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Pottery use by Early Holocene hunter-gatherers of the Korean Peninsula closely linked with the exploitation of marine resources

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S.S. and O.E.C. designed the research; S.S., A.L. and O.E.C. performed the research; J-H.A. and C-J.H. contributed contextual information to aid interpretation; S.S., A.L. and O.E.C. analyzed data; S.S., A.L. and O.E.C. wrote the paper; all authors were involved in reviewing the manuscript.

Abstract

The earliest pottery on the Korean peninsula dates to the early Holocene, notably later than other regions of East Asia, such as Japan, the Russian Far East and Southern China. To shed light on the function of such early Korean pottery and to understand the motivations for its adoption, organic residue analysis was conducted on pottery sherds and adhered surface deposit on the wall of pottery vessels (foodcrusts) excavated from the Sejuk shell midden (7.7-6.8ka calBP) on the southeastern coast and the Jukbyeon-ri site (7.9-6.9ka calBP) on the eastern coast of the Korean peninsula, that represents the earliest pottery assemblages with reliable radiocarbon dates. Through chemical and isotopic residue analysis, we conclude that the use of pottery at these sites was oriented towards marine resources, supported by lipid biomarkers typical of aquatic organisms and stable carbon isotope values that matched authentic marine reference fats. The findings contrast with other archaeological evidence, which shows that a wider range of available food resources were exploited. Therefore, we conclude pottery was used selectively for processing aquatic organisms perhaps including the rendering of aquatic oils for storage. Early pottery use in Korea is broadly similar to other prehistoric temperate hunter-gatherers, such as in Japan, northern Europe and northern America. However, it is also notable that elaborately decorated red burnished pottery excavated from isolated location at the Jukbyeon-ri site had a different usage pattern, which indicates that division of pottery use by vessel form was established even at this early stage.

Keywords
Eastern Asia, Lipid residue analysis, Aquatic biomarkers, Specific-compound stable isotope analysis, Ceramic vessels, Coastal Adaptation

Introduction

In East Asia, multiple and separate origins for pottery have been proposed, including southern China, Russian Far East and the Japanese archipelago (Keally et al. 2004), all dating to the Pleistocene and therefore significantly prior to ceramic production in any other part of the world (Jordan et al. 2016). Within this context, the Korean peninsula is notable for the later arrival of pottery which appeared only at the end of the early Holocene (Choe & Bale 2002), at around the 6th millennium BC, presumably via cultural transmission from one or more of these other regions. To shed light on the function of such early Korean pottery and to understand the motivations for its adoption, here we present the results of chemical and isotopic analysis to determine vessels contents. More specifically, we examine whether pottery had a general function and therefore dispersed as an 'adaptive' technology or a more restricted range of uses and therefore as a more specialized technology. The latter has been suggested for late Pleistocene ceramic vessels (Craig et al. 2013; Lucquin, Gibbs et al. 2016) reflecting motivations for pottery innovation. Therefore it is important to know whether a relaxing of function aided the later dispersal of ceramic technology to new regions as some have hypothesised (Jordan & Zvelebil 2009) or whether early pottery in Korea follows the same pattern as other parts of East Asia.

The Korean Neolithic (or *Chulmun* named after the pottery type name) is defined by the appearance of pottery (Kim 1986). Unlike the Western Neolithic complex, this key innovation is not synchronous with the appearance of domesticated plants and animals, sedentism or even polished stone tools. Animal husbandry in this area started much later, during the second half of the first millennium BC (Lee 2013). Domesticated plants appeared in the 4th millennium BC (Crawford & Lee 2003), represented by two types of millet—foxtail (*Setaria italica* ssp. *italica*) and broomcorn (*Panicum miliaceum*), accompanied with azuki bean (*Vigna angularis*) and soybean (*Glycine max*) (Lee 2011). Recent analysis of seed

impressions left imprinted on pottery vessels during their manufacture has tentatively suggested that millet may have been introduced as early as the Initial Chulmun (Obata & Manabe 2014) and therefore contemporary with the early adoption of pottery. However, the date of these vessels is disputed on typological grounds with the growing suspicion that they belong to the Middle to Late Chulmun (Kim 2016). So far there is no conclusive evidence showing that the adoption of pottery is in anyway linked to the introduction of crop cultivation in this part of the world.

Previously, Korean pottery was thought to be dated to as early as 11k calBP based on the typological comparison of the vessels from Gosan-ri on Jeju island, 50km south of the peninsula, with well dated Russian and Japanese assemblages (Kang 2006). However, recent re-dating indicates that this site is in fact much younger; ca. 9.5k calBP (Jeju Cultural Heritage Institute 2014). Interestingly, there is a lack of such early Holocene pottery on the Korean peninsula. The earliest securely dated pottery assemblages on the Korean peninsula are *Yunggimun* (clay strip decorated) vessels that appear widely (Figure 1) from ca. 7.8 to 6.4k calBP (Initial Chulmun), although pottery has been reported from pre-Yunggimun layers (Busan University Museum 1994) but their actual date requires confirmation.

As many early pottery sites on the Korean peninsula (Figure 1) are found in coastal locations, coinciding with the earliest shell middens, a reasonable hypothesis is that pottery was part of a broader maritime adaptation at this time. Sea-level on the east coast of the Korean peninsula stabilized during the period of pottery adoption 8-7ka calBP (Jang and Park 2001, p.143) which also broadly corresponds to the Holocene climate optimum and the stabilization of oak forests (Park et al. 2012). As a consequence of sea level change, it is quite possible earlier coastal sites, perhaps bearing pottery, are now submerged although the absence of any evidence for pottery even at earlier inland locations implies that the late adoption is a more reasonable interpretation (Lee 2011).

