# UNIVERSITY of BRADFORD

Library

# The University of Bradford Institutional Repository

http://bradscholars.brad.ac.uk

**Cooking fish and drinking milk? Patterns in pottery use in the southeastern Baltic, 3300-2400 Cal BC**

Carl Heron[1*], Oliver E. Craig[2], Alexandre Luquin[2], Valerie J. Steele[1], Anu Thompson[3] and Gytis Piličiauskas[4*]

[1]School of Archaeological Sciences, University of Bradford, Richmond Road, Bradford, West Yorkshire BD7 1DP, UK
[2]BioArCh, Department of Archaeology, University of York, Heslington, York YO10 5DD, UK
[3]School of Environmental Sciences, Nicholson Building, 4 Brownlow Street, The University of Liverpool, Liverpool L69 3GP, UK
[4]Lithuanian Institute of History, Kražių 5, 01108 Vilnius, Lithuania

*Corresponding authors (c.p.heron@bradford.ac.uk)

**Abstract**

A study of pottery vessel contents and use was undertaken in order to obtain information on food processed in Subneolithic and Neolithic vessels from Nida and Šventoji (3300-2400 cal BC). The aim is to assess pottery use during major changes in the coastal environment and in material culture. Bulk carbon and nitrogen isotope, lipid biomarker and compound specific carbon isotope analysis was undertaken on 'foodcrusts', charred deposits adhering to vessel surfaces, and absorbed residues from different vessel types. In addition, three archaeological seal bones were analysed for bulk collagen and compound specific carbon isotope analysis to establish collagen-lipid offsets to inform interpretation of the data. The results show that the majority of the vessels were used for processing aquatic products. At Nida the data suggest exploitation of freshwater resources and, in the later stages of occupation, dairying. Analysis of a small number of Subneolithic vessels from Šventoji produced results that are also consistent with processing of aquatic products. Other substances identified include Pinaceae sp. resin or tar and beeswax. These data demonstrate that identifying patterns in pottery use contributes to understanding Neolithisation processes.

**Highlights**:

- Analysis of Subneolithic and Neolithic pottery in the Southeastern Baltic confirms exploitation of aquatic resources.

- Analysis of Corded Ware Beakers, associated with domesticates, provides evidence of dairy products in the Neolithic.

- Analysis of collagen and lipids from seal bone enables offsets ($\Delta^{13}C_{\text{FA-COLL}}$) to be determined to aid interpretation.

## Introduction

Organic residue analysis of pottery vessels can be used to determine contents and use in diverse settings. Hunter-gatherer pottery use has been addressed in a number of studies (Isaksson, 2009; Papmehl-Dufay, 2010; Craig et al., 2011; Craig et al., 2013; Cramp et al., 2014a; Taché and Craig, 2015). Continuity and change in pottery vessel use before and after the introduction of domesticated plants and/or animals is helping to evaluate the relative importance of different foods during periods of cultural change at the transition to agriculture (Saul et al., 2014). Detecting marine resources in pottery vessels through organic residue analysis has only been achieved at the molecular level in the last 10 years (reviewed by Cramp and Evershed, 2014). The processing of marine foods in vessels associated with hunter-gatherers and early farmers has been demonstrated in the Western Baltic (Craig et al., 2011). In other contexts, marine resources have been shown to have a very minor presence in Neolithic pottery (Cramp et al., 2014b). In contrast the identification of freshwater resources in pots has received much less attention and there are no fixed criteria for authenticating this important resource. A combined approach based on lipid biomarker and compound specific carbon isotope measurements undertaken on absorbed residues and visible surface deposits, in some cases supported by bulk carbon and nitrogen isotope data obtained on visible surface deposits (often termed 'foodcrusts'), has been proposed in a number of studies (e.g. Craig et al., 2007; 2011; 2013; Hart et al., 2013; Taché and Craig 2015). The aim of the current paper is to determine patterns in pottery use through molecular and isotopic analysis of vessels from two Lithuanian Subneolithic and Neolithic coastal sites, i.e. Nida and Šventoji (3300-2400 cal BC). Extending knowledge of foods in the diet is appealing, especially at Nida, where other sources of information are largely absent - there are no human bones and only a few, poorly preserved animal bones.

Pottery technology reached the southeastern Baltic by around 5500-5000 cal BC and appears to have been strongly influenced by ceramic traditions to the east (Loze, 1998; Jussila and Kriiska, 2005). However, in coastal Lithuania the oldest ceramics are dated to 3900 cal BC. Human bone stable isotope and zooarchaeological data suggest that the diet of pottery-using communities may not have differed much from the diet of pre-ceramic hunter-gatherers. The term Subneolithic is used in this region to define pottery-using hunter-gatherers whereas Neolithic defines cultures with evidence of domesticated animals and/or plants. The dietary data highlight the importance of freshwater fish and forest game in subsistence practices (Eriksson, 2003; Antanaitis-Jacobs et al., 2009). The Globular Amphora Culture (GAC), from *c.* 3200 cal BC, and the Corded Ware Culture (CWC), from *c.* 2900/2800 cal BC, mark the beginning of the Neolithic period and stockbreeding in the southeastern Baltic. CWC and GAC pottery differs greatly from Subneolithic vessels with flat bases, a greater diversity of forms and sizes, better firing and new clay compositions. In inland areas, it is clear that the GAC and CWC groups introduce new subsistence practices and diet. CWC graves with bones of sheep or goat represent the earliest evidence of domesticated animals in most parts of the eastern Baltic. CWC graves from Latvia and Lithuania have been dated to 2900/2800-2500 cal BC (Eriksson, 2003; Antanaitis-Jacobs et al., 2009; Piličiauskas, unpublished data). However, the coastal area between Gdansk in Poland and Šventoji in Lithuania suggests a different pattern of Neolithisation. Zooarchaeological data shows that domesticated animals were successfully incorporated into a mainly fishing and seal hunting economy (Lasota-Moskalewska, 1997; Rimantienė, 1989). Pottery, instead of being simply adopted from inland neighbours, was highly elaborated in forms and ornamentation. This rather unique coastal adaptation together with its mixed economy is referred to as the Rzucewo

97   Culture (3200-2400 cal BC). The cultural sequence at Nida and Šventoji, compared to the inland
98   trajectory is shown in Figure 1.
99

## The archaeological sites
101  Nida and Šventoji are key sites in understanding coastal Neolithisation in Lithuania (Figures 2
102  and 3). Nida is situated on the SW edge of coastal Lithuania along the Curonian Spit, a narrow
103  band of land (1-3 km wide) separating the southeastern Baltic Sea from the Curonian Lagoon,
104  the largest lagoon in Europe. The spit and the lagoon emerged only after 5000/4750 cal BC,
105  during the regression of the Littorina sea (Damušytė, 2011). The oldest palaeosoils are dated to
106  3900/3700 cal BC (Dobrotin et al., 2013, table 1). The lagoon, fed by the Nemunas river, is
107  principally a freshwater body but it is also influenced by the brackish waters of the Baltic Sea.
108  Water salinity in the northern part of the lagoon today fluctuates between 0.1-7 psu (practical
109  salinity units) and this is tolerated by marine, brackish and freshwater species. The site of Nida
110  is located some 50 km south of the sea entrance (Klaipėda strait) to the Baltic and the waters in
111  the vicinity are likely to be dominated by the substantial freshwater discharges from the
112  Nemunas river 15 km to the east (Zemlys et al., 2013). The modern freshwater fish assemblage
113  in the Lithuanian part of the Curonian Lagoon is dominated by demersal species such as bream
114  (*Abramis brama*), roach (*Rutilus rutilus*), ruffe (*Gymnocephalus cernuus*) and silver bream
115  (*Blicca bjoerkna*). Predatory species are represented by perch (*Perca fluviatilis*) and zander
116  (*Sander lucioperca*; Repečka, 1997 cited in Žydelis and Kontautas, 2008).
117

118  Nida is the most intensively investigated site on the Curonian spit with a long research history
119  dating back to the 19th century (Hollack 1895). An area of 4640 m$^2$ was uncovered and more
120  than 100,000 sherds were collected during the 1974-1978 excavations (Rimantienė, 1989).
121  Between 2011-2013 excavations were resumed by one of us (GP). Most of materials belong to
122  Neolithic Rzucewo culture (3300/3200-2400 cal BC) although some fragments of Subneolithic
123  pointed-base vessels were also found (3500/3350 cal BC; Piličiauskas and Heron, 2015). Bones
124  are few and very badly preserved. Seal (*Phocidae* sp.), dog, beaver, red deer, elk, auroch/cattle,
125  boar/pig, fox, horse, sheep/goat, bream, pike (*Esox lucius*), and unidentified bird bones have
126  been found (Hollack, 1895; Rimantienė, 1989; Piličiauskas, unpublished data). However,
127  quantitative evaluation of the importance of these species in the subsistence practices is
128  impossible. Furthermore the domesticated animal bones cannot be assigned to any specific
129  phase of the Rzucewo Culture although it is likely that all or most of domesticated species
130  belong to the latest phase around 2500 cal BC.
131

132  Šventoji is situated on the NW edge of coastal Lithuania. Around 60 archaeological sites dating
133  to 4000-500 cal BC have been discovered or investigated there since 1966 (Rimantienė 2005;
134  Piličiauskas et al., 2012). Some of these sites are important because of the presence of
135  waterlogged cultural layers (gyttja) with good preservation of organic materials. At the Šventoji
136  1-6 sites, the horizon with Subneolithic pointed-base pottery belonging to the Narva culture is
137  overlain by Neolithic materials of the Globular Amphora Culture. The transition is dated to *c.*
138  2700 cal BC[1]. The Subneolithic and Neolithic sites, i.e. dwelling zones and fishing/dumping
139  zones, are situated on submerged slopes or on the shore of an ancient lagoon fed by the
140  Šventoji river. Diatom analysis and fish species studied indicate that the lagoon was principally a

---

[1]according to unpublished results of the excavations at Šventoji 4 in 2014 directed by G. Piličiauskas

141 freshwater body with limited and only occasional inflows of brackish waters from the eastern
142 Baltic. In a refuse layer at Šventoji 4, pike clearly dominates the fish bone assemblage although
143 rudd (*Scardinius erythrophthtalmus*), bream, perch, zander and wels catfish (*Silurus glanis*)
144 were also numerous. Marine fish are represented by flounder (*Pleuronectidae)* and four cod
145 (*Gadus morhua*) bones. Eel (*Anguilla anguilla*) is absent despite the many leister prongs
146 recovered. Among mammal bones, seals clearly predominate although boar, beaver and elk are
147 also numerous. Antanaitis-Jacobs et al., (2009) report collagen stable isotope data of a range of
148 the fauna from the Šventoji sites including marine and freshwater fish and marine mammals. At
149 around 2400 cal BC the Šventoji Palaeolagoon became overgrown and drained due to
150 significant isostatic land uplift (Piličiauskas et al., 2012).
151

152 **The pottery**
153 Figure 4 shows the vessel types compiled using data in Rimantienė (1989; 2005) and including
154 unpublished research by Piličiauskas (2011-2014). At Šventoji, 'Comb-like' vessels precede
155 Narva ware but in other areas (e.g., at the Narva type-site in Estonia), Narva ware is older than
156 'Comb-like' wares. There is very little information about this vessel type because the potsherds
157 are highly fragmented. It is possible that the vessels are similar to Narva ware although with a
158 rounded rather than a pointed base. One sherd from a 'Comb-like' ware vessel was included in
159 this study (L14, Šventoji 26). Two 'foodcrusts' from Subneolithic prolonged or extended bowls
160 were included in this study even though the sherds, recovered from older excavations, were
161 stabilized with consolidants (cf. Supplementary Table 1). The Globular amphora ware (GAW)
162 includes amphoras and other vessel forms with examples of the former included in this study.
163 Corded Ware vessels appear after GAW at the Šventoji 1 site. However, this type is known only
164 from older excavations. In this case, the potsherds were stabilised with consolidants and were
165 not included in this study.
166

167 At Nida, Narva ware is represented by only a small number of sherds and is replaced by
168 Rzucewo ware which persists for *c.* 800 years at the site (3300/3200-2400 cal BC). Most of the
169 finds were uncovered in 1974-78 and they have yet to be documented precisely. More recent
170 excavations and AMS dating provide some insights into the chronology of particular vessel
171 types. For example, the prolonged bowls exist only during the earliest phase of the Rzucewo
172 culture, i.e. 3200-2800 cal BC as do the tallest subtypes of large, wide-mouth vessels and less
173 profiled amphoras with larger handles. Beakers with high necks, short-wave moulded pots and
174 amphoras with very large bellies most likely belong to the later phase. Many of small and
175 medium sized vessels have soot and foodcrusts although they are usually absent from beakers.
176 Further work is underway to resolve the chronological sequence and the appearance and
177 disappearance of specific vessel types.
178

179 **Materials and Methods**
180 The samples analysed from both sites are presented in Table 1. In addition, three seal bones
181 (family Phocidae) were included in the study to evaluate the lipid isotope values of marine fauna
182 given the likely brackish conditions in the southeastern Baltic at this time. The seal bones were
183 recovered from Šventoji and are dated to the Subneolithic period.
184

| | Vessels sampled | Bulk $\delta^{13}C$ and $\delta^{15}N$ analyses | GC-MS analyses | GC-C-IRMS analyses |
|---|---|---|---|---|
| **Nida** | | | | |

| | | | | |
|---|---|---|---|---|
| Subneolithic | **1** | 1 | - | - |
| *Narva ware vessel = 1* | | | | |
| Neolithic | **28** | 30 | 26 | 16 |
| *Rzucewo ware vessels = 28, including Beakers = 5, Prolonged bowls = 4 and Wide mouth vessels = 2* | | | | |
| | | | | |
| **Šventoji** | | | | |
| Subneolithic | **17** | 17 | 2 | 1 |
| *Narva ware vessels = 16, including Prolonged bowls = 2. 'Comb-like' ware vessel = 1* | | | | |
| Neolithic | **8** | 8 | 2 | 2 |
| *Amphoras = 8* | | | | |

