

Volcanoes, medicine and monasticism: Investigating mercury exposure in medieval Iceland

Introduction

Research on the care and treatment of disease and disability in the past has undergone a resurgence over recent years. Theoretical and methodological developments have provided new insights into the treatment, perceptions and experiences of disease within past populations (e.g. Tilley, 2015; Powell et al, 2016). This paper adds to this growing corpus through the analysis of the medicinal use of mercury in an Augustinian monastery, Skriðuklaustur (AD 1494-1554), in the east of Iceland. Recent excavations revealed a hospital with evidence of a sophisticated array of herbs, surgical artefacts and medicinal preparations. Approximately half of the skeletons excavated (n=295) from the associated cemetery exhibited a range of pathological conditions, including treponemal disease, tuberculosis, hydatidosis and trauma (Zoëga, 2007; Kristjánsdóttir, 2011; Kristjánsdóttir and Collins, 2011; Sundman, 2011; Kristjánsdóttir, 2012). Skriðuklaustur monastery is the only excavated archaeological site in Iceland at which treponemal disease (probable venereal syphilis) has been confidently diagnosed according to the criteria described by Ortner (2003) and Hackett (1976); prior to the analysis of this assemblage many scholars had considered syphilis to have been present in Iceland only since modern times (Þorláksson, 2003). One other possible case of treponemal disease has since been described from Viðey, an island close to Reykjavík (Gestsdóttir, 2009). In Europe, during the medieval period, mercury (cinnabar) was used as a treatment for syphilis, as well as other venereal diseases, skin infections and leprosy (Dracobly, 2004). Mercury was also used for a wide range of other purposes, including ink and cosmetics, thus potentially exposing individuals engaged in a variety of activities to toxic concentrations (Parsons and Percival, 2005; Zuckerman, 2016). In the context of Iceland, it is also important to consider exposure to mercury via geothermal springs and volcanoes, which are serious sources of mercurial contamination (Roos-Barraclough et al., 2002; Gustin, 2003).

The current study builds upon an initial pilot analysis (2011) of mercury measurements conducted in two individuals found at Skriðuklaustur: one individual was found to have a normal mercury concentration and the other exhibited an elevated concentration. In the study presented here, mercury concentrations were measured in bone samples from 50 individuals excavated from Skriðuklaustur (n=36) (Figure 1). A minimum of nine individuals presented with skeletal lesions strongly suggestive of treponemal disease (e.g. gummatous periostitis, cranial stellate scarring (caries sicca), tibial bowing; see Hackett, 1976; Ortner, 2003; Aufderheide and Rodriguez-Martin, 2011 (Kristjánsdóttir, 2011)). An additional four individuals presented with skeletal lesions possibly suggestive of treponemal disease. These thirteen individuals were included in the sample set analysed in this research. Seven sampled individuals exhibited hydatid disease, based upon the presence of at least one hydatid cyst (see Kristjánsdóttir and Collins, 2011) (see Figure 2). Twelve individuals showed evidence of non-specific infections and five individuals demonstrated no pathological indicators for infectious disease. Associated soil and faunal samples were also analysed. A non-hospital site called Skeljastaðir, located in Þjórsárdalur valley in southern Iceland, was also included in this study (n=14) (Figure 1). This site was

occupied at least until the eruption of Hekla in AD 1104 (see Steffensen, 1943; Þórðarson, 1943), although some of the burials post-date the eruption (Sveinbjörnsdóttir et al., 2010). It was chosen as a comparative site due its proximity to volcanic activity and also because it was occupied prior to the increased movement of syphilis across northern Europe (Harper et al., 2011). The aim of this study was to examine whether skeletal samples from the monastic site of Skriðuklaustur exhibit elevated mercury concentrations as a consequence of its use in medicinal treatment. The research also aimed to assess whether high mercury within bone samples from Skeljastaðir could implicate chronic or acute mercury exposure resulting from nearby volcanic activity. Skeljastaðir is an important site for the evaluation of antemortem environmental exposure to volcanogenic heavy metals and post-mortem diagenesis. We hypothesise that there will be a correlation between skeletal lesions indicative of treponemal disease and mercury concentrations at Skriðuklaustur given its historically attested use in treating the disease in the sixteenth century. Likewise, we hypothesise that there will be a correlation between mercury concentrations in bone and the volcanic activities that occurred around Skeljastaðir.

