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The impact of environmental change on the use of early pottery by East Asian hunter-gatherers

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The invention of pottery was a fundamental technological advancement with far-reaching economic and cultural consequences. Pottery containers first emerged in East Asia during the Late Pleistocene in a wide range of environmental settings but became particularly prominent and much more widely dispersed following climatic warming at the start of the Holocene. Some archaeologists argue that this increasing usage was driven by environmental factors, as warmer climates would have generated a wider range of terrestrial plant and animal resources that required processing in pottery. However, this hypothesis has never been directly tested. Here, in one of the largest studies of its kind, we conducted organic residue analysis of over 800 pottery vessels selected from 46 Late Pleistocene and Early Holocene sites located across the Japanese archipelago to identify their contents. Our results demonstrate that pottery had a strong association with the processing of aquatic resources, irrespective of the ecological setting. Contrary to expectations, this association remained stable even after the onset of Holocene warming, including in more southerly areas where expanding forests provided new opportunities for hunting and gathering. Nevertheless, the results indicate that a broader array of aquatic resources were processed in pottery after the start of the Holocene. We suggest this marks a significant change in the role of pottery of hunter-gatherers, corresponding to an increased volume of production, greater variation in forms and sizes, the rise of intensified fishing, the onset of shellfish exploitation and reduced residential mobility.

archaeology | early pottery | organic residue analysis | stable isotopes | Jōmon

The production and use of hard, fired earthen containers represents a key technological development in human history. From its prehistoric origins at the end of the last Ice Age, pottery became a fundamental tool for transforming, mixing, storing, and serving foodstuffs almost globally, and was only replaced relatively recently by metal containers. Understanding the motivations for the emergence and wider adoption of pottery is a key question in world prehistory. Ceramic vessels were first invented by hunter-gatherers in East Asia during the Late Pleistocene in Southern China, Japan and the Russian Far East (1–3) during glacial climatic conditions (*ca*. 18,000-16,000 cal BP). With climatic warming in the Early Holocene (*ca*. 11,500 cal BP), pottery was produced in much more substantial quantities and became more widely adopted (4). Organic residue analysis of East Asian early pottery (5–7) is beginning to elucidate the motivations that lay behind early pottery innovation and its more widespread adoption. However, so far there has been no systematic investigation of pottery use across the transition from the Pleistocene to the Holocene.

One of the best areas to investigate the development of ceramic technology is the Japanese archipelago due to the intensively studied sequence of hunter-gatherer pottery, known as Jōmon (meaning cord marked). The Jōmon ceramic sequences not only offer the chance to study potential continuity or change in pottery function across the Pleistocene-Holocene transition (See SI Appendix, Figs. S1 and S2) but also offer scope to explore this process in a wide range of ecological settings (Fig. 1) because the main Japanese islands span a large latitudinal range (30°N-46°N), which ranged from steppe-tundra in the north to warm evergreen broadleaf forest in the south (Fig. 1). The transition from the Pleistocene to Holocene is clearly apparent in changes to the composition (see SI Appendix, Fig. S1) and extent of the pottery assemblages, although changes in volumes and sizes are more difficult to assess due to their highly fragmented nature. Firstly and most noticeably there is a substantial, 100-fold increase in the number of sherds recovered on early Initial Jōmon (Stage 4) sites compared to Final Incipient sites (Stage 3) across the archipelago (4). This cannot simply be explained by a greater intensity of occupation through time. Even large Incipient sites, such as Kuzuharazawa IV in Shizuoka, have less than a thousand sherds where similarly sized Initial Jōmon sites, such as Nakano B in Hokkaido or Jozuka in Kyushu have yielded tens to hundreds of thousands with the ratio of potsherd to other artefacts also dramatically increasing (8). Secondly, clearly defined regional styles and manufacturing techniques emerge in the Early Holocene that are thought to reflect a greater integration of production and use.

The emergence of regional pottery styles and greater scale of production corresponds to transformation of the local environment in many areas. These include the expansion of broadleaf forests, particularly in Southern Japan, (Fig. 1) with increased

Significance

The motivations for the widespread adoption of pottery is a key theme in world prehistory and is often linked with climate warming at the start of the Holocene. Through organic residue analysis, we investigated the contents of >800 ceramic samples from across the Japanese archipelago, a unique assemblage that transcends the Pleistocene-Holocene boundary. Against our expectations we found that pottery use did not fundamentally change in the Early Holocene. Instead aquatic resources dominated in both periods regardless of the environmental setting. Nevertheless, we found that a broader range of aquatic foods were processed in Early Holocene vessels corresponding to increased ceramic production, reduced mobility, intensified fishing and the start of significant shellfish gathering at this time.

