

Dangerous food. Climate change induced elevated heavy metal levels in Younger Stone Age seafood in Northern Norway.

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Abstract

Stable isotope and elemental analyses of the Atlantic cod (*Gadus morhua*) and harp seal (*Phoca groenlandica*) bone component of the marine food that dominated the Younger Stone Age (c. 6.1–3.5 ka BP) diet in Varanger, Arctic northern Norway, indicate, at times, climate change induced highly elevated levels of the heavy metals cadmium (Cd) and lead (Pb), and elevated levels of mercury (Hg). On average, the levels of cadmium and lead contamination in cod were up to 22 and 3 – 4 times, respectively, higher than today's recommended limits in soft tissue. The corresponding figures for seal were 15 and 3 – 4 times, respectively. The levels of Hg were generally below today's recommended limit in soft tissue, but still of considerable magnitude, almost similar to the measured values in modern fish in the Arctic. This shows that marine food in the Younger Stone Age was unhealthy, if not unsafe. We discuss this unexpected knock-on effect in terms of sea surface temperatures and sea level change. The elevated values may have been detrimental for humans, if

not for society; a balancing factor may have been a larger component of terrestrial resources than previously assumed. Concomitantly, this contribution to the paleo base-line record of toxicity may lead to predictions for seafood contamination in the future.

Keywords: Arctic seafood, heavy metal contamination, northern Norway, prehistory, Younger Stone Age.

1. Introduction

Climate change is bringing forth new issues for humankind and for our planet. One specifically vulnerable area is the Arctic. Ice sheets are melting and large biological and geological systems are being disturbed. In the Arctic Monitoring & Assessment Programme, it is stated that “though distantly located from industrialized centers and agricultural source regions, the Arctic is a sink for global pollutants” (AMAP 2017). Large areas of the Arctic are covered by peatlands and permafrost, which both act as reservoirs for many substances such as heavy metals. The mechanisms holding these in place are sensitive to climate change, sea level transgression, and erosion and could thus be involved in large future pollution events in the Arctic. The concept of climate change altering large ecological systems and causing changes in the world is, however, not new. In fact, the climate has been fluctuating throughout this planet’s history and has played a large part in shaping our present planetary environment. An aspect of recent climate change is anthropogenic involvement, which has consequences for some of these processes that extend far beyond those of past times. An example of this is heavy metal pollution, where it is well known that anthropogenic activities, such as, early mining of mercury from c. 7 ka BP and of lead from c. 5 ka BP (see Section 5) and the industrial revolution as from c. 0.2 ka BP have emitted heavy metals into the atmosphere and the sea, with subsequent input of heavy metals into food webs. It is believed, however, that there have been events with large, naturally sourced, inputs of, for example mercury, into marine ecosystems already in prehistoric times during the Holocene (Murray et al. 2015). Looking at mercury levels and other heavy metals in the remains of marine animals from the Holocene – before the time when

anthropogenic pollution could have begun to make an impact – is a way to try to understand both toxicity in seafood, and in what ways climate can affect such natural inputs of heavy metals into the food chain. It may also perhaps provide answers to how the heavy metal reservoirs of today might affect future food.

2. Background and study area

The study of prehistoric foodways has gained momentum over the past couple of decades, greatly helped by isotope and aDNA analyses (e.g., Eriksson and Lidén, 2012; Twiss, 2012; Schulting, 2014; Hastorf, 2017). However, comparatively little has surfaced concerning heavy metal toxins in the food chain of past societies and on how deep time-lines of toxin signals can be employed to predict future levels of contamination. Those non-observable toxins may have had, and most likely will continue to have, detrimental effect on individuals and society alike (e.g., Järup and Åkesson 2009; Trzcinka-Ochocka et al. 2010; Zara et al. 2015).

In a seminal study, Murray et al. (2015) found elevated mercury levels in pacific cod (*Gadus macrocephalus*) caught along the North American coast, dating back to c. 6.5 ka cal BP, and attributed this to erosion generated by rise in sea-level driven by postglacial climate change. In the present pilot study we investigate whether similar contamination, but from a broader range of heavy metals, could also be found in the prehistoric human food-web, notably among Atlantic cod and harp seal, along the shores of the Norwegian Arctic, northeast Atlantic Barents Sea, and if so, whether this could be related to similar causes.

Because of continuous isostatic uplift since the last glaciation and the formation of long series of paleo-beach ridges, the Varanger area in northeastern-most Norway (Figure 1a-c) offers an almost unique opportunity for the study of long-term trends in coastal human settlement - from c. 10 ka BP to the present. Of particular relevance for our study are archaeological sites with good preservation conditions for organic remains, providing an excellent opportunity to study economy, settlement and foodways. For the present study, we will only focus on heavy metal contamination in the Younger Stone Age from c. 6.1 – 3.5 ka BP, i.e. roughly the same time period as Murray et al.'s

(2015) older samples. The prehistoric tempo-cultural sequence in northern Norway is different from Southern Scandinavia with its adjacent parts of northern Europe and generally follows the scheme Older Stone Age (c. 12 – 7 ka BP), Younger Stone Age (c. 7 – 3.8 ka BP), and Early Metal Age (c. 3.8 – 2.0 ka BP). Put simply, the material culture in the Younger Stone Age, with its large slate-tool component, was rather different, there never was agriculture and during the Early Metal Age there never was a “true” Bronze Age (e.g. Olsen 1994).

The economy and settlement of the Younger Stone Age in Varanger has been dealt with *in extenso* (e.g., Simonsen, 1961; Renouf, 1989; Schanche, 1994; Hodgetts, 1999, 2010); suffice to say that settlement and food were predominantly coastal and marine, with household units living mostly in semi-sub-terranean pit-dwellings of varying size and shape. Among the faunal remains of seafood were fish (Atlantic cod, saithe (*Pollachius virens*), haddock (*Melanogrammus aeglefinus*) and ling (*Molva molva*)), and marine mammals such as harp seal and ringed seal (*Phoca hispida*), dolphins (*Lagenorhynchus*), and whales (*Cetacea*). Among terrestrial species we find reindeer (*Rangifer tarandus*), mountain hare (*Lepus timidus*), and beaver (*Castor fiber*); among birds, guillemot (*Uria aalge*), auk (*Alcidae*), and eider (*Somateria mollissima*) (e.g., Hodgetts, 1999, 2010), but seafood seemed invariably predominant. However, the relative proportion of species varies among and between household units. The reason for this, however, is not well understood. It may be incidental, but may also have been caused by differential access, seasonal fluctuations, different tastes, or differential consumption patterns relating to status (e.g., Schanche, 1994; Hodgetts, 1999, 2010).

Whichever the case, those were things prehistoric people knew about and could relate to. What they could not know about or relate to were toxic, non-observable aspects or properties of their food – aspects which may have had detrimental effects on both individuals and society and which are at the center of our investigation. With no human bones attainable for this study, we will focus on two of the main prey species – the next highest and highest trophic levels in the human seafood - Atlantic cod and harp seal.