Mercury in Historical and Bioarchaeological Context

Alchemy – and the importance of mercury within it – is inter-woven with the religious, philosophical and scientific ideologies of the Middle Ages and was practiced by a wide range of people, including kings, popes, doctors, clergy members and scientists (Parson and Percival, 2005). In the thirteenth century, alchemists began to use cinnabar (mercury sulfur) as a medicinal elixir; it was believed to impart long life due to its deep red color and philosophical associations with blood and the soul (Charlier et al., 2014). Historically, it was regarded to have anti-inflammatory properties (Latham, 1846; Fleming, 1997); however, modern clinical evidence has demonstrated its strong inflammatory (Houston, 2011) and anti-mitotic effects (Vallee and Ulmer, 1972). As Dr. Peter Mere Latham (1789-1875) once said, “Poisons and medicines are oftentimes the same substances given with different intents” (Latham, 1968). A number of cultures still make use of mercury today for spiritual, ritualistic, and medicinal applications: it is believed that mercury can ward off evil, purify the home, incite love, luck, or wealth (Parsons and Percival, 2005).

Cinnabar is known to have been mined in Spain as early as ca. 430 BCE and throughout the world thereafter (Parsons and Percival, 2005). The toxic side effects of working with cinnabar were observed from at least the Roman period (Hylander and Meili, 2003). Mercury is easily absorbed, poorly excreted and has no known beneficial properties or biological functions in living organisms (Miculescu et al., 2011). Despite this, Pliny the Elder, Galen, Paracelsus and John of St. Amand all espoused its medicinal uses (Hajdu, 2005; Parson and Percival, 2005; Ozuah, 2000). Later, it was suggested in *The Canon of Medicine* (AD 1025) that mercury could be used to treat skin diseases (Avicenna, 1999). Syphilis, and other conditions with similar signs and symptoms, were treated with compounds ranging from arsenic, bismuth, various vegetable-based remedies (e.g. guaiacum) and, most commonly, mercury (Crane-Kramer, 2000; Thomann, 2015). Medieval anti-syphilitic treatment was administered by inhaling mercurial vapors (fumigation) and by rubbing mercurial salves upon the lesions several times a day, ideally within a warmed, enclosed space (Dobson, 2007; Zuckerman, 2016). Some physicians also

prescribed pills made of mercury, mixed with other ingredients, such as honey, cinnamon, or senna (Thomann, 2015). Individuals with syphilis normally received long-term courses (months to years) of mercurial treatment (Dobson, 2007; Zuckerman, 2016). Physicians, patients and the general public were aware of the poisonous attributes of mercury and the symptoms caused by mercurial toxicity. Regardless, high dose mercurial treatments resulting in death or debilitation were not uncommon in the Middle Ages (O’Shea, 1990; Zuckerman, 2016).