185 *Table 1: Vessels sampled and number of each analysis performed on the sherds and foodcrusts*
186 *from Šventoji and Nida.*
187

188 *Bulk isotope ratio mass spectrometry* ($\delta^{13}$C and $\delta^{15}$N)
189 Collagen was extracted from the seal bones (0.5 g samples) using standard laboratory protocols
190 (Longin, 1971). Whole bone samples were demineralized in 0.5 M HCl at 4°C. The remaining
191 collagen was solubilized in pH 3 aqueous solution at 70°C for 48 h. The measurements ($\delta^{13}$C
192 and $\delta^{15}$N) were determined relative to the VPDB and AIR international standards, respectively.
193 Each sample of 'foodcrust' (*c.* 1mg) was weighed in duplicate into tin capsules and analysed
194 using an elemental analyser linked to a PDZ Europa 20/20 mass spectrometer (PDZ Europa
195 Ltd, Crewe, UK). Analytical precision was greater than 0.3‰ for both elements, as determined
196 by duplicate measurements.
197

198 *Solvent extraction of foodcrusts and ceramic samples*
199 All glassware and tools were triple washed in solvent, nitrile gloves were worn at all times and all
200 solvents and other reagents were analytical or HPLC grade. To detect any contamination
201 introduced during sample preparation and analysis a blank sample was prepared and analysed
202 with the samples. Sherds (2g) and 'foodcrusts' (*c.* 50 mg) were ground to a powder using an
203 agate pestle and mortar. All samples were accurately weighed into clean glass vials and
204 extracted in three aliquots of 5 mL dichloromethane (DCM):methanol (2:1 v/v) with
205 ultrasonication (15 minutes). Samples were then centrifuged for 10 minutes at 3,000 rpm to
206 separate the solid material from the extract. The extracts were pipetted into clean vials and the
207 solvent evaporated off under a gentle stream of dry nitrogen with gentle heating (30°C). Prior to
208 GC-MS, aliquots of the samples were derivatised by heating for 30 minutes at 70°C with *N,O*-
209 bis(trimethylsilyl)trifluoroacetamide (BSTFA) with 1% trimethylchlorosilane (TMCS). Excess
210 BSTFA was evaporated off and a measured amount of an internal standard (*n*-tetratriacontane)
211 was added to each sample before analysis. Samples were re-dissolved in DCM prior to analysis
212 by GC-MS. Prior to GC-C-IRMS aliquots of the lipid extracts were methylated using $BF_3$-
213 methanol complex (14% w/v, 200 μL, 1 h, 70°C) and extracted into hexane (3 x 1 mL).
214

215 *Acidified methanol extraction*
216 The seal bones (*c.* 1-2 g) together with a small number of sherds and foodcrusts were extracted
217 using the acidified methanol procedure (Correa-Ascencio and Evershed, 2014; Colonese et al.,
218 2015). After adding 4 mL of methanol, the samples were ultrasonicated for 15 min.
219 Subsequently, 800 μL of concentrated $H_2SO_4$ was added and the samples were heated at 70°C
220 for 4 h. The samples were then centrifuged at 3,000 rpm for 5 min. The supernatant was

221 extracted with hexane (3 × 2 mL) and neutralised with $K_2CO_3$. The extracts were then dried
222 under a gentle stream of $N_2$. An internal standard (*n*-tetratriacontane) was added to each sample
223 before analysis by GC-MS and GC-C-IRMS.
224
225 *Gas Chromatography-mass spectrometry*
226 GC-MS analysis was carried out on an Agilent 7890A series GC attached to an Agilent 5975C
227 Inert XL mass selective detector. The split-splitless injector was operated in splitless mode and
228 the injector and interface were maintained at 300°C and 340°C, respectively. Helium was the
229 carrier gas at constant inlet pressure. The column was inserted directly into the ion source of the
230 mass spectrometer. The ionisation energy was 70eV and spectra were obtained by scanning
231 between *m/z* 50 and 800. All samples were analysed using an Agilent DB5-ms 30m x 2.5mm x
232 2.5 µm column. The oven temperature was programmed to be isothermal at 50°C for 2 minutes,
233 followed by a rise of 10°C per minute up to 350°C and an isothermal hold for 10 minutes.
234 Compounds were identified by comparison with the NIST library of mass spectral data and
235 published data. Peak area measurements for quantification were carried out using the
236 interactive RTE integrator within the Agilent Chemstation enhanced data analysis software.
237 Abundances were calculated as mg of compound per gram of sample (mg/g).
238
239 *Gas Chromatography-combustion-isotope ratio mass spectrometry*
240 Carbon stable isotope ratios were determined on two fatty acid methyl esters, methyl palmitate
241 ($C_{16:0}$) and methyl stearate ($C_{18:0}$), in each extract using a Delta V Advantage isotope ratio mass
242 spectrometer (Thermo Fisher Scientific, Bremen, Germany) linked to a Trace Ultra gas
243 chromatograph (Thermo Fisher) with a ConFlo IV interface (Cu/Ni combustion reactor held at
244 1000°C; Thermo Fisher). All samples were diluted with hexane and subsequently 1 µL of each
245 sample was injected into a DB5 fused-silica column (30m x 0.25mm id x 0.25 µm film thickness).
246 The temperature was set for 1 min at 45°C, and raised by 6°C $min^{-1}$ to 295°C, where it was held
247 for 15 min. The carrier gas was ultra-high-purity grade helium at a flow rate of 1.4 mL $min^{-1}$. The
248 eluted products were combusted to $CO_2$ and ionised in the source of the mass spectrometer by
249 electron ionisation. The ion intensities of *m/z* 44, 45, and 46 were monitored in order to
250 automatically compute the $^{13}C/^{12}C$ ratio of each peak in the extracts. Computations were
251 performed with Isodat 3.0 Gas Isotope Ratio MS Software (version 3.0; Thermo Fisher) and
252 were based on comparisons with a standard reference gas ($CO_2$) of known isotopic composition
253 that was repeatedly measured. The results from the analysis are reported in ‰ relative to an
254 international standard (V-PDB). Replicate measurements of each sample and a mixture of fatty
255 acid methyl esters (FAMEs) with $\delta^{13}C$ values traceable to international standards were used to
256 determine the instrument precision (<0.3‰) and accuracy (<0.5‰). The values were also
257 corrected subsequent to analysis to account for the methylation of the carboxyl group. The
258 corrections were based on comparisons with a standard mixture of $C_{16:0}$ and $C_{18:0}$ fatty acids of
259 known isotopic composition processed in each batch as a sample.
260
261 **Results**
262 The bulk isotope data are presented in Figure 5. Table 2 compares the data obtained on the
263 foodcrusts. The prolonged bowls from both sites are presented separately as these values differ
264 from the remaining vessels.
265

| | Nida Neolithic | | Šventoji sites Subneolithic | | Šventoji sites Neolithic |
|---|---|---|---|---|---|
| | Prolonged | Other vessels | Prolonged | Other vessels* | All vessels |

|  | bowls (n = 3) | (n = 19) | bowls (n = 2) | (n = 15) | (n = 7) |
|---|---|---|---|---|---|
| $\delta^{13}C$ | -33.1 ± 0.3 | -29.8 ± 1.8 | -30.6 ± 1.5 | -26.5 ± 0.9 | -26.1 ± 0.8 |
| $\delta^{15}N$ | 11.2 ± 0.8 | 9.6 ± 1.3 | 10.5 ± 0.9 | 11.3 ± 0.8 | 7.5 ± 2.9 |
| C/N ratio | 22.0 ± 10.1 | 13.9 ± 6.9 | 22.8 ± 7.5 | 7.3 ± 1.7 | 10.9 ± 4.5 |

266 *Table 2: Bulk carbon and nitrogen isotope data and atomic C/N ratios for Subneolithic and Neolithic*
267 *'foodcrusts' from Nida and Šventoji. n – the number of vessels with foodcrusts sampled. Only one*
268 *Subneolithic foodcrust was available from a Narva pot from Nida. It is not included in these data but is*
269 *listed in Supplementary Table 1.*

270

271 The GC-MS data are presented in Supplementary Table 2. Most of the residues are dominated
272 by saturated fatty acids ($C_{16:0}$ and $C_{18:0}$) although some samples have more complex lipid
273 distributions, especially from Šventoji, with higher than usual relative abundances of unsaturated
274 fatty acids, cholesterol, cholesterol oxidation products and terpenoid fractions.

275

276 TMTD (4,8,12-trimethyltridecanoic acid) was identified in association with all the vessels from
277 Šventoji and all vessels from Nida with the exception of two of the four beakers sampled. The
278 co-occurrence of TMTD, pristanic acid (2,6,10,14-tetramethylpentadecanoic) and phytanic acid
279 (3,7,11,15-tetramethylhexadecanoic acid) is consistent with the processing of aquatic tissues in
280 the vessels. These acyclic isoprenoids are degradation products of phytol (Rontani and
281 Volkman, 2003). TMTD has been described as "a compound exclusive to the marine
282 environment" Corr et al. (2008, 2106), although it is also found in freshwater tissues (Ackman
283 and Hooper, 1970). Phytanic and pristanic acids are found in the tissues of ruminant animals but
284 in combination with TMTD supports the presence of aquatic resources. ω-(*o*-
285 alkylphenyl)alkanoic acids were detected in many of the residues although the C22 isomers
286 were not routinely found. These are thermal alteration products of unsaturated fatty acids and
287 the presence of longer chain (C20 and C22) isomers supports the presence of aquatic
288 resources (Cramp and Evershed, 2014). Vicinal dihydroxyfatty acids were detected in several
289 extracts but the carbon chain lengths were typically the less diagnostic molecules of C16 and
290 C18 chain length. These arise from the oxidative degradation of unsaturated fatty acids (Hansel
291 and Evershed, 2009; Hansel et al., 2011). Recent studies have shown that they occur largely in
292 the 'bound' fraction of organic residues associated with pottery sherds and in very low quantities
293 requiring the use of selected ion monitoring for detection (Cramp et al., 2014b).

294

295 The lipid biomarker data, especially the presence of long-chain unsaturated fatty acids (>C18:1),
296 isoprenoid fatty acids, particularly TMTD (4,8,12-trimethyltridecanoic acid), ω-(*o*-
297 alkylphenyl)alkanoic acids (>C18) and abundant cholesterol and cholesterol-oxidation products
298 is consistent with the processing of aquatic resources in the majority of the pottery vessels.
299 Lipids were also extracted from all three seal bones. The excellent preservation of endogenous
300 lipid (Supplementary Figure 1) is exemplified by the unusually high abundance of hexadecenoic
301 (palmitoleic, $C_{16:1}$) acid relative to two saturated fatty acids ($C_{14:0}$ and $C_{18:0}$), a feature observed
302 in modern seal blubber (Shahidi and Zhong, 2005, 263). One extract contained traces of TMTD.

303

304 The compound specific isotope data are plotted in Figure 6 alongside reference data obtained
305 on modern organisms from the UK (Dudd and Evershed, 1998) and the western Baltic (Craig et
306 al., 2011). Additional data (published in Cramp et al., 2014a) obtained on eight Late Comb Ware
307 vessels and five Corded Ware Beakers from southern Finland are included for comparison. The
308 clear marine signals (higher $\delta^{13}C$ values) found in the Late Comb Ware vessels from southern
309 Finland are not seen in the vessels from Nida or Šventoji. The correspondence between

310   depleted carbon isotope values (< -30‰), the presence of aquatic biomarkers and the high δ$^{15}$N
311   values in the foodcrusts strongly supports the processing of freshwater resources in these
312   vessels (Craig et al., 2007) or organisms (e.g., seals, birds) subsisting on freshwater and
313   brackish resources. The more depleted carbon isotope values result from a variety of causes
314   such as diet, habitat and the varying input of $^{13}$C-depleted terrestrial organic matter although
315   there is considerable variation leading to large differences in the tissues of freshwater organisms
316   (e.g., Dufour et al., 1999; Katzenberg et al., 1999). Few compound specific carbon isotope
317   values from modern freshwater fish have been published to date. Craig et al. (2007) include
318   values obtained on fish from a UK river. Outram et al. (2012) report the analysis of 16 Kazakh
319   riverine freshwater fish. Cramp and Evershed (2014, Figure 6) plot a wide range of authentic
320   fats and oils including the aforementioned samples as well as roach and perch from a UK lake.
321   The depleted values for the fatty acids from the modern freshwater lipids in all of the above
322   studies are notable and show clear isotopic separation from marine resources.
323
324   The relationship between bulk and compound specific carbon isotope values has not been
325   investigated comprehensively to date. The bulk carbon isotope value of the foodcrust may
326   represent carbon sources from the different compound classes present (fats, proteins and
327   carbohydrates) whereas the compound specific carbon isotope value represents the carbon
328   sources of the principal lipid components only. There is a strong positive correlation ($r = 0.76$)
329   between the bulk carbon isotope value of foodcrusts and the mean of the $C_{16:0}$ and $C_{18:0}$ values
330   from GC-C-IRMS analysis of the residues absorbed into the interior surface of the same vessel.
331   The mean offset between bulk and compound specific values is -1.6‰ reflecting the greater
332   depletion in carbon-13 of lipid compared to other food components. Comparing these two values
333   may be questionable especially as the compound specific value is derived from fatty acids in the
334   absorbed residue rather than the foodcrust. The strong positive correlation merits further
335   consideration. Firstly it suggests that the lipid absorbed into the vessels is very similar in origin
336   to the charred food residue adhering to the vessel surface. Secondly, it suggests that, in certain
337   cases, the bulk carbon isotope value can give an indication of the compound specific isotope
338   value, perhaps where the vessel has been used to process the same food throughout its use-
339   life. Clearly further consideration of these issues is needed from a wider range of sites and
340   situations.
341
342   **Discussion – the relationship between vessel type and residue identification**
343   *Nida*
344   In general, the high nitrogen isotope values in the foodcrusts are consistent with high trophic
345   level resources such as aquatic products. The δ$^{13}$C values for the foodcrusts from Nida are
346   more depleted than those at Šventoji, both for the prolonged bowls and the other vessels
347   sampled. Whilst this might represent different products processed in these vessels, an
348   alternative explanation is that the more depleted values from Nida indicate foods from a
349   freshwater body only minimally influenced by marine/brackish water from the Baltic Sea. In
350   contrast the Šventoji lagoon may have been more susceptible to influxes of saline water with
351   wider resource availability resulting in less depleted isotope values. Very few faunal remains
352   from Nida are available to evaluate this possibility. One seal phalanx (Phocidae sp.) from the
353   site has a δ$^{13}$C value of -15.9‰, which is consistent with values from 13 seals at Šventoji (see
354   Table 3). Although the sample size is small, modern freshwater fish from the Curonian Lagoon
355   have lower δ$^{13}$C values than Neolithic fish bone from Šventoji by an average of -2.3‰
356   (Piličiauskas, unpublished results) but the species are different and any differences will, in part,
357   be the result of the fossil fuel effect.