3. Atlantic cod and Harp seal

Atlantic cod. The northeast Atlantic cod stock inhabits the Barents Sea, i.e., the area between the Lofoten Islands in the south and the Svalbard archipelago in the north, and roughly between Svalbard to the west and Novaja Zemlja to the east (Figure 2). Cod is a demersal fish, but seasonally pelagic (Bogstad 2016). Older cod, above 2 years, mainly feed on fish, often herring (*Clupea herengus*) or capelin (*Mallotus villosus*) and benthic organisms, while the younger cod feed on zooplankton.

The northeast Atlantic cod consists of two stocks: the migrating cod called *skrei* and coastal cod (*kysttorsk*). The *skrei* migrate between their spawning grounds and their home habitat in the Barents Sea. The archipelagoes of Lofoten and Vesterålen are the main spawning grounds for *skrei* (February to April). During the spring and summer, the young drift with the Norwegian Atlantic Current and the North Cape Current towards the north and east into the Barents Sea; the first *skrei* usually arrive in Varanger by March or April and remain until June. The coastal cod is genetically somewhat different from the *skrei*, and it does not migrate. Most fjords, including the Varanger Fjord seem to have its own cod stock (Hodgetts, 1999; Bogstad, 2016).

Harp seal is exclusively found in the northern parts of the North Atlantic, where it occurs in three separate populations with distinct breeding grounds: the White Sea, around Jan Mayen, and around Newfoundland (Figure 2). It is an ice dependent and highly migratory species. The young are born on the (drift) ice in March and weaned after approximately 12 days. They are opportunistic hunters, but seem to prefer capelin, herring, and Atlantic cod (Kovacs and Lydersen, 2015).

The harp seals found in the Varanger area stem from the White Sea. Immature seals mostly appear in February – March, whereas mature seals migrate to the area in greater quantities between March and April, and pups even later (Hodgetts, 1999).

Although we currently have no data on prehistoric migration patterns for Atlantic cod and harp seal, we anticipate that there have been ecological amplitudes, just like in palaeobotanical and dendrochronological studies, also during the Younger Stone Age (e.g., Helema et al. 2013).

4. Cadmium, lead and mercury as pollutants

While some chemical elements, such as, selenium, copper and chrome, are vital in low concentrations for humans, but poisonous in high doses, the heavy metals cadmium, lead and mercury have no vital function in humans and are, according to the Norwegian Food Safety Authority, among the most detrimental to human health (NFSA).

Cadmium occurs naturally in small amounts in soils, particularly in areas abundant with alum-slate and enters the environment by volcanic eruptions and the erosion of bedrock (NIPH1). It was known to the Romans c. 2 ka BP and its extraction is generally associated with the mining of zinc (ISE 2019). It occurs in seawater in a variety of dissolved and solid forms, originating from river drainage, erosion in coastal areas, and the atmosphere. How it is circulated by sea-currents and concentrated in different parts of the oceans is less understood, but concentrations are expected to be higher in close geographical proximity to rich, natural sources. Cadmium is bio-accumulative; with a biological half-life of 10-30 years, it accumulates in the body with age, particularly in the liver and kidneys (NIPH1). It has been shown to cause cancer, kidney, liver and lung disease, and may lead to weakening of the skeleton (osteoporosis). Pregnant women and children are most at risk. It may also provoke deficiencies in reproduction (NIPH1). For those reasons, the European Food Safety Authority (EFSA) has imposed maximum limits for consumption of cadmium-rich foods. For fish and seal the maximum limit is 50 ng/g in soft tissue (edible parts), with a tolerable weekly intake of 2.5 micrograms per kilo (2.5 ng/g) bodyweight per week (NIMR1). Cd values have been measured on human skeletal elements in a healthy modern Spanish population where the mean value was 40 ng/g (Garcia et al. 2006).

Lead occurs naturally in small amounts in soils, and has been mined for more than 5000 years (Rich 1994). It enters the oceans by river drainage, erosion of coastal zones, and the atmosphere (NIPH2). However, its circulation and concentration in different ecological regimes of the oceans is not well understood. The half-life of lead varies from about one month in blood, 1-1.5 months in soft tissue, to about 25-30 years in bone (MDH), thus it tends to accumulate in the human skeleton with age. It is

also bio-accumulative and known to cause detrimental effects on the human brain and nervous system and have negative effects on learning abilities. It may do serious harm to fetuses and cause reduced capacity of reproduction. Based on the toxicity of lead, EFSA has imposed a maximum limit for lead in fish (and seal) of 300 ng/g for soft tissue (NIMR2). Pb values have been measured on human skeletal elements in a healthy modern Spanish population with a mean value of between 280 and 3500 ng/g (Garcia et al. 2006).

Mercury has been mined for about 7000 years (Hernández et al., 1999, Gajic-Kvašcevic et al., 2012). It can be a local, regional and global pollutant and is toxic to most animals. It occurs naturally in the earth's crust and it can be naturally emitted and re-emitted in volcanic eruptions, but also in forest fires and from water columns (AMAP 2011). However, in conjunction with the industrial revolution and the burning of fossil fuels that it brought about, anthropogenic-caused mercury release has been increasing (AMAP, 2011). When mercury is transformed into methylmercury (MeHg) it becomes bioavailable and can accumulate in food webs. Once mercury has been methylated it will have the capability to accumulate and bio-magnify in the food web by trophic transport from lower to higher levels. MeHg can enter marine food webs through sediments or dissolved organic matter in the ocean (Murray et al., 2015).

The Arctic regions have large areas covered in permafrost and peatlands (Gorham 1991). Permafrost soils are the biggest reservoir for mercury. When the soil is frozen, mercury bound in the soil becomes immobilised. However, Klaminder et al. (2008) regard thawing palsas (pingo) bogs, and the subsequently mobilized mercury, as a considerable future source of mercury emissions. In humans, mercury may lead to dietary indigestion and affect the nervous system, leading to neurological symptoms (starting at concentrations above 1.6 mg/kg of body weight) such as: nervousness, anxiety or mood changes, numbness, memory problems, depression and, physical tremors (e.g., Azevedo et al. 2012). Based on the toxicity of mercury, EFSA has imposed a maximum limit for mercury in fish (and seal) of 500 ng/g for soft tissue (NIMR2). In a healthy contemporary

Spanish human population Hg values in skeletal elements were measured to a mean value of 10 ng/g (Garcia et al. 2006).