It is difficult to determine whether mercury conferred any beneficial effects upon recipients. As mercury is biocidal to the type of bacteria responsible for syphilis (spirochetes) (Pound and May, 2005), it is possible that mercurial treatments successfully treated infections in the primary and tertiary stages when the bacterial load is low. According to the nineteenth century accounts of Dr. Jón Hjaltalín (John Hjaltelin), the Inspecting Medical Officer of Iceland at the time, calomel and mercurial ointments successfully cured some cases of hydatid disease and syphilis (Hjaltelin, 1868; Hjaltelin, 2013). Mercury remained one of the most common treatments for syphilis until the arsenic compound, arsphenamine (1909), and later the antibiotic, penicillin (1940), were introduced (Ozuah, 2000). “Blue mass,” a substance composed of mercury, honey and licorice, was routinely administered by hospitals to any patient requiring purgative or cathartic treatment between 1800 and 1940. Since then, physicians have renounced the medical benefits of mercury and officially recognized it as a poisonous substance (Graeme and Pollack, 1998; Ozuah, 2000). In recent years, the bioarchaeological record has been able to provide independent verification of the historic evidence for the widespread medicinal use of mercury. Rasmussen et al. (2013b, 2015) found elevated mercury concentrations in skeletal remains from several medieval Danish and German cemeteries and monastic sites, likely a result of mercurial treatments. Another study from Poland also identified elevated bone mercury in individuals dating from the 14th-19th centuries AD with pathological changes consistent with venereal syphilis (see Kępa et al., 2012).

Uptake of heavy metals by bone during diagenetic processes is an important consideration when examining post-mortem mercury concentrations. Rasmussen et al. (2015) measured soil samples associated with skeletons that had high mercury concentrations and these showed no correlation with mercury levels, thus indicating that diagenesis was not a factor (Rasmussen et al., 2013a; 2015). Yamada et al. (1995) and Zuckerman (2017a) likewise found no evidence for diagenetic transfer of mercury between bone and soil. While lead can be passively absorbed by bone hydroxyapatite (the mineral and matrix component of bone and teeth) (Swanston et al., 2012) mercury is uncommon in the natural environment and humans are not prone to post-mortem uptake (Avila et al., 2014; Rasmussen et al., 2015). Bone hydroxyapatite retains mercury by replacing calcium and bonding with carbonates during life (Lee et al., 2005, Avila et al., 2014). Therefore, high concentrations of mercury in skeletal remains are considered to be a robust indicator of *in vivo* exposure (Schwarz et al., 2013; Rasmussen et al., 2015). The bone matrix acts as a long term (>2 years) heavy metal reservoir until remodeling or resorption occur (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 does not absorb well through the skin itself as it easily vaporizes during handling; however, the body can retain at least 74 to 80 percent of inhaled mercury vapour as it is dispersed to the brain, kidneys, lungs and gastrointestinal tract (Syversen and Kaur, 2012). Its toxic properties inhibit cellular, enzymatic and membrane activity and transport functions (Ozuah, 2000; Bradberry, 2012). Individuals exposed to mercury vapor may develop a cough, fever, chills, nausea, dyspnea and general weakness within a matter of hours after exposure (Graeme and Pollack, 1998; Syversen and Kaur, 2012). Although some historical medical texts emphasise the importance of dosage in the prevention of mercurial toxicity (O’Shea, 1990), pre-symptomatic toxicity can occur at very low levels of exposure (Zahir et al., 2005). The United States Environmental Protection Agency estimates that the maximum daily exposure to mercury should not exceed 0.001 milligrams per kilogram of body weight. The levels of mercury included within nineteenth century treatments are documented to far exceed these limits, with 2 grains of mercuric solution equating to 129.6 milligrams (Ioannou et al., 2016, p. 2). For example, a standard dose in 17-19th century England was approximately 5 grains (325 mg) daily for up to two years, although the mercurial form (i.e. calomel) used during this period was likely less toxic than the mixtures of mercuric chloride, metallic mercury and pure cinnabar used in the 16th century (Zuckerman, 2016). Non-adult bones are especially prone to biogenic uptake (Ziola-Frankowska, 2017), so such doses were likely to have been particularly harmful when administered to children during treatment for congenital syphilis.