At Nida, thirteen of the 15 vessels analysed by GC-MS and GC-C-IRMS show evidence of aquatic products despite the fact that these all date to the Neolithic when domesticated resources were available. A further twelve vessels sampled for bulk isotope analysis only also indicate aquatic products based on high $\delta^{15}N$ and low $\delta^{13}C$ values. An outlier in this group is L11i ($C_{16:0}$ = -25.8‰; $C_{18:0}$ = -28.3‰). This residue includes the full range of aquatic biomarkers but is the most enriched of the $C_{16:0}$ and $C_{18:0}$ values. The bulk carbon isotope value is also among the most enriched of the foodcrusts from Nida (-27.7‰). This residue could represent a marine source (plotting closely to one of the archaeological seal bone residues from Šventoji) or a mixture of aquatic and terrestrial resources.

Aquatic biomarkers are absent from the remaining two vessels from Nida analysed by GC-MS. Both of these vessels are beakers and 'foodcrusts' were also absent. Compound specific carbon isotope analysis of the absorbed residues showed significant offsets ($\Delta^{13}C$ value) between $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ of -4.1‰ and -5.4‰. This suggests the presence of dairy fats in these vessels (Copley et al., 2003). As a caveat, modern wild ruminant adipose (red deer) has been shown to have $\Delta^{13}C$ values of up to -4.3‰ (Craig et al., 2012) so this possibility cannot be discounted in one case, especially as red deer bone occurs in the faunal assemblage. The type of beaker with the putative dairy residues is well known among Corded Ware assemblages across Central and Eastern Europe (especially in the Middle Dnieper region of Ukraine, Artemenko, 1967), but is also very common in the latest stage of the Rzucewo Culture (including at the Rzucewo type site, Zurek, 1954). It is very rare at Nida and differs from other pots by a dark fabric, polished surface and very specific ornamentation, namely intermediate zones of horizontal lines and oblique hatches. The date of the cultural layer that these were found in is dated to 2700-2400 cal BC, i.e. towards the end of the settlement's occupation; they are absent from the earliest phase (3300-2700 cal BC). Figure 6 shows that dairy products have also been reported in three Corded Ware beakers of similar date from southern Finland (Cramp et al., 2014a) and our data support the association made in that study. DNA evidence (Haak et al., 2015) equates a 'massive migration' of genetically distinct Steppe pastoralists associated with Corded Ware pottery. Another study (Allentoft et al., 2015) found the highest lactose tolerance frequency, among a sample of Bronze Age Europeans, in Corded Ware and closely related Scandinavian cultures. The data presented here provide complementary evidence based on the occurrence of dairy residues in some, but not all, of the Corded Ware beakers at Nida. Corded Ware beakers may have served as drinking vessels and, if so, imply the consumption of fresh milk supporting the DNA evidence that at least some of the early pastoralists in the region were lactose tolerant. However, other possibilities, such as the use of milk to seal the permeable vessel walls prior to use, or the presence of processed dairy products such as yoghurt, butter or cheese must also be considered. Two Corded Ware beakers from Nida contain freshwater resources, supporting the results in Cramp et al. (2014a) that these vessels are not used exclusively for terrestrial foods. A foodcrust from another beaker from Nida has bulk isotope values consistent with freshwater resources ($\delta^{13}C$ value, -32.9‰; $\delta^{15}N$ value, 9.9‰).

*Šventoji*

The foodcrusts from Šventoji are less depleted in carbon-13 (-25.1 to -28.7‰) compared to the samples from Nida. Nevertheless, the biomarker profiles and high $\delta^{15}N$ values support the presence of aquatic resources. There is no significant difference in the carbon isotope ratios between the Subneolithic and Neolithic foodcrusts (excluding the prolonged bowls) at Šventoji.

405    However, there is a significant difference in the nitrogen isotope ratios (heteroscedastic t test;
406    $0.02 > p > 0.01$) with the Neolithic foodcrusts having lower $\delta^{15}N$ values (see Table 2).
407
408    To further explore the relationship between the pottery residues and the fauna from the site
409    three seal bones were analysed following the approach outlined in Colonese et al. (2015). Table
410    3 shows molecular and isotopic data obtained on three archaeological seal bones from Šventoji
411    (see also Supplementary Table 3). The offset between collagen and lipids ($\Delta^{13}C_{FA\text{-}COLL}$) from the
412    same bone ranges from 10.5 to 11.6‰. This is lower than the data from two pinnipeds from
413    Cnoc Caig, Oronsay (UK) which had higher $\Delta^{13}C_{FA\text{-}COLL}$ values up to 17.3‰ (Colonese et al.,
414    2015). In another study, Fernandes et al. (2014) showed that modern fishbone collagen from
415    two marine fish is *c.* 10‰ heavier than bulk tissue lipids which accords well with the data
416    presented here. The compound specific values for the seal bones range from -25.5 to -27.7‰. If
417    bone lipid values can be used as a proxy for tissue lipid, then this provides an indication of the
418    range expected for pottery vessel residues used to process seal tissues in this region. The
419    mean bulk $\delta^{13}C$ value of 15 Subneolithic foodcrusts from Šventoji is -26.5 ± 0.9‰. Compound
420    specific data was obtained on only one Subneolithic vessel (L14i) giving $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$
421    values of -29.8 and -29.5‰ respectively.
422
423    The high $\delta^{15}N$ values in the Subneolithic foodcrusts (11.3 ± 0.8‰) supports the presence of
424    aquatic products in the vessels. The lipid extracts from two Subneolithic pointed-base vessels
425    from Šventoji 3 and 26 (L13 and L14 respectively) are remarkable for the high abundance of
426    cholesterol and cholesterol oxidation products. The presence of TMTD and C16-C20 ω-(*o*-
427    alkylphenyl)alkanoic acids strongly suggests an aquatic contribution to the residues. Figure 7
428    shows the total ion current (TIC) chromatogram of an absorbed residue from a Subneolithic
429    pointed-base vessel from Šventoji 26. The abundance of cholesterol in the residue is
430    noteworthy. The peak area ratio of cholesterol to hexadecanoic acid ($C_{16:0}$) is 0.77, which is
431    highly unusual in archaeological lipid residues. The Narva vessel from Šventoji 3 (L13) has an
432    even higher ratio of 0.85. The remaining samples from Šventoji have cholesterol/$C_{16:0}$ ratios of
433    only 0.03 and 0.09 consistent with all the residues from Nida (≤0.01 to 0.08). A high abundance
434    of cholesterol together with a range of cholesterol oxidation and reduction products has been
435    identified in residues considered to have an aquatic origin, especially vessels in ceramic and
436    stone considered to have been used as lamps (Heron et al.. 2013, supplementary information;
437    Solazzo and Erhardt, 2007). A similar suite of cholesterol oxidation products has been found in
438    experiments conducted on the pyrolysis of cholesterol in hydrous conditions below 200°C
439    (Rushdi et al., 2003).
440
441    The two Neolithic globular amphoras from Šventoji 4 have complex residues. The amphora, L1,
442    produced $C_{16:0}$ and $C_{18:0}$ carbon isotope values of -29.1‰ and -28.8‰ respectively with a
443    substantial lipid extract of *c.* 3 mg g$^{-1}$, which includes abundant unsaturated fatty acids in the
444    range $C_{16:1}$-$C_{20:1}$. All three isoprenoid fatty acids are present suggesting an aquatic product.
445    Abundant Pinaceae sp. diterpenoids are also present, including abietic and pimaric acids as well
446    as the oxidation products, dehydroabietic and 7-oxodehydroabietic acids. Their presence could
447    suggest a lining or sealant of resin or tar applied to the vessel to facilitate storage of liquid
448    contents. A second Neolithic globular amphora, L17 produced a residue considered to be highly
449    degraded beeswax with no aquatic biomarkers. The identification of beeswax is supported by an
450    *n*-alkane (C23-C29) and *n*-alkanol (C24-C30) fraction together with traces of the lower molecular
451    weight palmitate wax ester fraction (C40-C42) and a high abundance of tetracosanoic acid
452    ($C_{24:0}$) relative to other long-chain fatty acids (Heron et al., 1994; Regert et al., 2001). Beeswax

453  could also have served as a sealant although other uses of the vessel (e.g., to store honey) may
454  have resulted in the accumulation of beeswax absorbed into or on the surface of the sherd. In
455  this case a visible deposit was present and gave a $\delta^{13}C$ value of -25.8‰ and a $\delta^{15}N$ value of
456  1.5‰ consistent with low trophic level terrestrial resources. The charred deposit has *c.* 3%
457  nitrogen by weight, which hints at a contribution from another organic product to the residue as
458  beeswax has very low levels of nitrogen.
459

460

| Sample (lipid yield) | Aquatic Biomarkers | Other Lipids | Bone Collagen | | | Bone Lipid | | Collagen-lipid spacing [$\Delta^{13}C_{FA-COLL}$]* (‰) |
|---|---|---|---|---|---|---|---|---|
| | | | $\delta^{13}C$ (‰) | $\delta^{15}N$ (‰) | C/N | $\delta^{13}C_{16:0}$ (‰) | $\delta^{13}C_{18:0}$ (‰) | |
| Seal bone 1 (29 µg/g) | TMTD (tr) | Fatty acids ($C_{12:0}$-$C_{24:0}$, tr $C_{16:1}$-$C_{22:1}$), sterols | -16.3 | 15.5 | 3.4 | -27.3 | -28.4 | -11.6 |
| Seal bone 585 (67 µg/g) | - | Fatty acids ($C_{14:0}$-$C_{24:0}$, tr $C_{16:1}$-$C_{24:1}$), sterols | -15.3 | 13.1 | 3.2 | -27.7 | -25.6 | -11.4 |
| Seal bone 696 (322 µg/g) | - | Fatty acids ($C_{12:0}$-$C_{24:0}$, tr $C_{16:1}$-$C_{22:1}$) | -16.6 | 12.0 | 3.2 | -25.5 | -28.6 | -10.5 |
| Mean values for ten seal bones from Šventoji (from Antanaitis-Jacobs *et al.* 2009) | | | -16.7 ± 0.9 | 12.4 ± 1.0 | 3.4 ± 0.1 | nd | nd | nd |

461

*Table 3: Lipid and collagen data from three seal bones from Šventoji. nd – not determined. *Collagen-lipid offset determined by taking the mean of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ and subtracting from the bone collagen $\delta^{13}C$ value. The lipids were extracted by the acid-methanol method which results in modification of cholesterol hence the generic label 'sterols' is given here.*

465

466
467   *The prolonged bowls from Nida and Šventoji*
468   Five prolonged bowls (three from Neolithic Nida and two from Subneolithic Šventoji) were
469   analysed. These vessels have among the lowest $\delta^{13}C$, the highest $\delta^{15}N$ values and the highest
470   C/N ratios in each assemblage (Figure 5 and Table 2). GC-MS analysis of the Šventoji bowls
471   was not possible as they had been subject to conservation treatment. Two of the Nida bowls
472   were analysed by GC-C-IRMS and confirm the highly depleted carbon isotope values
473   determined by bulk isotope analysis (Figure 6, Supplementary Table 2). The high C/N ratios are
474   consistent with a substance rich in fat/oil and the lipid biomarker data confirm the presence of
475   TMTD and C18-C20 ω-(*o*-alkylphenyl)alkanoic acids, which suggests an aquatic rather than a
476   terrestrial source. Ethnographic analogy and experimental archaeology indicates that these
477   vessels may have been used as lamps for illumination and the analytical data support this
478   (Heron et al., 2013). In the latter study, analysis of 'blubber lamps', similar in shape to the
479   prolonged bowls but usually smaller, from western Baltic coastal sites identified lipids consistent
480   with marine oils. The only inland find sampled produced much more depleted GC-C-IRMS
481   values < -30.0‰. Analysis of the two Nida prolonged bowls produced $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$
482   values < -32.0‰. These data are considerably more depleted than would be expected for
483   marine oils and suggest an alternative source for the contents at inland Western Baltic and
484   coastal southeastern Baltic locations.
485
486   Seal blubber oil is commonly assumed to have been the fuel for these vessels but this seems
487   unlikely unless the seals were consuming abundant freshwater foods in their diet. Some modern
488   seal populations do consume, exclusively or partially, freshwater fish (e.g., Auttila et al., 2015,
489   Lundström et al., 2010). Oil extracted from freshwater organisms is another possibility. Historical
490   accounts (e.g., Svenson, 1985, 171) record the extraction of oil from the threespine stickleback
491   (*Gasterosteus aculeatus*). The threespine stickleback is found in freshwater, brackish and
492   coastal marine habitats and was caught when the fish formed huge shoals during late autumn
493   and winter. The oil was then used for many purposes including illumination. Collagen extracted
494   from modern freshwater fish from the Curonian Lagoon has $\delta^{13}C$ values ranging from -23.5 to -
495   24.3‰ (without correction for the fossil fuel effect; Piličiauskas, unpublished results). Applying a
496   similar collagen-lipid offset demonstrated for the seals from Šventoji would produce the highly
497   depleted lipid signals in the prolonged bowls from Nida. In summary, the data suggest that the
498   shallow, oval or prolonged bowls, in the western and eastern Baltic, contained heated aquatic
499   oils that may have served as illuminants.
500
501   **Conclusions**
502   This study has demonstrated the presence of a range of organic residues present in the pottery
503   vessels from two coastal prehistoric sites in the southeastern Baltic. There is clear evidence of
504   the processing of aquatic resources in the vessels. Distinguishing freshwater, brackish and
505   marine signals in this environment is complex and further work is needed, particularly through
506   investigating compound specific faunal reference lipid signals. The identification of freshwater
507   resources is suggested especially in the more carbon-13 depleted environment manifested in
508   the residues at Nida. Subsistence based on freshwater foods from the Curonian Lagoon, as well
509   as terrestrial resources, is in a good agreement with the very limited zooarchaeological data
510   from Nida. However this contrasts with the evidence from the type site of Rzucewo where
511   marine fish (e.g. *G. morhua*) and seals clearly dominate the faunal assemblage (Makowiecki,
512   2003, 88-91). The data presented here contrast with exploitation of marine resources along the
513   coast of southern Finland at this time (Cramp et al., 2014a). At least one Corded Ware beaker

shows clear evidence of a dairy residue. Although there is evidence of change in the mid-3rd millennium cal BC, coastal populations continued to exploit aquatic resources and to make use of the same fishing tools and fishing stations (Piličiauskas et al., 2012). Cultural and ideological 'neolithisation' can be seen as a response to a pan-European Neolithic idea but with modification according to local natural environments such as in areas with extraordinarily rich aquatic resources (e.g., the Curonian Lagoon) and that are especially demanding for agricultural activities (e.g., the sand dunes of the Curonian spit). This trajectory could be also characteristic to a large extent for the Šventoji sites.