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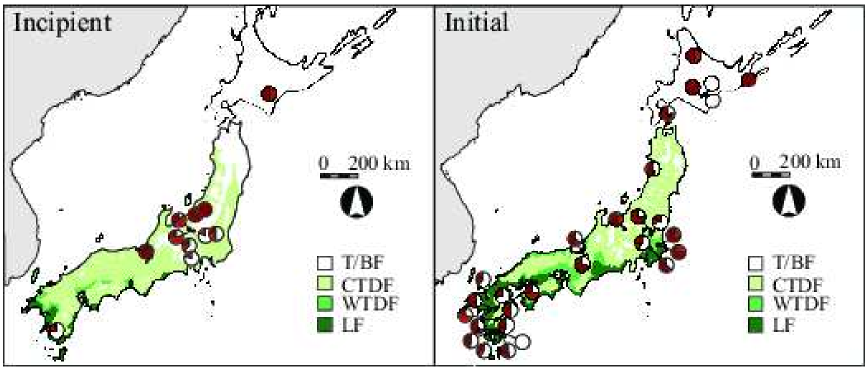

Fig. 1. *Locations of the sampling sites, distributions of the aquatic biomarkers/phytanic acid SRR ratios (from Table 1) and change in vegetation cover (9, 10) across the Japanese archipelago from the Late Pleistocene/Incipient Jōmon (A) to Early Holocene/Initial Jōmon (B). The maps account for changes in sea level across these periods (44). Key: Dark red, complete suite of aquatic biomarkers and/or phytanic acid SRR ratio >75.5%; Red, partial suite of aquatic biomarkers and/or phytanic acid SRR ratio >75.5%; Open, absence of aquatic biomarkers and/or phytanic acid SRR ratio <75.5%; T/BF, tundra/boreal forest; CTDF, cool temperature deciduous forest; WTDF, warm temperature deciduous forest; LF, lucidophyllous forest.*

Table 1. The frequency of aquatic derived residues associated with Incipient and Initial Jōmon pottery from Japan.

Period	# Samples (with lipid)	Full suite of aquatic biomarkers* % (n)	Partial suite of aquatic biomarkers** % (n)	Phytanic acid % (n)	>75.5% SSR-phytanic % (n)	Minimum number of aquatic vessels† % (n)
Incipient (ca. 14,460-11,310 cal BP)	179 (156)	30.8% (48)	7.1% (11)	93.6% (146)	43.6% (68)	46.8% (73)
Initial (ca. 11,500-8,000 cal BP)	622 (566)	10.8% (61)	6.9% (39)	77.0% (436)	42.0% (238)	45.2% (256)
Total	801 (722)	15.1% (109)	6.9% (50)	80.6% (582)	42.4% (306)	45.6% (329)

[*] Presence of C_{18} and C_{20} APAAs together with one of three isoprenoid fatty acids.

[**] Presence of C_{18} APAAs and TMTD.

[†] Having either phytanic acid SSR ratio >75.5% or containing aquatic biomarkers.

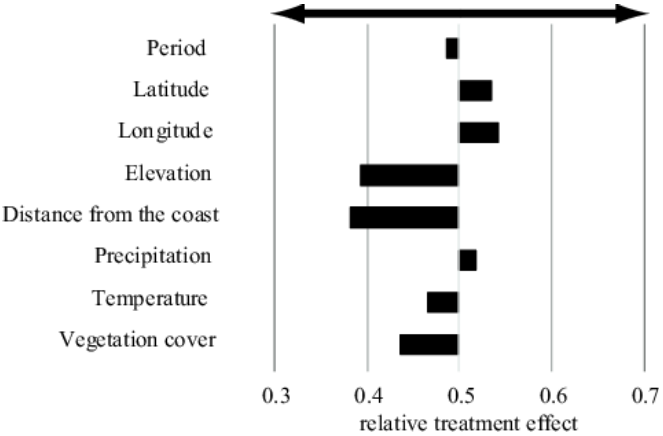

Fig. 2. *Relative treatment effects (RTE) of different geographical and temporal variables on the presence of aquatic derived lipids in Incipient and Initial pottery using a non-parametric multivariate test.RTE of treatment "k" is defined as the probability that a randomly chosen subject from treatment "k" displays a higher response than a subject that is randomly chosen from any of the treatment groups, including treatment "k". The range of possible effect is 0.27 to 0.73.*

opportunities for the exploitation of terrestrial resources, such as forest game, acorns and chestnuts, (9, 10) but also greater access to marine resources through expansion of the coastal shelf (11). Increased pottery production at this time is often seen as a response to the need for processing these newly available resources, as well as intensification and increased sedentism (12) in response to the ameliorated climate and changing coastline. There is, however, little direct evidence to support this view. The analysis of animal and plant remains tentatively show a broadening of the available resources exploited in the Holocene (13) but the data are severely constrained due to generally poor organic preservation in Japan's prevailing acidic soils (14). Some of the best palaeoeconomic data derives from coastal and lacustrine shell middens that commence during the Initial Jōmon period and while these point to a broad economic base, with terrestrial plant and animal remains well represented in addition to fish remains and shell (e.g. (15, 16), it is unknown whether pottery use also broadened at this point.

Organic residue analysis provides the only approach for directly examining the contents of pottery vessels, and in the absence of quantifiable numbers of faunal and floral remains at the majority of sites (17), it is also a valuable tool for examining palaeoeconomic change through this critical period in East Asian prehistory. Previous studies have already shown that Incipient Jōmon vessels dating to the Late Pleistocene (*ca*. 15,000–11,500 cal BP) were predominantly used for processing aquatic species, particularly seasonally abundant marine and anadromous fish (5, 6). Produced in low numbers compared to other artefacts (4), it has been suggested that pottery did not have a major economic function at this time and may have been prestige items associated with the collective procurement of aquatic foods during periods of sedentism by otherwise largely mobile Pleistocene hunter-gatherer groups (5, 6, 18). In contrast, the only organic residue analysis of Initial Jōmon pottery is limited to a small number of sherds from the site of Torihama in Western Japan (5). It is therefore not known whether the function of pottery fundamentally changed in the Early Holocene, as a consequence of

