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

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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.

Keywords: Southeastern Baltic; Subneolithic; Neolithic; Pottery use; Organic residues; Aquatic resources; Dairy products.

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 ($\Delta^{13}\text{C}_{\text{FA}-\text{COLL}}$) 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

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

The archaeological sites

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

Nida is the most intensively investigated site on the Curonian spit with a long research history dating back to the 19th century (Hollack 1895). An area of 4640 m² was uncovered and more than 100,000 sherds were collected during the 1974-1978 excavations (Rimantienė, 1989). Between 2011-2013 excavations were resumed by one of us (GP). Most of materials belong to Neolithic Rzućewo culture (3300/3200-2400 cal BC) although some fragments of Subneolithic pointed-base vessels were also found (3500/3350 cal BC; Piličiauskas and Heron, 2015). Bones are few and very badly preserved. Seal (*Phocidae* sp.), dog, beaver, red deer, elk, auroch/cattle, boar/pig, fox, horse, sheep/goat, bream, pike (*Esox lucius*), and unidentified bird bones have been found (Hollack, 1895; Rimantienė, 1989; Piličiauskas, unpublished data). However, quantitative evaluation of the importance of these species in the subsistence practices is impossible. Furthermore the domesticated animal bones cannot be assigned to any specific phase of the Rzućewo Culture although it is likely that all or most of domesticated species belong to the latest phase around 2500 cal BC.

Šventoji is situated on the NW edge of coastal Lithuania. Around 60 archaeological sites dating to 4000-500 cal BC have been discovered or investigated there since 1966 (Rimantienė 2005; Piličiauskas et al., 2012). Some of these sites are important because of the presence of waterlogged cultural layers (gyttja) with good preservation of organic materials. At the Šventoji 1-6 sites, the horizon with Subneolithic pointed-base pottery belonging to the Narva culture is overlain by Neolithic materials of the Globular Amphora Culture. The transition is dated to c. 2700 cal BC¹. The Subneolithic and Neolithic sites, i.e. dwelling zones and fishing/dumping zones, are situated on submerged slopes or on the shore of an ancient lagoon fed by the Šventoji river. Diatom analysis and fish species studied indicate that the lagoon was principally a

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

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

The pottery

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

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

Materials and Methods

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

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

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

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

Bulk isotope ratio mass spectrometry ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$)

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

Solvent extraction of foodcrusts and ceramic samples

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

Acidified methanol extraction

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

extracted with hexane (3 × 2 mL) and neutralised with K₂CO₃. The extracts were then dried under a gentle stream of N₂. An internal standard (*n*-tetratriacontane) was added to each sample before analysis by GC-MS and GC-C-IRMS.

Gas Chromatography-mass spectrometry

GC-MS analysis was carried out on an Agilent 7890A series GC attached to an Agilent 5975C Inert XL mass selective detector. The split-splitless injector was operated in splitless mode and the injector and interface were maintained at 300°C and 340°C, respectively. Helium was the carrier gas at constant inlet pressure. The column was inserted directly into the ion source of the mass spectrometer. The ionisation energy was 70eV and spectra were obtained by scanning between *m/z* 50 and 800. All samples were analysed using an Agilent DB5-ms 30m x 2.5mm x 2.5 µm column. The oven temperature was programmed to be isothermal at 50°C for 2 minutes, followed by a rise of 10°C per minute up to 350°C and an isothermal hold for 10 minutes. Compounds were identified by comparison with the NIST library of mass spectral data and published data. Peak area measurements for quantification were carried out using the interactive RTE integrator within the Agilent Chemstation enhanced data analysis software. Abundances were calculated as mg of compound per gram of sample (mg/g).

