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1 **Title:**

2 Investigating the function of prehistoric stone bowls and griddle stones in the Aleutian Islands by lipid
3 residue analysis

4

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14 **ABSTRACT**

15

16 The earliest durable cooking technologies found in Alaska are stone bowls and griddle stones recovered
17 from the Aleutian Islands. This paper aims to identify the function of these artefacts. Molecular and
18 chemical analysis of carbonised residues found on their surfaces confirm that these artefacts were used
19 to process marine resources. Both artefacts have high lipid content and C:N ratios, suggesting they were
20 used to process oily substances. Stable isotope results of individual lipids suggest that they were used to
21 process different sets of resources within the aquatic spectrum as griddle stones have slightly more ¹³C
22 depleted lipids than stone bowls, possibly indicating more variable use. Integration of these results with
23 archaeological and ethnographic data lead us to infer that griddle stones were used for the cooking of a
24 diversity of aquatic resources, possibly with the addition of plant foods, whereas stone bowls were
25 specifically used to render marine mammal fats. We further hypothesize that a sudden peak in stone bowl
26 frequencies at 4000-3000 cal yr BP was connected to a Neoglacial cold spell bringing sea-ice conditions
27 to the Aleutian Islands. This may have led to new subsistence strategies in which the rendering of marine
28 mammal fats played a central role.

29

30 **Keywords:** durable container technologies, Aleutian Islands, stone bowls, griddle stones, oil rendering,
31 cooking, organic residue analysis, lipids, compound specific isotopes, maritime adaptation, Neoglacial

32

33 **INTRODUCTION**

34

35 The use of durable container technologies in prehistory has often been connected to increasingly
36 sedentary lifestyles, generally linked to agriculture. But it was not only the introduction of farming that led
37 people to stay in one place. Seasonal abundance of aquatic resources in specific parts of the landscape
38 can also facilitate increasing sedentism (Jordan and Zvelebil, 2009). The sub-Arctic Aleutian Islands are
39 an ecological hotspot where early sedentism occurred based on the year-round abundance of marine
40 resources. Through exploiting marine mammals and fish the Aleutian tradition grew out to become "...one
41 of the world's most highly specialized and successful maritime hunter-gatherer adaptations" (Corbett and

42 Yarborough, 2016: p.607). Terrestrial resources were scarce with only a limited range of plant and
43 terrestrial animals available. However, birds such as ptarmigan and waterfowl were abundant.
44 Nevertheless, people focused their main efforts on the sea.

45
46 Heavy stone vessels such as bowls and flat cooking stones known as griddle stones were a technology
47 central to this subsistence economy. The procurement, manufacture and maintenance of these tools
48 required investment of time and effort (Jeanotte et al., 2012). But despite their apparent importance, the
49 function of these artefacts remains unclear. Carbonised deposits on griddle stone surfaces, bowl rims and
50 exteriors hint at the use of these artefacts as food processing tools using direct heating methods. Knecht
51 et al. (2001: p.49) and Knecht and Davis (2008: p.73) suggested that stone bowls were used for the hot
52 rendering of sea mammal oil, one of the most important commodities in the life of Northern peoples.
53 However, this has never been tested. Little is known regarding the use of griddle stones although it has
54 been suggested that they were used for cooking sea food (Jeanotte et al., 2012).

55
56 In this paper we aim to identify the function of stone bowls and griddle stones. The organic residues
57 preserved on these artefacts offer the opportunity to identify different cooking and storage practices.
58 Through organic residue analysis we test the hypotheses that 1) these artefacts were used for the
59 processing of aquatic resources, and 2) stone bowls and griddle stones may have been used for different
60 purposes. Building on our finds, our second aim is to explore why stone bowl frequencies peak so
61 suddenly and to explore the role of climate change in the emergence and abundance of this artefact type.

62

63

64 **Culture historical phasing**

65

66 Humans first arrived in the Aleutian Islands around 9000 cal yr BP. Their subsistence practice is
67 considered to have been focused on maritime resources despite the terrestrial character of their toolkit.
68 Possibly these people were late Paleoarctic terrestrial game hunters that came to the Aleutian Islands
69 using a route across landfast sea ice (Davis et al., 2016: p.293). Knecht and Davis (2001) divided the
70 Anangula tradition in an early (9000-7000 cal yr BP) and a late stage (7000-4000 cal yr BP). It has been
71 argued that an influx of Ocean Bay I people from Kodiak Island (Fig.1) around 7000 cal yr BP further
72 added to the foundation of the specialized maritime adaptation that the Unangaġ people (better known as
73 the Aleut) are known for (Dumond, 1977; Dumond and Bland, 1995). People were attracted to the region
74 because of an abundance of fish (cod and halibut) and sea mammals such as: harbor seal, whales,
75 porpoise, sea lion and sea otter but also a variety of bird species. The earliest stone bowls (n=2) and
76 griddle stones (n=1) are found in low numbers at a few sites dating to the Early Anangula phase. They
77 become more numerous during later phases.

78

79 The Margaret Bay phase (4000-3000 cal yr BP) is a period of both climatic and cultural change. Based on
80 the faunal assemblages of the Margaret Bay (3800-3000 cal yr BP) and Amaknak Bridge (3500-2500 cal
81 yr BP) sites, Davis (2001) and Crockford and Frederick (2007) argue for the presence of sea-ice in the
82 region generated by the onset of the colder sea-surface temperatures of the Neoglacial. This induced
83 marine productivity and new species appeared in the region such as walrus, ringed seal and polar bear.
84 The new situation presented challenges and opportunities for the Unangaġ. Subsistence practices were
85 adapted to the new circumstances and focused more on ice-bound marine mammal hunting (Knecht and
86 Davis, 2008). Stone bowls peak during this phase (Table 1) with high occurrences at the Margaret Bay
87 (n=434), and Amaknak Bridge (n=71) sites, which suggests the substantial importance of these artefacts
88 in Aleutian daily life at these sites.