5. Materials and methods

We selected eight archaeological sites, or contextual units from the sites, dating to the Younger Stone Age; 1) Skjåvika (Gjessing, 1938), 2) Høyvikhaugen, the midden between Houses 1 and 2 (Niemi and Oppvang, 2014), 3) Mortensnes, House 52 and 57, field A (Schanche, 1988), 4) Gropbakkeengen, House 3 (Simonsen, 1961), 5) Gressbakken Nedre Vest, House 4 (Simonsen, 1961), 6) Advik, Houses B and F (NE-midden) (Simonsen, 1961), 7) Nyelv Nedre Vest, House 3 (Simonsen, 1961; Renouf, 1989) and 8) Bergeby, House 18 (Schanche, 1994) (Figure 1b). The sites/units span the time 6.3 – 3.8 ka cal BP. We opted for as many bones as possible of Atlantic cod and harp seal, respectively, from the food refuse from each site or household. Being the generally most consumed seafoods and representing the next-highest and highest trophic level in the human diet, respectively, they will serve as proxies for toxicity in human seafood consumption. We have chosen to analyse bones, not only because of a lack of preserved soft tissue from the species, but because we are interested in long-term accumulation of toxins.

5.1 Sampling

We used a set protocol for sampling, and isotope and element analyses. The bones were sampled for stable carbon $\delta^{13}\text{C}$ and nitrogen $\delta^{15}\text{N}$ isotope analysis, to address bioaccumulation (see Järup and Åkesson 2009; Trzcinka-Ochocka et al. 2010; Zaza et al. 2015), and for elemental analysis (Cd, Pb, Hg). We aimed to sample compact cortical bones from which the collagen is usually well preserved; dentary and vertebrae for cod, lower jaws and femurs for seal. However, in some cases where such bones were absent, other bones were used. We made sure we only sampled each individual once, meaning that all samples represent one individual.

Based on the bones, it proved impossible to distinguish with any certainty between the usually larger migratory cod (*skrei*) and the local non-migratory coastal cod. At sites with no

identified harp seal bones (Gropbakkeengen, Skjåvika), remains attributed to the general seal family (*Phocidae*) or ringed seal (*Phoca hispida*) were used. We did not target ringed seal as part of our study *per se*, only as occasional substitutes for harp seal. Even though there are some differences in the behavior between the two stocks of Atlantic cod and between the different seal species, we consider that to be of no significant consequence for the main purpose of this paper. Also, by comparing the averages of the harp seal data with the other seal data it appears that $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ remain constant and although the values for cadmium and lead appear slightly higher when all seals are included, the general magnitudes of contamination remain the same.

Prior to drilling the samples for bone powder, the bones were cleaned ultrasonically in deionized water and the surface layer was removed by a dental drill. Then another dental drill was used to obtain the bone powder (around 100 mg from each specimen) to be subjected to analyses. The bones were retrieved from the collections at the Bergen University Zoological Museum and at the Tromsø University Museum, UiT The Arctic University of Norway. All subsequent sampling took place at the Archaeological Research Laboratory at Stockholm University and the Tromsø University Museum. Stable isotope analysis was performed at the Stable Isotope Laboratory (SIL) at the Department of the Geological Sciences, Stockholm University and the element analyses were performed by ALS Scandinavia, Luleå, Sweden.

5.2 Stable isotope analyses.

Bone powder (c. 50-80 mg) was obtained by means of a dentist's drill, discarding the surface layer to avoid contamination. Collagen was subsequently extracted by the method of Brown et al. (1988), which includes ultrafiltration to remove the <30 kDa fraction potentially containing contaminants of low molecular weight. As cod and seal bones contain high levels of fat, the samples were also subjected to a lipid removal procedure (Lidén et al. 1995). This was done right after the demineralization of the bone powder by immersing the remainder in a chloroform/methanol solution (2:1), shaking it on a shaking unit for two hours, filtering and finally leaving it in a fume hood for 24

hours to evaporate the chloroform/methanol solution. All the sampling and extraction was performed at the Archaeological Research Laboratory, and the subsequent EA-IRMS analysis took place at the Stable Isotope Laboratory (SIL), Dept. of Geological Sciences, both at Stockholm University. Collagen was weighed into tin capsules (c. 0.5 mg for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ analysis) for combustion in a Carlo Erba NC2500 elemental analyser connected to a continuous flow isotope ratio mass spectrometer – a Finnigan MAT Delta+ for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. The precision of the measurements was $\pm 0.15\%$ or better for both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. Collagen quality criteria for the samples include carbon and nitrogen C/N ratios of 2.9-3.6 (DeNiro 1985) as well as C and N concentrations between 15.3-47% and 5.5-17.3% respectively (Ambrose 1990). We have only included bones that followed the quality criteria for collagen in the elemental analyses since, at present, there is no consensus regarding quality criteria for bioapatite.

5.3 Elemental analyses.

About 20 mg of bone powder from each specimen was sent to ALS Scandinavia for element analyses. Dissolution took place in a microwave oven in a closed teflon container with $\text{HNO}_3/\text{H}_2\text{O}_2$. Analysis with ICP-SFMS was according to the Swedish Standards Institute SS EN ISO 17294-1, 2 and the EPA-method 200.8, and only values above the limit of quantification (LOQ) are reported, here defined as 10 times the standard deviation for the blank, i.e. approximately three times higher than the detection limit. Values are reported in nanograms per gram (ng/g).

5.4 Radiocarbon dating.

The ^{14}C -dates are presented in Table 1. We have aimed to date one out of eight samples per species and site. In cases where this was not possible, we have used single ^{14}C -dates or an average of multiple ^{14}C dates from the given archaeological context. We have re-calibrated all non-AMS dates using IntCal13 and the Marine13 radiocarbon age calibration curves 0 – 50 ka cal BP (Reimer et al., 2014). All age-determinations made on bones from marine mammals have been corrected for the

marine reservoir effect by applying the average value of weighted mean ΔR and standard deviation offset ($\Delta R = -8$, Std. = 65), derived from the five nearest points in and around the Varangerfjord area (points 55, 679, 1061, 1070, 1074) found in the CHRONO Marine Reservoir Database (CMRD), to the OxCal v4.3.2, IntCal13 and Marine13 curve (Brown et al., 2019).

6. Results and discussion

A total 124 bones (76 cod, 48 seal) were sampled, but only 40 (16 cod, 24 seal) produced enough collagen of good quality for isotope and elemental analyses.

All data is listed in Table 1, which includes only samples with collagen of good quality for analyses.

6.1 Stable Isotope analyses

The descriptive statistics for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ are listed in Table 2, and plotted in Figure 3.

6.1.1 Cod

The data generally seem to be in agreement with the variation found in Atlantic cod by Petersen et al. (2015). Compared to Barrett et al.'s (2011, Figures 2 and 3) distributions for Atlantic cod, however, our data have a much wider distribution for both isotopes. Both geographical differences and variations in diets and thus fish size may contribute to isotope variation. Among the latter one may mention migration patterns as a likely contributing factor. We consider it likely that the two groups found in our data represent coastal cod (low $\delta^{13}\text{C}$, low $\delta^{15}\text{N}$) and migratory *skrei* (high $\delta^{13}\text{C}$, high $\delta^{15}\text{N}$), respectively. This, however, does not have any immediate consequences for our results on the elemental analysis.