**References**
Ackman, R.G., Hooper, S.N., 1970. Branched-chain fatty acids of four fresh-water fish oils. Comparative Biochemistry and Physiology 32, 117–25.

Allentoft, M.E., Sikora, M., Sjögren, K.G., Rasmussen, S., Rasmussen, M., Stenderup, J., Damgaard, P.B., Schroeder, H., Ahlström, T., Vinner, L., Malaspinas, A.S., Margaryan, A., Higham, T., Chivall, D., Lynnerup, N., Harvig, L., Baron, J., Della Casa, P., Dąbrowski, P., Duffy, P.R., Ebel, A.V., Epimakhov, A., Frei, K., Furmanek, M., Gralak, T., Gromov, A., Gronkiewicz, S., Grupe, G., Hajdu, T., Jarysz, R., Khartanovich, V., Khokhlov, A., Kiss, V., Kolář, J., Kriiska, A., Lasak, I., Longhi, C., McGlynn, G., Merkevicius, A., Merkyte, I., Metspalu, M., Mkrtchyan, R., Moiseyev, V., Paja, L., Pálfi, G., Pokutta, D., Pospieszny, Ł., Price, T.D., Saag, L., Sablin, M., Shishlina, N., Smrčka, V., Soenov, V.I., Szeverényi, V., Tóth, G., Trifanova, S.V., Varul, L., Vicze, M., Yepiskoposyan, L., Zhitenev, V., Orlando, L., Sicheritz-Pontén, T., Brunak, S., Nielsen, R., Kristiansen, K., Willerslev, E., 2015. Population genomics of Bronze Age Eurasia. Nature 522, 167–172.

Antanaitis-Jacobs, I., Richards, M., Daugnora, L., Jankauskas, R., Ogrinc, N., 2009. Diet in early Lithuanian prehistory and the new stable isotope evidence. Archaeologica Baltica 12, 12–30.

Artemenko, I.I., 1967. Plemena Verkhnego i Srednego Podneprov'ia vepokhu bronzy. 1967. Materialy i issledovaniia po ar-kheologii SSSR 148 (In Russian).

Auttila, M., Sinisalo, T., Valtonen, M., Niemi, M., Viljanen, M., Kurkilahti, M., Kunnasranta, M., 2015. Diet composition and seasonal feeding patterns of a freshwater ringed seal (*Pusa hispida saimensis*). Marine Mammal Science 31 (1), 45-65.

Colonese, A.C., Farrell, T., Lucquin, A., Firth, D., Charlton, S., Robson, H.K., Alexander, M., Craig, O.C., 2015. Archaeological bone lipids as palaeodietary markers. Rapid Communications in Mass Spectrometry 29 (7), 611–18.

Copley, M.S., Berstan, R. Dudd, S.N. Docherty, G., Mukherjee, A.J., Straker, V., Payne, S., Evershed, R.P., 2003. Direct chemical evidence for widespread dairying in prehistoric Britain. Proceedings of the National Academy of Sciences of the USA 100, 1524–29.

Corr, L.T., Richards, M.P., Jim, S., Ambrose, S.H. Mackie, A., Evershed, R.P., 2008. Probing dietary change of the Kwäday Dän Ts'ìnchi individual, an ancient glacier body from British Columbia: I. Complementary use of marine lipid biomarker and carbon isotope signatures as novel indicators of a marine diet. Journal of Archaeological Science 35, 2102–10.

Correa-Ascencio, M., Evershed, R.P., 2014. High throughput screening of organic residues in archaeological potsherds using direct acidified methanol extraction. Analytical Methods 6 (5), 1330–40.

Craig, O.E., Allen, R.B., Thompson, A., Stevens, R.E., Steele, V.J., Heron, C., 2012. Distinguishing wild ruminant lipids by GCCIRMS. Rapid Communications in Mass Spectrometry 26, 2359–64.

Craig, O.E., Forster, M., Andersen, S.H., Koch, E., Crombé, P., Milner, N.J., Stern, B., Bailey, G.N., Heron, C.P., 2007. Molecular and isotopic demonstration of the processing of aquatic products in northern European prehistoric pottery. Archaeometry 49, 135–52

Craig, O.E., Saul, H., Lucquin, A., Nishida, Y., Taché, K., Clarke, L., Thompson, A., Altoft, D.T., Uchiyama, J., Ajimoto, M., Gibbs, K., Isaksson, S., Heron, C.P., Jordan, P., 2013. Earliest evidence for the use of pottery. Nature 496, 351–54

Craig, O.E., Steele, V.J., Fischer, A., Hartz, S., Andersen, S.H., Donahue, P., Glykou, A., Saul, H., Jones, D.M., Koch, E., Heron C., 2011. Ancient lipids reveal continuity in culinary practices across the transition to farming in Northern Europe. Proceedings of the National Academy of Sciences, USA. 108, 17910–15.

Cramp, L.J.E., Evershed, R.P., 2014. Reconstructing aquatic resource exploitation in human prehistory using lipid biomarkers and stable isotopes. In: Holland, H.D., Turekian, K.K. (Eds) Treatise on Geochemistry: Archaeology and Anthropology. Elsevier, Oxford, pp. 319–39.

Cramp, L.J.E., Evershed, R.P., Lavento, M., Halinen, P., Mannermaa, K., Oinonen, M., Kettunen, J., Perola, M., Onkamo, P., Heyd, V., 2014a. Neolithic dairy farming at the extreme of agriculture in northern Europe. Proc. R. Soc. B 281, DOI:10.1098/rspb.2014.0819.

Cramp, L.J.E., Jones, J., Sheridan, A., Smyth, J., Whelton, H., Mulville, J., Sharples, N., Evershed, R.P., 2014b. Immediate replacement of fishing with dairying by the earliest farmers of the northeast Atlantic archipelagos. Proceedings of the Royal Society B - Biological Sciences 281, DOI: 10.1098/rspb.2013.2372.

607

608  Damušytė, A., 2011. Lietuvos Pajūrio geologinė Raida Poledynmečiu. Daktaro disertacija (Post-
609  Glacial Geological History of the Lithuanian Coastal Area. Doctoral Dissertation). Vilnius
610  University, Vilnius.

611

612  Dobrotin, N., Bitinas, A., Michelevičius, D., Damušytė, A., Mažeika J., 2013. Reconstruction of
613  the Dead (Grey) Dune evolution along the Curonian Spit, Southeastern Baltic. Bulletin of the
614  Geological Society of Finland 85 (1), 53–64.

615

616  Dudd, S.N., Evershed, R.P., 1998. Direct demonstration of milk as an element of archaeological
617  economies. Science 282, 1478–81.

618

619  Dufour, E., Bocherens, B., Mariotti, A., 1999. Investigation of isotopic composition of European
620  lacustrine fish: implications for dietary reconstructions. Journal of Archaeological Science 26,
621  617–27.

622

623  Eriksson, G., Lõugas, L., Zagorska, I., 2003. Stone Age hunter-fisher-gatherers at Zvejnieki,
624  northern Latvia: stable isotope and archaeozoological data. Before Farming 1, 1–25.

625

626  Fernandes, R., Meadows, J., Dreves, A., Nadeau M.-J., Grootes, P., 2014. A preliminary study
627  on the influence of cooking on the C and N isotopic composition of multiple organic fractions of
628  fish (mackerel and haddock). Journal of Archaeological Science 50, 153–59.

629

630  Haak, W., Lazaridis, I., Patterson, N., Rohland, N., Mallick, S., Llamas, B., Brandt, G.,
631  Nordenfelt, S., Harney, E., Stewardson, K., Fu, Q., Mittnik, A., Bánffy, E., Economou, C.,
632  Francken, M., Friederich, S., Pena, R.G., Hallgren, F., Khartanovich, V., Khokhlov, A., Kunst,
633  M., Kuznetsov, P., Meller, H., Mochalov, O., Moiseyev, V., Nicklisch, N., L. Pichler, S.L., Risch,
634  R., Rojo Guerra, M.A., Roth, C., Szécsényi-Nagy, A., Wahl, J., Meyer, M., Krause, J., Brown, D.,
635  Anthony, D., Cooper, A., Alt, K.W., Reich, D., 2015. Massive migration from the steppe was a
636  source for Indo-European languages in Europe. Nature doi:10.1038/nature14317.

637

638  Hansel, F.A., Evershed, R.P., 2009. Formation of dihydroxyacids from Z-monounsaturated
639  alkenoic acids and their use as biomarkers for processing of marine commodities in
640  archaeological pottery vessels. Tetrahedron Letters 50, 5562–64.

641

642  Hansel, F.A., Bull, I.D., & Evershed, R.P., 2011. Gas chromatographic mass spectrometric
643  detection of dihydroxy fatty acids preserved in the 'bound' phase of organic residues of
644  archaeological pottery vessels. Rapid Communications in Mass Spectrometry 25 (13), 1893–98.

645

646  Hart, J.P., Lovis, W.A., Urquhart, G.R., Reber, E.A., 2013. Modelling freshwater reservoir offsets
647  on radiocarbon-dated charred cooking residues. American Antiquity 78 (3), 536–52.

648

649  Heron, C., Andersen, S., Fischer, A., Glykou, A., Hartz, S., Saul, H., Steele, V.J., Craig, O.E.,
650  2013. Illuminating the Late Mesolithic: residue analysis of 'blubber' lamps from Northern Europe.
651  Antiquity 87, 178–88.

652

Heron, C., Nemcek, N., Bonfield, K.M., Dixon, D., Ottaway, B.S., 1994. The chemistry of Neolithic beeswax. Naturwissenschaften 81, 266–69.

Hollack, E., 1895. Bericht des Herrn Lehrer Hollack über seine. Untersuchungen und Ausgrabungen auf der Kurischen Nehrung. Sitzungsberichte der Altertumsgesellschaft Prussia 19, 146–61.

Isaksson, S., 2009. Vessels of change. A long-term perspective on prehistoric pottery-use in southern and eastern middle Sweden based on lipid residue analyses. Current Swedish Archaeology 17, 131–149.

Jussila, T., Kriiska, A., 2005. Shore displacement chronology of the Estonian Stone Age. Estonian Journal of Archaeology 8 (1), 3–32.

Katzenberg, M.A., Weber, A.W., 1999. Stable isotope ecology and paleodiet in the Lake Baikal region of Siberia. Journal of Archaeological Science 26, 651–65.

Lasota-Moskalewska, A., 1997. Animal remains from the Neolithic settlement at Rzucewo. In, D Król (ed) The built environment of coast areas during the Stone Age, Gdańsk, pp. 162–166.

Longin, R., 1971. New method of collagen extraction for radiocarbon dating. Nature 230, 241–242.

Loze, I., 1988. Poseleniâ kamennogo veka Lubanskoj niziny. Mezolit, rannij i srednij neolit. Riga: Zinatne (in Russian).

Lundström, K., Hjerne, O., Lunneryd, S.G., Karlsson, O., 2010. Understanding the diet composition of marine mammals: grey seals (*Halichoerus grypus*) in the Baltic Sea. ICES Journal of Marine Science 67 (6), 1230–1239

Makowiecki, D., 2003. Historia ryb i rybołówstwa w holocenie na niżu polskim w świetle badań archeoichtiologicznych. IAE PAN, Poznań.

Outram, A. Kasparov, A., Stear, N.A. Varfolomeev, V., Usmanova, E., Evershed R.P., 2012. Patterns of pastoralism in later Bronze Age Kazakhstan: New evidence from faunal and lipid residue analyses. Journal of Archaeological Science 39 (7), 2424–35.

Papmehl-Dufay, L., 2010. Pitted Ware culture ceramics: Aspects of pottery production and use at Ottenby Royal Manor, Öland, Sweden. In: Jordan, P., Zvelebil, M. (Eds) Ceramics Before Farming. The Dispersal of Pottery Among Prehistoric Eurasian Hunter-Gatherers. Left Coast press, Walnut Creek, pp. 421–446.

Piličiauskas, G., Mažeika., J, Gaidamavičius, A., Vaikutienė, G., Bitinas, A., Skuratovič, Ž., Stančikaitė, M., 2012. New archaeological, paleoenvironmental, and 14C data from Šventoji Neolithic sites, NW Lithuania. Radiocarbon 54 (3-4), 1017–31.