89

90 The Amaknak phase of 3000-1000 cal yr BP can be considered the start of the florescence of the
91 Aleutian tradition (Davis et al., 2016: p.286) with a complex and varied toolkit representing the continuous
92 further development of the long established maritime adaptation. Stone bowls seem to go out of use
93 during this period (Davis, et al., 2016: p. 286), while the occurrence of griddle stones increases from this
94 time onwards (Knecht and Davis, 2003; Jeanotte et al., 2012). These two technologies are hardly ever
95 found together. Temperatures fluctuated and possibly influenced the human populations in the area.
96 Colder temperatures led to increased marine productivity, which could have induced cultural expansion
97 as suggested by Maschner (2016: p.340). During the Late Aleutian phase of 1000 to 2000 cal yr BP
98 tensions rose along the Pacific coast of SW Alaska. Fortified sea stacks and refuge sites indicate warfare,
99 possibly with the newly established Koniag tradition of Kodiak Island, but also among neighbouring
100 Unanga groups (Davis et al., 2016: p.286).

101

102 **MATERIALS AND METHODS**

103

104 **Stone Bowls**

105

106 These heavy, non-portable artefacts are made of ground volcanic tuff and come in different textures and
107 colours. Although no complete specimens have been recovered to date, (partial) reconstructions show
108 that shapes varied from oval to rectangular and sizes range from 12 to 45 cm in diameter, and 3 to 12 cm
109 in depth (Figs. 2 and 3). Bowls are distinguished from lamps mainly by their relative depth and base
110 thickness. Where lamps are often shallow with a thick base, bowls have higher walls with a base that is
111 always thinner than the walls and which allows for cooking using a direct heating source. Another
112 distinction is the absence of a wick in bowls whereas some lamps have a raised platform for the wick.
113 Stone bowls occur in large numbers during the colder Margaret Bay phase (4000-3000 cal yr BP). At the
114 Margaret Bay site a total of 434 fragments were recovered, 75% of which dated to around 3300-3100 cal
115 yr BP, the final phase of occupation. At the Amaknak Bridge site 71 fragments were found dating towards
116 the very end of the phase around 2780 cal yr BP. Six fragments were reported from the base of the
117 Chaluka mound dated 3700 cal yr BP (Denniston, 1966: p.108). A few fragments (n=6) were found at the

118 lower levels of the Tanaxtaxak site, also ascribed to the Margaret Bay phase based on artefact
119 assemblage (Knecht and Davis, 2003: p.45). Stone bowls are scarce outside this period though a few
120 older fragments were found at the earlier levels of Margaret Bay (Knecht et al., 2001) and at the Anangula
121 Blade site (n=1) (McCartney and Veltre, 1996) and the Oiled Blade site (n=1) on Hog Island at 9000 cal yr
122 BP (Knecht and Davis, 2001: p.273). With the abandonment of the Margaret Bay and Amaknak Bridge
123 sites stone bowls also seem to disappear from the Aleutian Islands archaeological record (Knecht et al.,
124 2001: p.49).

125

126 **Griddle Stones**

127

128 Referred to as “stone frying-pans” by (Jochelson, 1925: p.109), the presence of these grease-covered
129 stone slabs goes back 9000 years in the Aleutians (Fig. 4b). No complete specimens of griddle stones
130 are known. Like the stone bowls, they are all fragmented, perhaps fractured during use, or purposefully
131 broken after their use-life was completed. Jeanotte et al. (2012) showed that at the ADK-011 site on Adak
132 Island the majority of griddle stone raw material was carefully selected from a source some 5 km away
133 from the site, while a lesser quality source was also available much closer to site. This indicates that
134 these artefacts were not just flat stones selected randomly. Acquiring them would have been costly both
135 in time and effort.

136

137 Despite the importance of these food processing techniques in the Aleutian subsistence economy, the
138 subject has received little attention in current archaeological literature. Jeanotte et al. (2012) were the first
139 to perform analysis on the residues associated with the griddle stones by using bulk carbon isotope
140 analysis and visible/near infrared spectrometry but were not able to offer any specific identifications. Here
141 we aim to investigate the function of stone bowls and griddle stones through the structural and isotopic
142 analysis of lipids that are preserved in the greasy crusts on the artefact surfaces. This approach has been
143 shown to be highly effective distinguishing marine and terrestrial products formed during the use of
144 archaeological artefacts (Colonese et al., 2017; Craig et al., 2013; Farrell et al., 2014; Shoda et al., 2017).

145

146 **Lipid Extraction of archaeological food crusts**

147

148 Twenty charred surface residue samples of approximately 100mg were collected of stone bowls from the
149 Margaret Bay (n=11), Amaknak Bridge (n=8), and Tanaxtaxak (n=1) sites. Where available multiple
150 samples were taken to compare interior with exterior residues or base with rim residues. Most of the
151 bowls however only had encrustations on the exterior. Charred surface deposits were also collected from
152 eight griddle stones (~100mg). One sample dates to the Early Anangula phase (9000-8000 cal yr BP)
153 Oiled Blade site, while the other sampled griddle stones were much younger with two specimens from the
154 Tanaxtaxak site on Unalaska (around 500 cal yr BP) and five samples from the Ulyagan site on Carlisle
155 Island, part of the Islands of the Four Mountains (around 400 cal yr BP). The Tanaxtaxak griddle stones
156 were sampled on both sides for comparative reasons. Samples were acquired by scraping off surface
157 residues using a sterile scalpel, and homogenized by grinding the samples to a fine powder using a
158 mortar and pestle.

159

160 Approximately 20 mg of the sample was weighed out for lipid extraction using acidified methanol and
161 following established protocols (Colonese et al., 2017; Papakosta et al., 2015). This approach has been
162 extremely efficient in extracting lipids from carbonised deposits, especially where intact and partially
163 degraded acyl lipids are unlikely to survive (Craig et al., 2007; Lucquin et al., 2016b).