6.1.2 Seal

The descriptive statistics for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ are listed in Table 2. A scatter plot is provided in Figure 3. The stable isotope values fit well with Nelson et al.'s (2012) analyses of harp seal consumed by the Medieval Norse in Greenland, except for three values. All three exceptions, two from Skjåvika and one from Bergeby, have $\delta^{15}\text{N}$ values above -17.0‰ and one from Skjåvika has a $\delta^{13}\text{C}$ value close to -

18.0‰. Very low $\delta^{13}\text{C}$ values (c. -17.8‰ to c. -19.4‰) have, for instance, also been found among adult harp seals in the northwest Atlantic, including the Gulf of St. Lawrence and the Scotian shelf (Tucker et al., 2013).

7. Elemental analyses

Data from the elemental analyses is listed in Table 1 and illustrated in Figures 4a-c.

A relatively high degree of variation among the element data is readily apparent from the table and figures. All sites, except for Høyvikhaugen and Skjåvika are old excavations from which no geochemical samples were taken to evaluate background noise. However, at Skjåvika it is clear that the concentrations of Cd and Pb vary independently of age and layer. Consequently, the variation cannot have been caused by the natural levels in the earth in which they were deposited. Also, as to the older excavations, the micro-topography does not suggest the possibility of influx of contaminants from the surroundings. Moreover, the sites are all placed on beach ridges that formed only shortly before settlement leaving virtually no time for pollution from the air. Further, we have only included bones that comply with the quality criteria for well preserved collagen. Thus, we are quite confident that our samples have not been contaminated from their surrounding soil matrices.

Inter-site variability may be ascribed to variations in climate and temperature, whereas intra-site variability may derive from differences in age, skeletal element, stock or subspecies variation or variation in feeding or migration pattern. Brown et al. (2016:503) state that " .., both natural geographical differences and diet variability among regions explain the spatial patterns for THg and TCd concentrations in ringed seals" – and this probably goes for harp seals as well – and that "high TCd concentrations are related to seals feeding more on invertebrates than fish" (Brown et al. 2016:503). Although there is considerable variation in Cd among the three seals with deviating isotope values, we suggest that these three examples indicate a migration and/or feeding pattern somewhat different from the rest. Again, this does not have any immediate consequences for our results below.

Differences in age (when killed) and average contents of heavy metals, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ derived from seal femurs from Bergeby and Gressbakken, are presented in Table 3. Although the small number of samples does not invite statistical analysis, the figures indicate that juveniles contain 1.6 times more Cd and 3 times more Pb than adults, but roughly the same amount of Hg. This is somewhat surprising as it is generally assumed that older animals or fish, affected by bioaccumulation, contain higher values of toxins (Nascimento et al. 2016). A likely explanation in this case is the possible differing feeding and/or migration patterns, but also the effect of lactation in the juveniles, where the mother transfer her elevated values through the placenta or with the milk.

At Skjåvika, the two isotopic cod groups may indicate that they represent *skrei* and coastal cod, respectively. The Cd and Pb values for sub-group 1 ($12\text{‰} > \delta^{15}\text{N} < 14\text{‰}$) are 2887 ng/g and 1220 ng/g, and for sub-group 2 ($14\text{‰} > \delta^{15}\text{N} < 16\text{‰}$) 2935 ng/g and 1525 ng/g, respectively. Again, given the few cases in each group, these data do not encourage statistical treatment. We observe, however, that the difference in Cd contents is minimal; it is much larger for Pb with sub-group 2 showing the highest figure. This is interesting since Cd and Pb contents seem to correlate positively with $\delta^{15}\text{N}$ for both, indicating bioaccumulation in both these elements, since higher nitrogen values indicate higher trophic levels. Again, one possible explanation could be differential feeding and/or migration patterns for *skrei* and coastal cod. It is interesting that all the Varanger Fjord sites show “*skrei*” isotopic values, while Skjåvika on the outer coast also has the specimens with coastal cod isotopic values. However, Skjåvika faces a major fjord to the west. The evidence from the Varanger Fjord sites may suggest that those sites were mainly used – or at least used – during the late winter or spring when the *skrei* appears.

What also points to differential feeding/migration patterns are the disparate Cd and Pb values for the three seal samples from Skjåvika (Cd = 2000 ng/g, 1880 ng/g, and 257 ng/g, and Pb = 584 ng/g, 4420 ng/g, and 425 ng/g, respectively). Given that Cd and Pb contamination of seafood was severe, the obvious question is: did this have a (long-term) effect on humans, and if so, what effects?

Lacking information from human skeletons, this is difficult to assess, but with the very high values of toxins one would expect effects to have been detrimental for humans, if not for society. A balancing factor may have been extensive use of terrestrial resources. However, reindeer generally make up less than 5% of the mammalian remains on the Varanger sites (Hodgetts 2010) and unfortunately it is unknown to what extent they used plants or fruits.

7.1 Cod

Cadmium shows average values significantly above today's critical limit for soft tissue (50 ng/g) for all sites or units (Table 4), as does lead for Høyvikhaugen and Skjåvika, whereas Nyelv has slightly higher values above today's critical limit of 300 ng/g for consumption of soft tissue. For Hg, although some sites have elevated values, no sites show average values above today's critical limit at 500 ng/g for soft tissue. The data clearly indicate that Atlantic cod, for a period of about 2.2 ka (c. 6.3 - 4.1 ka cal BP) during the Younger Stone Age, show highly elevated, dangerously toxic, levels of Cd and Pb and Hg concentrations.

7.2 Seal

Also for the seals, the average cadmium values for all sites or units are way above today's critical limit at 50 ng/g for consumption of soft tissue (Table 4), as are the mean values for lead for the sites or units Bergeby, Mortensnes, and Skjåvika (300 ng/g), with Gressbakken coming close at 282 ng/g. As for the cod, the mercury values are elevated but fall below today's critical limit of 500 ng/g for consumption of soft tissue. Again, it is striking that harp seals also show very high, dangerously toxic, levels of Cd and Pb and elevated levels of Hg during 1.7 ka (5.5 – 3.8 ka cal BP) of the Younger Stone Age.

8. Heavy metals, temperatures and sea-levels

As indicated at the outset, the above variation in levels of contamination may be related to Holocene climate and ecosystems change. With often wide differences in chronological and spatial resolution, differences in calculation methods and smoothing techniques, the correlation of natural scientific

climate and biogeochemical proxies is challenging (e.g., Blankholm 2011, 2018). Many variables may have contributed to the elevated levels of toxicity we have found, such as, volcanic activity, raising sea-surface temperatures, and erosion caused by rising sea-levels. A perusal of volcanic activity (Pilcher et al. 2005, Helema et al. 2013, Vorren et al. 2007) show six events predating different sites with the shortest possible time-span (see Table 1). Those time-ranges are 22, 56, and 83 years, respectively; the longest interval is 348 years. As far as we know, there are no exact estimates of discharge of heavy metals from volcanoes into the Barents Sea. From Helema et al. (2013) and Baillie (2010), the climatic effect of eruptions tends to fade within a year or two. It would thus seem reasonable to suggest that downpour of heavy metals did not exceed a couple of years and would probably have lost effect when the occupations took place. Moreover, it must be considered that tephra layers in Northern Fennoscandia (and the equivalent deposition in the sea) are extremely thin indicating very little *de facto* accumulation with potential to enter into the biosphere. Even if huge amounts of heavy metals were discharged on Jan Mayen or on Iceland, those would probably not have reached the Barents Sea.