699 Piličiauskas, G., Heron, C., 2015. Aquatic radiocarbon reservoir offsets in Lithuania.
700 Radiocarbon (in press).
701
702 Regert, M., Colinart, S., Degrand, L., Decavallas, O., 2001. Chemical alteration and use of
703 beeswax through time: Accelerated ageing tests and analysis of archaeological samples from
704 various environmental contexts. Archaeometry 43 (4), 549–69.
705
706 Rimantienė, R., 1989. Nida. Senųjų baltų gyvenvietė. Vilnius: Mokslas.
707
708 Rimantienė, R., 2005. Die Steinzeitfischer an der Ostseelagune in Litauen. Litauisches
709 Nationalmuseum, Vilnius.
710
711 Rontani, J.-F., Volkman, J.K., 2003. Phytol degradation products as biogeochemical tracers in
712 aquatic environments. Organic Geochemistry 34, 1–35.
713
714 Rushdi, A.I., Ritter, G., Grimalt, J.O., Simoneit, B.R.T., 2003. Hydrous pyrolysis of cholesterol
715 under various conditions. Organic Geochemistry 34 (6), 799–812.
716
717 Saul, H., Glykou, A., Craig, O.E., 2014. Stewing on a theme of cuisine: biomolecular and
718 interpretive approaches to culinary changes at the transition to agriculture. In: Whittle, A., Bickle,
719 P. (Eds.) Early Farmers: The View from Archaeology and Science. Proceedings of the British
720 Academy 198. Oxford University Press/British Academy, Oxford, pp. 197-213.
721
722 Shahidi, F., Zhong, Y., 2005. Marine mammal oils. In: Shahidi, F. (Ed), Bailey's Industrial Oil and
723 Fat Products. 6th edition, volume 3. Wiley, New York, pp. 259–78.
724
725 Solazzo, C., Erhardt, D., 2007. Analysis of lipid residues in archaeological artifacts: marine
726 mammal oil and cooking practices in the Arctic. In: Barnard, H., Eerkens, J.W. (Eds.) Theory and
727 Practice of Archaeological Residue Analysis (British Archaeological Reports international series
728 1650). Archaeopress, Oxford, pp. 161–178
729
730 Stančikaitė, M., Daugnora, L., Hjelle, K., Hufthammer, A.K., 2009. The environment of the
731 Neolithic archaeological sites in Šventoji, Western Lithuania. Quaternary International 207 (1-2),
732 117–29.
733
734 Svensson, R., 1985. Skärgårdsliv i gången tid, en skildring av arbete och lek i Stockholms
735 skärgård. 2nd edition. Streiffert, Stockholm.
736
737 Taché, K., Craig, O.E., 2015. Cooperative harvesting of aquatic resources triggered the
738 beginning of pottery production in north-eastern North America. Antiquity 89, 177–90.
739
740 Zemlys, P., Ferrarin, C., Umgiesser, G., Gulbinskas, S., Bellafiore, D., 2013. Investigation of
741 saline water intrusions into the Curonian Lagoon (Lithuania) and two-layer flow in the Klaipėda
742 Strait using finite element hydrodynamic model. Ocean Science 9, 573–84.
743

744 Žydelis, R.R., Kontautas, A.A., 2008. Piscivorous birds as top predators and fishery competitors
745 in the lagoon ecosystem. Hydrobiologia 611, 45–54.
746
747 Zurek, J. 1954. Osada w Rzucewie i kultura rzucewska. Fontes Archaeologici Posnanienses 4,
748 15–22.
749
750
751
752

**Figures**

Figure 1: An overview of the cultural sequence at Nida and Šventoji during the Subneolithic and Neolithic periods compared to the inland trajectory. GAC – Globular Amphora Culture; CWC – Corded Ware Culture.

Figure 2: Location of Nida and Šventoji in the SE Baltic. Environmental data indicates that the Curonian Lagoon was a large freshwater body in the period 3200-2500 cal BC, i.e. during the Rzucewo Culture occupation at Nida.

Figure 3: Location of the Šventoji Subneolithic and Neolithic sites within the reconstructed palaeolandscape around 3000 cal BC. The water table is considered to 1.4 m a.s.l. The hypothetical sandy spit would have been destroyed by the formation of the Baltic sea; lagoonal gyttja is found today in boreholes made directly on the beach and is sometimes washed up onto the beach by wave action. Erosional channels appeared in the deepest parts of the lagoonal ecosystem and were used as the main fishing sites.

Figure 4: Subneolithic and Neolithic vessel types from Nida and Šventoji compiled using data in Rimantienė (1989; 2005) and unpublished research by Piličiauskas.

Figure 5: Plot of $\delta^{15}N$ versus $\delta^{13}C$ values for 'foodcrusts' adhering to Subneolithic and Neolithic vessels from Nida and Šventoji. The prolonged bowls are plotted separately to aid recognition on the plot. The asterisk (*) above three of the data points corresponds to pottery samples from earlier excavations that were consolidated post-excavation.

Figure 6: Bivariate plot of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ for 18 lipid extracts from vessels from Nida and Šventoji and three seal bone lipid extracts from Šventoji. The plot also includes data from Typical/Late Comb Ware (4th millennium BC) and Corded Ware beakers (3rd millennium BC) from southern Finland published in Cramp et al. (2014a). The symbols labelled * are residues without aquatic biomarkers. The archaeological data are plotted alongside modern reference fats and oils reproduced from Dudd and Evershed 2008 and Craig et al., 2011. All reference data plotted with 95% confidence intervals.

Figure 7 (a): Partial TIC (total ion current) chromatogram showing the lipid extract of sample L14i (absorbed residue from Šventoji 26, Pointed-base vessel, Subneolithic 'Comb-like' ware). Key: C$n$:0 – saturated fatty acids with $n$ carbon atoms; C$n$:1 – monounsaturated fatty acids with $n$ carbon atoms; chol – cholesterol; IS – internal standard. (b) Expanded chromatogram (*c.* 25-30 mins) showing the range and diversity of cholesterol alteration products.
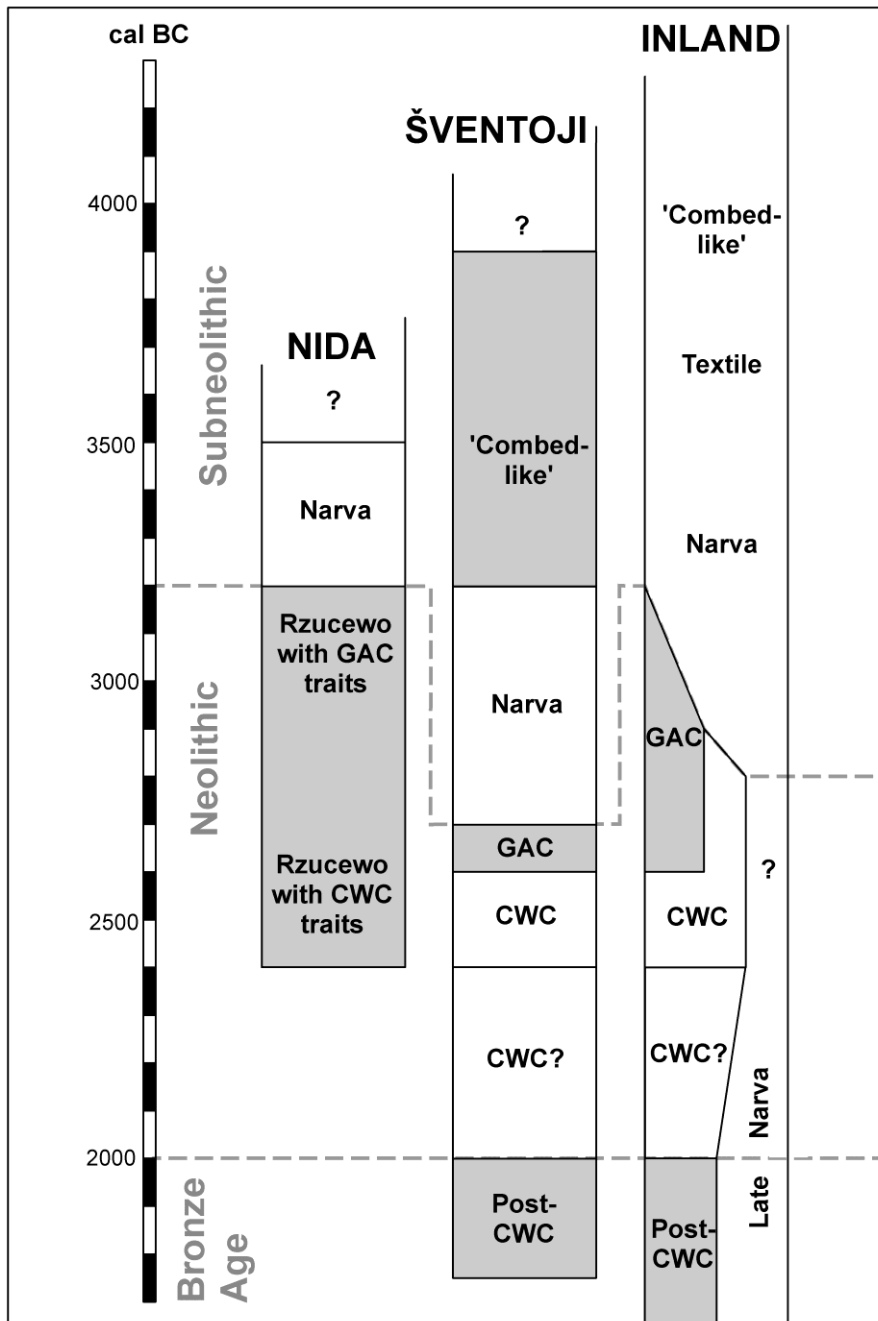
792    Figure 1
793
794
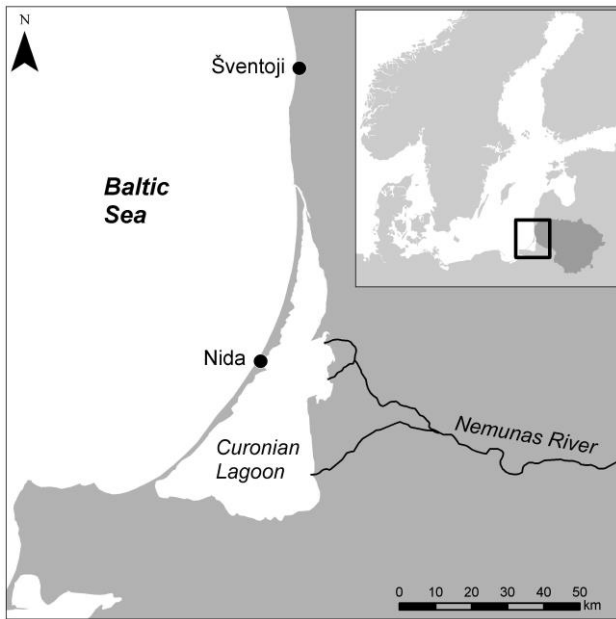
795
796    Figure 2
797
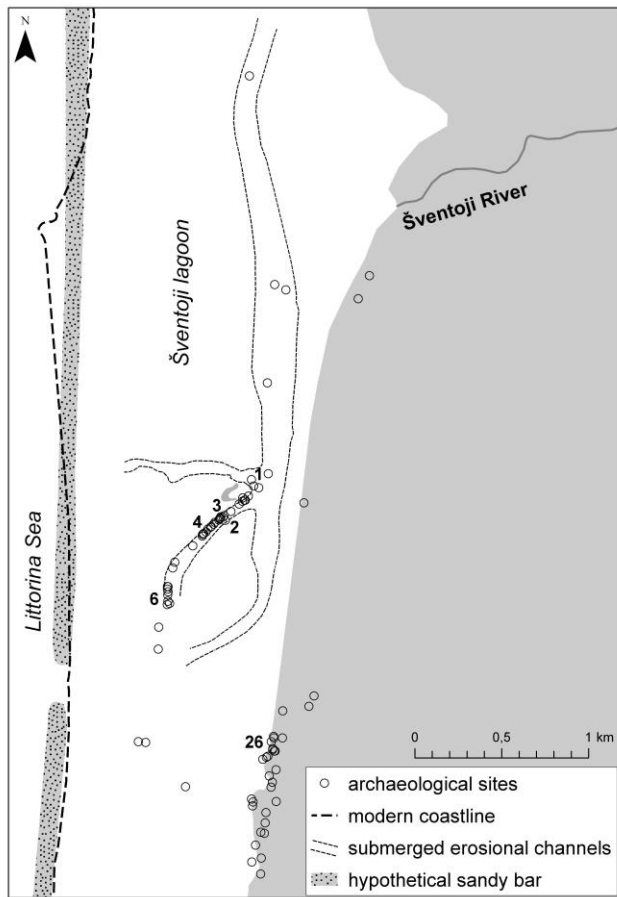
798
799    Figure 3
800
801
802

Figure 4

δ¹⁵N

◇ Nida Neolithic - Other vessels

◆ Nida Neolithic - Prolonged bowls

△ Sventoji Subneolithic - Other vessels

▲ Sventoji Subneolithic - Prolonged bowls

■ Nida Subneolithic vessel

▲ Sventoji Neolithic - Other vessels

Figure 5

(a)