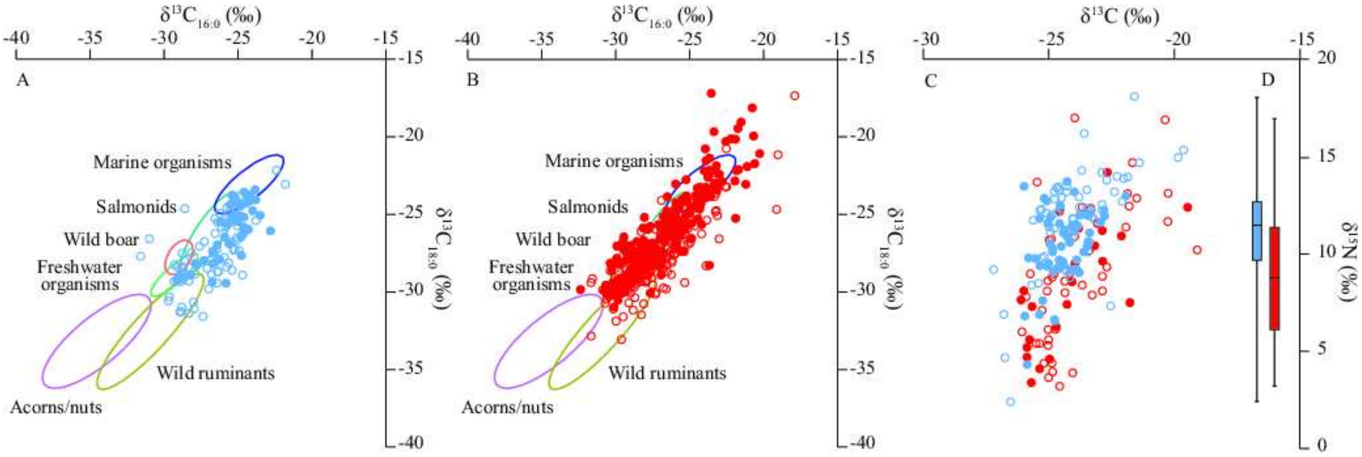

Fig. 3. *Bulk and single compound stable isotope data from Late Pleistocene/Incipient Jōmon (blue) and Early Holocene/Initial Jōmon (red) ceramic vessels. δ^{13}C values of $C_{16:0}$ and $C_{18:0}$ n-alkanoic acids extracted from Late Pleistocene/Incipient (A) and Early Holocene/Initial (B) Jōmon pottery, which show a broadening of aquatic resources. The 95% confidence ellipses are based on modern Japanese authentic reference fats (5, 6, 21–23). Bulk δ^{13}C and δ^{15}N stable isotope data (C) obtained from carbonised residues adhering to Incipient and Initial Jōmon vessels (some data previously reported in (5, 6, 40, 41). (D) Box plot of the δ^{15}N values, which also demonstrate a broadening of aquatic resources. Key: Filled circle, sample with aquatic biomarkers and/or phytanic acid SRR ratio >75.5%; Open circle, absence of aquatic biomarkers and/or phytanic acid SRR ratio <75.5%.*

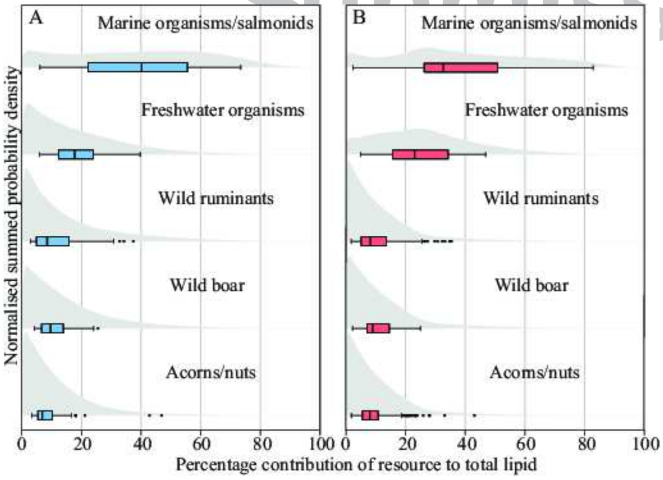

Fig. 4. *Estimated percentage contributions of lipid from different food sources to (A) Late Pleistocene/Incipient and (B) Early Holocene/Initial Jōmon pottery using a concentration dependent mixing model. The model parameters are described in the SI Appendix. Box plots show the range of median % contributions estimated from each pot for each food source. The summed probability density distributions (grey) shows the relative likelihood of the contribution of each food resource summed across the two samples groups and normalised to account for differences in samples size.*

the ameliorating climates, nor how responses varied across the archipelago.