With all this uncertainty we exclude volcanic activity for further consideration for the present.

We will, however, explore two causes that may have had a direct bearing on the quality of seafood: sea surface temperature (SST) and sea-level change, and focusing on the peaks in the trends for Cd and Pb for cod around 5.5 ka cal BP and Hg for cod around 4.6 ka cal BP (Figures 4a-c).

8.1 Sea surface temperature

Given that higher temperatures generally provide higher levels of nutrition, biomass, and, not least, emission and release of heavy metals, we investigated how temperature correlates with levels of heavy metals. In Table 1 we have listed the SSTs at 100 m depth from the PSh-5159N core monitoring the North Cape current at Ingøy dypet at 79° 21.80N/22° 38.77E (Risebrobakken et al., 2011), recalculated by Katrine Husum of the Norwegian Polar Institute and following the method developed by Husum and Hald (2012). Within our time-brackets for cod and seal, which begin approximately 1.8

and 2.5 ka, respectively, after the peak of the Holocene climatic optimum at c. 8.0 ka BP, it appears that SST variation was limited; between 5.1 and 6.1 °C for cod and 5.7 and 6.1 °C for seal. This is not the same as saying that small changes may not have been ecologically significant, but sources of error inherent to the dating and temperature calculations may have had some impact. Scatterplots of SST and chemical element per species are provided in Figure 5a-f.

The data do not lend themselves to parametric correlations, but the Spearman Rank Correlation test is applicable in four of the six cases (see Table 5). No correlation is statistically significant, but there does, however, seem to be some apparent trends. For cod, SST and Cd seem to have a positive, but weak correlation. SST and Pb show an apparent moderate and positive trend. For seal, SST and Cd one may perceive a weak negative, for SST and Pb a weak positive trend, but in both of the latter cases we consider the nature of the distribution inadequate for meaningful correlation.

Although the above apparent trends may be indicative, they do not invite more than suggestive conclusions. To this end we need more data from both species, not only to fill in gaps in the data series, but also to allow us to investigate a fuller chronological range covering both the early and mid-Holocene (c. 10.0 – 6.5 ka cal BP) and the time period c. 3.7 – 2.0 ka cal BP. Also, delayed effects of temperature rise in heavy metal content in the sea and seafood is not well understood. It may very well have taken a good while before effects became apparent. If or when we become able to demonstrate more powerful, statistical significant, relationships based on a broader range of data, this could then serve for predictions about future levels of heavy metals in seafoods.

8.2 Sea-level change.

The mid-Holocene transgression, culminating at c. 6.6 ka cal. BP, is supposed to have caused the release of huge quantities of heavy metals by erosion of landmasses (e.g., Murray et al., 2015).

Relative sea-levels (sea-levels relative to isostatic postglacial rebound of land) have been studied in the Varanger area for about a century (e.g., Blankholm, 2018). By chance, one of the most thorough studies was made on the palaeo-shoreline formations at Brannsletta, in the vicinity of the sites Gressbakken, Nyelv and Advik (Fletcher et al., 1993). The drawing in Blankholm (2018, Figure 4),

shows that the spike for Cd and Pb in cod at c. 5.5 ka cal BP (Figure 4a-b) coincides with the beginning of a plateau-like interval of iso/eustatic (near) equilibrium. This means a phase where sea-level rise kept pace with or maybe even exceeded land rebound, and an extended period for erosion at the same absolute height. Another spike of cod and Hg at c. 4.6 ka cal BP (Figure 4c) also occurred during this interval. This may corroborate Murray et al.'s (2015) finding that erosion of landmasses may form part of an explanation for high values of heavy metals, at the least in cod. However, again we need a broader range of data to investigate this further.

9. Conclusions

In this study we found highly elevated levels of the toxic heavy metals cadmium, lead, and mercury in well preserved skeletal remains of Atlantic cod and harp seal from the north Norwegian coast, dating to the Younger Stone Age. For cod those levels of cadmium and lead were up to 22 and 3 – 4 times higher than today's recommended limits for consumption of soft tissue, respectively; for seal the corresponding figures were 15 and 3 – 4 times, respectively. For mercury, levels were generally below today's recommended limit, but still of substantial magnitude. We have used cod and seals as proxies for understanding how the load of these heavy metals might have been on the contemporary human population in the area. Marine food during the Younger Stone Age in Varanger was unhealthy if not unsafe.

This is surprising considering that a heavy load of these metals into the arctic marine fauna is generally associated with the onset of the Anthropocene and the following pollution caused by the industrial revolution. We have found a correlation in shoreline displacement and the elevated values for Atlantic cod. We have also found a positive, but weak (non-significant) correlation for SST and cadmium and a positive, but moderate (non-significant) correlation for SST and lead for cod. For seal, the nature of the distributions for SST, Cadmium and lead appears inadequate for meaningful correlation. From the outset, one would expect all the metals in both species to co-vary with SST. However, with our present data we cannot pursue this problem further.

Further studies from the entire Holocene, at both regional and circumpolar scales are warranted for a better understanding of both how climate change induced heavy metal levels linked with prehistoric hunter-gatherer health, economies and culture change, and with various aspects of marine ecosystems, such as, hot-spots for heavy metal outwash and variations in sea-currents and species migration patterns.

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Data availability

The data generated by this research is available on request to the corresponding author.

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List of figures.

Figure 1. Maps of a) the northeastern Atlantic, b) northern Fennoscandia, and c) the study area with sites mentioned in the text. Map by Johan Arntzen.