The background concentrations of mercury in archaeological bone have been established in femoral bone to between 0.08 (cortical) – 0.3 ppm (trabecular) (see Rasmussen et al., 2015). Since Iceland has extensive mercury emitting volcanic activity (e.g. Mt. Hekla) (see Coderre and Steinthorsson, 1970; Thordarson and Larsen, 2007), and therefore a generally higher atmospheric mercury concentration than locales where background levels were established in bone (i.e. Denmark, Germany), it is also likely that normal concentrations established in archaeological bone from Iceland begin at a higher threshold. A study that analysed contemporary bone samples demonstrated an average concentration of 0.9 ppm in older individuals (>65), but it was unable to report a concentration for younger individuals (see Miculescu et al., 2011). Following global industrial pollution, and the cumulative properties of heavy metals in bone, concentrations in contemporary human bone are generally higher than in historical populations (Ericson et al., 1991). Furthermore, heavy metal toxicity and increasing age are both linked with impaired renal and liver function, which limit the excretion of toxic substances like mercury (Ziola-Frankowska et al., 2016). Although trabecular bone was not used in this study, only concentrations of >0.3 ppm or greater were deemed to be elevated.

Syphilis in Iceland

The origins and spread of treponemal diseases remains a topic of ongoing debate (Baker and Armelagos, 1988; Powell and Cook, 2005; Harper et al., 2011), as does the emergence of venereal syphilis (*Treponema pallidum pallidum*) in Iceland (Kristjánisdóttir, 2011). Some scholars believe that venereal syphilis appeared in Europe in the late fifteenth century, post-contact with the New World, while others suggest an Old World origin (Meyer et al., 2002; Harper et al., 2011; Mays et al., 2011). It has also been suggested that the Vikings could have played a role in the spread of syphilis, following the establishment

of their settlements in the New World; however, the skeletal record cannot yet attest to this (Meyer et al., 2002; Mays et al. 2003). Determining the origin of syphilis in the Old World is complicated by difficulties in assigning a definitive diagnosis from skeletal lesions in all cases and the lack of reliable radiocarbon dating (Harper et al., 2011). Likewise, historical records are often not specific enough in their pathological descriptions for an unequivocal diagnosis (Mitchell, 2011). Syphilis may have also been mistaken by past physicians for a number of other diseases (e.g. leprosy) that cause aggressive skin lesions (Lefort and Bennike, 2007). However, some scholars have indicated that diagnostic confusion between leprosy and syphilis cases was unlikely during the Medieval period (see Crane-Kramer, 2000).

According to historical evidence from sixteenth century Iceland, one barber-surgeon was paid to provide treatment for 100 individuals suffering from syphilitic infections (Diplomatarium Islandicum IX, p. 290). Such practitioners likely focused on treating external skin lesions with surgery and ointments and may have also administered mercurial treatments (i.e. fumigation, salivation) (Dracobly, 2004; Rasmussen et al., 2008; Kristjánsdóttir, 2011; Zuckerman, 2016). Infection with syphilis was associated with shame and deviant or promiscuous behaviour (Zuckerman, 2017b) and many individuals may have avoided seeking treatment, particularly at religious institutions. Within medieval monasteries, monks prepared medicines based upon Galenic formulations, which were often composed of a mixture of herbs, minerals, metals and animal parts (Hajdu, 2005). The Icelandic church maintained close ties with the Catholic Church on the European continent, thus medical knowledge and practices were exchanged. Similarly, during the fifteenth century, German, Dutch, English and Danish merchants engaged in trade between Iceland and the mainland, spreading both diseases and their purported remedies. The remains of imported objects and food found at Skriðuklaustur (e.g. fruits, exotic plants) also highlight Iceland's international cultural integration at this time (Kristjánsdóttir et al., 2014; Kristjánsdóttir, 2016). As monasteries were obliged to provide burial rites to all those who died within their care, individuals excavated from Skriðuklaustur may have been very diverse in terms of origin and social status, including foreign merchants, elite members of the church and the common people of Iceland (Kristjánsdóttir, 2012).