Stable isotope analysis of human diets also show that the Chulmun hunter-gatherers were heavily dependent on marine resources as a source for their dietary protein, especially individuals buried on the south coast of the Korean peninsula (An 2006; Choy & Richards 2010; Choy et al. 2010; Choy et al. 2012; Shin et al. 2013). However, this technique is insensitive to low protein plant foods, unless they are consumed in great quantity and the analysis of the faunal and botanical remains from these sites generally show that broad spectrum of food resources was exploited despite the coastal settings (Ahn et al. 2007). In any case, neither the faunal remains nor stable isotope data necessarily relate to the use of pottery, especially if used highly selectively. The most promising approach to reveal pottery contents, and thus direct insight into the motivation for its adoption, is through organic residue analysis. While this technique has been applied widely to early pottery assemblages in East Asia (Craig et al. 2013; Lucquin, Gibbs et al. 2016), much less attention has been paid in Korea with only three studies of much later prehistoric material so far undertaken (Kwak & Marwick 2015; Heron, Shoda et al. 2016; Kwak et al. 2017).

Here we targeted both ceramic matrices and adhering surface deposits (hereafter foodcrust) from the Sejuk shell midden site on the southeast coast and the Jukbyeon-ri site on the east coast of the Korean peninsula, dated to ca. 7.7-6.8k calBP and 7.6-6.9k calBP, respectively. Pottery from these sites represent the earliest examples on the Korean peninsula. Moreover, the geographical location (Figure 1) and nature of the sites, i.e shell midden vs. open site, provide a basis to examine the variability of the earliest pottery phases for this region.

Materials

The Sejuk shell midden

Sejuk, one of the earliest known shell midden sites in Korea, is located at an inner bay on the southeast coast of the Korean peninsula (Figure 1); 35°27′50″N, 129°21′15″E. An area of approximately 10,000 square metres was excavated in 2000. The stratigraphy was established which divides the midden into three cultural layers. However, the evidence from pottery typology and radiocarbon dates show that the materials were deposited very quickly and probably belonged to a continuous singular phase (Ahn et al. 2007).

A large amount of ecofacts have been recovered from this site supporting a broad subsistence base. Seed and fruit remains are represented by wild fruits such as *Acitinidia*, *Rubus* and *Vitis*, nuts like *Quercus* and *Styrax*. Animal remains are represented both by the terrestrial and aquatic remains. These include terrestrial (*Sus scrofa*, *Cervus nippon hortulorum*, *Hydropotes inermis*) and sea mammals (*Zalophus californianus japonicus*), birds (*Gavia* sp., *Phalacrocorax filamentosus* and *Phalacrocorax pelagicus*), marine fish (*Pagrus major*, *Thunnus* sp. and *Mugilidae* sp.), and shells (*Mytilus coruscus*, *Litiopidae sp.*, *Ruditapes philippinarum*, *Crassostrea gigas*, *Lunella coreensis*, *Cyclina sinensis*). Pollen analysis of sediments showed the dominance of *Quercus* in the surrounding forest accompanied with other deciduous broadleaf trees as well as *Pinus* (Ahn et al. 2007), which corresponds to the remains of nuts and terrestrial mammals in the archaeological deposits. Other than pottery, bone/stone fishing hooks and stone arrowheads have been excavated, which also support the interpretation that a wide range of food resources were exploited.

In total, we collected 30 pottery sherds from Sejuk. Each sherd was associated with a foodcrust (Figure 2) adhering to either the interior or the exterior rim, seemingly as a result of food boiling over during cooking. Such charred surface deposits are a feature of early pottery from this region. In total, 29 potsherds and 33 foodcrusts (12 exterior and 21 interior samples) were selected for the analysis (Table 1). It was difficult to find any significant difference of size or volume of pottery vessels based on relatively small size of the fragments.

The Jukbeyon-ri site

The Jukbyeon-ri site, a waterlogged non-shell midden site, is located at the top of a cape on the east coast of the Korean Peninsula (Figure 1). Four locations at the site were excavated by the Samhan Institute of Cultural Properties from 2009 to 2013, as well as general surveys for the surrounding areas to find two more Chulmun sites in this area (Figure 3). Significant amount of pottery and stone tools including hunting, fishing and plant processing tools are recovered from these locations while no ecofacts were reported other than a putative wooden dugout and other timbers (The Samhan Institute of Cultural Properties 2012; 2015).

The samples are from the two locations at the Jukbyeon-ri site; 1) *Location Jung-ro 3-3*; 37° 03′ 28″ N, 129° 25′ 42″ E and 2) *Location 15-68*; 37° 03′ 32″ N, 129° 25′ 37″ E. The excavation areas are 2,870 and 140 square metres, respectively. It is noteworthy that Jung-ro 3-3 is located in commanding position facing the ocean compared to 15-68 (Figure 3). Although broadly contemporaneous (Table S1), Jung-ro 3-3 contains much higher frequency of pottery decorated with a red pigment slip, so-called ‘red burnished pottery’ (Figure 4A). Whereas at location 15-68, the *Yunggimun* style coarse pottery (Figure 4B) is dominant, very similar to the coarse wares from the Sejuk site (Samhan Institute of Cultural Properties 2015: 101). The different styles of pottery found at Jukbyeon-ri are themselves intriguing and implies that a diverse ceramic repertoire was adopted by hunter-gatherers of this region. Indeed,

"the Jukbyeon-ri style pottery" is viewed as a separate assemblage in this period, based on the difference from *Yunggimun* style, that are distributed in the southern area (Lim 2012). It is unknown whether these different pottery typologies relate to a difference in function.

Twenty two pottery sherds and four foodcrust were sampled from the Location Jung-ro 3-3 and 33 pottery sherds and eight foodcrust were sampled from the Location 15-68. These consist of plain pottery with coarse clay (type CW in the Table S5) and red burnished pottery with fine-clay (type RBW). Rim, body, bottom of vessels, as well as the attached 'knobs' were selected as samples and the last is especially used as blank control to ensure that extracted lipids are originated from contents of pottery rather than surrounding soils as has been already discussed (Heron et al. 1991). The details of these samples are shown in Table S4 and S5.