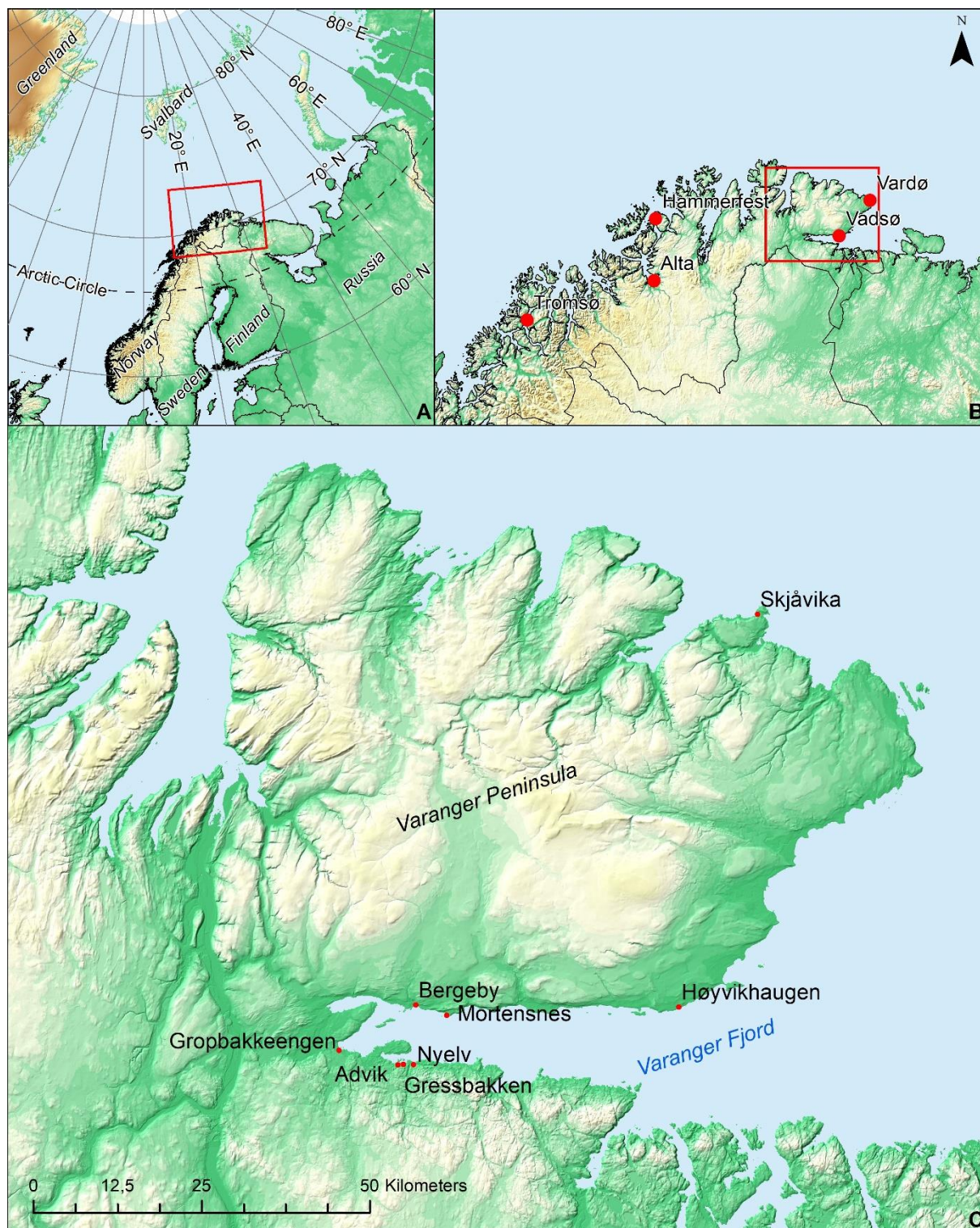


Figure 2. Spawning grounds for northeast Atlantic Arctic cod (modified from Bogstad 2016 with permission from the Institute of Marine Research) and breeding areas and range of the harp seal (modified with permission from C. W. Sanger 1977). Cod: light blue = foraging area, black arrow = spawning migration route, orange = spawning area. Harp seal: solid red = breeding ground, red line = migration route.



Figure 3. Isotope values for Atlantic cod and harp seal in the Varanger area, northern Norway.

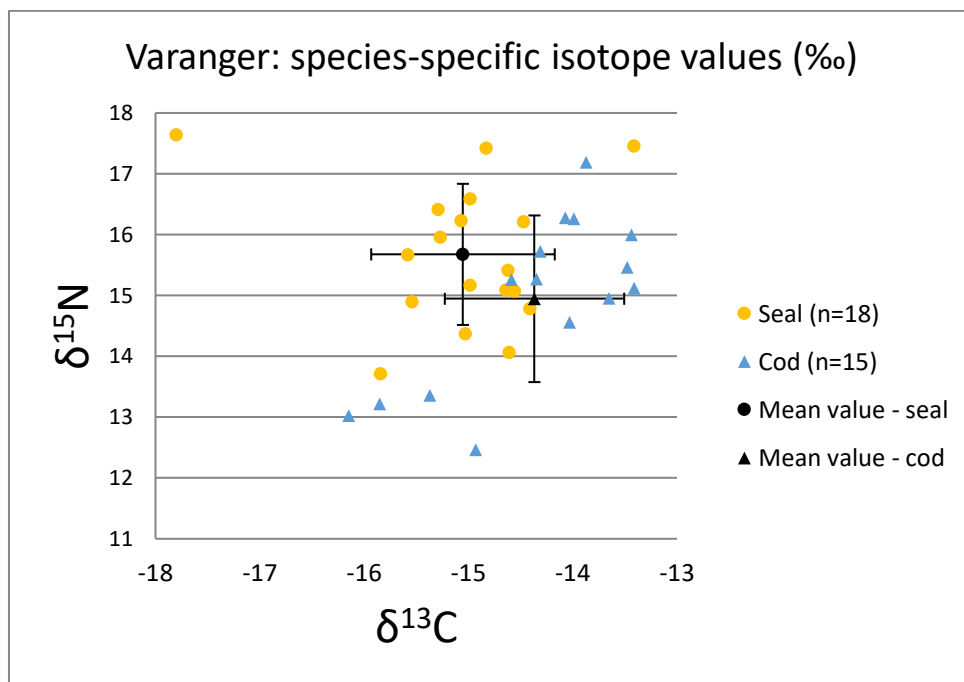


Figure 4. (a) Cadmium concentration (ng/g) for Atlantic cod and harp seal. Limit value = European Food Safety Authority (EFSA) maximum limits for consumption of cadmium, (b) lead concentration (ng/g) for Atlantic cod and harp seal. Limit value = European Food Safety Authority (EFSA) maximum limits for consumption of lead, and (c) mercury concentration (ng/g) for Atlantic cod and harp seal.