164

165 One mL of methanol was added to the sample which was subsequently ultrasonicated for 15 min. Then
166 200 μ L sulphuric acid (H_2SO_4) was added after which the samples were heated for 4 hours at 70°C. The
167 samples were then centrifuged at 3000 rpm for 5 minutes. The supernatant was transferred to a sterile
168 vial and then extracted three times by adding 2 mL of hexane, mixing, separating and removing the
169 supernatant. The sample was neutralized by passing through a pipette with glass wool and potassium
170 carbonate (K_2CO_3). Eventually the extracts were dried under a gentle stream of nitrogen (N_2) and an
171 internal standard (10 μ L C36 alkane) was added to all samples (lipid quantities ranging from 40 to 8600
172 μ g/g) before further analysis by GC (gas chromatography), GC-MS (GC-mass spectrometry) and GC-c-
173 IRMS (GC-combustion-isotope ratio MS). The majority of acid extracts was also silylated after acid

174 extraction by adding 100 μ L of BSTFA (N,O-bis(trimethylsilyl)trifluoroacetamide) and heating the sample
175 at 70°C for 60 min in order to determine the presence of dihydroxy acids (Hansel and Evershed, 2009).

176

177 **Collagen extraction of archaeological bones**

178

179 A selection of archaeological bone material from the Tanaxtaxak, Margaret Bay and Summer Bay sites as
180 well as the Brooks River area on the Alaska Peninsula was collected to serve as collagen reference
181 material for bulk isotope analysis. Species were: Fin whale (n=2), Porpoise (n=2), Right whale (n=2),
182 Narwhal/Beluga whale (n=3) (all determined using ZooMS, courtesy of the University of York), and sea
183 lion (n=2), seal (n=4), sea otter (n=2), eagle (n=1), bear (n=2), caribou (n=5), anadromous fish (n=2) and
184 marine fish (n=5). Sampling was done by removing a small section of mechanically cleaned bone using a
185 sterile Dremel saw.

186

187 Collagen of 32 bone samples was extracted using a modified Longin method (Brown et al., 1988).

188 Samples (200-300 mg) were demineralized using 0.6 M hydrochloric acid (HCl) at 4°C for several days

189 depending on the sample. Samples were rinsed with distilled water after demineralization. Then they

190 were gelatinised with 0.001 M HCl at 80°C for 48 hours after which the samples were first filtered using

191 Poly-ethylene Ezee filters (Elkay Laboratories Ltd., 9 mL, pore size 60-90 μ m). Subsequently the samples

192 were ultrafiltered (30 kDa, Amicon® Ultra-4 centrifugal filter units, Millipore, MA, USA). Finally the

193 samples were frozen and lyophilised.

194

195 **GC-MS**

196

197 The equipment used for GC-MS analysis was an Agilent 7890A series chromatograph attached to an

198 Agilent 5975C Inert XL mass-selective detector with a quadrupole mass analyser (Agilent technologies,

199 Cheshire, UK). A splitless injector was used and kept at 300 °C. The GC column was inserted

200 into the ion source of the mass spectrometer directly. The carrier gas used was Helium with a constant

201 flow rate of 3 mL min⁻¹. The ionisation energy of the MS was 70eV and spectra were obtained by

202 scanning between m/z 50 and 800. A DB-5ms (5%-phenyl)-methylpolysiloxane column (30 m x 0.250 mm
203 x 0.25 mm; J&W Scientific, Folsom, CA, USA) was used for scanning. The temperature was set at 50 °C
204 for 2 min, then raised by 10 °C min⁻¹ until it reached 325 °C where it was held for 15 min.

205
206 All extracts were also analysed on a DB-23 (50%-Cyanopropyl)- methylpolysiloxane column (60 m x
207 0.250 mm x 0.25 mm; J & Scientific, Folsom, CA, USA) in SIM mode to identify isoprenoid fatty acids and
208 ω - (o-alkylphenyl) alkanolic acids as aquatic biomarkers (Cramp and Evershed, 2014) and to resolve the
209 mixture of phytanic acid diastereomers (Lucquin et al., 2016a). The temperature was set at 50 °C for 2
210 min, then raised by 10 °C min⁻¹ until it reached 100 °C, then raised by 4 °C min⁻¹ to 140 °C, then by 0.5 °C
211 min⁻¹ to 160 °C, then by 20 °C min⁻¹ to 250 °C where it was maintained for 10 min. The first group of ions
212 (m/z 74, 87, 213, 270) corresponding 4,8,12- trimethyltridecanoic acid (TMTD) fragmentation, the second
213 group of ions (m/z 74, 88, 101, 312) corresponding to pristanic acid, the third group of ions (m/z 74, 101,
214 171, 326) corresponding to phytanic acid and the fourth group of ions (m/z 74, 105, 262, 290, 318, 346)
215 corresponding to ω -(o-alkylphenyl) alkanolic acids of carbon length C16 to C22 were monitored,
216 respectively. Helium was used as the carrier gas with a flow rate of 2.4 mL min⁻¹. The relative abundance
217 of two diastereomers of phytanic acids was obtained by the integration of the ion m/z 101.