Sampling and Analysis

A total of fifty rib samples were selected (see Table 1) according to pathological descriptions, age, sex, preservation and completeness (>50%). These bioarchaeological parameters were recorded using standard anthropological (e.g. Brothwell, 1981; Buikstra and Ubelaker, 1994; White et al., 2011; Mitchell and Brickley, 2017) and palaeopathological descriptions of the skeletal remains (e.g. Ortner, 2003; Aufderheide and Rodriguez-Martin, 2011; Roberts and Connell, 2004). Cortical bone samples from non-pathologically altered ribs were selected here for conservation and ethical reasons. Thirty-six samples were taken from individuals buried at the Skriðuklaustur cemetery, five of which were taken from individuals without pathological changes associated with infectious disease. From the site at Skeljastaðir, fourteen rib samples were taken from individuals buried in the cemetery, but only six individuals had bone changes indicative of infectious disease.

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

Mercury (Hg) concentrations in the bone and soil samples were determined by ICP-MS (inductively coupled plasma mass spectrometry) after mineralisation with closed vessel acid digestion.¹ Portions (up to 200 mg weighed to 0.1 mg) of pulverised samples together with 3 ml HNO₃ 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). ¹¹⁵In 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.

In order to control for diagenetic factors and evaluate environmental baselines for mercury, 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. At Skriðuklaustur, animal bones (n=23) representing dog and fox (*Canidae* sp.), inland fish, sea fish, seals (*Phocidae* sp.), cattle (*Bos taurus*), sheep (*Ovis aries*), goat (*Capra hircus*), swan (*Cygnus* sp.) and horses (*Equus* sp.), were measured. Soil samples (n=14) from several locations within the cemetery and just outside of the site were also measured. At Skeljastaðir, soil samples (n=9) from within the cemetery were analysed, however, no animal bones were preserved or available for study from this site.

Results and Discussion

¹ The analysis was conducted at Matís, the Icelandic Institute of Research and Development in food and bio-technology.

Regardless of species, all of the animal bone samples (n=23) from Skriðuklaustur exhibited a mercury concentration of <0.06 ppm indicating that diagenesis did not contribute to the concentrations established in the human remains. However, animal bones were not available for analysis from Skeljastaðir. Mercury levels in the soil is relatively low (average 0.035 ppm in subsoil and 0.061 ppm in topsoil) where anthropogenic origin does not occur (Salmien et al., 2005). The normal mercury concentrations (all <0.06 ppm) determined in the 22 soil samples from both sites also demonstrates that post-mortem diagenesis or contamination were not issues amongst the well preserved bones analysed in this research.

Mercury exposure at Skriðuklaustur

The presence of elevated mercury levels (>0.3 ppm) in eleven samples analysed from individuals excavated from Skriðuklaustur indicates that mercury was likely used as a medicinal treatment (see Table 1 and Figures 3-4). The mean concentration amongst those without pathological skeletal changes associated with infectious disease (n=6) was 0.190 ppm, while for those with pathological skeletal lesions (n=30) it was 0.540 ppm. The mean concentration, excluding non-adults, was lower amongst women (n=18) at 0.399 ppm than men (n=11) at 0.394 ppm. With one male outlier (SKR 174) excluded, the average for men (n=11) was 0.427 ppm. This slight difference may be correlated primarily with the small sample size; however, access to medical treatment may have differed between the sexes and social classes. Amongst older adults (>36 years) (n=16) the mean concentration was 0.327 ppm, while the younger adults (17-35 years) (n=14) had a mean concentration of 0.482 ppm. In a study by Zuckerman (2016), the same pattern, that younger adults exhibited a higher average concentration than older adults, was noted. Mercurial treatments usually began with the emergence of dermatological lesions (Zuckerman 2017a), which normally occur during the primary and secondary stages of syphilis (Baughn and Musher, 2005; Nyatsanza and Tipple, 2016). It is therefore possible that older adults, who may have entered the latent or tertiary stages (e.g. latent stage may last for more than 30 years) (Nyatsanza and Tipple, 2016), were no longer receiving mercurial treatments at the time of death. Again, this difference is also likely connected with the small sample size, but it is important to note that the lower average could potentially reflect advanced bone remodeling and the elimination of mercury over time. For example, one skeleton (SKR 23) (Figures 5-6) with skeletal changes consistent with treponemal disease (probable venereal syphilis) (i.e. cranial stellate scarring (caries sicca) and anteriorly bowed tibiae with gummatous lesions) also had one of the lowest mercury concentrations (0.069 ppm). Despite a small sample size (n=7), research conducted by Tucker (2007) found that individuals with the most extreme bone reactions also had lower mercury concentrations, potentially suggesting that mercurial treatment functioned to reduce bone activity. It is also possible that this individual refused treatment, did not survive long enough to receive treatment, or died due to iatrogenic complications or a secondary condition. One mature adult male (SKR 130), which also exhibits the skeletal features of treponemal disease (probable venereal syphilis), presented with a mercury concentration of 0.598 ppm. Two adult individuals (SKR 150 and 201) exhibited the highest mercury concentrations amongst those analysed at Skriðuklaustur. While neither showed gummatous lesions or stellate scarring on the superior cranial vault, both presented with extensive pathological changes to the tibiae. A young adult female (SKR 201) exhibited the highest concentration (1.823