Methods

Bulk Isotope analysis of foodcrust samples

Thirty three foodcrust samples from Sejuk and twelve from Jukbyeon-ri were analyzed by elemental analysis - isotope ratio mass spectrometry (EA-IRMS). The samples were crushed into fine homogenised powder (~2mg) and duplicated into two tin capsules. Their bulk stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotope value were determined based on the method previously described (Craig et al. 2007). Samples with less than 1% nitrogen were omitted considering reliability of measurement. Precision of instrument on repeated measurement was ±0.2‰ (s.e.m.), $\delta^{13}C$, $\delta^{15}N$ = [(Rsample/Rstandard−1)] × 1,000, where R = $^{13}C/^{12}C$ and $^{15}N/^{14}N$. All values are expressed in per mill (‰) relative to the standards, Vienna PeeDee Belemnite for $\delta^{13}C$ and air N_2 for $\delta^{15}N$, respectively.

Lipid residue extraction

Lipids were extracted from 29 pottery sherds and 30 foodcrusts from Sejuk and 55 sherds and 12 foodcrusts from Jukbyeon-ri. Lipid extraction and methylation were conducted in one-step according to established method (Correa-Ascencio & Evershed 2014). In short, methanol was added to the powdered samples (pottery sherd powder: 4ml to 1g, foodcrust: 1ml to 10-30mg) and the mixture was sonicated for 15 minutes followed by acidification with concentrated sulphuric acid (800µl and 200µl, respectively). The sealed acidified samples were heated at 70°C for four hours then cooled to the room temperature. Lipids were extracted from centrifuged samples with *n*-hexane (3 × 2mL) and directly analyzed by GC, GC-MS and GC-c-IRMS.

Gas Chromatography Mass Spectrometry

GC-MS analysis was undertaken using an Agilent 7890A series chromatograph attached to an Agilent 5975C Inert XL mass-selective detector with a quadrupole mass analyser (Agilent technologies, Cheadle, Chershire, UK). A splitless injector was used and kept at 300°C. The GC column was inserted into the ion source of the mass spectrometer directly. Helium was used as the carrier gas, its flow was constant at a rate of 3mL min^{-1}. The ionisation energy of the MS was 70eV and spectra were obtained by scanning between *m/z* 50 and 800.

A DB-5ms (5%-phenyl)-methylpolysiloxane column (30m × 0.250mm × 0.25µm; J&W Scientific, Folsom, CA, USA) was used for scanning and SIM. For scanning, the temperature was set at 50°C for 2 minutes, then raised by 10°C min^{-1} until it reached 325°C where it was held for 15 minutes. The same

chromatographic conditions were used in SIM mode. The first group of ions (*m/z* 189, 204, 231, 425, 440) corresponding to major ion fragments derived from miliacin, a pentacylic triterpenoid methyl ether found in common/broomcorn millet (Heron et al. 2016). After 16 minutes, the second group of ions (*m/z* 57, 71, 85, 478, 506) were detected to record internal standard (n-hexatriacontane).

All extracts were also analysed on a DB-23 (50%-Cyanopropyl)-methylpolysiloxane column (60m × 0.250mm × 0.25µm; J & Scientific, Folsom, CA, USA) to identify isoprenoid fatty acids and ω-(o-alkylphenyl) alkanoic acids as aquatic biomarkers (Cramp & Evershed 2014) and to resolve the mixture of phytanic acid diastereomers (Lucquin, Colonese, et al. 2016). The temperature was set at 50°C for 2 minutes, 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 min^{-1} to 160°C , then by 20°C min^{-1} to 250°C where it was maintained for 10 minutes. The first group of ions (*m/z* 74, 87, 213, 270) corresponding 4,8,12-trimethyltridecanoic acid (TMTD) fragmentation, the second group of ions (*m/z* 74, 88, 101, 312) corresponding to pristanic acid, the third group of ions (*m/z* 74, 101, 171, 326) corresponding to phytanic acid and the fourth group of ions (*m/z* 74, 105, 262, 290, 318, 346) corresponding to *ω*-(*o*-alkylphenyl) alkanoic acids of carbon length C_{16} to C_{22} were monitored, respectively. The carrier gas used was helium with a flow rate of 2.4mL min^{-1}. The relative abundance of two diastereomers of phytanic acids is obtained by the integration of the ion *m/z* 101.

Gas Chromatography combustion Isotope Ratio Mass Spectrometry

Stable carbon isotope values of methyl palmitate ($C_{16:0}$) and methyl stearate ($C_{18:0}$) derived from precursor fatty acids were measured by GC-c-IRMS, following existing procedure (Craig et al. 2012). An Isoprime 100 (Isoprime, Cheadle, UK) linked to a Hewlett Packard 7890B series GC (Agilent Technologies, Santa Clara, CA, USA) with an Isoprime GC5 interface (Isoprime Cheadle, UK) was used for the analysis. One microlitre of each sample was injected into DB-5MS ultra-inert fused-silica column. The temperature was set at 50°C for 0.5 minutes and raised by 0.5°C min^{-1} to 50°C, then raised by 10°C min^{-1} to 300°C where it was held for 10 minutes. The carrier gas used was ultra high purity grade helium with a flow rate of 3 mL min^{-1}. The gas flows eluting from the column were split into two streams. One was directed into an Agilent 5975C inert mass spectrometer detector (MSD), for the sake of sample identification and quantification, while the other was directed through the GC5 furnace kept at 850°C to oxidise all the carbon species to CO_2. A clear resolution and a baseline separation of the analysed peaks were achieved.

Eluted products were ionized in the mass spectrometer by electron impact and ion intensities of *m/z* 44, 45 and 46 were recorded for automatic computing of the $^{13}C/^{12}C$ ratio of each peak in the extracts. Computation was made with IonVantage and IonOS softwares (Isoprime, Cheadle, UK) and was based on comparisons with standard reference gas (CO_2) of known isotopic composition that was repeatedly measured. The results of the analysis were expressed in per mill (‰) relative to an international standard, V-PDB.