218

219 **Bulk isotope analysis: carbon/nitrogen**

220

221 Thirty-one surface residue samples of which 21 stone bowls, seven griddle stones and three lamps as
222 well as 32 bone collagen samples were analysed by elemental analysis - isotope ratio mass spectrometry
223 (EA-IRMS). The residue samples were ground into a homogenised powder. The residue and collagen
224 samples were weighed out in duplicate into tin capsules (~0.9 mg). The bulk stable nitrogen ($\delta^{15}\text{N}$) and
225 carbon ($\delta^{13}\text{C}$) isotope values were measured based on previously described methods (Craig et al., 2007).
226 Precision of instrument on repeated measurement was $\pm 0.2\%$ (s.e.m.), $\delta^{13}\text{C}$, $\delta^{15}\text{N} = [(R_{\text{sample}}/$
227 $R_{\text{standard}} - 1)] \times 1000$, where $R = {}^{13}\text{C}/{}^{12}\text{C}$ and ${}^{15}\text{N}/{}^{14}\text{N}$. Accuracy was determined by measurements of
228 international standard reference materials within each analytical run. These were IAEA 600 $\delta^{13}\text{C}_{\text{raw}} = -$
229 27.69 ± 0.02 , $\delta^{13}\text{C}_{\text{true}} = -27.77 \pm 0.04$, $\delta^{15}\text{N}_{\text{raw}} = 1.49 \pm 0.38$, $\delta^{15}\text{N}_{\text{true}} = 1.0 \pm 0.2$; IAEA N2 $\delta^{15}\text{N}_{\text{raw}} = 20.9$

230 ± 0.33 , $\delta^{15}\text{N}_{\text{true}} = 20.3 \pm 0.2$; IA Cane, $\delta^{13}\text{C}_{\text{raw}} -11.76 \pm 0.10$; $\delta^{13}\text{C}_{\text{true}} = -11.64 \pm 0.03$. Data were normalised
231 to these international standards. All samples with %N values below 1% and %C below 10% were
232 excluded.

233

234 **Gas chromatography combustion isotope ratio mass spectrometry**

235

236 Eleven stone bowl and seven griddle stone samples were measured in duplicate for stable carbon isotope
237 values of methyl palmitate ($\text{C}_{16:0}$) and methyl stearate ($\text{C}_{18:0}$) derived from precursor fatty acids by GC-c-
238 IRMS, following existing procedure (Craig et al., 2012). The instrument used for the analysis was a Delta
239 V Advantage isotope ratio mass spectrometer (Thermo Fisher, Bremen, Germany) linked to a Trace Ultra
240 gas chromatograph (Thermo Fisher) with a GC Isolink II interface (Cu/Ni combustion reactor held at
241 $1000\text{ }^\circ\text{C}$; Thermo Fisher) to oxidise all the carbon species to CO_2 . The carrier gas used was ultra-high
242 purity grade helium with a flow rate of 2 mL min^{-1} and parallel acquisition of the molecular data was
243 realised by deriving a small part of the flow to an ISQ mass spectrometer (Thermo Fisher). Samples were
244 diluted in hexane and $1\text{ }\mu\text{L}$ of each sample was injected into DB-5MS ultra-inert fused-silica column
245 ($60\text{ m} \times 0.25\text{ mm} \times 0.25\text{ }\mu\text{m}$; J&W Scientific). The temperature was set at $50\text{ }^\circ\text{C}$ for 0.5 min and raised by
246 $25\text{ }^\circ\text{C min}^{-1}$ to $175\text{ }^\circ\text{C}$, then raised by $8\text{ }^\circ\text{C min}^{-1}$ to $325\text{ }^\circ\text{C}$ where it was held for 20 min. A clear resolution
247 and a baseline separation of the analysed peaks were achieved. Eluted products were ionized in the
248 mass spectrometer by electron impact and ion intensities of m/z 44, 45 and 46 were recorded for
249 automatic computing of the $^{13}\text{C}/^{12}\text{C}$ ratio of each peak in the extracts. Computation was made with Isodat
250 software (version 3.0; Thermo Fisher) and was based on comparisons with standard reference gas (CO_2)
251 of known isotopic composition that was repeatedly measured. The results of the analysis were expressed
252 in per mill (‰) relative to an international standard, VPDB.

253

254 The accuracy of the instrument was determined on *n*-alkanoic acid ester standards of known isotopic
255 composition (Indiana standard F8-3). The mean \pm S.D. values of these were $-29.60 \pm 0.21\text{‰}$ and $-23.02 \pm$
256 0.29‰ for the methyl ester of $\text{C}_{16:0}$ (reported mean value vs. VPDB $-29.90 \pm 0.03\text{‰}$) and $\text{C}_{18:0}$ (reported
257 mean value vs. VPDB $-23.24 \pm 0.01\text{‰}$) respectively. Precision was determined on a laboratory standard

258 mixture injected regularly between samples (28 measurements). The mean \pm S.D. value of *n*-alkanoic
259 acid esters were $-31.65 \pm 0.27\text{‰}$ for the methyl ester of C_{16:0} and $-26.01 \pm 0.26\text{‰}$ for the methyl ester of
260 C_{18:0}. Each sample was measured in replicate (average s.d. 0.07‰ for C_{16:0} and 0.13‰ for C_{18:0}). Values
261 were also corrected subsequent to analysis to account for the methylation of the carboxyl group that
262 occurs during acid extraction. Corrections were based on comparisons with a standard mixture of C_{16:0}
263 and C_{18:0} fatty acids of known isotopic composition processed in each batch under identical conditions.

264

265 **RESULTS**

266

267 **Lipid preservation**

268

269 The preservation of organic residues on the artefacts is very good in general. Both griddle stones and
270 stone bowls provided high quantities of lipids per sample. Thirty of 33 samples ranged from 400 to 8600
271 $\mu\text{g g}^{-1}$, indicative of exceptional preservation. The griddle stones (n=8, mean = 4200 $\mu\text{g/g}$), were richer in
272 lipids than stone bowls (n=22, mean = 2477 $\mu\text{g g}^{-1}$). However, the majority of stone bowls sampled here
273 are about 1500 years older than the griddle stones so this may be the result of degradation. Both of these
274 artefact types contained a much greater amount of lipids than commonly found on charred deposits
275 associated with ceramic cooking pots. For example, the mean lipid concentration from 14 charred
276 deposits on pottery from the sub-Arctic Sakhalin Islands extracted under identical conditions was 298 μg
277 g^{-1} (Gibbs et al. 2017). Only two stone bowl samples showed lower lipid preservation with lipid quantities
278 ranging from 40 to 130 $\mu\text{g g}^{-1}$ (Supplementary Table 1). The oldest stone bowl sample in the Aleutian
279 Islands from the Anangula Blade site (Quimby, 1945; McCartney and Veltre, 1996) yielded no lipid
280 biomarker results and based on the associated %N value of 0.71 we discarded this sample.