To investigate further, here we present new chemical and isotopic analysis of 638 sherds and 77 charred deposits from 39 Incipient (*ca*. 14,460-11,310 cal BP) and Initial Jōmon sites (*ca*. 11,500-8,000 cal BP). The sites were chosen to examine variability over an ecological transect through Japan (see SI Appendix, Datasets S1 and S2), including inland and coastal localities (Fig. 1) at variable elevations (0-1,500 m). The majority of Incipient Jōmon sherds have cord-marked decoration corresponding to Phase 3a (*ca*. 14,460-12,000 cal BP) and 3b (*ca*. 12,030-11,310 cal BP) (see SI Appendix, Fig. S2) as defined by Taniguchi (12), with the majority corresponding to the Younger Dryas chronozone. When combined with previous data (5, 6), we have a comprehensive corpus of over 800 samples from 46 sites making this one of the largest studies of its kind. We hypothesised that environmental factors (e.g. site location, ecological zone, elevation) would largely determine the use of pottery with an increase in the processing of aquatic organisms in the cooler northern regions where terrestrial resources were less available. Further, we may expect to see a clear increase in oil rich plant products, such as nuts and seeds, and ruminant products, such as sika deer (*Cervus nippon*), and a shift away from aquatic resources in all but coastal sites at the start of the Holocene associated with climate amelioration.

Results and Discussion

Overall, interpretable amounts of lipids were readily extractable from fragments of Jōmon pottery and adhering charred deposits using an acid/methanol extraction procedure (see Methods Summary). In total, 94% (611/652) of the potsherds and 74% (111/149) of the carbonised deposits yielded appreciable quantities of lipids i.e. that were either above the minimum amount required for interpretation (>5μg g^{-1} for potsherds and >100μg g^{-1} for charred deposits) (6, 19) or contained distinctive lipids traceable to a specific source.

Evidence for the processing of aquatic foods

Although the procedure deployed is suitable for identifying fats, oils and waxes from a wide range of plant and animal products (20), a distinctive feature of many of the Jōmon sherds analysed was the presence of aquatic derived lipids. In total, 15.1% (109/722; Table 1) of the samples analysed satisfy the established criteria for the presence of 'aquatic biomarkers' in pottery (5, 20), which includes the presence of ω-(*o*-alkylphenyl) alkanoic acids (APAAs) with C_{18} and C_{20} carbon atoms and isoprenoid fatty acids (either phytanic, pristanic or 4,8,12-trimethyl tridecanoic acid (TMTD)). Notably the C_{20} APAAs are formed during the protracted heating of the $C_{20:x}$ mono- and polyunsaturated fatty acids which are only found in appreciable concentrations in freshwater and marine animals (21, 22). The presence of APAAs implies that the pottery vessels were subjected to prolonged heating (typically >270°, >17h; (21, 22)), easily achieved through boiling or roasting of their contents, which is consistent with the presence of charred 'foodcrusts' on many vessels. Multi-branched isoprenoid fatty acids originate from the breakdown of phytol, a constituent of chlorophyll, but only accumulate at high concentrations in ruminant and aquatic animal tissues. In particular, TMTD is considered more of a characteristic of aquatic oils (23).

These 'aquatic biomarker' estimates should be considered as a minimum percentage, since APAAs are not always formed during food preparation and both APAAs and isoprenoids may be lost in the burial environment relative to other lipid molecules with higher relative concentrations. A further 6.9% (50/722) have C_{18} APAAs and TMTD which are most likely aquatic in origin (i.e. 'partial aquatic biomarkers'; Table 1), while the majority of samples (81%; 582/722) contained phytanic acid, the most frequent isoprenoid acid. Among the resources available to Japanese Pleistocene and Holocene hunter-gatherers, wild ruminants such as sika deer offer the only other major source of phytanic acid other than aquatic oils (7, 24). To distinguish these, we examined the ratio of the two naturally occurring configurations, or diastereomers, of phytanic acid (3S,7R,11R,15-phytanic acid ∼ SRR, and 3R,7R,11R,15-phytanic acid ∼ RRR (see Methods Summary)). Despite considerable overlap, the SRR isomer tends to dominate in aquatic oils compared to ruminant fats (7, 24) and a SRR% above 75.5% can be assigned to this source, using a conservative limit (95% confidence). Over 52% (306/582) of the samples with phytanic acid met this criteria, for the remainder the source of phytanic acid is uncertain as it fell within both the aquatic and ruminant range.

Using the SRR ratio and presence of aquatic biomarkers, we conservatively assigned a minimum number of vessels analysed that were used to process aquatic foods across the Pleistocene-Holocene transition (Table 1). Overall, there was a striking consistency in the use of pottery throughout the Japanese archipelago regardless of period or environmental setting (Fig. 1). A non-parametric multivariate inference test (25) showed significant ($p=<0.001$) effects of period, latitude, longitude, elevation, distance from the coast, precipitation, temperature and vegetation cover on the frequency of aquatic resources in the vessels (Fig. 2; see SI Appendix). However, when the relative effects were quantified (Fig. 2) the site's distance from the coast and its elevation had the greatest effect but even this effect was not strong (i.e. the relative effect value does not approach the minimum or maximum effect). The other environmental variables and the period classification of the vessels had no or very weak effects (0.43-0.54) on the presence/absence of aquatic derived lipids. Pots were used to process aquatic resources at an equally high frequency throughout the archipelago from Hokkaido and Northern Honshu to Kyushu.

There was only a slight decrease in pottery used for processing aquatic resources between the Incipient (47%, 73/156) and Initial (45%, 256/566; Table 1) Jōmon. These results refute the expectation of a dramatic change in the function of pottery at start of the Holocene when terrestrial resources were more available, even accounting for potential biases in site location between periods (Fig. 2). These data corroborate what we have suggested previously (5, 20) that pottery production for the exploitation of aquatic resources was embedded as a cultural norm in the social memories of these foragers.