The accuracy and precision of the instrument was determined on *n*-alkanoic acid ester standards of known isotopic composition (Indiana standard F8-3). The mean ± S.D. values of these were -29.82 ± 0.16‰ and -23.28 ± 0.19‰ for the methyl ester of $C_{16:0}$ (reported mean value vs. VPDB -29.90 ± 0.03‰) and $C_{18:0}$ (reported mean value vs. VPDB -23.24 ± 0.01‰) respectively. Each sample was measured in replicate (mean of S.D. 0.11‰ for $C_{16:0}$ and 0.10‰ for $C_{18:0}$). Values were also corrected subsequent to analysis to account for the methylation of the carboxyl group that occurs during acid extraction. 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 under identical conditions.

Results and Discussion

Evidence for the processing of marine resources

Several complementary lines of evidence point to the processing of marine resources in many of the pottery vessels from the Sejuk shell midden and Jukbyeon-ri sites.

Firstly, with just a single exception discussed below, the range of $\delta^{15}N$ values (9-13‰; median = 11.0‰) obtained from bulk isotope analysis of foodcrusts from both sites are similar to tissues measured in modern marine fish, and shellfish from Japan (Yoneda et al. 2004) and foodcrusts on ceramics from the coastal Jomon site of Torihama strongly implicated with the processing of aquatic resources (e.g. Lucquin, Gibbs et al. 2016; Fig 5A). Interestingly, $\delta^{13}C$ values (-21 to -27‰; median = -23.2‰) from the Korean foodcrusts are generally more enriched in ^{13}C compared to those from Torihama (Fig 5A), perhaps reflecting a greater preference for processing marine products. However, these values are difficult to compare with modern fish due to preferential survival of the ^{13}C depleted lipid components compared to proteins and carbohydrates (Heron & Craig 2015). Some thermal alteration to the isotope values may also be expected. A comparison of two exterior deposits (USJ11, USJ18) with their respective interiors showed a slight enrichment in ^{15}N (1.1‰ and 1.5‰ respectively, Table 1) similar in magnitude to experimentally charred seeds (Nitsch et al. 2015). These deposits most likely formed when the vessel's contents spilled over the rim and became exposed directly to the fire.

Secondly, diagnostic compounds for aquatic foods, including isoprenoid alkanoic acids and ω-(*o*-alkylphenyl) alkanoic acids (APAAs) containing 18 to 22 carbon atoms (Hansel et al. 2004; Evershed et al. 2008) were observed in many of the samples analysed by GC-MS (Table 1; Figure 6, Table S2,S3,S4,S5). At Sejuk, 61% of potsherds and 62% of the foodcrust which contained interpretable amounts of lipids (>5μg g^{-1} for pottery sherds and >100μg g^{-1} for foodcrust) had APAAs of carbon length C_{18}-C_{22}. Generally the GCMS results showed good correspondence between foodscrusts and ceramic matrices at Sejuk, although there were some discrepancies. Notably the isoprenoid alkanoic acids were observed more frequently in the ceramic samples compared to foodcrusts. This attributable to differential preservation of these lipid classes in the different sample types. At Jukbyeon-ri, where the lipid preservation in the potsherds was generally poorer, 26% of the potsherds and 89% of the foodcrusts containing lipids yielded these markers. APAAs are formed only by protracted heating of mono- and polyunsaturated fatty acids at high temperature (Hansel et al. 2004) which supports the inference that these compounds are formed through pottery use, rather than migration of lipids from the post-depositional environment. Even the samples that do not satisfy the full criteria of aquatic oil identification, features typical of degraded aquatic oils were also observed. A further 10 potsherds and 4 foodcrust had partial aquatic biomarkers, that is, C_{18} APAAs accompanied by at least one isoprenoid. Alone however this analysis is not sufficient to distinguish marine or freshwater fish, aquatic mammals or shellfish.

Thirdly, the ratio of the two natural diastereomers of phytanic acid, 3S,7R,11R,15-phytanic acid (SRR) and 3R,7R,11R,15-phytanic acid (RRR), were determined by GC-MS operating in SIM mode. Phytanic acid is found in both aquatic and ruminant animals but a higher SRR/RRR ratio (SRR%) is characteristic of aquatic organisms (Lucquin, Colonese, et al. 2016; Schröder & Vetter 2010). Of the samples that contained phytanic acid, over 90% had SRR% within the distribution of the modern fish oils analysed (Figure 7). However, as the range of SRR% values of the fish oil overlaps with ruminant fats (Figure

7), a proportion of sherds cannot be securely assigned to either source. Nevertheless, over 80% samples from Sejuk and 90% from Jukbyeon-ri had SRR% greater than 75% of the modern ruminant fats sampled whilst 54% from Sejuk and 85% from Jukbyeon-ri had values greater than 95% of the ruminant fats sampled, indicating that the origin of phytanic acid extracted from the majority of samples is from an aquatic rather than ruminant source.

Finally, GC-c-IRMS was used to investigate the origin of the most abundant saturated fatty acids ($C_{16:0}$ and $C_{18:0}$) based on their $\delta^{13}C$ values. The result of GC-c-IRMS analysis for 18 samples of foodcrusts and 59 samples of potsherds shows that the majority of vessels have fatty acid isotope values matching authentic oils extracted from marine products, including salmonids feeding in the marine environment (Figure 8A). It is interesting that there is very little evidence of terrestrial animal fat in the pottery samples considering that terrestrial animal bones such as wild boar (*Sus scrofa*), Sika deer (*Cervus nippon hortulorum*) and Korean water deer (*Hydropotes inermis*) have been recovered from the Sejuk and Hwangsong-dong site nearby (Choi 2012). Presumably these animals were prepared in other ways that did not involve ceramic vessels. Where both a foodcrust and potsherd were measured, there was very little difference in the carbon isotope measurements of extracted fatty acids (typically less than 1‰, Table S2,S4). Lipids absorbed within the walls of the potsherds form over multiple cooking 'events', while foodcrusts generally represents the last cooking event (Mukherjee et al. 2008), this implies that the vessel was used in similar manner throughout their use history.