281

282 **GC-MS analysis**

283

284 Thirty of 33 samples of both the stone bowls (20 of 23) and the griddle stones (10 of 10) contained
285 isoprenoid acids: TMTD (4,8,12-trimethyltridecanoic acid), pristanic acid (2,6,10,14-

286 tetramethylpentadecanoic acid), and phytanic acid (3,7,11,15-tetramethylhexadecanoic acid) (Fig. 5), as
287 well as ω -(*o*-alkylphenyl) alkanolic acids (APAAs) of carbon length 16 to 22 (Fig. 7). These meet the
288 established criteria for the identification of aquatic resources in archaeology (Hansel et al., 2004;
289 Evershed et al., 2008; Hansel and Evershed, 2009; Lucquin et al., 2016a). Interestingly, APAAs are only
290 formed during the prolonged heating of tri-unsaturated fatty acids at a temperature of at least 270°C and
291 therefore the aquatic oils must have been heated on these artefacts presumably during their processing.
292 These data rule out the contamination of degraded aquatic oils that may be present in the soils as these
293 are unlikely to have been heated. Additionally, the presence of APAAs on these stone artefacts suggests
294 that formation of these compounds is not necessarily dependent on the presence of a ceramic matrix as
295 stated by Evershed, et al. (2008: p.111). To date no evidence of pottery has been found in the Aleutian
296 Islands. Isoprenoid acids are degradation products of phytol, a constituent of chlorophyll, and occur
297 widely in marine organisms. Phytanic acid also occurs in the tissues of ruminant animals. The contribution
298 SSR:SRR diastereomers of phytanic acid (SSR%) provides a means to discriminate these sources
299 (Lucquin et al., 2016a). As expected, because of the lack of ruminants in the area, the data obtained
300 confirms the aquatic origin of phytanic acid in all the Aleutian samples as compared to modern references
301 (Fig. 6).

302
303 Saturated fatty acids range from C₈ to C₃₂ and unsaturated fatty acids, even numbered from C_{16:1} to C_{24:1},
304 with some extending up to C_{26:1}. All samples contain longer chain FA and UFAs with the exception of the
305 three badly preserved samples. Dicarboxylic acids (or diacids), most likely degradation products of
306 unsaturated fatty acids, are also widely present in all samples ranging mostly from 7 to 15 carbon length
307 (Fig. 5). Experimental work by Evershed et al. (2008) showed that diacids of carbon length 8 to 11 form
308 during the heating of aquatic oils. Following derivatization of the acid extract with BSTFA, trace amounts
309 of long chain *n*-alkanols (C₂₂ - C₃₂) were also present in many of the samples with an even number of
310 carbon atoms. As these were found in trace amounts there is a possibility they are derived from the burial
311 environment, as they are a common lipid component of soils (van Bergen et al., 1998), derived from wax
312 compounds in higher plants. However, these compounds were much more abundant in the charred
313 deposits from several of the griddle stones from the Ulyagan site, along with the matching distribution of

314 long chain fatty acids (C₂₂-C₃₂). In this case, it is conceivable that plant products were directly processed
315 on these artefacts.

316

317 **Stable isotope analysis of individual fatty acids**

318

319 Based on the lipid residue analysis results, integrated with contextual archaeological information of the
320 materials, it seems very likely that the tested artefacts were used for the processing of aquatic resources.
321 But what kind of aquatic resources were processed? Were the different artefacts used for different
322 purposes? To further differentiate within the aquatic spectrum we analysed stable isotopes of individual
323 fatty acids C₁₆ and C₁₈ using GC-c-IRMS. This approach serves as a means to discriminate aquatic
324 animals based on their habitat (marine, anadromous, and freshwater), with the marine species relatively
325 enriched in ¹³C compared to the others (Fig 8).

326

327 The carbon isotope values show some difference between the two technological groups (Fig. 8). In
328 general griddle stones are more depleted in both $\delta^{13}\text{C}_{16}$ and $\delta^{13}\text{C}_{18}$ than stone bowls. Two stone bowls
329 are more depleted than the others. Of the seven tested griddle stones, five are separated from the stone
330 bowls and two have similar fatty acid isotope values to the stone bowls. One of these is the only
331 exceptionally old specimen from the Early Anangula phase (9000-8000 cal yr BP) site of Oiled Blade
332 (UNL-318). It is unlikely that the isotopic approach deployed here can be used to distinguish between
333 marine mammal and marine fish oil in this case. Although no authentic lipid carbon isotopes values have
334 been measured from the Aleutian Islands, the $\delta^{13}\text{C}$ values of collagen extracted from 17 marine mammals
335 (mean $\delta^{13}\text{C}_{\text{coll}} = -14.47 \pm 0.04$) and five marine fish (mean $\delta^{13}\text{C}_{\text{coll}} = -11.96 \pm 0.03$) from SW Alaska
336 (Supplementary Table 2) are similar. More depleted lipid sources could include salmon ($\delta^{13}\text{C}_{\text{coll}} = -$
337 15.44 ± 0.05) or potentially terrestrial resources including plants. The latter would be consistent with
338 degraded wax esters found on the griddle stones. However, we stress that overall both the stone bowls
339 and griddle stones have strong marine isotope signatures and aquatic lipid profiles, so any other products
340 are only a minor component in these residues.