Pottery from sites more distant from the palaeocoastline tended to have fewer aquatic derived lipids but the effect was marginal and co-varied with site elevation. Indeed, aquatic products were frequently identified in ceramics from inland riverine and lacustrine sites (Fig. 1) most likely pointing to the exploitation of freshwater resources and/or migratory species, such as salmonids. To distinguish the source of these residues further we examined the carbon isotope ($\delta^{13}C$) values of the major saturated fatty acids ($C_{16:0}$ and $C_{18:0}$) extracted from the sherds, and bulk carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope values of any adhering carbonised residues (see SI Appendix, Dataset S2). In Fig. 3, the fatty acid data are compared with $\delta^{13}C$ values obtained from modern authentic Japanese plants and animals (5, 6, 26–28). These generally support the lipid biomarker data with many vessels plotting in the reference ranges for aquatic resources. Interestingly, there was only a weak negative correlation between distance from the coast and the $\delta^{13}C_{16:0}$ value (Spearman $\rho(560)=-0.25\ p=<0.001$) and no correlation with the bulk $\delta^{13}C$ value (Spearman $\rho(190)=0.03\ p=0.6667$) as may have been expected if marine resources were preferentially processed at coastal sites compare to inland localities. This may be explained by the exploitation of migratory fish, such as salmonids, which have $\delta^{13}C$ values that approach the marine range (Fig. 3).

Fully marine species, beyond the isotopic range of reference salmonids (Fig. 3; see SI Appendix, Dataset S3) were identified in pottery from sites located >15 km from the prehistoric coastline, the maximum logistical walking distance for a logistical day trip (29), which suggests that aquatic resources were not only acquired locally for direct consumption but could also have been preserved (e.g. dried) and transported. These include an Incipient vessel from Taisho 3 and 13 Initial vessels from Haizuka, Higashimyou, Nishinojo, Nisshin 3 and Taisho 3. Although site elevation has the greatest effect on the presence/absence of aquatic derived lipids, even pottery from remote mountainous areas were also used to process aquatic foods. At Yukura Cave (at elevation of *ca.* 1534 m) almost half the vessels had residues typical of salmonids with the nearest source, the Shinano river (30), located *ca.* 15 km away. Conversely at these remote hunting sites and more broadly in Japan's warmer forested areas, a surprisingly low number of residues could be attributed to forest game species, such as sika deer and wild boar (*Sus scrofa*) (Fig. 3), implying they were processed in other ways.

There is also surprisingly limited data to suggest that plant foods were processed in Incipient or Initial Jōmon pottery across Japan. Low to trace amounts of leafy plant-derived lipids, including phytosterol, long-chain even-numbered fatty acids or long-chain odd-numbered alkanes, were present in some samples, most notably at Kenshojo in Southern Kyushu (see SI Appendix, Dataset S2). Isotopic analysis of the foodcrusts adhering to the potsherds also generally had lower (<22) C:N atomic ratios (mean = 12.0 ± 5.1) more typical of carbonised terrestrial animal and marine tissues than plant remains (27) (See SI Appendix, Dataset S2). Plant resources, particularly acorns and chestnuts, and artefacts associated with plant processing are frequently found on Incipient and Initial sites (9, 31–33) suggesting they were an important feature of the Jōmon economy. While the organic residue evidence cannot rule out the presence of plants in pottery entirely, the data clearly show that Incipient and Initial Jōmon vessels were not extensively used for this purpose. Rather we contend that Incipient and Initial Jōmon hunter-gatherers had a clear preference for preparing aquatic resources over terrestrial animal and plant products in pottery. Moreover, we assert that this cooking practice was pervasive over a wide range of environmental settings and persistent through time and through significant climate change.

Holocene pottery used for processing of a wider array of aquatic resources

Although there is strong evidence that aquatic resources were exploited in both periods we found evidence across the Japanese archipelago of diversification in the types of aquatic foods processed in the pottery at the start of the Holocene. A much narrower range of $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values were obtained from Late Pleistocene (Incipient Jōmon) pottery when compared to those from the Early Holocene (Initial Jōmon) (Figs. 2*A* and 2*B*). During the Incipient Jōmon, $\delta^{13}C_{16:0}$ and $\delta^{13}C_{18:0}$ values are relatively homogenous i.e. have low variances ($\sigma^2=3.5$, n = 119, mean =-26.3‰). Regardless of the geographic setting, the majority of values fall within the ranges established from the analysis of modern marine organisms and salmonids (Fig. 3*A*), which was corroborated by the presence of aquatic biomarkers in many of the samples. These data support the general model proposed previously (6, 34, 35) that the earliest phases of pottery use are highly specialised and focused on seasonally available aquatic resources.