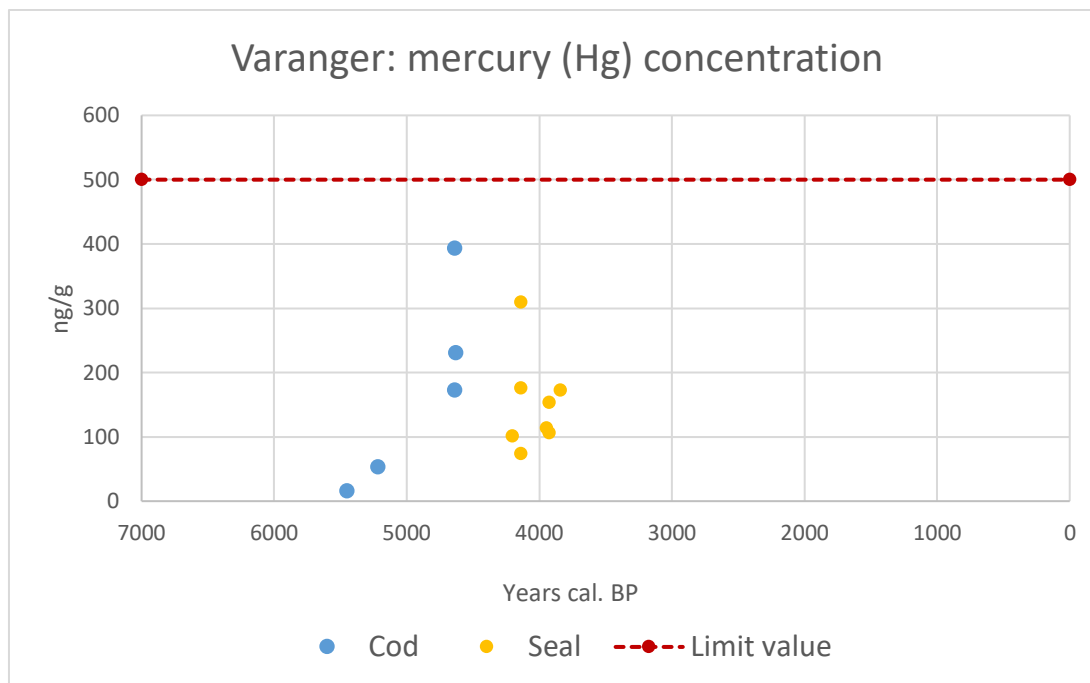
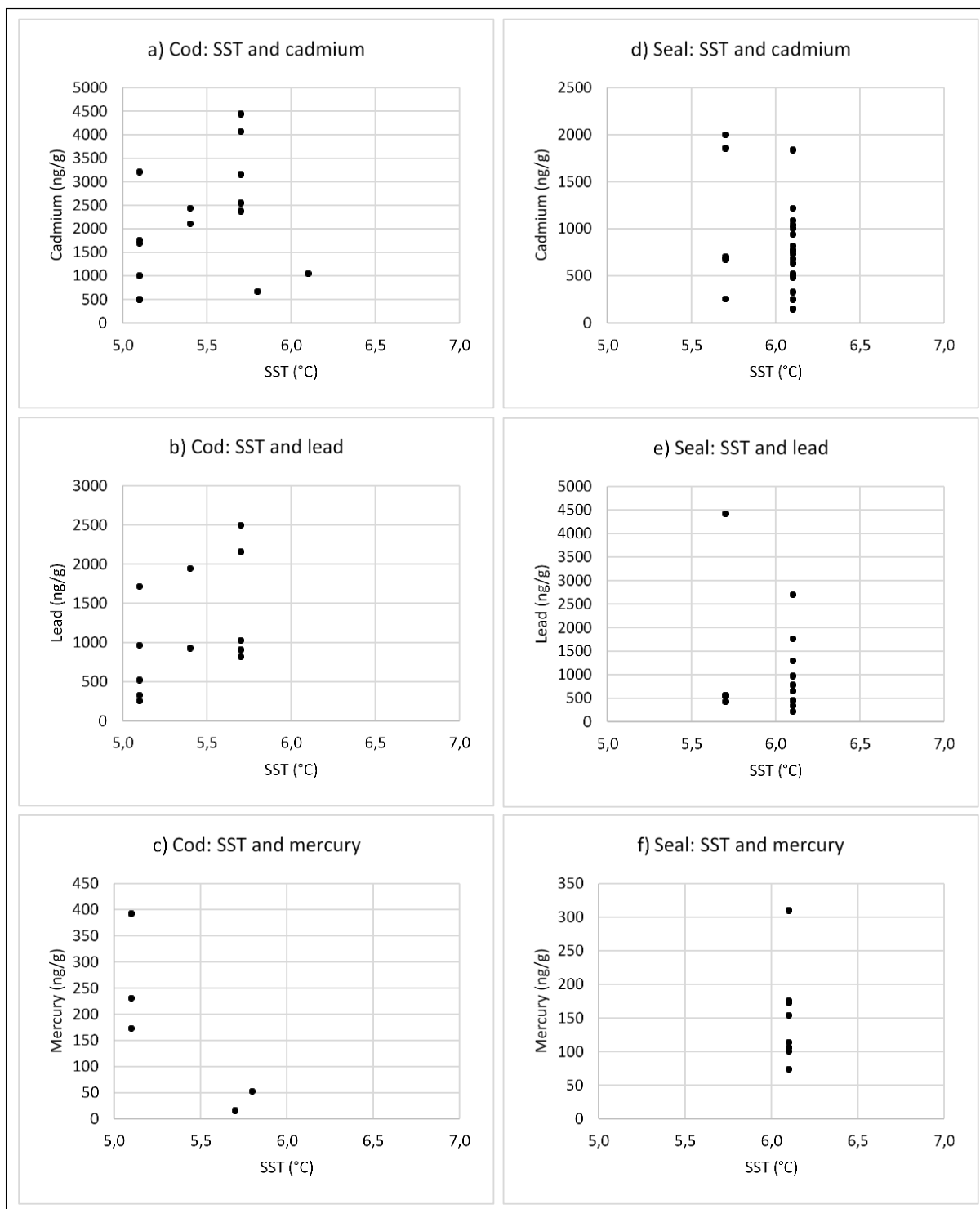


Figure 5. Scatterplots of SST and chemical element for Atlantic cod and seal: a) cod, SST and kadmium, b) cod, SST and lead, c) cod, SST and mercury, d) seal, SST and cadmium, e) seal, SST and lead, f) seal, SST and mercury.



List of tables.

Table 1. Cd, Pb, Hg, $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$, values for cod and seal. Elemental concentration in ng/g, stable isotopes in ‰. * = below limit of quantification (LOQ).