It is not possible to do determine the types of marine resources exploited in the vessels especially considering a wide range of fish (tuna, mullet and bream) and marine mammals were found at Sejuk and other Initial Chulmun sites, including whale (Choi 2012). Shellfish also cannot be ruled out, although the $\delta^{15}N$ values of foodcrusts are too elevated to be consistent with the authentic tissue values so far analysed (Yoneda et al. 2004). As the $\delta^{13}C$ values of the bulk sample and of the fatty acids ($C_{16:0}$ and $C_{18:0}$) are highly correlated (Pearson R= 0.66 and 0.65, respectively) and very similar (Table S2,S4), we suggest that the foodcrusts are largely derived aquatic oils, rather than degradation of other food components, such as proteins, which are relatively enriched in ^{13}C.

Interestingly the atomic C:N ratios of the Korean foodcrusts (range = 7-30; median = 13.3; Figure 5B, Table S2,S4) are higher than those from Torihama in Japan (Figure 5B), especially in the Sejuk assemblage. Since many of these samples have high $\delta^{15}N$ values and lipid profiles typical of aquatic products it is tempting to relate these organic signatures specifically to the production and processing of marine oils. Similar bulk isotope and element characteristic have been observed in ceramic vessels with typologies associated with the burning of oils for illumination (Heron et al. 2013). Selective loss of proteinaceous compounds during burial leading to a relative enrichment in ^{13}C depleted lipids may produce a similar signal but it is interesting to note that such characteristics were not observed in the much larger sample from Torihama. The partial rendering oils from marine fish or mammals to provide high-energy greasy soups, or even storable commodities for exchange or gifting, as documented in historic coastal hunter-gatherers (Kuhnlein et al. 1982), would have been difficult to achieve without advanced container technology, such as pottery.

Variation in pottery use by location and vessels type

At Jukbyeon-ri there was an opportunity to compare pottery use by vessel function and the location of deposition. Whilst the majority of residues from this site had fatty acids $\delta^{13}C$ values indicative of marine foods (Fig 8C and D), a number of pots from the Jung-ro 3-3 location, including all the red-burnished wares (RBWs) had more depleted values (Fig 8D) consistent with a greater input of terrestrial foods or

freshwater fish, although the latter is less likely given the site's coastal setting. The range of concentration observed in RBWs is more restricted than in the coarsewares (CWs; values) where some lipids rich samples have been founds. None contained APAAs produced by thermal alteration, indicative of a different use perhaps as storage containers. Such complex culinary practices are remarkable during the initial phase of pottery adoption. It seems that foods were carefully separated and specific pottery vessels were selected for their preparation. It is difficult to explain this practice in purely functional terms and it is more likely linked with specialised activities, such as elaborate feasting, as previously hypothesised as a general motivation for pottery adoption and innovation (Hayden 1995). This is reinforced by the prominent location of Jung-ro 3-3 in the coastal landscape and the importance of the site for producing valuable (granite-gneiss) raw materials for lithic manufacture that were distributed to the southern coastal areas of the peninsula (Hwang 2016).

In addition, ergostanol, hydrogenation by-product of ergosterol, a sterol specific to fungi (Isaksson et al. 2010), was identified as a noticeable component in extracts from four of the coarseware pottery (JBR34, 37, 40 and 45) sherds from loc. Jung-ro 3-3 whilst it was absent in pottery from loc. 15-68 and at Sejuk. Fungal lipids are surprisingly rarely encountered in organic residue analysis of ceramics and it has been suggested they are derived from deliberate fermentation of sugars rather than from the burial environment (Isaksson et al. 2010). The preservation of seasonally available wild plants foods through fermentation would have been a highly attractive strategy for hunter-gatherers but this hypothesis is tentative given that fungal growth may occur following discard of the pottery and that, in this case, the marker is associated with animal rather than plant derived lipids. A further source from ergosterol could be directly from edible fungi that were surely available to temperate boreal hunter-gatherers.

Evidence for plant processing?

Despite the abundance of macro-plant remains at Sejuk, the evidence for plant processing in pottery is extremely scant. One foodcrust (USJ02F) was relatively depleted in ^{15}N (1.5‰) and ^{13}C (-26.8‰) and had the highest C:N ratio (57.6; Figure 5); characteristics comparable with measurements on charred acorns (Kudo 2014) and charred plant deposits from Japanese Jomon sites (Heron, Habu et al. 2016). The $\delta^{13}C$ values of fatty acids from this vessel were also relatively depleted compared to all others; ($\delta^{13}C_{16:0}$ = -31.0‰ and $\delta^{13}C_{18:0}$ = -31.4‰; Figure 8B, Table S2) and no biomarkers associated with aquatic foods were identified. These values are consistent with a plant source and this interpretation is supported by the presence of visible fiber-like remains encased with the charred crust. Furthermore, the radiocarbon date of this sample was much younger than those obtained from pots with aquatic biomarkers and more enriched fatty acid $\delta^{13}C$ values (Table S2). This discrepancy in dates is most likely explained by the incorporation of 'old' carbon from the marine reservoir, lending further compelling support to the presence of marine products in the majority of pottery vessels.

Miliacin, a compound found in broomcorn millet, *Panniacum millacin* (Heron, Shoda et al. 2016) was absent in all the samples analysed. This contrasts with evidence of millet seed impressions on *Yunggimun* pottery sherds (Obata & Manabe 2014) and with the results from the Bronze Age Korean site of Majeon-ri (ca 2.8-2.5k calBP), where seven from fifteen potsherds analysed contained miliacin (Heron, Shoda et al. 2016). Although, an absolute absence of plants in the early Korean pottery cannot be inferred, particularly due to the low N and lipid content of many wild plant species available at this time, so far there is little evidence to support the notion that plant processing was a major driver for the adoption of pottery in Korea or indeed elsewhere in East Asia (Lucquin, Gibbs et al. 2016; Craig et al. 2013).

