitical conditions, and disaster mitigation protocols in regions with volcanic risks rather than focus on concerns over the severity of the immediate, temporary effects resulting from volcanic eruptions (see Thesis Summary).

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