In contrast, the variance of $\delta^{13}C_{16:0}$ values significantly increased (Brown–Forsythe test F(1,558)=10.42 *p<0.005*) during the Initial Jōmon (σ^2=6.9, n = 441, mean =-27.0‰ for $C_{16:0}$; Fig. 3*B*). This most likely reflects a broadening of the aquatic foods processed to encompass a greater range of both marine and freshwater species (Fig. 3*B*). The high frequency of the other aquatic derived lipids on Initial Jōmon sherds supports this contention but mixing with terrestrial animal and even plant resources, also relatively depleted in ^{13}C, cannot be ruled out entirely. In order to investigate the effects of mixing different resources in the vessels we applied a concentration dependent Bayesian mixing model (36) that used the $\delta^{13}C_{16:0}$, $\delta^{13}C_{18:0}$ and SRR% values as proxies, with priors based on the presence of isoprenoid and APAAs (see SI Appendix). This model was used to examine the likely probability of different proportions of lipids derived from plants (acorns and chestnuts), freshwater organisms (fish), wild boar, wild ruminants (sika deer) and marine organisms/salmonids to each pot. By summing the probabilities for each period and examining their densities (Fig. 4), only aquatic organisms can be reliably considered to have made a substantial contribution (i.e. >25% of total lipid) in either periods (Fig. 4) based on the assumptions used in the model. Noticeably, however, the percentage contribution of lipid from freshwater organisms is predicted to increase from the Incipient to Initial Jōmon (Fig. 4) consistent with a broadening of pottery use at this time.

Surprisingly few vessels contained substantial amounts of non-aquatic products. Ruminant, wild boar and acorn/chestnut were estimated by the model to have made a contribution of >25% in 21, 1 and 7 samples respectively. It should be noted that their contribution to the remaining vessels cannot be ruled out entirely; between 0 to 25% lipid contributions from non-aquatic sources were most likely, although depending on their lipid content, these could have had a greater relative contribution by total weight. Further distinction is not possible using the isotope approach and SRR% alone. Even where prior information form the biomarker evidence is deployed there is a high degree of equifinality regarding the source contributions.

A broadening of the range of aquatic resources processed in pottery during the Holocene and across the Japanese archipelago can also be seen from the bulk nitrogen ($\delta^{15}N$) stable isotope values of carbonised residues adhering to pottery (Fig. 3*C* and 3*D*). Nitrogen stable isotope values of charred deposits are often used to crudely distinguish between high trophic level aquatic resources and lower trophic level terrestrial organisms (37), although ^{15}N enrichment due to charring also needs to be accounted for (38, 39). In total, $\delta^{13}C$ and $\delta^{15}N$ values were obtained on 157 samples from 21 sites (Fig. 3*C*), which were complemented with previously published data undertaken as part of AMS radiocarbon (^{14}C) dating programs (40, 41). As with $\delta^{13}C_{16:0}$, a broader range of nitrogen isotope values were obtained from the Initial Jōmon pottery (Incipient variance, σ^2=5.6, n = 119; Initial variance, σ^2=10.9, n = 71; Brown–Forsythe test F(1,188)=13.49 *p*=<0.005). A decrease in $\delta^{15}N$ values (Fig. 3*D*) was also observed between the Incipient (median = 11.5‰) and Initial Jōmon (median = 8.8‰) and overall the distributions of the $\delta^{15}N$ values were significantly different (Mann–Whitney *U* test; *U*=6024; *p*=<0.005). In contrast, the range of $\delta^{13}C$ values is similar (U=4128; *p*=0.79) between the Incipient (-27 to -20‰, median = -24‰) and Initial Jōmon (-26 to -19‰, median = -24‰).

Interestingly, aquatic biomarkers were frequently observed in charred deposits with lower $\delta^{15}N$ values (Fig. 3*C*) ruling out predominantly terrestrial input. Although outside the range of marine finfish and marine mammals (>12‰, 17, 40), these $\delta^{15}N$ values are within the range of values obtained on lower trophic level freshwater fish and marine/freshwater shellfish (17, 40), accounting for a 1‰ increase with charring (38, 40). Therefore, a more likely explanation is that the observed broadening and decrease in $\delta^{15}N$ values of charred deposits observed in the Holocene (Fig. 3*D*) is due to a diversification of aquatic resource exploitation to encompass freshwater fishing and/or shellfish collection. This explanation is also consistent with the establishment of shell middens in Japan at this time (16) but currently we are unable to unequivocally distinguish shellfish derived residues with the methods at our disposal.

Conclusion

There is a dramatic increase in the scale of pottery use across Japan after the onset of Early Holocene warming. We have investigated the extent to which these environmental changes drove diversification in pottery function as a broader range of resources became readily available. The earliest pottery in Japan was used to process aquatic resources, but contrary to expectations, we found no evidence that its function expanded in the Early Holocene to include the processing of terrestrial animal and plant resources. Instead, our results show remarkable continuity and consistency in the function of pottery across the Pleistocene-Holocene transition, pointing towards a strong cultural association between pottery and the processing of aquatic resources. This pattern also holds throughout the different ecological zones of the Japanese archipelago. As a result, we suggest that after its first invention, pottery developed particular cultural associations linked to processing aquatic resources, and that these were robust enough to withstand the effects of major climatic and environmental transformations at the Pleistocene-Holocene transition. Moreover, these 'culinary' preferences persisted across Japan, even in warmer southern areas where abundant nut and plant resources were increasingly available. Our earlier study from the Torihama shell midden site (5–7) in Japan indicates that this cognitive association persisted until at least the Middle Holocene, and may only have been truncated by the arrival of rice and millet agriculture *ca*. 2,500 cal BP. A similar association between early pottery and aquatic resources has also been identified in adjacent regions of East Asia such as Sakhalin Island (6, 34, 35) and the Korean Peninsula (5–7). Here, pottery appears in the Early and Middle Holocene and from the outset demonstrates close association with processing of marine foods.