Motivations for early pottery production

The motivations for hunter-gatherer pottery invention, innovation and uptake are still debated. Previously, it has been hypothesised that pottery production was promoted by the anticipation of seasonally abundant aquatic resources (Taché & Craig 2015), a feature typical of delayed-return economies with storage technologies (Jordan 2014, p.127). In fact, the strong association between pottery use and aquatic resources is not unique at these sites, period or region and is a common feature of the early phases of Northern hunter-gatherer pottery use, with similar findings in Japan (Craig et al. 2013), Northern Europe (Craig et al. 2007; Oras et al. 2017), North America (Taché & Craig 2015), although the initial results from North African Early Holocene pottery suggest a different adaptation (Dunne et al. 2016). It is reasonable to conclude that similar motivations for the adoption of pottery by other Northern Eurasian and American hunter-gatherers were at play, perhaps related to the intensive and cooperative exploitation of marine resources at sites like Sejuk and Jukbyeon-ri. However, unlike Japan, where pottery first appeared during a cool period (Kawahata et al. 2017) when terrestrial resources were scarce, the arrival of pottery in Korea corresponds to a climate optimum with widespread deciduous forests when terrestrial resources were plentiful. The stabilization of coastlines and the appearance of shell middens provides the only indication that the earliest pottery in Korea was linked with environmental change, although the loss of earlier coastal sites to rising sea-levels makes it difficult to assess how 'novel' such environments were.

Whilst we can't speculate on the 'value' of pottery to prehistoric hunter-gatherers, economic or otherwise, the strong association with aquatic oils despite the availability of other terrestrial and plant resources (Ahn et al. 2007; Choi 2012) indicates that their use was highly specialized. On the Korean peninsula, early pottery is implicated in the production of oily marine foods that were set apart from utilitarian culinary activities or else ceramic containers were embedded in routine subsistence practices, but as specialized 'tools'. More detail of the scale of production and context of use is needed to distinguish these scenarios both in Korea and more generally (Cohen 2013). At Jukbyeon-ri, there are indications that pottery had a more elaborate role within hunter-gatherer society linked with the use of space and involving specific vessel forms. But for the most part, the specialized use of pottery for processing aquatic foods was transmitted along with knowledge of pottery production, as part of the same cultural tradition.

Finally, the precise cultural influences and processes which stimulated the adoption of ceramic container technology in the Korean peninsula are still unknown. During this period in Japan, ceramic vessels were produced at such as scale that they must have been prominent in many spheres of life, from the utilitarian to the ritual, as well as important expressions of cultural identity in their own right. Nevertheless, the albeit limited residue data available from Jomon pottery shows a strong association with aquatic foods during this period (Lucquin, Gibbs et al. 2016). The timing of adoption of pottery on the Korean peninsula corresponds with such large-scale and diversified pottery production in Japan and the widespread appearance of shell middens (Initial Jomon), however it is important to note that typological differences in pottery between Korea and Japan does not support the simple extension of this tradition. Combined organic residue analysis and radiocarbon dating of pottery from other adjacent regions dating to this period, is clearly called for to tackle this question adequately.

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Table 1. *Summarized results of lipid residue analysis of Sejuk and Jukbyeon-ri sites by this study. CW=coarseware, RBW=red burnished ware, PS=potsherds, FC=foodcrust. Aquatic biomarkers are interpreted from the presence of isomers of APAAs (C_{20} and C_{22}) and at least one isoprenoid fatty acids (pri, phy or TMTD) (Evershed et al. 2008) while partial biomarkers are interpreted when they have C_{18}APAAs with at least one isoprenoid fatty acids. Number with lipids preserved is determined by lipid concentration (>5μg g-1 for pottery sherds and >100μg g-1 for foodcrust).*

Site	Location	Typology	No samples analysed		No with lipids preserved		Aquatic biomarkers		Partial biomarkers		% aquatic biomarkers		% biomarkers including partial	
			PS	FC	PS	FC	PS	FC	PS	FC	PS	FC	PS	FC
Sejuk	All	CW	29	33	29	30	12	14	5	2	41%	47%	59%	53%
Jukbyeon-ri	Jung-ro 3-3	CW	16	4	13	4	7	4	0	0	54%	100%	54%	100%
		RBW	6	0	6	n/a	0	n/a	0	n/a	0%	n/a	0%	n/a
		All	22	4	19	4	7	4	0	0	37%	100%	37%	100%
Jukbyeon-ri	15-68	CW	27	8	20	5	2	4	1	0	10%	80%	15%	80%
		RBW	6	0	2	n/a	0	n/a	0	n/a	0%	n/a	0%	n/a
		All	33	8	22	5	2	4	1	0	9%	80%	14%	80%
Jukbyeon-ri	Total	CW	43	12	33	9	9	8	1	0	27%	89%	30%	89%
		RBW	12	0	8	n/a	0	n/a	0	n/a	0%	n/a	0%	n/a
		All	55	12	41	9	9	8	1	0	22%	89%	24%	89%
Total	All	All	84	45	70	39	21	22	6	2	30%	56%	39%	62%

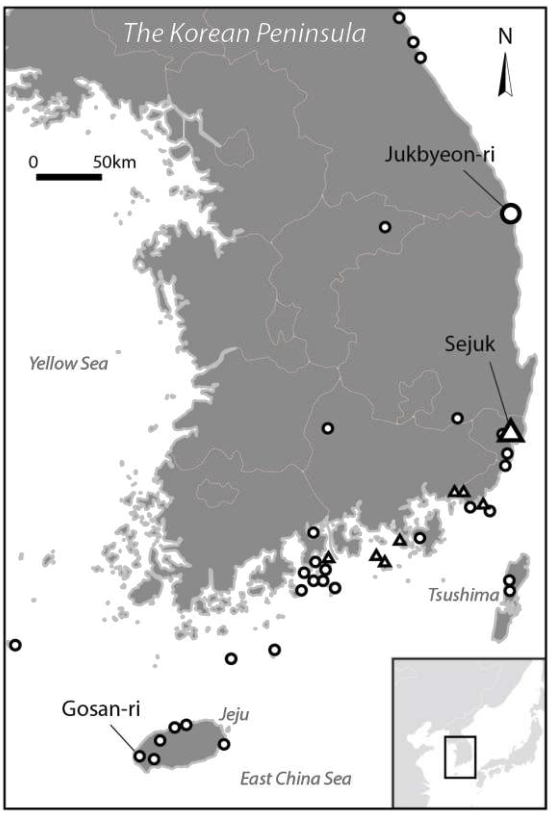


Figure 1: *A map showing the distribution of Yunggimun style and earlier pottery on and around the Korean peninsula, indicating the locations of some sites mentioned in this paper. Triangles - shell midden sites, Circles - open sites. Adapted from Dongsamdong shell midden museum 2004.*