(b)
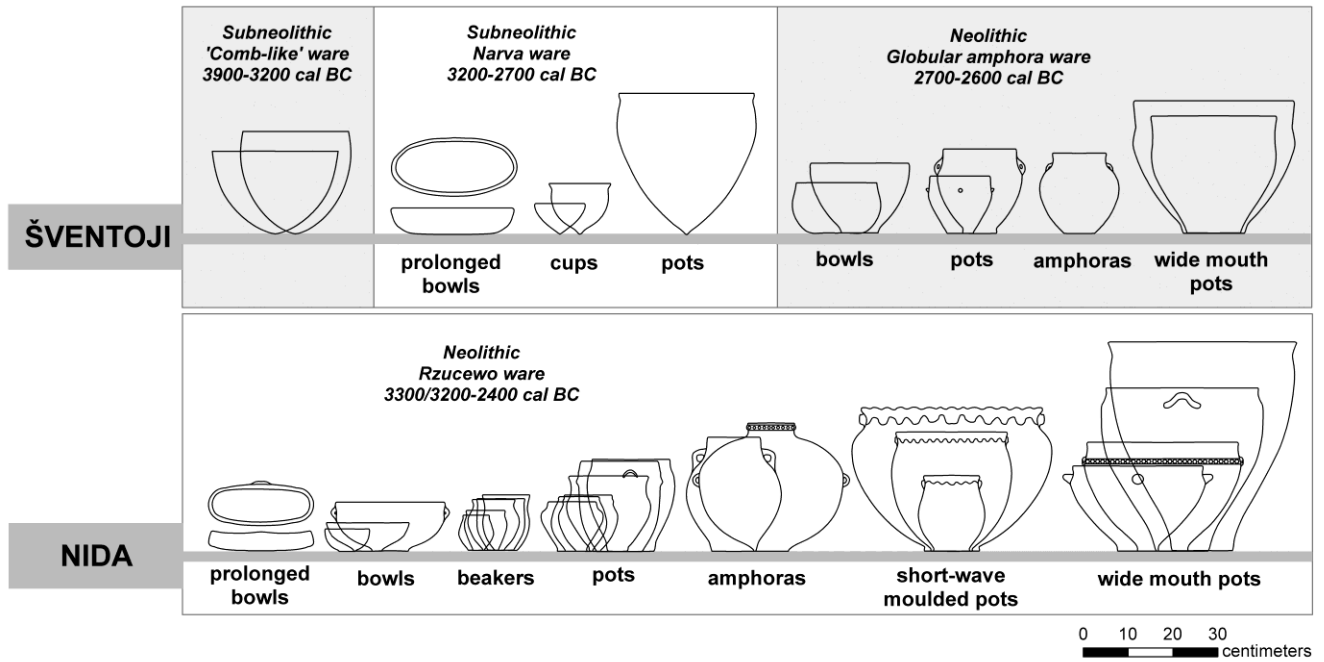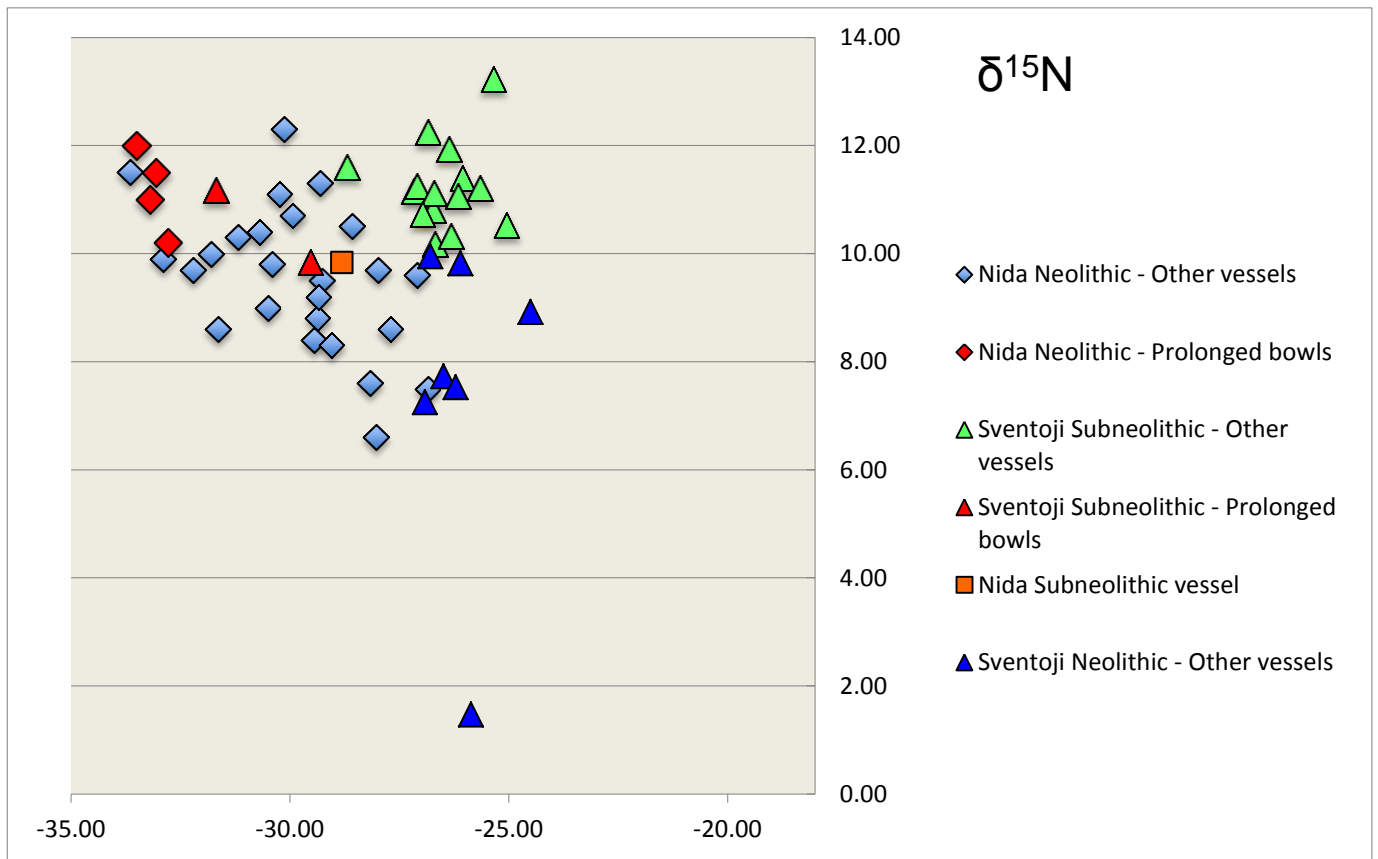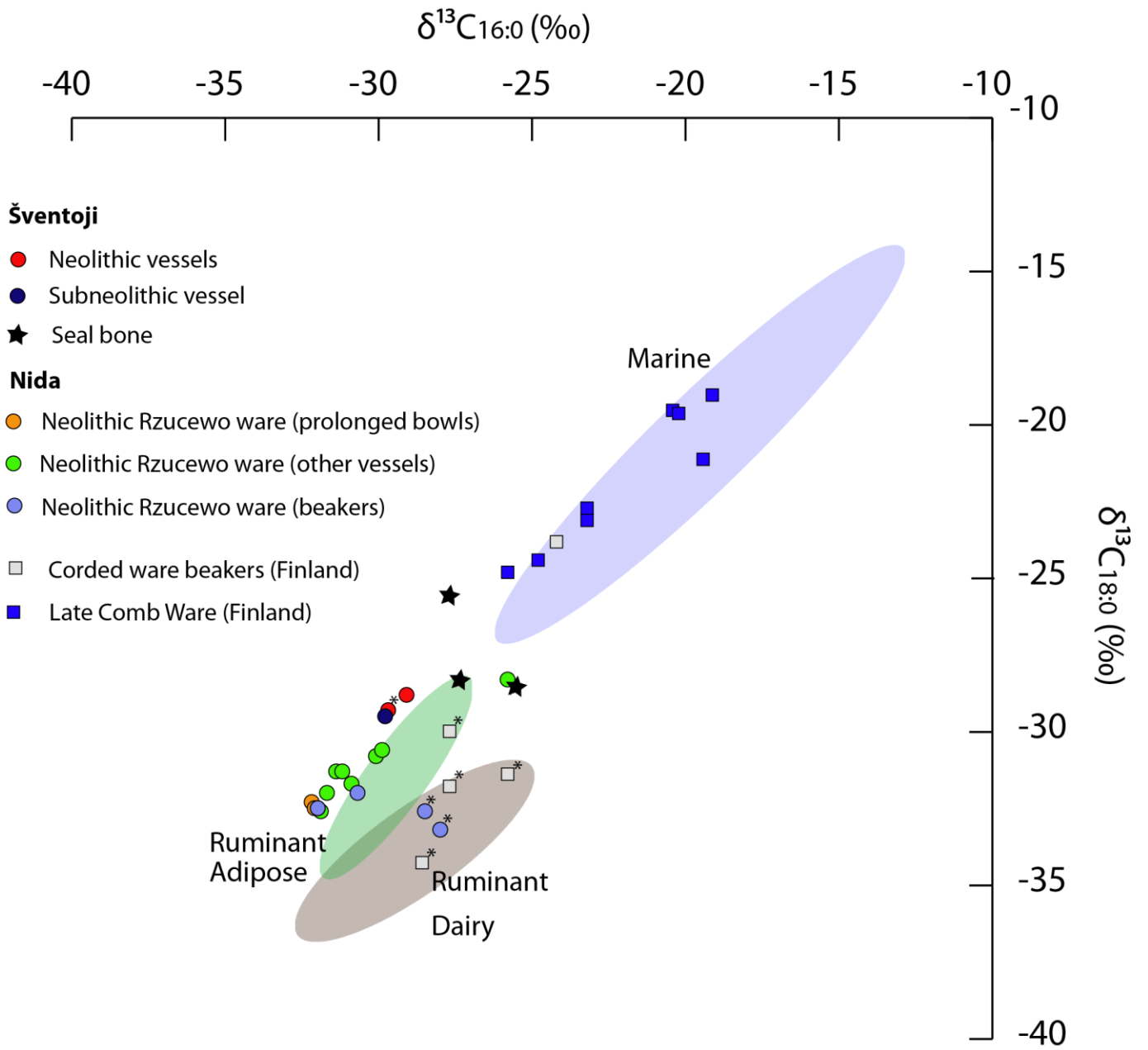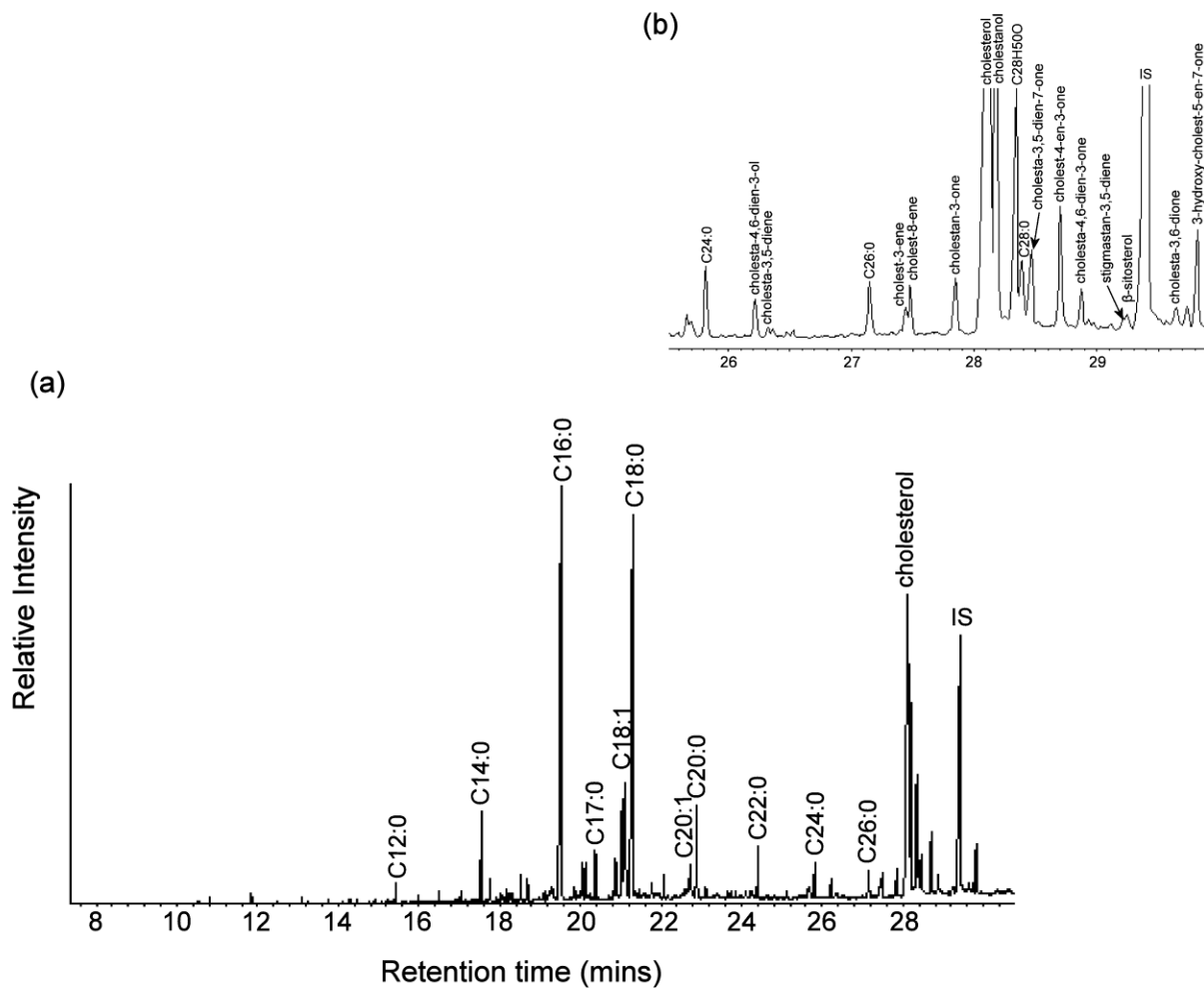
815
816    Figure 7

817 **Supplementary data**
818
819 Supplementary Figure 1
820 Partial total ion current (TIC) chromatogram of lipid extract of seal bone (sample 696)
821 highlighting the excellent preservation of unsaturated fatty acids, especially hexadecenoic acid
822 ($C_{16:1}$).
823
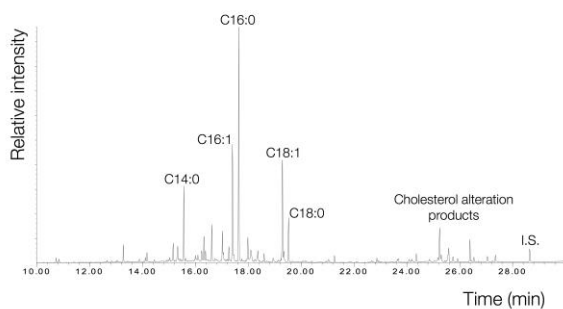