Our current research also identified an important new pattern, which is that pottery was used to process a broader spectrum of aquatic foods in the Early Holocene, including shellfish, freshwater fish and a greater range of marine taxa. This corresponds to significant climate warming *ca*. 11,500 years ago which may have reduced salmonid stocks in Northern Japan (42, 43) prompting a switch to other aquatic species, but also created greater opportunities for inshore fishing and shellfish gathering through the expansion of the marine shelf (11). Also at this time, pottery traditions began to flourish, with greater variation in forms and volumes reflecting intensified usage. We suggest that this represents an important change away from the small-scale and specialised use of pottery in the Late Pleistocene to a greater utilitarian function in the Early Holocene as fishing and shellfish gathering intensified. Whether this change served as a driving force for the wider-range dispersal of hunter-gatherer pottery from East Asia into surrounding areas along aquatic ecotones (2), needs to be tested through further organic residue analysis and greater AMS radiocarbon dating of early pottery sites.

Finally, we are unable to explain either the invention of pottery in the Late Pleistocene or its more varied and intensified use in the Holocene in purely functional terms. Indeed, aquatic foods were undoubtedly exploited by maritime East Asian hunter-gatherers well before pottery appeared (45). Social and demographic factors, indirectly linked to economic change, provide a more compelling argument. We suggest that pottery was initially developed as a novel, prestige technology during periods of seasonal population aggregation focused on cooperative harvesting of migratory fish, such as salmonids. From the start of the Holocene, however, it was produced in significantly larger quantities, associated with intensification of aquatic resource exploitation and increasing sedentism.

Methods summary

We obtained 652 ceramic sherds and 172 adhering carbonised residues from 46 archaeological sites throughout the Japanese archipelago. Assignation to the Incipient or Initial Jōmon was based on pottery typology or independently through the AMS radiocarbon (^{14}C) dating of associated organic materials.

Organic residue analysis: Lipids were directly extracted and methylated with acidified methanol according to established methods (6, 7). Briefly, methanol (1 or 4 mL) was added to homogenised carbonised residues (10-20 mg) or ceramic powders (0.5-1.0 g) drilled (2-5 mm depth) from the interior or exterior surface of the sherd. The sample was sonicated in a water bath for 15 min, and acidified with concentrated sulphuric acid (200 or 800 µL). The acidified suspension was heated in a block for 4 h at 70 °C. Lipids were extracted *n*-hexane (3 × 2 ml), and subsequently analysed by GC-MS and GC-c-IRMS (see SI Appendix). Interior foodcrusts or exterior carbonised residues were also analysed by Elemental Analysis-Isotope Ratio Mass Spectrometry (see SI Appendix) using previously reported protocols (5, 6).

Statistical and GIS: All statistical tests were performed using *R studio* (version 1.0.136) and *Past* (version 3.18). Mapping was undertaken with QGIS (version 2.18.9).