Gas Chromatography-combustion-isotope ratio mass spectrometry

Carbon stable isotope ratios were determined on two fatty acid methyl esters, methyl palmitate (C_{16:0}) and methyl stearate (C_{18:0}), in each extract using a Delta V Advantage isotope ratio mass spectrometer (Thermo Fisher Scientific, Bremen, Germany) linked to a Trace Ultra gas chromatograph (Thermo Fisher) with a ConFlo IV interface (Cu/Ni combustion reactor held at 1000°C; Thermo Fisher). All samples were diluted with hexane and subsequently 1 µL of each sample was injected into a DB5 fused-silica column (30m x 0.25mm id x 0.25 µm film thickness). The temperature was set for 1 min at 45°C, and raised by 6°C min⁻¹ to 295°C, where it was held for 15 min. The carrier gas was ultra-high-purity grade helium at a flow rate of 1.4 mL min⁻¹. The eluted products were combusted to CO₂ and ionised in the source of the mass spectrometer by electron ionisation. The ion intensities of *m/z* 44, 45, and 46 were monitored in order to automatically compute the ¹³C/¹²C ratio of each peak in the extracts. Computations were performed with Isodat 3.0 Gas Isotope Ratio MS Software (version 3.0; Thermo Fisher) and were based on comparisons with a standard reference gas (CO₂) of known isotopic composition that was repeatedly measured. The results from the analysis are reported in ‰ relative to an international standard (V-PDB). Replicate measurements of each sample and a mixture of fatty acid methyl esters (FAMES) with δ¹³C values traceable to international standards were used to determine the instrument precision (<0.3‰) and accuracy (<0.5‰). The values were also corrected subsequent to analysis to account for the methylation of the carboxyl group. The corrections were based on comparisons with a standard mixture of C_{16:0} and C_{18:0} fatty acids of known isotopic composition processed in each batch as a sample.

Results

The bulk isotope data are presented in Figure 5. Table 2 compares the data obtained on the foodcrusts. The prolonged bowls from both sites are presented separately as these values differ from the remaining vessels.

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)
$\delta^{13}\text{C}$	-33.1 ± 0.3	-29.8 ± 1.8	-30.6 ± 1.5	-26.5 ± 0.9	-26.1 ± 0.8
$\delta^{15}\text{N}$	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

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

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

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

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

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

depleted carbon isotope values ($< -30\text{‰}$), the presence of aquatic biomarkers and the high $\delta^{15}\text{N}$ values in the foodcrusts strongly supports the processing of freshwater resources in these vessels (Craig et al., 2007) or organisms (e.g., seals, birds) subsisting on freshwater and brackish resources. The more depleted carbon isotope values result from a variety of causes such as diet, habitat and the varying input of ^{13}C -depleted terrestrial organic matter although there is considerable variation leading to large differences in the tissues of freshwater organisms (e.g., Dufour et al., 1999; Katzenberg et al., 1999). Few compound specific carbon isotope values from modern freshwater fish have been published to date. Craig et al. (2007) include values obtained on fish from a UK river. Outram et al. (2012) report the analysis of 16 Kazakh riverine freshwater fish. Cramp and Evershed (2014, Figure 6) plot a wide range of authentic fats and oils including the aforementioned samples as well as roach and perch from a UK lake. The depleted values for the fatty acids from the modern freshwater lipids in all of the above studies are notable and show clear isotopic separation from marine resources.

The relationship between bulk and compound specific carbon isotope values has not been investigated comprehensively to date. The bulk carbon isotope value of the foodcrust may represent carbon sources from the different compound classes present (fats, proteins and carbohydrates) whereas the compound specific carbon isotope value represents the carbon sources of the principal lipid components only. There is a strong positive correlation ($r = 0.76$) between the bulk carbon isotope value of foodcrusts and the mean of the $\text{C}_{16:0}$ and $\text{C}_{18:0}$ values from GC-C-IRMS analysis of the residues absorbed into the interior surface of the same vessel. The mean offset between bulk and compound specific values is -1.6‰ reflecting the greater depletion in carbon-13 of lipid compared to other food components. Comparing these two values may be questionable especially as the compound specific value is derived from fatty acids in the absorbed residue rather than the foodcrust. The strong positive correlation merits further consideration. Firstly it suggests that the lipid absorbed into the vessels is very similar in origin to the charred food residue adhering to the vessel surface. Secondly, it suggests that, in certain cases, the bulk carbon isotope value can give an indication of the compound specific isotope value, perhaps where the vessel has been used to process the same food throughout its use-life. Clearly further consideration of these issues is needed from a wider range of sites and situations.