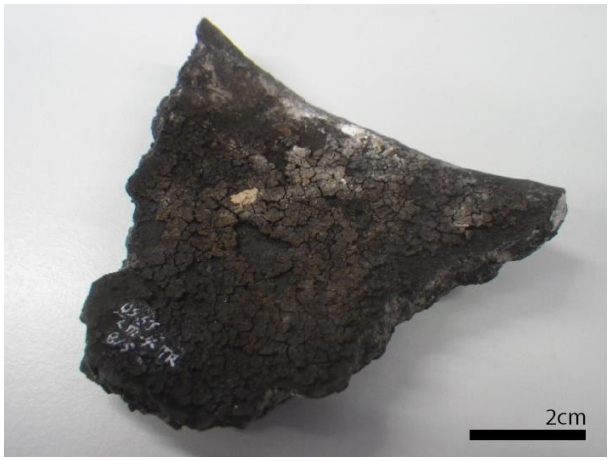


Figure 2: *An example of a typical coarseware 'Chulmun' pottery sherd with attached 'foodcrust' excavated from the Sejuk site (USJ08).*

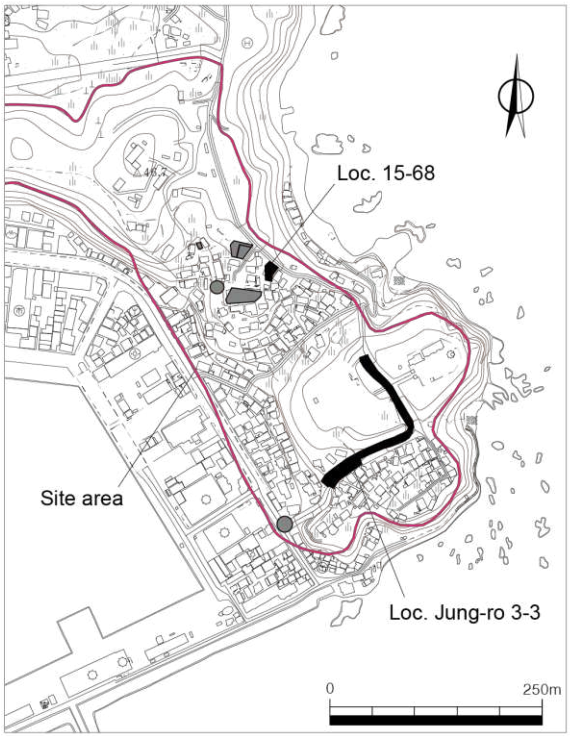


Figure 3: *A map indicating the locations and site area of the Jukbyeon-ri site. Locations from where the samples were obtained are shown in black and other locations where Chulmun assemblages were confirmed in grey.*



Figure 4: *Examples of red burnished pottery (A, height= 18.0cm) and coarse pottery (B, height= 14.8cm) from the Jukbyeon-ri site.*

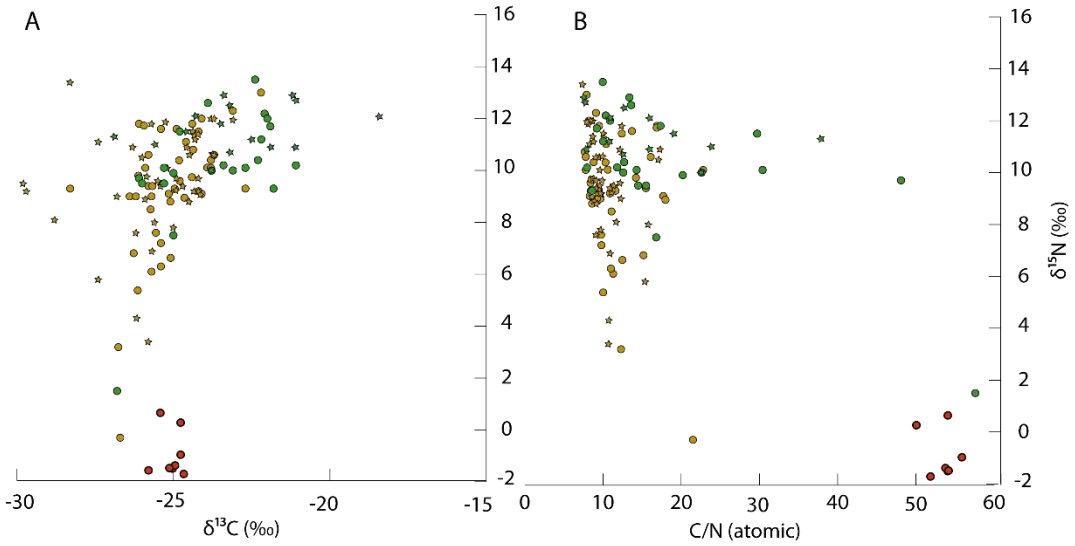


Figure 5: *Plots of the $\delta^{13}C$ to $\delta^{15}N$ values (A) and atomic C:N ratios to $\delta^{15}N$ (B) of foodcrusts from the Korean Peninsula Initial Neolithic (this study, Green), Torihama Jomon shell midden ((Lucquin, Gibbs, et al. 2016); Beige) and charred (plant) material from the middle Jomon site of Sannai Maruyama ((Heron & Habu 2016); Red). Stars indicate samples with distinctive lipid biomarkers derived from aquatic foods (Evershed et al. 2008).*