ppm) of all the adults and presented with destructive bone changes resulting in several palatal perforations. It warrants consideration that a toxically elevated mercury concentration may also contribute to a younger age at death, particularly considering the unstandardised dosage used for treatment in the 16th century (see Ioannou et al., 2016; Zuckerman, 2016). An older adult male (SKR 174) presented with the lowest mercury concentration (<0.03 ppm), likely due to the exaggerated, abnormal bone remodeling associated with Paget's disease. Six individuals (SKR 10, 65, 115, 144, 174 and 221) exhibited no pathological changes associated with infectious disease, with normal mercury concentrations ranging between <0.03-0.283 ppm. It is also worth noting a female skeleton (SKR 65) without pathological bone changes who also had an elevated level of mercury (0.476 ppm). This individual was buried within the church itself, which potentially indicates that she maintained a special status within the monastery (Kristjánsdóttir, 2010). Benefactors, laymen and others residing at Skriðuklaustur may have also been at risk of exposure to mercurial vapors. Since treatment for syphilis involved frequent and extensive rubdowns with mercurial ointments, medical practitioners were consistently exposed to it. They also had to prepare fumigation rooms with mercurial mixtures and heat to initiate vaporisation, undoubtedly resulting in exposure to the vapors (Beck, 1997; O'Shea, 1990; Zuckerman, 2016). It is thereby possible that some of the individuals analysed in this research could represent the medical practitioners that provided these treatments.

Non-adults, particularly infants and young children, require special consideration during this type of research as they are prone to rapid respiratory failure when acutely exposed to mercurial vapor. Additionally, children have a lower threshold to toxicity than adults and retain far higher concentrations of mercury in their bodies than adults (Ozuah, 2000; Guzzi and La Porta, 2008). At Skriðuklaustur, approximately 90 out of the c. 295 individuals excavated were non-adults (<18 years). In this study, six non-adults were analysed, two of whom (SKR 146 and 163) were neonates that exhibited non-specific evidence of infectious disease. Congenital syphilis is often diagnosed based upon specific skeletal changes (see Ortner, 2003; Aufderheide and Rodriguez-Martin, 2011) and characteristic dental enamel abnormalities (see Ioannou et al., 2016). In children, treatment with mercury also causes enamel abnormalities, which are different from those caused by congenital syphilis (Ioannou et al., 2016). Both of these individuals exhibited elevated mercury concentrations (see Table 1, Figures 3-4), possibly resulting from transplacental transfer during gestation, or from treatments beginning shortly after birth. No dental enamel defects were present in the observable dentition, but dental changes only occur in a still unknown percentage (circa 10%-65%) of affected individuals (Ioannou et al., 2016).