Discussion – the relationship between vessel type and residue identification

Nida

In general, the high nitrogen isotope values in the foodcrusts are consistent with high trophic level resources such as aquatic products. The $\delta^{13}\text{C}$ values for the foodcrusts from Nida are more depleted than those at Šventoji, both for the prolonged bowls and the other vessels sampled. Whilst this might represent different products processed in these vessels, an alternative explanation is that the more depleted values from Nida indicate foods from a freshwater body only minimally influenced by marine/brackish water from the Baltic Sea. In contrast the Šventoji lagoon may have been more susceptible to influxes of saline water with wider resource availability resulting in less depleted isotope values. Very few faunal remains from Nida are available to evaluate this possibility. One seal phalanx (*Phocidae* sp.) from the site has a $\delta^{13}\text{C}$ value of -15.9‰ , which is consistent with values from 13 seals at Šventoji (see Table 3). Although the sample size is small, modern freshwater fish from the Curonian Lagoon have lower $\delta^{13}\text{C}$ values than Neolithic fish bone from Šventoji by an average of -2.3‰ (Piličiauskas, unpublished results) but the species are different and any differences will, in part, 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 $\delta^{15}\text{N}$ and low $\delta^{13}\text{C}$ values. An outlier in this group is L11i ($\text{C}_{16:0} = -25.8\text{‰}$; $\text{C}_{18:0} = -28.3\text{‰}$). This residue includes the full range of aquatic biomarkers but is the most enriched of the $\text{C}_{16:0}$ and $\text{C}_{18:0}$ 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 ($\Delta^{13}\text{C}$ value) between $\delta^{13}\text{C}_{16:0}$ and $\delta^{13}\text{C}_{18:0}$ 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 $\Delta^{13}\text{C}$ 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 ($\delta^{13}\text{C}$ value, -32.9‰ ; $\delta^{15}\text{N}$ 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 $\delta^{15}\text{N}$ 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.

However, there is a significant difference in the nitrogen isotope ratios (heteroscedastic t test; $0.02 > p > 0.01$) with the Neolithic foodcrusts having lower $\delta^{15}\text{N}$ values (see Table 2).

To further explore the relationship between the pottery residues and the fauna from the site three seal bones were analysed following the approach outlined in Colonese et al. (2015). Table 3 shows molecular and isotopic data obtained on three archaeological seal bones from Šventoji (see also Supplementary Table 3). The offset between collagen and lipids ($\Delta^{13}\text{C}_{\text{FA-COLL}}$) from the same bone ranges from 10.5 to 11.6‰. This is lower than the data from two pinnepeds from Cnoc Caig, Oronsay (UK) which had higher $\Delta^{13}\text{C}_{\text{FA-COLL}}$ values up to 17.3‰ (Colonese et al., 2015). In another study, Fernandes et al. (2014) showed that modern fishbone collagen from two marine fish is c. 10‰ heavier than bulk tissue lipids which accords well with the data presented here. The compound specific values for the seal bones range from -25.5 to -27.7‰. If bone lipid values can be used as a proxy for tissue lipid, then this provides an indication of the range expected for pottery vessel residues used to process seal tissues in this region. The mean bulk $\delta^{13}\text{C}$ value of 15 Subneolithic foodcrusts from Šventoji is $-26.5 \pm 0.9\text{‰}$. Compound specific data was obtained on only one Subneolithic vessel (L14i) giving $\delta^{13}\text{C}_{16:0}$ and $\delta^{13}\text{C}_{18:0}$ values of -29.8 and -29.5‰ respectively.