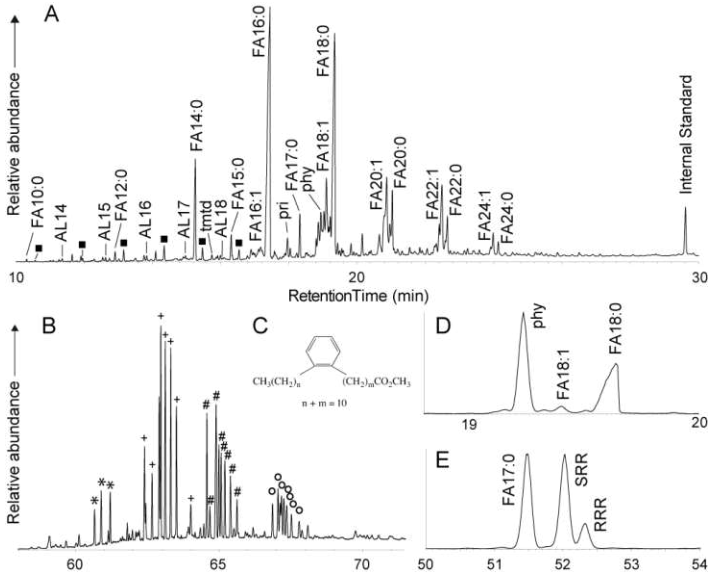


Figure 6: *Typical partial gas chromatogram of Jukbyonn-ri potsherd (JBR35) lipid extract. A, the total ion chromatogram is characteristic of heated and degraded aquatic oil and is dominated by medium- and long-chain saturated and mono-unsaturated fatty acids and isoprenoid fatty acids. α,ω-dicarboxylic acids (■) with carbon chain ranges of C_8-C_{13} were also observed and are the likely oxidation products from long-chain unsaturated fatty acids. n-hexatriacontane is used as internal standard. B, The summed m/z 105 ion chromatogram shows the presence of ω-(o-alkylphenyl) alkanoic acids with 16(*), 18(+), 20(#) or 22(o) carbon atoms. The presence of the third and fourth components are thermally produced from $C_{20:x}$ and $C_{22:x}$ polyunsaturated fatty acids found only at high abundance in aquatic organisms (Evershed et al. 2008). C, Chemical structure of APAAs (adapted from Hansel et al. 2004). D, the m/z 101 ion chromatogram shows that phytanic acid appears as a unique peak on the DB-5MS column. E, the m/z 101 ion chromatogram shows the diastereomers of phytanic acid (SRR and RRR) are resolved on the DB-23 column.*

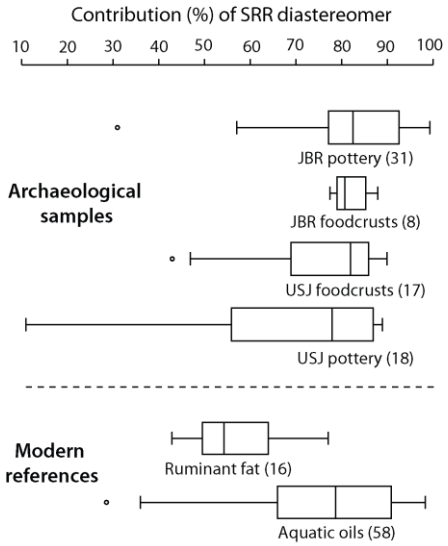


Fig 7: Boxplots of the SRR diastereomer in total phytanic acid (SRR%) extracted from archaeological pottery and foodcrusts form Sejuk (USJ) and Jukbyeon-ri (JBR) sites. These values are compared with published SRR% from modern ruminant and aquatic animals (Lucquin, Colonese, et al. 2016)

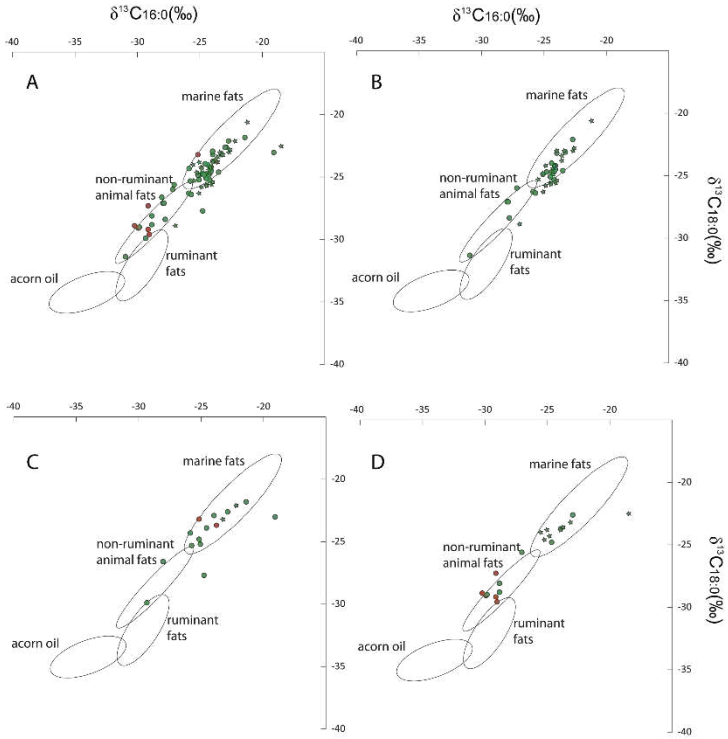


Figure 8: Plot of the $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ n-alkanoic acids extracted from foodcrusts and ceramics from coarse wares (green) and red-burnished wares (red) from the Korean Initial Neolithic. Samples containing aquatic biomarkers are marked with a star. (A) All samples; (B) Sejuk; (C) loc. 15-68 and (D) loc. Jung-ro 3-3 at Jukbyeon-ri. The data are compared with reference ranges for authentic reference lipids and archaeological bone (Craig et al. 2013; Taché & Craig 2015; Craig et al. 2012;Spangenberg et al 2006: Cramp et al. 2014; Outram et al. 2009; Colonese et al. 2015; Dudd 1999; Miyata et al. 2015, Lucquin et al 2016) plotted at 66.7% confidence.