824
825
826
827
828

829 Supplementary Table 1
830 Bulk isotope data of all foodcrusts – additional foodcrust data are presented in Supplementary Table 2 along with
831 corresponding lipid data on these deposits and absorbed residues from the same vessel.
832

| No | Site | Field_No | Weight | Museum_No | Sample | LabCode | Date_BP | Pollution | Temper | Thickness mm | Style | period | $\delta^{15}N$ | $\delta^{13}C$ | C/N |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | Nida 1 | 189 | 0,008 | | interior | Poz-49775 | 5225±35 | clean | very abundant crushed stone | 12 | Rzucewo | Neolithic | 9.81 | -30.40 | 10.12 |
| 2 | Nida 1 | 6921 | 0,005 | | interior near bottom | Poz-64683 | 4540±30 | clean | abundant crushed stone (1-2 mm) | 13 | Rzucewo | Neolithic | 8.27 | -29.05 | 7.92 |
| 3 | Nida 1 | | 0,004 | EM2243:3778 | interior | Hela-2469 | 4946±34 | clean | very abundant crushed stone (<3 mm) | 11 | Rzucewo | Neolithic | 8.81 | -29.36 | 9.49 |
| 4 | Nida 1 | | 0,009 | EM2243:2321 | interior | Hela-2468 | 4917±34 | clean | sand | 7 | Rzucewo | Neolithic | 9.15 | -29.34 | 9.09 |
| 5 | Šventoji 4 | 34 | 0,009 | | exterior of the rim | Poz-61563 | 4500±60 | clean? | mollusc shells | 9 | Narva | Subneolithic | 10.17 | -26.68 | 8.61 |
| 7 | Šventoji 4 | sherd 1436 (09.14) | 0,012 | | interior | Hela-2464 | 4805±33 | clean | mollusc shells | 11 | Narva | Subneolithic | 11.16 | -27.16 | 5.74 |
| 8 | Šventoji 1 | ? | 0,009 | | interior | Hela-2476 | 4625±32 | clean? | abundant crushed stone (<5 mm) | 8 | Globular Amphora | Neolithic | 9.95 | -26.79 | 7.52 |
| 11 | Šventoji 6 | | 0,015 | EM2138:1410 | exterior of the rim | Poz-61585 | 4520±35 | polymer | mollusc shells and plants | 10 | Narva | Subneolithic | 12.25 | -26.85 | 10.19 |
| 12 | Nida 1 | | 0,006 | EM2243:3760 | interior | Hela-2728 | 4818±43 | clean | abundant crushed stone (<3 mm) | 13 | Rzucewo | Neolithic | 8.55 | -31.64 | 11.06 |

| # | Site | | | | Location | Lab ID | Date | | Temper | | Culture | Period | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 13 | Nida 1 | | 0,007 | EM2243:4331 | interior and exterior of the rim | Hela-2467 | 5041±34 | clean | abundant crushed stone (<4 mm) | 9 | Rzucewo | Neolithic | 9.55 | -27.09 | 6.09 |
| 17 | Šventoji 4 | 333 | 0,012 | - | interior | - | - | clean | mollusc shells | 9 | Narva | Subneolithic | 10.32 | -26.32 | 5.87 |
| 18 | Šventoji 4 | 530 | 0,010 | - | interior | - | - | clean | mollusc shells | 9 | Narva | Subneolithic | 11.94 | -26.36 | 5.36 |
| 19 | Šventoji 4 | 44/1 | 0,009 | - | interior | - | - | clean | mollusc shells | 8 | Narva | Subneolithic | 13.23 | -25.35 | 6.13 |
| 20 | Šventoji 4 | 1677 | 0,010 | - | interior | - | - | clean | mollusc shells | 9 | Narva | Subneolithic | 10.80 | -26.72 | 6.50 |
| 21 | Šventoji 4 | 1234 | 0,012 | - | interior | - | - | clean | abundant crushed stone (<3 mm) | 6 | Globular Amphora | Neolithic | 7.25 | -26.91 | 7.39 |
| 22 | Šventoji 4 | 1398 | 0,018 | - | interior | - | - | clean | abundant crushed stone (<3 mm) | 7 | Globular Amphora | Neolithic | 8.92 | -24.50 | 9.95 |
| 34 | Nida 1 | 6930 | 0,030 | | exterior of the neck | - | - | clean | abundant crushed stone (1-2 mm) | 7 | Rzucewo | Neolithic | 11.54 | -33.65 | 35.66 |
| 35 | Nida 1 | 5472 | 0,029 | | exterior of the rim | - | - | clean | abundant crushed stone (1-2 mm) | 9 | Rzucewo | Neolithic | 11.05 | -30.23 | 16.77 |
| 38 | Šventoji 2 | | 0,006 | EM135:141 | exterior of the rim | Hela-2477 | 4507±32 | clean | abundant crushed stone (<4 mm) | 8 | Globular amphora | Neolithic | 9.82 | -26.10 | 18.24 |
| 39 | Nida 1 | 2030 | 0,014 | | interior | - | - | clean | abundant crushed stone (1-2 | 8 | Rzucewo | Neolithic | 9.46 | -29.27 | 9.49 |

mm)

| # | Site | Sample | | | Location | | | Surface | Temper | N | Culture | Period | | | | |
|---|------|--------|---|---|----------|---|---|---------|--------|---|---------|--------|---|---|---|---|
| 40 | Nida 1 | 17 | 0,012 | | exterior of the rim | - | - | clean | abundant crushed stone (1-2 mm) | 9 | Rzucewo | Neolithic | 11.30 | 29.30 | - | 14.40 |
| 41 | Šventoji 4 | 2 | 0,007 | | interior | - | - | clean | crushed stone (2-4 mm) | 8 | Globular amphora | Neolithic | 7.73 | 26.49 | - | 7.72 |
| 42 | Šventoji 4 | 111 | 0,030 | | interior | - | - | clean | mollusc shells | 10 | Narva | Subneolithic | 11.40 | 26.05 | - | 5.71 |
| 43 | Šventoji 3 | 179 | 0,004 | | interior | - | - | clean | mollusc shells | 11 | Narva | Subneolithic | 11.60 | 28.69 | - | 8.33 |
| 44 | Šventoji 3 | 114 | 0,011 | | exterior of the rim | Hela-2462 | 4756±32 | clean | mollusc shells | 10 | Narva | Subneolithic | 11.07 | 26.16 | - | 8.66 |
| 45 | Šventoji 4 | 1413 (09.14) | 0,007 | | exterior of the rim | - | - | clean | abundant crushed stone (<4 mm) | 7 | Globular amphora | Neolithic | 7.53 | 26.21 | - | 9.13 |
| 46 | Nida 1 | | 0,007 | | interior | - | - | clean | mollusc shells | 7 | Narva | Subneolithic | 9.84 | 28.81 | - | 5.79 |
| 47 | Šventoji 3 | 18 (08.16) | 0,012 | | exterior of the rim | - | - | clean | mollusc shells | 9 | Narva | Subneolithic | 11.22 | 25.65 | - | 7.54 |
| 48 | Šventoji 3 | 35 (08.17) | 0,013 | | interior | - | - | clean | mollusc shells | 10 | Narva | Subneolithic | 10.53 | 25.05 | - | 6.09 |
| 49 | Šventoji 3 | 246 (09.13) | 0,004 | | interior | - | - | clean | mollusc shells | 8 | Narva | Subneolithic | 11.24 | 27.09 | - | 6.17 |
| 54 | Šventoji 4 | Prk2/115 | - | | inside of bowl/lamp | - | - | polymer | mollusc shells | | Narva | Subneolithic | 11.17 | 31.69 | - | 19.95 |
| 55 | Šventoji 6 | 31g | - | | inside of bowl/lamp | - | - | clean/polymer? | mollusc shells | | Narva | Subneolithic | 9.85 | 29.53 | - | 29.6 |
| 56 | Nida 1 | 2/10A | - | EM 2243:2949 | inside of bowl/lamp | - | - | clean | abundant crushed stone (1-2 mm) | | Rzucewo | Neolithic | 11.02 | 33.23 | - | 14.8 |

| 57 | Nida 1 | 5/6A | - | EM 2243:6121 | inside bowl/lamp | - | - | clean | abundant crushed stone (1-2 mm) | | Rzucewo | Neolithic | 11.96 | -33.51 | 13.95 |
| 58 | Nida 1 | 1978, 2/28R | - | | interior | - | - | clean | abundant crushed stone (1-2 mm) | | Rzucewo Beaker | Neolithic | 9.9 | -32.9 | 29.1 |

## Supplementary Information 2 (GC-MS, IRMS and GC-C-IRMS data)

834 GC-MS, IRMS and GC-C-IRMS data. The bulk isotope data on the foodcrusts presented here enables comparison with other
835 datasets on the foodcrusts and on absorbed residues from the same vessel and is in addition to the data in Supplementary
836 Table 1.

837

838 Key:

839 Shaded boxes – analysis not conducted; nd – not detected; TMTD – 4,8,12-trimethyltridecanoic acid;

840 DHAA – Dihydroxyalkanoic acids

841

842

843 Šventoji sites

844

| Sample (Yield) | Aquatic Biomarkers | Other Lipids | Bulk isotope data | | | Single-compound isotope data | |
|---|---|---|---|---|---|---|---|
| | | | Bulk $\delta^{13}C$ | Bulk $\delta^{15}N$ | C/N ratio | $\delta^{13}C$ $C_{16:0}$ | $\delta^{13}C$ $C_{18:0}$ |
| L1i (3.1 mg g$^{-1}$) Sherd, Šventoji 4 <br> Interior absorbed residue from Neolithic Globular Amphora ware 2003, 1603(1608?) | TMTD | Fatty acids ($C_{6:0}$-$C_{25:0}$, $C_{16:1}$-$C_{22:1}$), phytanic acid, tr pristanic acid, C5-C10 diacids, C18-C20 DHAA, tr cholesterol and cholesterol oxidation products. diterpenoids | | | | -29.1 | -28.8 |
| L13i (0.1 mg g$^{-1}$) Sherd, Šventoji 3 <br> Interior absorbed residue from Pointed-base vessel Subneolithic Narva ware, 2005, 85 | nd | Abundant cholesterol and cholesterol oxidation products. Fatty acids ($C_{7:0}$-$C_{26:0}$, $C_{14:1}$-$C_{20:1}$), phytanic acid, C18 DHAA, | | | | nd | nd |
| L13e (<0.001 mg g$^{-1}$) Sherd, Šventoji 3 <br> Exterior absorbed residue from Pointed-base vessel Subneolithic Narva ware, 2005, 85 | nd | Fatty acids ($C_{12:0}$-$C_{18:0}$, $C_{18:1}$), C8-C10 diacids | | | | nd | nd |
| L13s (<0.1 mg g$^{-1}$) 'Foodcrust', Šventoji 3 <br> Exterior deposit from Pointed-base vessel Subneolithic Narva ware 2005, 85 | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic acids | Abundant cholesterol and cholesterol oxidation products. Fatty acids ($C_{8:0}$-$C_{24:0}$, $C_{16:1}$-$C_{20:1}$), phytanic acid, C7-C9 diacids | -26.7 | 11.1 | 7.4 | nd | nd |
| L14i (1.2 mg g$^{-1}$) Sherd, Šventoji 26 | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic | Abundant cholesterol and cholesterol oxidation products. | | | | -29.8 | -29.5 |

| Sample | Aquatic Biomarkers | Other Lipids | Bulk δ13C | Bulk δ15N | C/N ratio | δ13C C16:0 | δ13C C18:0 |
|---|---|---|---|---|---|---|---|
| Interior absorbed residue from Pointed-base vessel Subneolithic 'Comb-like' ware 2005, test-pit | acids | Fatty acids ($C_{6:0}$-$C_{30:0}$, $C_{14:1}$-$C_{24:1}$), phytanic acid, tr pristanic acid, C4-C10 diacids, C16-C18 DHAA | | | | | |
| L14s (0.2 mg g[-1]) 'Foodcrust', Šventoji 26 Exterior deposit from Pointed-base vessel Subneolithic 'Comb-like' ware 2005, test-pit | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic acids | Abundant cholesterol and cholesterol oxidation products. Fatty acids ($C_{9:0}$-$C_{28:0}$, $C_{16:1}$-$C_{18:1}$), phytanic acid, C8-C11 diacids | -27.0 | 10.7 | 10.9 | | |
| L17i (0.03 mg g[-1]) Sherd, Šventoji 4 Interior absorbed residue from Amphora Neolithic Globular Amphora ware Šv 4, 14, 982 | nd | Fatty acids ($C_{9:0}$-$C_{32:0}$, $C_{16:1}$-$C_{20:1}$, $C_{18:2}$), tr phytanic acid, *n*-alcohols (C24 - C30); *n*-alkanes (C23 - C29); palmitate wax esters (C40-C42), cholesterol; squalene; β-sitosterol; possible sugars; traces vanillin, 4-hydroxy-benzaldehyde, 4-hydroxy-benzoic acid, 3,5-dimethoxy-4-hydroxy-benzaldehyde; tr ketone (C31); oleanolic acid; | | | | -29.7 | -29.8 |
| L17f 'foodcrust', Šventoji 4 Interior absorbed residue from Amphora Neolithic Globular Amphora ware Šv 4, 14, 982 | | | -25.8 | 1.5 | 16.2 | | |