Sample ID	Site	Unit	Species	Age cal BP	Temp. °C	Cd (ng/g)	Pb (ng/g)	Hg (ng/g)	$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)	% C	% N	C:N
HVH 12	Høyvikhaugen	Midden between Houses 1 and 2	Atlantic cod	6258	5.4	2440	1950	*	-13.6	15.0	37.21	12.86	3.38
HVH 14	Høyvikhaugen	Midden between Houses 1 and 2	Atlantic cod	6258	5.4	2110	930	*	-13.9	17.2	40.85	14.48	3.29
SKJ 4	Skjåvika	Layer 4	Atlantic cod	5450	5.7	2550	1030	*	-15.8	13.2	33.08	11.88	3.25
SKJ 6	Skjåvika	Layer 4	Atlantic cod	5450	5.7	3160	821	*	-14.9	12.5	39.53	12.95	3.56
SKJ 9	Skjåvika	Layer 4	Atlantic cod	5450	5.7	4450	2500	*	-13.4	15.1	41.17	15.01	3.20
SKJ 20	Skjåvika	Layer 4	Atlantic cod	5450	5.7	4070	2160	16	-15.4	13.4	32.72	11.16	3.42
SKJ 22	Skjåvika	Layer 4	Atlantic cod	5450	5.7	2380	909	*	-14.4	15.3	36.77	12.83	3.34
ADV 17	Advik	House F, NE midden	Atlantic cod	5216	5.8	673	*	53	-13.5	15.5	33.96	11.62	3.41
SKJ 7	Skjåvika	Layer 3	Atlantic cod	4890	5.1	1770	334	*	-16.2	13.0	32.52	11.68	3.25
SKJ 24	Skjåvika	Layer 3	Atlantic cod	4890	5.1	1700	970	*	-14.0	14.6	36.76	13.04	3.29
SKJ 26	Skjåvika	Layer 3	Atlantic cod	4890	5.1	-	-	-	-14.3	15.7	41.41	14.76	3.27
SKJ 28	Skjåvika	Layer 3	Atlantic cod	4890	5.1	3210	1720	*	-14.6	15.3	41.93	14.67	3.33
NYV 3	Nyelv	House 3	Atlantic cod	4640	5.1	500	524	173	-13.4	16.0	35.42	12.12	3.41
NYV 5	Nyelv	House 3	Atlantic cod	4640	5.1	1010	259	393	-14.0	16.3	36.90	12.61	3.41
NYV 6	Nyelv	House 3	Atlantic cod	4629	5.1	504	*	231	-	-	-	-	-
GRN 14	Gressbakken	House 4	Atlantic cod	4139	6.1	1050	*	*	-14.1	16.33	34.59	12.21	3.31
GKN 2	Gropbakkeengen	House 3	Ringed seal	5454	5.7	704	*	*	-	-	-	-	-
SKJ 14	Skjåvika	Layer 4	Seal family	5450	5.7	257	426	*	-13.4	17.5	39.40	14.07	3.27
SKJ 16	Skjåvika	Layer 4	Ringed seal	5450	5.7	679	561	*	-15.5	14.9	39.24	13.91	3.29
SKJ 17	Skjåvika	Layer 4	Seal family	5450	5.7	2000	564	*	-15.8	13.7	42.52	15.10	3.28
SKJ 18	Skjåvika	Layer 4	Ringed seal	5450	5.7	1860	4420	*	-17.8	17.6	42.75	15.36	3.25
GRN 4	Gressbakken	House 4	Harp seal	4204	6.1	529	*	101	-	-			
GRN 1	Gressbakken	House 4	Harp seal	4139	6.1	777	225	74	-15.0	14.4	39.14	13.75	3.32
GRN 3	Gressbakken	House 4	Harp seal	4139	6.1	737	*	176	-14.	14.0	39.91	13.88	3.35
GRN 5	Gressbakken	House 4	Harp seal	4139	6.1	1040	340	310	-14.6	15.1	37.17	12.89	3.36
BRY 4	Bergeby	House 18	Harp seal	3998	6.1	253	*	*	-14.4	14.8	37.51	12.96	3.38
ADV 10	Advik	House B	Harp seal	3973	6.1	634	*	*	-	-	-	-	-
GRN 2	Gressbakken	House 4	Harp seal	3962	6.1	510	*	*	-	-	-	-	-
ADV 8	Advik	House B	Harp seal	3947	6.1	686	*	114	-15.3	16.4	38.76	13.35	3.39
ADV 9	Advik	House B	Harp seal	3947	6.1	1090	*	*	-15.6	15.7	32.62	11.07	3.44
ADV 11	Advik	House B	Harp seal	3947	6.1	820	*	*	-15.1	16.2	40.51	13.99	3.38
BRY 2	Bergeby	House 18	Harp seal	3925	6.1	146	*	*	-14.6	15.1	37.24	12.98	3.35
BRY 3	Bergeby	House 18	Harp seal	3925	6.1	332	653	*	-14.6	17.4	36.43	12.78	3.33
BRY 5	Bergeby	House 18	Harp seal	3925	6.1	1220	786	*	-15.0	16.6	38.93	13.38	3.39
BRY 6	Bergeby	House 18	Harp seal	3925	6.1	484	453	*	-15.0	15.2	35.19	12.23	3.36
BRY 8	Bergeby	House 18	Harp seal	3925	6.1	943	980	106	-15.3	16.0	38.36	12.98	3.45
BRY 9	Bergeby	House 18	Harp seal	3925	6.1	153	*	*	-14.6	15.4	36.47	12.71	3.37

BRY 16	Bergeby	House 18	Harp seal	3925	6.1	767	1770	154	-14.5	16.2	37.48	13.08	3.34
MOR 2	Mortensnes	House 52 and 57, field A	Harp seal	3870	6.1	1840	1300	*	-	-	-	-	-
MOR 1	Mortensnes	House 52 and 57, field A	Harp seal	3841	6.1	1010	2710	173	-	-	-	-	-

Table 2. Descriptive statistics for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values in cod and seal.

Species	Isotope	N	Range (‰)	Min. (‰)	Max. (‰)	Mean (‰)	Std. (‰)	Var. (‰)
Atlantic cod	$\delta^{13}\text{C}$	15	2.7	-16.2	-13.4	-14.4	0.86	0.74
Atlantic cod	$\delta^{15}\text{N}$	15	4.7	12.5	17.2	14.9	1.37	1.87
Seal	$\delta^{13}\text{C}$	18	4.4	-17.8	-13.4	-15.1	0.88	0.77
Seal	$\delta^{15}\text{N}$	18	3.9	13.9	17.6	15.7	1.16	1.34

Table 3. Average contents of elements (ng g⁻¹) for juvenile and adult seal femurs at Bergeby.

Element	Cd (ng/g)	Pb (ng/g)	Hg (ng/g)	$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)
Juvenile (< 1 year)	816 (n=4)	1047 (n=4)	130 (n=2)	-14.9 (n=4)	16.5 (n=4)
Adult	513 (n=7)	339 (n=3)	187 (n=3)	-14.7 (n=7)	14.9 (n=7)

Table 4. Average and standard deviation of Cd, Pb, and Hg in Atlantic cod and harp seal bones per site in ng/g.

Species	Element	N	rho	Sign. 95%-level
Atlantic cod	Cadmium	15	0.296	No
Atlantic cod	Lead	12	0.476	No
Atlantic cod	Mercury	5 *)	-	-
Seal	Cadmium	24	-0.156	No
Seal	Lead	13	0.045	No
Seal	Mercury	8*)	-	-

Table 5. Spearman Rank correlation (rho) of SST and element levels per species. *) indicate N is too low for valid application.

Species	Element	N	rho	Sign. 95%-level
Atlantic cod	Cadmium	15	0.296	No
Atlantic cod	Lead	12	0.476	No
Atlantic cod	Mercury	5 *)	-	-
Seal	Cadmium	24	-0.156	No
Seal	Lead	13	0.045	No
Seal	Mercury	8*)	-	-