The high $\delta^{15}\text{N}$ values in the Subneolithic foodcrusts ($11.3 \pm 0.8\text{‰}$) supports the presence of aquatic products in the vessels. The lipid extracts from two Subneolithic pointed-base vessels from Šventoji 3 and 26 (L13 and L14 respectively) are remarkable for the high abundance of cholesterol and cholesterol oxidation products. The presence of TMTD and C16-C20 ω -(*o*-alkylphenyl)alkanoic acids strongly suggests an aquatic contribution to the residues. Figure 7 shows the total ion current (TIC) chromatogram of an absorbed residue from a Subneolithic pointed-base vessel from Šventoji 26. The abundance of cholesterol in the residue is noteworthy. The peak area ratio of cholesterol to hexadecanoic acid ($\text{C}_{16:0}$) is 0.77, which is highly unusual in archaeological lipid residues. The Narva vessel from Šventoji 3 (L13) has an even higher ratio of 0.85. The remaining samples from Šventoji have cholesterol/ $\text{C}_{16:0}$ ratios of only 0.03 and 0.09 consistent with all the residues from Nida (≤ 0.01 to 0.08). A high abundance of cholesterol together with a range of cholesterol oxidation and reduction products has been identified in residues considered to have an aquatic origin, especially vessels in ceramic and stone considered to have been used as lamps (Heron et al., 2013, supplementary information; Solazzo and Erhardt, 2007). A similar suite of cholesterol oxidation products has been found in experiments conducted on the pyrolysis of cholesterol in hydrous conditions below 200°C (Rushdi et al., 2003).

The two Neolithic globular amphoras from Šventoji 4 have complex residues. The amphora, L1, produced $\text{C}_{16:0}$ and $\text{C}_{18:0}$ carbon isotope values of -29.1‰ and -28.8‰ respectively with a substantial lipid extract of c. 3 mg g^{-1} , which includes abundant unsaturated fatty acids in the range $\text{C}_{16:1}$ - $\text{C}_{20:1}$. All three isoprenoid fatty acids are present suggesting an aquatic product. Abundant Pinaceae sp. diterpenoids are also present, including abietic and pimaric acids as well as the oxidation products, dehydroabietic and 7-oxodehydroabietic acids. Their presence could suggest a lining or sealant of resin or tar applied to the vessel to facilitate storage of liquid contents. A second Neolithic globular amphora, L17 produced a residue considered to be highly degraded beeswax with no aquatic biomarkers. The identification of beeswax is supported by an *n*-alkane (C_{23} - C_{29}) and *n*-alkanol (C_{24} - C_{30}) fraction together with traces of the lower molecular weight palmitate wax ester fraction (C_{40} - C_{42}) and a high abundance of tetracosanoic acid ($\text{C}_{24:0}$) 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 $\delta^{13}\text{C}$ value of -25.8‰ and a $\delta^{15}\text{N}$ 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.
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Sample (lipid yield)	Aquatic Biomarkers	Other Lipids	Bone Collagen			Bone Lipid		Collagen-lipid spacing [$\Delta^{13}\text{C}_{\text{FA-COLL}}$]*
			$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)	C/N	$\delta^{13}\text{C}_{16:0}$ (‰)	$\delta^{13}\text{C}_{18:0}$ (‰)	
Seal bone 1 (29 µg/g)	TMTD (tr)	Fatty acids ($\text{C}_{12:0}$ - $\text{C}_{24:0}$, tr $\text{C}_{16:1}$ - $\text{C}_{22:1}$), sterols	-16.3	15.5	3.4	-27.3	-28.4	-11.6
Seal bone 585 (67 µg/g)	-	Fatty acids ($\text{C}_{14:0}$ - $\text{C}_{24:0}$, tr $\text{C}_{16:1}$ - $\text{C}_{24:1}$), sterols	-15.3	13.1	3.2	-27.7	-25.6	-11.4
Seal bone 696 (322 µg/g)	-	Fatty acids ($\text{C}_{12:0}$ - $\text{C}_{24:0}$, tr $\text{C}_{16:1}$ - $\text{C}_{22:1}$)	-16.6	12.0	3.2	-25.5	-28.6	-10.5
Mean values for ten seal bones from Šventoji (from Antanaitis-Jacobs <i>et al.</i> 2009)			-16.7 ± 0.9	12.4 ± 1.0	3.4 ± 0.1	nd	nd	nd