845

846  Nida

847

| Sample (Yield) | Aquatic Biomarkers | Other Lipids[1] | Bulk δ13C | Bulk δ15N | C/N ratio | δ13C C16:0 | δ13C C18:0 |
|---|---|---|---|---|---|---|---|
| L2i (0.8 mg g[-1]) Sherd Interior absorbed residue from Beaker Neolithic Rzucewo ware 2013, 6311 | TMTD | Fatty acids ($C_{8:0}$-$C_{24:0}$, tr $C_{16:1}$-$C_{19:1}$), phytanic acid, pristanic acid, C7-C12 diacids, C18 DHAA, tr cholesterol and cholesterol oxidation products; tr diterpenoids | | | | -32.0 | -32.5 |
| L3i (0.6 mg g[-1]) Sherd Interior absorbed Beaker | nd | Fatty acids ($C_{9:0}$-$C_{24:0}$, $C_{16:1}$-$C_{18:1}$), phytanic acid, C7-C12 diacids, C18 DHAA, tr diterpenoids | | | | -28.5 | -32.6 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Neolithic Rzucewo ware 2013, 6308 & 6309 | | | | | | | |
| L4i (0.3 mg g$^{-1}$)<br>Sherd<br>Interior absorbed from Wide-mouthed pot<br>Neolithic Rzucewo ware 2013, 6910 | TMTD, C16-C22 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{7:0}$-C$_{30:0}$, C$_{16:1}$-C$_{19:1}$), phytanic acid, C6-C11 diacids, C18 DHAA, tr cholesterol and cholesterol oxidation products | | | | -31.4 | -31.3 |
| L5i (0.5 mg g$^{-1}$)<br>Sherd<br>Interior absorbed residue from pot<br>Neolithic Rzucewo ware 2013, 5483 | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{8:0}$-C$_{28:0}$, C$_{16:1}$-C$_{18:1}$), phytanic acid, C5-C8 diacids, tr cholesterol | | | | -31.2 | -31.3 |
| L5f (0.05 mg g$^{-1}$)<br>'foodcrust'<br>Interior deposit from pot<br>Neolithic Rzucewo ware 2013, 5483 | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{9:0}$-C$_{24:0}$, C$_{16:1}$-C$_{20:1}$), phytanic acid | -30.5 | 9.0 | 8.8 | | |
| L5s (0.2 mg g$^{-1}$)<br>'foodcrust'<br>Exterior deposit from pot<br>Neolithic Rzucewo ware 2013, 5483 | nd | Fatty acids (C$_{14:0}$-C$_{20:0}$, tr C$_{18:1}$), tr phytanic acid, C8-C9 diacids | -30.1 | 12.3 | 14.5 | | |
| L6i (1.4 mg g$^{-1}$)<br>Sherd<br>Interior absorbed residue from Wide-mouthed pot<br>Neolithic Rzucewo ware 2013, 6948 | TMTD | Fatty acids (C$_{9:0}$-C$_{24:0}$, C$_{16:1}$-C$_{19:1}$), phytanic acid, tr pristanic acid, tr C8-C9 diacids, tr cholesterol and cholesterol oxidation products | | | | -30.9 | -31.7 |
| L6f (<0.005 mg g$^{-1}$)<br>'foodcrust'<br>Interior deposit from Wide-mouthed pot<br>Neolithic Rzucewo ware 2013, 6948 | nd | nd | -29.5 | 8.4 | 10.4 | | |
| L6s (<0.005 mg g$^{-1}$)<br>'foodcrust'<br>Exterior deposit from Wide-mouthed pot<br>Neolithic Rzucewo ware 2013, 6948 | nd | nd | -28.0 | 9.7 | 22.3 | | |
| L7i (2.2 mg g$^{-1}$)<br>Sherd<br>Interior absorbed residue from pot<br>Neolithic Rzucewo ware 2013, 5474, 5, 6 | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{9:0}$-C$_{24:0}$, C$_{16:1}$-C$_{18:1}$), phytanic acid, tr pristanic acid, tr C8-C9 diacids, C18 DHAA, tr cholesterol and cholesterol oxidation products | | | | -30.1 | -30.8 |
| L7f (0.05 mg g$^{-1}$)<br>'foodcrust'<br>Interior deposit from pot<br>Neolithic Rzucewo ware 2013, 5474, 5, 6 | nd | Fatty acids (C$_{14:0}$-C$_{24:0}$, C$_{18:1}$), diterpenoids | -28.2 | 7.6 | 12.6 | | |
| L7s<br>'foodcrust' | | | -28.6 | 10.5 | 15.1 | | |

| Sample | TMTD / alkanoic acids | Lipid composition | | | | | |
|---|---|---|---|---|---|---|---|
| Exterior deposit from pot Neolithic Rzucewo ware 2013, 5474, 5, 6 | | | | | | | |
| L8iA (1.9 mg g⁻¹) Sherd Interior absorbed residue above clear 'tide mark' from large pot Neolithic Rzucewo ware 2013, 6165 | nd | Fatty acids ($C_{9:0}$-$C_{26:0}$, $C_{16:1}$-$C_{24:1}$), tr phytanic acid, C8-C9 diacids, C16-C18 DHAA, tr cholesterol and cholesterol oxidation products | | | | -32.2 | -32.6 |
| L8iB (10.8 mg g⁻¹) Sherd Interior absorbed residue below clear 'tide mark' from large pot Neolithic Rzucewo ware 2013, 6165 | TMTD, C18-C22 ω-(o-alkylphenyl)alkanoic acids | Fatty acids ($C_{8:0}$-$C_{24:0}$, $C_{16:1}$-$C_{18:1}$), phytanic acid, C6-C9 diacids, C18 DHAA, cholesterol and cholesterol oxidation products | | | | -31.1 | -31.3 |
| L8f (0.02 mg g⁻¹) 'foodcrust' Interior deposit from large pot Neolithic Rzucewo ware 2013, 6165 | TMTD, C18-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids ($C_{9:0}$-$C_{24:0}$, $C_{16:1}$-$C_{18:1}$), phytanic acid, C8-C11 diacids, tr cholesterol and cholesterol oxidation products | -31.2 | 10.3 | 11.1 | | |
| L8s 'foodcrust' Exterior deposit from large pot Neolithic Rzucewo ware 2013, 6165 | | | -29.9 | 10.7 | 13.5 | | |
| L9i (0.8 mg g⁻¹) Sherd Interior absorbed residue from pot Neolithic Rzucewo ware 2012, 1985 | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids ($C_{7:0}$-$C_{26:0}$, $C_{16:1}$-$C_{20:1}$), phytanic acid, tr pristanic acid, tr C8-C9 diacids, tr cholesterol and cholesterol oxidation products | | | | -31.9 | -32.6 |
| L9f (0.01 mg g⁻¹) 'foodcrust' Interior deposit from Pot Neolithic Rzucewo ware 2012, 1985 | nd | tr cholesterol and cholesterol oxidation products | -30.7 | 10.4 | 9.4 | | |
| L9s 'foodcrust' Exterior deposit on pot Neolithic Rzucewo ware 2012, 1985 | | | -32.2 | 9.7 | 23.1 | | |
| L10i (2.2 mg g⁻¹) Sherd Interior absorbed residue from Wide-mouthed pot Neolithic Rzucewo ware 2012, 629 | TMTD, C16-C22 ω-(o-alkylphenyl)alkanoic acids | Fatty acids ($C_{6:0}$-$C_{24:0}$, $C_{14:1}$-$C_{19:1}$), phytanic acid, tr pristanic acid, C8-C9 diacids, tr cholesterol and cholesterol oxidation products | | | | -29.9 | -30.6 |
| L10f (0.04 mg g⁻¹) 'foodcrust' Interior deposit from Wide-mouthed pot Neolithic Rzucewo ware 2012, 629 | TMTD, C16-C22 ω-(o-alkylphenyl)alkanoic acids | Fatty acids ($C_{9:0}$-$C_{26:0}$, $C_{16:1}$-$C_{22:1}$), phytanic acid, tr cholesterol and cholesterol oxidation products | -28.0 | 6.6 | 10.7 | | |
| L10s | | | -26.9 | 7.5 | 9.8 | | |

| Sample | Lipids / biomarkers | Fatty acids | | | | | |
|---|---|---|---|---|---|---|---|
| 'foodcrust'<br>Exterior deposit from Wide-mouthed pot<br>Neolithic Rzucewo ware 2012, 629 | | | | | | | |
| L11i (0.2 mg g$^{-1}$)<br>Sherd<br>Interior absorbed residue from Wide-mouthed pot<br>Neolithic Rzucewo ware 2012, 1231 | TMTD, C18-C22 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{9:0}$-C$_{26:0}$, tr C$_{16:1}$-C$_{20:1}$), phytanic acid, tr pristanic acid, C6-C9 diacids, tr C18 DHAA, tr cholesterol | | | | -25.8 | -28.3 |
| L11f (<0.01 mg g$^{-1}$)<br>'foodcrust'<br>Interior deposit from Wide-mouthed pot<br>Neolithic Rzucewo ware 2012, 1231 | nd | Fatty acids (C$_{16:0}$-C$_{18:0}$, tr C$_{18:1}$) | -27.7 | 8.6 | 11.3 | | |
| L12i (2.3 mg g$^{-1}$)<br>Sherd<br>Interior absorbed residue from Prolonged bowl/lamp<br>Neolithic Rzucewo ware 2012, 2020 | tr TMTD, C18-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{14:0}$-C$_{24:0}$, C$_{16:1}$-C$_{18:1}$), phytanic acid, C4-C11 diacids, C16-18 DHAA, tr cholesterol and cholesterol oxidation products | | | | -32.2 | -32.3 |
| L12f (0.4 mg g$^{-1}$)<br>'foodcrust'<br>Interior deposit from Prolonged bowl/lamp<br>Neolithic Rzucewo ware 2012, 2020 | tr TMTD, C18-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{8:0}$-C$_{26:0}$, tr C$_{16:1}$-C$_{19:1}$, C$_{18:2}$), phytanic acid, C6-C11 diacids, tr C18 DHAA, tr cholesterol and cholesterol oxidation products, diterpenoids | -32.8 | 10.2 | 23.6 | | |
| L12s<br>'foodcrust'<br>Exterior deposit from Prolonged bowl/lamp<br>Neolithic Rzucewo ware 2012, 2020 | | | -33.1 | 11.6 | 35.7 | | |
| L16i (<0.001 mg g$^{-1}$)<br>Sherd<br>Interior absorbed residue from Wide-mouthed pot<br>Neolithic Rzucewo ware 2013, 5824 | ?? | ?? | | | | -32.0 | -32.5 |
| L16iM (0.01 mg g$^{-1}$)<br>Sherd<br>Interior absorbed from wide-mouthed pot<br>Neolithic Rzucewo ware 2013, 5824<br>*Acid-methanol extraction* | TMTD, C18-C20 ω-(o-alkylphenyl)alkanoic acids | Fatty acids (C$_{12:0}$-C$_{26:0}$, tr C$_{16:1}$-C$_{18:1}$), phytanic acid | | | | | |
| L16 (0.02 mg g$^{-1}$)<br>Sediment | nd | Fatty acids (C$_{14:0}$-C$_{24:0}$, tr C$_{16:1}$-C$_{18:1}$), tr phytanic acid | | | | | |
| L19i (0.2 mg g$^{-1}$)<br>Sherd<br>Interior absorbed residue from | TMTD, C16-C20 ω-(o-alkylphenyl)alkanoic | Fatty acids (C$_{9:0}$-C$_{26:0}$, tr C$_{16:1}$-C$_{20:1}$) | | | | -30.7 | -32.2 |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Beaker, Neolithic Rzucewo ware 1977 *Acid-methanol extraction* | acids | | | | | | |
| L19f 'foodcrust' Interior deposit from Beaker Neolithic Rzucewo ware 1977 | | | -31.8 | 10.0 | 15.2 | | |
| L21i *foodcrust* Interior absorbed residue from Beaker, Neolithic Rzucewo ware 1975, i(i?) 20a *Acid-methanol extraction* | nd | Fatty acids ($C_{14:0}$-$C_{24:0}$, tr $C_{14:1}$-$C_{22:1}$) | | | | -28.0 | -33.4 |
| L25i Sherd Interior absorbed residue from Prolonged bowl/lamp Neolithic Rzucewo ware 1977, 6 | TMTD, C18 ω-(o-alkylphenyl)alkanoic acids | Fatty acids ($C_{6:0}$-$C_{26:0}$, tr $C_{16:1}$-$C_{22:1}$), tr phytanic acid, C7-C11 diacids, C16-C18 DHAA, tr cholesterol and cholesterol oxidation products, | | | | -32.1 | -33.1 |

851 Supplementary Table 3
852 Seal bone lipid data showing comparison between samples subjected to prior solvent extraction before treatment with
853 acidified methanol.
854
855
856 Supplementary Table 3.
857

| Sample (lipid yield) | Aquatic Biomarkers | Other Lipids | Bone Collagen | | | Bone Lipid | | Collagen-lipid offset [$\delta^{13}C_{collagen}$ - mean of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$] (‰) |
|---|---|---|---|---|---|---|---|---|
| | | | $\delta^{13}C$ (‰) | $\delta^{15}N$ (‰) | C/N | $\delta^{13}C_{16:0}$ (‰) | $\delta^{13}C_{18:0}$ (‰) | |
| Seal bone 1 (29 µg/g) | TMTD (tr) | Fatty acids ($C_{12:0}$-$C_{24:0}$, tr $C_{16:1}$-$C_{22:1}$), sterols | -16.3 | 15.5 | 3.4 | -27.3 (**-27.5**) | -28.4 (**-28.9**) | -11.6 |
| Seal bone 585 (67 µg/g) | nd | Fatty acids ($C_{14:0}$-$C_{24:0}$, tr $C_{16:1}$-$C_{24:1}$), sterols | -15.3 | 13.1 | 3.2 | -27.7 (**-26.4**) | -25.6 (**-25.1**) | -11.4 |
| Seal bone 696 (322 µg/g) | nd | Fatty acids ($C_{12:0}$-$C_{24:0}$, tr $C_{16:1}$-$C_{22:1}$) | -16.6 | 12.0 | 3.2 | **-25.5** | **-28.6** | -10.5 |
| Mean values for ten seal bones from Šventoji (from Antanaitis-Jacobs *et al.* 2009) | | | -16.7 ± 0.9 | 12.4 ± 1.0 | 3.4 ± 0.1 | nd | nd | nd |

858
859 Lipid and collagen data from three seal bones from Šventoji. nd – not determined. The compound specific values in bold were
860 obtained on separate extracts without a prior solvent extraction step before acid-methanol extraction. *Collagen-lipid offset
861 determined by taking the mean of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ and subtracting from the bone collagen $\delta^{13}C$ value. The differences in
862 compound specific isotope data between the seal bones with and without prior solvent extraction are very small in the two
863 cases where both methods were employed. The seal bone proved to be lipid rich and well preserved so presumably
864 dominated over any effects from exogenous lipid.
865