341

342 Bulk $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ isotopes

343

344 The $\delta^{15}\text{N}$ ratios of the carbonised deposits associated with the stone bowls and griddle stones are
345 generally within the range expected for marine tissues (i.e. $>10\text{‰}$; Fig 9). These values can be tentatively
346 compared to $\delta^{15}\text{N}$ values of collagen from associated fauna (fish, marine mammals, terrestrial fauna) by
347 assuming that any of the $\delta^{15}\text{N}$ in the surface residues are derived from animal tissue protein and the
348 $\Delta^{15}\text{N}_{\text{tissue-collagen}} = \text{ca. } +2\text{‰}$ (Fernandes et al., 2015). Interestingly, the range of $\delta^{15}\text{N}$ values observed in
349 the carbonised deposits is at the lower end of the marine mammal and fish range (Supplementary Table
350 1).

351

352 Compared to pottery vessels from the Sakhalin Islands, which are assumed to have been used for
353 cooking a range of marine tissues (Gibbs et al. 2017), all the Aleutian artefacts have higher C:N ratios,
354 indicative of a relatively higher lipid content. These data are more comparable to the so called 'blubber
355 lamps' of the European Mesolithic where it is thought that marine mammal oil was burned for illumination
356 (Heron et al., 2013). One caveat to this interpretation is that extensive microbial degradation or
357 percolation with groundwater might lead to a preferential loss of protein, effectively increasing the C:N
358 ratio (Heron and Craig, 2015) although this would seem less likely given the environment is so conducive
359 to molecular preservation. The $\delta^{13}\text{C}$ values in all but one case are less than -25‰ (Supplementary Table
360 1) which are consistent with values reported from pottery from coastal sites with clear marine lipid
361 signatures (Craig et al., 2011, 2013).

362

363 DISCUSSION

364

365 Our main question centred on the specific function of griddle stones and stone bowls within the Aleutian
366 subsistence economy. We hypothesized that these artefacts were used to process aquatic resources but
367 were used in different ways. The results of this research cautiously support our hypotheses. All artefacts
368 with sufficient preservation ($n=30$) show strong evidence for the processing of aquatic resources. Minor
369 isotopic differences between stone bowls and griddle stones may indicate a more variable use of the

370 latter. However, the lipid concentrations and C:N ratios from both artefacts are consistent with their use
371 for processing aquatic oils and fats. By integrating the organic residue results with information of
372 archaeological contexts, ethnography and climate, we discuss the possible function and role of these
373 artefacts placed in a framework of the wider subsistence strategies of the ancient Unangâ.

374

375 **Griddle stone function**

376

377 Griddle stone charred residues show a clear aquatic signal. This is visible in the presence of all aquatic
378 biomarkers as well as in compound specific-, and bulk isotope results. We hypothesized that griddle
379 stones had a more general use for cooking foodstuffs. The residue results are cautiously supportive of
380 this notion. Interpretation is based on comparisons with stone bowls that we assume were used for a very
381 specific purpose, namely the rendering of marine oil. The residues on griddle stones seem to be derived
382 from a bigger diversity of resources. Despite this, aquatic oils still make up the majority of the sample.

383 Depleted $\delta^{13}\text{C}$ values may indicate the contribution of salmon and plant products to the sample.

384 Furthermore the presence of *n*-alkanols on the Ulyagan griddle stones supports the possibility that plant
385 products contributed to the otherwise predominantly aquatic sample. The lower C:N ratio values attest to
386 a higher presence of proteins possibly caused by the cooking of flesh as opposed to fats.

387

388 Not all griddle stones show the same consistent residue results. We analysed the earliest griddle stone in
389 the Aleutian Islands, a partial specimen from the Oiled Blade (UNL-318) site, dating to the Early Anangula
390 phase at 9000-8000 cal yr BP. The compound specific $\delta^{13}\text{C}$ isotope data and C:N ratio value of this
391 particular griddle stone are closer to stone bowl values. This possibly indicates a less diverse use on this
392 ancient griddle stone as opposed to griddle stones from later periods.

393

394 Ethnographic resources are of great value when considering function because griddle stones were still
395 widely in use by the Unangâ during early contact times. A report by C.I. Shade describes the traditional
396 Unangan way to prepare cod soup using a griddle stone: *“The traditional method of making soup was to
397 dig a fire pit and place over it a stone, flush with the ground. Then a very thin beach stone was placed on*

398 *the fire stone and clay walls built upon this base. The liquid was cooked in this. A bluish clay called qudii*
399 *u was used for the walls of this vessel which turned white when heated. This kind of fire pit was called*
400 *unaalu. The same vessel was used more than once. One way of preparing the cod soup was with*
401 *seaweed and seal oil” (in: Johnson, 2004: p.52).*

402

403 This is an interesting notion suggesting griddle stones were actually also used as containers. It also
404 attests to the use of marine mammal oil in cooking practices, agreeable with our findings. No evidence for
405 the use of clay has ever been detected in the archaeological record of the Aleutians. However, some
406 griddle stones are clean in the centre and have a thick edge of greasy and carbonized material around
407 this clean area (Fig. 4a). This use-wear pattern could represent the process described above. Not all
408 griddle stones show this residue distribution though, some show residues in the centre (Fig. 4b). This may
409 suggest different methods of cooking with the use of griddle stones.

410

411 Use patterns possibly changed through time with the earlier griddle stone used for the processing of a
412 single commodity, while griddle stones of the Late Aleutian phase were probably used to cook dishes of a
413 more diverse character, although still predominantly aquatic.

414

415 **Stone bowl function**

416

417 The charred residues distributed mainly along the rims on both interior and exterior, but also on the bases
418 of the bowls, seem to be solely aquatic in origin. The stone bowls have comparable lipid distribution, C:N
419 ratios and bulk isotope characteristics to prehistoric oil lamps from Europe that were unequivocally used
420 to burn aquatic oils for fuel (Heron et al., 2013; Heron and Craig, 2015; Piezonka et al., 2016; Oras et al.,
421 2017).