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

The prolonged bowls from Nida and Šventoji

Five prolonged bowls (three from Neolithic Nida and two from Subneolithic Šventoji) were analysed. These vessels have among the lowest $\delta^{13}\text{C}$, the highest $\delta^{15}\text{N}$ values and the highest C/N ratios in each assemblage (Figure 5 and Table 2). GC-MS analysis of the Šventoji bowls was not possible as they had been subject to conservation treatment. Two of the Nida bowls were analysed by GC-C-IRMS and confirm the highly depleted carbon isotope values determined by bulk isotope analysis (Figure 6, Supplementary Table 2). The high C/N ratios are consistent with a substance rich in fat/oil and the lipid biomarker data confirm the presence of TMTD and C18-C20 ω -(α -alkylphenyl)alkanoic acids, which suggests an aquatic rather than a terrestrial source. Ethnographic analogy and experimental archaeology indicates that these vessels may have been used as lamps for illumination and the analytical data support this (Heron et al., 2013). In the latter study, analysis of 'blubber lamps', similar in shape to the prolonged bowls but usually smaller, from western Baltic coastal sites identified lipids consistent with marine oils. The only inland find sampled produced much more depleted GC-C-IRMS values $< -30.0\text{‰}$. Analysis of the two Nida prolonged bowls produced $\delta^{13}\text{C}_{16:0}$ and $\delta^{13}\text{C}_{18:0}$ values $< -32.0\text{‰}$. These data are considerably more depleted than would be expected for marine oils and suggest an alternative source for the contents at inland Western Baltic and coastal southeastern Baltic locations.

Seal blubber oil is commonly assumed to have been the fuel for these vessels but this seems unlikely unless the seals were consuming abundant freshwater foods in their diet. Some modern seal populations do consume, exclusively or partially, freshwater fish (e.g., Auttila et al., 2015, Lundström et al., 2010). Oil extracted from freshwater organisms is another possibility. Historical accounts (e.g., Svenson, 1985, 171) record the extraction of oil from the threespine stickleback (*Gasterosteus aculeatus*). The threespine stickleback is found in freshwater, brackish and coastal marine habitats and was caught when the fish formed huge shoals during late autumn and winter. The oil was then used for many purposes including illumination. Collagen extracted from modern freshwater fish from the Curonian Lagoon has $\delta^{13}\text{C}$ values ranging from -23.5 to -24.3‰ (without correction for the fossil fuel effect; Piličiauskas, unpublished results). Applying a similar collagen-lipid offset demonstrated for the seals from Šventoji would produce the highly depleted lipid signals in the prolonged bowls from Nida. In summary, the data suggest that the shallow, oval or prolonged bowls, in the western and eastern Baltic, contained heated aquatic oils that may have served as illuminants.

Conclusions

This study has demonstrated the presence of a range of organic residues present in the pottery vessels from two coastal prehistoric sites in the southeastern Baltic. There is clear evidence of the processing of aquatic resources in the vessels. Distinguishing freshwater, brackish and marine signals in this environment is complex and further work is needed, particularly through investigating compound specific faunal reference lipid signals. The identification of freshwater resources is suggested especially in the more carbon-13 depleted environment manifested in the residues at Nida. Subsistence based on freshwater foods from the Curonian Lagoon, as well as terrestrial resources, is in a good agreement with the very limited zooarchaeological data from Nida. However this contrasts with the evidence from the type site of Rzucewo where marine fish (e.g. *G. morhua*) and seals clearly dominate the faunal assemblage (Makowiecki, 2003, 88-91). The data presented here contrast with exploitation of marine resources along the 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.