Mercury exposure at Skeljastaðir

At Skeljastaðir, all of the individuals analysed showed elevated mercury concentrations, some of which were exceedingly high (see Table 1 and Figures 7-8). The mean concentration amongst those without pathological changes (n= 8) was 4.945 ppm, while amongst those with pathological changes (n=6) it was 3.151 ppm. These averages are drastically higher than those determined at Skriðuklaustur primarily because of approximately five individuals with exceedingly high mercury concentrations. As previously noted, Skeljastaðir is located close to Mt. Hekla, which erupted in AD 1104, during the occupation of the site (c. AD 1000-1104) (Þórðarson, 1943; Steffensen, 1943;

Gestsdóttir, 2014). Hekla is a known mercury emitter (Coderre and Steinthorsson, 1977), which includes passive degassing between and after eruptions (D'Alessandro, 2006), therefore, it was expected that those settled in the region would exhibit increased mercury concentrations in bone. According to historical records, nearly all 23 eruptions since, and including the AD 1104 eruption, resulted in a toxic fallout responsible for mass mortalities of livestock (Dugmore and Véststeinsson, 2012). Less commonly, some eruptions were documented to have caused poisoning or mass mortality to the human population (Grattan, 2006). Four individuals (PSK 17, 32, 34 and 54) presented with raised concentrations ranging from 1.585 ppm – 3.340 ppm, while another four (PSK 4, 29, 37 and 44) presented with remarkably high concentrations ranging from 10.134 ppm – 13.059 ppm. As some of the individuals buried at Skeljastaðir post-date the eruption, according to radiocarbon dating (see Sveinbjörnsdóttir et al., 2010), heavy mercury exposure during the eruption could be implicated. It is important to consider these extreme elevations because mercury toxicity itself can result in pathological skeletal changes, including brittle teeth, dental enamel defects, periodontitis, increased ante-mortem tooth loss and dental attrition, as well as new bone formation on the mandible and maxilla (Zuckerman, 2016). For example, one individual (PSK 29) presented with ante-mortem tooth loss of all mandibular teeth (the maxilla was unobservable), marked alveolar resorption, lingual wear and enamel defects in the anterior teeth (mottling), and a mercury concentration of 12.860 ppm. Another individual (PSK 51) presented with several distally rotated teeth, dental crowding, non-eruption of all permanent canines, mandibular periosteal new bone formation and considerable alveolar resorption, although with the less severely elevated mercury concentration of 0.438 ppm. While these skeletal changes could have occurred due to mercury exposure, fluorosis could also be implicated, along with numerous other specific and non-specific differential diagnoses.

Trade, treatment and exposure

The aetiology of the elevated mercury concentrations amongst individuals buried at Skriðuklaustur may represent a mixture of exposure points, including medicinal treatments, exposure via pigments used in scholarly work within the monastery (e.g. vermilion) and the regular consumption of marine fish and mammals (see Parsons and Percival, 2005; Mehler, 2015). Vermilion or other mercuric compounds have not yet been identified at Skriðuklaustur. The amount of mercury contamination in the oceans increased five-fold during the 19th century and ten-fold in the 20th century (Hylander and Meili, 2003; Parsons and Percival, 2005). Mercury contamination in marine animals was therefore far lower in the past than it is today, hence the consumption of marine resources was unlikely to be a serious contributor of mercury exposure in medieval populations in Iceland. As Skeljastaðir is located near the base of Mt. Hekla, the aetiology of the elevated mercury concentrations is most likely only correlated with volcanogenic emissions. Although mercury was used in a number of ways at this time, including as a medicine for skin conditions (Swiderski, 2008), its use significantly increased when venereal syphilis became prominent in Europe (Parsons and Percival, 2005). Additionally, isotopic analyses demonstrated that marine protein, a potential source of mercury, was not a primary dietary component at Skeljastaðir (Sveinbjörnsdóttir et al., 2010), unlike at Skriðuklaustur (Kristjánssdóttir, 2016). Since volcanic eruptions can last for months or even years (Simkin and Siebert, 1994) and volcanic systems passively release emissions almost continuously

(D'Alessandro, 2006), it is not possible to determine whether these elevations were caused by an eruption or simply due to chronic exposure through residency in such close proximity to Mt. Hekla. Substantially elevated bone mercury concentrations in rib samples could indicate toxic exposure relatively close (weeks to years) to the time death.