422

423 Aquatic oil played an important role in the lives of prehistoric and historic peoples of the (sub)Arctic. Not
424 only was the substance used as a fuel to burn in oil lamps, it is known to be an important part of the
425 Unanga diet (Unger, 2014), and critical for the storage of various foodstuffs (Frink and Giordano, 2015).

426 Knecht (2001, 2008) has suggested multiple times that stone bowls were used for the purpose of
427 rendering aquatic oils. And despite the absence of stone bowls in archaeological sites dating to later
428 phases, one ethnographic source refers to an artefact used during contact times which description
429 sounds remarkably like that of a stone bowl: *"The stone of which these lamps are made is very soft, and
430 may be hollowed out with others of greater hardness, not merely for this purpose, but also for deep pots,
431 in which they boil their fish. They use them however, but seldom, preferring mostly the iron and copper
432 kettles, which they procure from the Russians"* (Sarychev, 1806: p.73).

433

434 Another argument supporting the use of stone bowls for the rendering of aquatic oils is the residue
435 distribution on the rims but not on the bottom (Fig. 2). Oil may have been rendered by placing cuts of fat
436 in boiling water. The rendered oil could be scooped off the surface leaving the bottom of the bowl clean
437 but the rims stained. It seems probable that the oil came from marine mammals because they yield much
438 more fat than fish do and were readily available as is evident from archaeological faunal material.

439

440 **Explaining the peak in stone bowl frequencies**

441

442 The high stone bowl occurrence during the Margaret Bay phase at the Margaret Bay, and Amaknak
443 Bridge sites is remarkable. What were the driving forces behind the sudden spike in the occurrence of
444 such a specialist artefact type? It is possible that this change in stone bowl frequency is the product of a
445 sampling error. After all, the Margaret Bay (13,500 artefacts, 434 stone bowls) and Amaknak Bridge
446 (3000 artefacts, 71 stone bowls) sites were extensively excavated in comparison to sites of earlier phases
447 (e.g. Oiled Blade: 800 artefacts, 1 stone bowl). However, other sites have also seen extensive
448 investigation and yielded no evidence of stone bowls. For example the Summer Bay site where 564 m²
449 was excavated, yielding 3300 artefacts but no stone bowls, the same goes for the upper levels of
450 Tanaxtaxak, 3500 artefacts total but no stone bowls after the Margaret Bay phase (Knecht and Davis,
451 2001: p.270, Table 1). The only exception of stone bowl occurrence after the Margaret Bay phase is a
452 surface find that may or may not belong to the Late Aleutian Eider Point site (Fig. 3).

453

454 Here we explore the notion that stone bowl frequencies spike during the Margaret Bay phase and go out
455 of use after this period ends. Why did frequencies peak at this specific time? What changed?
456 Furthermore, if aquatic oil was rendered using stone bowls then why does this artefact only occur at this
457 frequency during the Margaret Bay phase (4000-3000 cal yr BP)? Assuming oil rendering was important
458 throughout the entire prehistoric and historic sequence of the Aleutian Islands, one would expect to see
459 high frequencies of stone bowls throughout the whole sequence. Were they replaced by another tool
460 type, was the method for rendering oil changed?

461
462 At around 3500 cal yr BP a cold spell ascribed to the Neoglacial brought colder temperatures to the
463 Aleutian Islands (Fig. 10). Marine mammals became more abundant as lower sea surface temperatures
464 increased marine productivity. This induced cultural expansion and increased marine mammal hunting
465 practices. Archaeological bone material is preserved for the first time in the Aleutian sequence and it
466 shows the presence of sea-ice dependent species such as polar bear, ringed seal, and walrus that are
467 not present in later phases when there is no sea-ice (Crockford and Frederick, 2007; Davis, 2001; Knecht
468 and Davis, 2008).

469
470 The increased presence of marine mammals rich in fats during this period of high marine productivity may
471 have increased the rendering of marine oil. On the other hand the unpredictability of climate change could
472 have posed problems for the rendering of oil using a cold method where pieces of fat were stored in a
473 cleaned seal skin, referred to as a *seal poke*, and left to slowly self-render into oil (Frink and Giordano,
474 2015). Temperature was of the utmost importance to this process. Under no circumstances was the
475 substance allowed to freeze, nor should it become too warm. Therefore it was stored in a cool and dark
476 place, often a submerged pit, to prevent the oil from becoming rancid (Frink and Giordano, 2015). Semi-
477 subterranean houses in the Aleutians dating to before 3000 cal yr BP often had subfloor storage pits lined
478 with stone slabs (Knecht and Davis, 2008). It is possible that these pits were used for this purpose.

479
480 We contend that decreasing temperatures in the Aleutians as demonstrated by faunal remains of the
481 Margaret Bay and Amaknak Bridge sites (Davis, 2001; Crockford and Frederick, 2007), may have

482 induced a change in the method for rendering oil. The hot rendering of oil is not only more controlled but
483 also quicker. Although stone containers could be used for cold rendering as well, for hot rendering the
484 use of a durable container such as a stone bowl was a necessity. Fat was cut up and boiled in water
485 using a container, either by means of stone boiling, or by heating the container directly over a fire. The
486 latter seems to have been the case for the examined specimens as evidenced by thick carbonated
487 encrustations stuck to the bases of the bowls.

488

489 **The adoption of durable cooking technologies in the Circumpolar North**

490 Aleutian stone bowls and griddle stones are among the earliest durable, non-portable cooking
491 technologies in the circumpolar North. We argue here that stone bowls were used for the rendering of
492 marine fats while griddle stones were probably used for cooking food with high contributions of marine
493 oils. But why were they adopted in the first place? Were stone bowls unique in light of their function?
494 Were they replaceable? How does the adoption of stone bowls relate to the wider debate of early durable
495 container technologies in the circumpolar North?