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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 $\delta^{15}\text{N}$ versus $\delta^{13}\text{C}$ 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 $\delta^{13}\text{C}_{16:0}$ and $\delta^{13}\text{C}_{18:0}$ 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.

Figure 1

Figure 2

Figure 3

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Figure 6

815
816 Figure 7

817 **Supplementary data**

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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 ($C_{16:1}$).

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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

N o	Site	Field_N o	Weig ht	Museum_N o	Sample	LabCod e	Date_B P	Pollution	Temper	Thickne ss mm	Style	period	$\delta^{15}\text{N}$	$\delta^{13}\text{C}$	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	Švento ji 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	Švento ji 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	Švento ji 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	Švento ji 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

13	Nida 1		0,007	EM2243:43 31	interior and exterior of the rim	Hela- 2467	5041±3 4	clean	abunda nt crushed stone (<4 mm)	9	Rzucew o	Neolithic	9.55	- 27.09	6.09
17	Švento ji 4	333	0,012	-	interior	-	-	clean	mollusc shells	9	Narva	Subneolith ic	10.3 2	- 26.32	5.87
18	Švento ji 4	530	0,010	-	interior	-	-	clean	mollusc shells	9	Narva	Subneolith ic	11.9 4	- 26.36	5.36
19	Švento ji 4	44/1	0,009	-	interior	-	-	clean	mollusc shells	8	Narva	Subneolith ic	13.2 3	- 25.35	6.13
20	Švento ji 4	1677	0,010	-	interior	-	-	clean	mollusc shells	9	Narva	Subneolith ic	10.8 0	- 26.72	6.50
21	Švento ji 4	1234	0,012	-	interior	-	-	clean	abunda nt crushed stone (<3 mm)	6	Globula r Amphor a	Neolithic	7.25	- 26.91	7.39
22	Švento ji 4	1398	0,018	-	interior	-	-	clean	abunda nt crushed stone (<3 mm)	7	Globula r Amphor a	Neolithic	8.92	- 24.50	9.95
34	Nida 1	6930	0,030		exterior of the neck	-	-	clean	abunda nt crushed stone (1-2 mm)	7	Rzucew o	Neolithic	11.5 4	- 33.65	35.6 6
35	Nida 1	5472	0,029		exterior of the rim	-	-	clean	abunda nt crushed stone (1-2 mm)	9	Rzucew o	Neolithic	11.0 5	- 30.23	16.7 7
38	Švento ji 2		0,006	EM135:141	exterior of the rim	Hela- 2477	4507±3 2	clean	abunda nt crushed stone (<4 mm)	8	Globula r amphor a	Neolithic	9.82	- 26.10	18.2 4
39	Nida 1	2030	0,014		interior	-	-	clean	abunda nt crushed stone (1-2 mm)	8	Rzucew o	Neolithic	9.46	- 29.27	9.49

									mm)							
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

833 **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

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843 Šventoji sites

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			Bulk isotope data			Single-compound isotope data	
Sample (Yield)	Aquatic Biomarkers	Other Lipids	Bulk $\delta^{13}\text{C}$	Bulk $\delta^{15}\text{N}$	C/N ratio	$\delta^{13}\text{C}$ C _{16:0}	$\delta^{13}\text{C}$ C _{18:0}
L1i (3.1 mg g ⁻¹) Sherd, Šventoji 4 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 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 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 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

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, <i>n</i> -alcohols (C24 - C30); <i>n</i> -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