While high levels of atmospheric mercury occur in Iceland with volcanic emissions (Coderre and Steinthorsson, 1977), there are no sources of cinnabar ore (personal correspondence: Kristján Jónasson, Geologist, Icelandic Institute of Natural History). Therefore, mercury for medicinal purposes was imported. Archaeological evidence has made it clear that monastic practices and knowledge, as well as foreign objects, food and even medical materials (e.g. surgical instruments and refined sulphur) were frequently exchanged across active trade routes between Iceland and mainland Europe (Mehler, 2011; Kristján dóttir, 2016; Kristján sdóttir, 2017). Refined sulphur, which was found at Skriðuklaustur (Kristján sdóttir, 2012), was often traded for imported foods, materials and other commodities (Mehler, 2011; 2015). It is likely that substances and information about their collection, processing and uses were exchanged across both monastic and secular trade arrangements through nearby ports, such as Gautavík (Mehler, 2015) (see Figure 1).

While it remains somewhat unclear whether mercurial treatments are able to remediate syphilis infections on any level (Ortner, 2003; Tucker, 2007; Zuckerman, 2016), it is evident that it results in antemortem tooth and hair loss, severe kidney damage, lung damage, neurotoxicity and a number of vitamin deficiencies. Mercury toxicity can cause death or lead to permanent physical and psychological impairment, potentially resulting in marked disability (Graeme and Pollack, 1998; Syversen and Kaur, 2012). The identification and diagnosis of venereal syphilis remains complicated due to the overlapping skeletal changes associated with differing treponemal infections (Ortner, 2003; Harper et al., 2011). In some monastic/hospital contexts, elevated mercury levels in individuals with limited or no osseous changes could indicate treatment for treponemal infections at earlier stages of the disease. Such interpretations become much more complex in Iceland due to geothermal volatility.

Conclusions

This analysis has made important contributions to the understanding of disease and medical treatment, as well as the impact of geothermal exposure to mercury, in medieval Iceland. At Skriðuklaustur, eleven individuals with skeletal changes indicative of infectious diseases, including treponematoses, exhibited elevated mercury concentrations which were likely to have been associated with medical treatment. Individuals with pathological bone changes as well as normal or unsubstantially elevated mercury concentrations may, for example, represent people that were no longer receiving treatment, those who refused or could not afford treatment or died due to acute mercury toxicity. Alternatively, low concentrations in individuals with extensive bone change could indicate resolution or cessation of treatment followed by the elimination of mercury content through bone remodeling. The probable use of mercury at Skriðuklaustur as a medicinal treatment, in addition to other therapeutic devices (i.e. surgical tools, medicinal herbs and refined sulphur) indicates that Iceland was far from isolated in terms of medical materials and practices, via trade and monastic networks (see Kristján sdóttir, 2012; 2016; 2017).

Infections with treponemal disease, as well as many other infectious diseases, were present in Iceland during the same time period that venereal syphilis reached epidemic levels throughout Western Europe. However, this research has demonstrated that bone mercury elevations in Icelandic skeletons are not only found in individuals suffering from infectious diseases. Indeed, significantly higher concentrations were observed at the earlier, non-hospital site of Skeljastaðir, which were much more likely to originate from a volcanic source. As hypothesised, the individuals at Skeljastaðir appear to have been chronically exposed to volcanogenic and geothermal emissions and were potentially heavily exposed to high levels of mercury following the eruption. On the other hand, the less severe elevations determined at Skriðuklaustur might be explained by controlled dosage and the temporary nature of mercurial treatment. In summary, this research has demonstrated the interplay of environmental emissions, infectious disease, medico-cultural practices and the complex origin of mercury exposure in historical Iceland.

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