496

497 Stone bowls were a means to an end, to render marine oil. Alternative methods could be used to reach
498 that same end. The seal poke system allowed for the rendering of fat into oil using a cold method that
499 could have been employed before and after the period when bowls became abundant. Climate change
500 may have made this system to render oil more prone to failure and induced the introduction of the stone
501 bowl. However, sedentism also plays an important role here. It was sedentism that allowed for the
502 manufacture and maintenance of this expensive container technology. The fact that a lot of early pottery
503 is associated with the processing of aquatic resources (Jordan and Zvelebil, 2009; Craig et al., 2013) may
504 be closely linked to the notion that a stable abundance of aquatic resources induced sedentism, instead
505 of the idea that aquatic resource processing demanded the use of durable containers such as stone
506 bowls and pottery.

507

508 That said, climatic circumstances could very well have encouraged the local invention of the stone bowl in
509 the Aleutian Islands. The hot rendering of aquatic oil would have been difficult, or even impossible with

510 different, less durable technologies such as basketry or seal pokes. Therefore we ascribe the sudden
511 peak in stone bowl frequencies to a change in oil rendering methods brought on by the Neoglacial cold
512 spell. The introduction and use of griddle stones as a cooking technology is more gradual and consistent.
513 The demand for a container or grill plate to cook on would have easily led to the utilization of these flat
514 stones that are abundant in the Aleutian Islands.

515

516 **CONCLUSIONS**

517 In this paper our aim was to identify the function of stone bowls and griddle stones. We argue that stone
518 bowls were used for the rendering of marine oil using direct heating. We ascribe the sudden peak in stone
519 bowl occurrence to a shift in temperature caused by the Neoglacial cold spell that subsequently induced a
520 change in oil rendering methods and brought an abundance of sea-ice dependent species, rich in fats, to
521 the area. Our results also suggest that griddle stones were used for the processing of a slightly more
522 diverse set of resources although still predominantly marine. We ascribe this diversity to cooking
523 practices as opposed to oil rendering in stone bowls.

524

525 This is the first systematic research into the function of Aleutian cooking technologies employing
526 molecular and chemical analysis of carbonised residues. Future work could test the hypotheses raised in
527 this paper by analysing a larger set of samples, especially regarding griddle stones to more firmly
528 establish trends. The differences between the two technological groups are minor but apparent.
529 Differentiation within the aquatic spectrum is still poorly understood and advances in experimental
530 techniques involving APAA isomer ratios and phytanic acid ratios (Fig. 6), but also an expansion of
531 archaeological reference data from, for example, bone lipids, are necessary to further understand the
532 origin of varying signals.

533

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689 **List of Tables + captions:**

690 Table 1: Prehistory of the Eastern Aleutian Islands: dates, site characteristics, subsistence trends
691 and climatic influences. Based on: 1: Davis et al, (2016), 2: Hatfield (2010), 3: Maschner (2016), 4: Mason
692 (2001), 5: Magny and Haas (2004), 6: Knecht and Davis, 2008, 7: Corbett and Yarborough (2016)

693 Supplementary Table 1: Summary of organic residue analysis results on stone bowls and griddle
694 stones from the Aleutian Islands. Blank cells = no analysis, - = compound not present

695 Supplementary Table 2: Bone collagen data from archaeological contexts of the Aleutian Islands
696 and the Alaska Peninsula. Species were identified by ZooMS at the University of York for the following
697 samples: 201, 204-5, 209-10, 310-12, 316, and 319.

698 **List of Figures + captions**

699 Figure 1: map of the Aleutian Islands, Alaska Peninsula and Kodiak Island with emphasis on site
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701 Tanaxtaxak), Umnak Island (Anangula Blade), and Carlisle Island (Ulyagan).

702 Figure 2: Example of two different types of stone bowls fashioned out of differing textured volcanic
703 tuff and varying in size and shape: a) UNL48-57: red, more crude textured tuff, thick rim, thin base; b)
704 UNL50-51: sand colored fine tuff, finely ground both inside and out c) UNL50-51: base with carbonized
705 encrustations (Photographs by M. Admiraal, courtesy of the Museum of the Aleutians).

706 Figure 3: Stone bowl with encrustation on the interior. Surface find from Eider Point site (UNL-19)
707 probably dating to the Late Aleutian phase (1,000-2,000 cal BP) (Photograph by M. Admiraal, courtesy of
708 the Museum of the Aleutians).

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712 Figure 5: a typical total ion current (TIC) of an acid/methanol extract of a stone bowl from the

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721 downward triangles) (Bell et al., 2007; Outram et al., 2009; Craig et al., 2011, 2013; Debono Spiteri, 2012;
722 Cramp et al., 2014; Colonese et al., 2015; Horiuchi et al., 2015; Taché and Craig, 2015; Choy et al.,
723 2016). b. GC-c-IRMS results showing isotopic values of C_{16:0} and C_{18:0} fatty acids of stone bowls (green
724 circles), griddle stones (yellow squares) and one lamp (blue triangle).

725 Figure 9: Bulk isotope results of stone bowls (green circles), griddle stones (yellow squares) and
726 lamps (blue triangles) compared to Sakhalin pottery (open diamonds) (Gibbs et al., 2017) and European
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728 2017) against archaeological bone collagen data from the Aleutian Islands and the Alaska Peninsula. The
729 collagen $\delta^{15}\text{N}$ values were adjusted by +2‰ to correct for the collagen to tissue offset in order to make
730 these values more comparable with the food crusts (Fernandes et al., 2015).

731 Figure 10: Schematic diagram of stone bowl and griddle stone relative abundance against the
732 Margaret Bay phase sea-ice presence in the Aleutian Islands as inferred by (Crockford and Frederick,
733 2007) on the basis of archaeological faunal assemblages.