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Sample (Yield)	Aquatic Biomarkers	Other Lipids ¹	Bulk δ ¹³ C	Bulk δ ¹⁵ N	C/N ratio	δ ¹³ C C16:0	δ ¹³ C 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) Sherd Interior absorbed from Wide-mouthed pot 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) Sherd Interior absorbed residue from pot 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) 'foodcrust' Interior deposit from pot 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) 'foodcrust' Exterior deposit from pot 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) Sherd Interior absorbed residue from Wide-mouthed pot 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) 'foodcrust' Interior deposit from Wide-mouthed pot Neolithic Rzucewo ware 2013, 6948	nd	nd	-29.5	8.4	10.4		
L6s (<0.005 mg g ⁻¹) 'foodcrust' Exterior deposit from Wide-mouthed pot Neolithic Rzucewo ware 2013, 6948	nd	nd	-28.0	9.7	22.3		
L7i (2.2 mg g ⁻¹) Sherd Interior absorbed residue from pot 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) 'foodcrust' Interior deposit from pot 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 'foodcrust'			-28.6	10.5	15.1		

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		

'foodcrust' Exterior deposit from Wide-mouthed pot Neolithic Rzucewo ware 2012, 629							
L11i (0.2 mg g ⁻¹) Sherd Interior absorbed residue from Wide-mouthed pot 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) 'foodcrust' Interior deposit from Wide-mouthed pot 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) Sherd Interior absorbed residue from Prolonged bowl/lamp 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) 'foodcrust' Interior deposit from Prolonged bowl/lamp 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 'foodcrust' Exterior deposit from Prolonged bowl/lamp Neolithic Rzucewo ware 2012, 2020			-33.1	11.6	35.7		
L16i (<0.001 mg g ⁻¹) Sherd Interior absorbed residue from Wide-mouthed pot Neolithic Rzucewo ware 2013, 5824	??	??				-32.0	-32.5
L16iM (0.01 mg g ⁻¹) Sherd Interior absorbed from wide-mouthed pot Neolithic Rzucewo ware 2013, 5824 <i>Acid-methanol extraction</i>	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) 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) Sherd 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

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Beaker, Neolithic Rzucewo ware 1977 <i>Acid-methanol extraction</i>	acids						
L19f 'foodcrust' Interior deposit from Beaker Neolithic Rzucewo ware 1977			-31.8	10.0	15.2		
L21i <i>'foodcrust'</i> <i>Interior absorbed residue from</i> <i>Beaker,</i> <i>Neolithic Rzucewo ware 1975, i(i?)</i> <i>20a</i> <i>Acid-methanol extraction</i>	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.

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856 Supplementary Table 3.
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Sample (lipid yield)	Aquatic Biomarkers	Other Lipids	Bone Collagen			Bone Lipid		Collagen-lipid offset [$\delta^{13}\text{C}_{\text{collagen}}$ - mean of $\delta^{13}\text{C}_{16:0}$ and $\delta^{13}\text{C}_{18:0}$] (‰)
			$\delta^{13}\text{C}$ (‰)	$\delta^{15}\text{N}$ (‰)	C/N	$\delta^{13}\text{C}_{16:0}$ (‰)	$\delta^{13}\text{C}_{18:0}$ (‰)	
Seal bone 1 (29 µg/g)	TMTD (tr)	Fatty acids ($\text{C}_{12:0}$ - $\text{C}_{24:0}$, tr $\text{C}_{16:1}$ - $\text{C}_{22:1}$), 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 ($\text{C}_{14:0}$ - $\text{C}_{24:0}$, tr $\text{C}_{16:1}$ - $\text{C}_{24:1}$), 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 ($\text{C}_{12:0}$ - $\text{C}_{24:0}$, tr $\text{C}_{16:1}$ - $\text{C}_{22:1}$)	-16.6	12.0	3.2	-25.5	-28.6	-10.5
Mean values for ten seal bones from Šventoji (from Antanaitis-Jacobs <i>et al.</i> 2009)			-16.7 ± 0.9	12.4 ± 1.0	3.4 ± 0.1	nd	nd	nd

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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 $\delta^{13}\text{C}_{16:0}$ and $\delta^{13}\text{C}_{18:0}$ and subtracting from the bone collagen $\delta^{13}\text{C}$ 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.
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