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1. Kuzmin YV (2017) The origins of pottery in East Asia and neighboring regions: An analysis based on radiocarbon data. *Quat Int* 441:29–35.
2. Jordan P, Gibbs K, Hommel P, Piezonka H, Silva F (2016) Modelling the diffusion of pottery technologies across Afro-Eurasia: emerging insights and future research. *Antiquity* 90(351): 590-603.
3. Yanshina OV (2017) The earliest pottery of the eastern part of Asia: Similarities and differences. *Quat Int* 441:69–80.
4. Taniguchi Y (2017) The Beginning of Pottery Technology in Japan: The Dating and Function of Incipient Jomon Pottery. *The Emergence of Pottery in West Asia*, eds Tsuneki A, Nieuwenhuyse O, Campbell S (Oxbow Books, Oxford).
5. Lucquin A, et al. (2016) Ancient lipids document continuity in the use of early hunter–gatherer pottery through 9,000 years of Japanese prehistory. *Proc Natl Acad Sci USA* 113(15):3991–3996.
6. Craig OE, et al. (2013) Earliest evidence for the use of pottery. *Nature* 496(7445):351–354.
7. Shoda S, Lucquin A, Ahn J-H, Hwang C-J, Craig OE (2017) Pottery use by early Holocene hunter-gatherers of the Korean peninsula closely linked with the exploitation of marine resources. *Quat Sci Rev* 170:164–173.
8. Aira Town Educational Board (2005) Excavation Report: The Kenshojo Site (in Japanese).
9. Sakaguchi T (2009) Storage adaptations among hunter–gatherers: A quantitative approach to the Jomon period. *J Anthropol Archaeol* 28(3):290–303.
10. Melvin Aikens C, Akazawa T (1996) The Pleistocene—Holocene Transition in Japan and Adjacent Northeast Asia. *Humans at the End of the Ice Age, Interdisciplinary Contributions to Archaeology*, eds Straus LG, Eriksen BV, Erlandson JM, Yesner DR (Springer, New York, NY).
11. Sato H, Izuho M, Morisaki K (2011) Human cultures and environmental changes in the Pleistocene–Holocene transition in the Japanese Archipelago. *Quat Int* 237(1):93–102.
12. Kaner S, Taniguchi Y (2017) The Development of Pottery and Associated Technological Developments in Japan, Korea, and the Russian Far East. *Handbook of East and Southeast Asian Archaeology*, eds Habu J, Lape P, Olsen J (Springer, New York, NY).
13. Matsui A, Kanehara M (2006) The question of prehistoric plant husbandry during the Jomon period in Japan. *World Archaeol* 38(2):2590273.
14. Matsui A (2014) Shuryou no Taisho (Hunting game). *Jomon Jidai (Ge) (Jomon Period (Second Volume), The Archaeology of Japan Lecture* (Aoki Shoten, Tokyo), pp 3–35.
15. Saga City Educational Board (2016) The Higashimyou site cluster synthesis report (in Japanese).
16. Habu J, Matsui A, Yamamoto N, Kanno T (2011) Shell midden archaeology in Japan: Aquatic food acquisition and long-term change in the Jomon culture. *Quat Int* 239(1–2):19–27.
17. Yoneda M, et al. (2004) Isotopic evidence of inland-water fishing by a Jomon population excavated from the Boji site, Nagano, Japan. *J Archaeol Sci* 31(1):97–107.
18. Hayden B (1995) The emergence of prestige technologies and pottery. *The Emergence of Pottery: Technology and Innovation in Ancient Societies* (Smithsonian Institution Press, Washington, DC), pp 257–266.
19. Evershed RP (2008) Experimental approaches to the interpretation of absorbed organic residues in archaeological ceramics. *World Archaeol* 40(1):26–47.
20. Evershed RP (2008) Organic residue analysis in archaeology: the archaeological biomarker revolution. *Archaeometry* 50(6):895–924.
21. Hansel FA, Copley MS, Madureira LAS, Evershed RP (2004) Thermally produced ω-(o-alkylphenyl) alkanoic acids provide evidence for the processing of marine products in archaeological pottery vessels. *Tetrahedron Lett* 45(14):2999–3002.
22. Evershed RP, Copyley MS, Dickson L, Hansel FA (2008) Experimental evidence for the processing of marine animal products and other commodities containing polyunsaturated fatty acids in pottery vessels. *Archaeometry* 50(1):101–113.
23. Ackman RG, Hooper SN (1968) Examination of isoprenoid fatty acids as distinguishing characteristics of specific marine oils with particular reference to whale oils. *Comp Biochem Physiol* 24:549–565.
24. Lucquin A, Colonese AC, Farrell TFG, Craig OE (2016) Utilising phytanic acid diastereomers for the characterisation of archaeological lipid residues in pottery samples. *Tetrahedron Lett* 57(6):703–707.
25. Burchett W, Ellis A, Harrar S, Bathke A (2017) Nonparametric Inference for Multivariate Data: The R Package npmv. *J Stat Softw* 76(4):1–18.
26. Horiuchi A, Miyata Y, Kamijo N, Cramp L, Evershed RP (2015) A Dietary Study of the Kamegaoka Culture Population during the Final Jomon Period, Japan, Using Stable Isotope and Lipid Analyses of Ceramic Residues. *Radiocarbon* 57(4):721–736.
27. Heron C, et al. (2016) Molecular and isotopic investigations of pottery and "charred remains" from Sannai Maruyama and Sannai Maruyama No. 9, Aomori Prefecture. *Japanese Journal of Archaeology* 4(1):29–52.
28. Miyata Y, Horiuchi A, Takada H, Nakamura T (2015) Evaluation of sea mammals as marine resource by lipid analysis in pottery excavated from Mawaki archaeological site, Ishikawa, Japan. *The 32nd Annual Meeting of Japan Society for Scientific Studies on Cultural Property Abstracts*: 40–41.
29. Kelly RL (2013) *The Lifeways of Hunter-Gatherers: The Foraging Spectrum* (Cambridge University Press, Cambridge).
30. Kaner S (2009) Long-term innovation: The appearance and spread of pottery in the Japanese Archipelago. *Ceramics Before Farming: The Dispersal of Pottery Among Prehistoric Eurasian Hunter-Gatherers*, eds Jordan P, Zvelebil M (Left Coast Press, Walnut Creek, CA).
31. Crawford GW (2011) Advances in Understanding Early Agriculture in Japan. *Curr Anthropol* 52(S4):S331–S345.
32. Noshiro S, Sasaki Y (2014) Pre-agricultural management of plant resources during the Jomon period in Japan—a sophisticated subsistence system on plant resources. *J Archaeol Sci* 42:93–106.
33. Shibutani A (2009) Late Pleistocene to Early Holocene Plant Movements in Southern Kyushu, Japan. *Arch* 5(1):124–133.
34. Taché K, Craig OE (2015) Cooperative harvesting of aquatic resources and the beginning of pottery production in north-eastern North America. *Antiquity* 89(343):177–190.
35. Gibbs K, et al. (2017) Exploring the emergence of an "Aquatic" Neolithic in the Russian Far East: organic residue analysis of early hunter-gatherer pottery from Sakhalin Island. *Antiquity* 91(360):1484–1500.
36. Fernandes R, et al. (2018) Reconstruction of prehistoric pottery use from fatty acid carbon isotope signatures using Bayesian inference. *Org Geochem* 117:31-42.
37. Heron C, Craig OE (2015) Aquatic Resources in Foodcrusts: Identification and Implication. *Radiocarbon* 57(4): 707-719.
38. Nitsch EK, Charles M, Bogaard A (2015) Calculating a statistically robust δ^{13}C and δ^{15}N offset for charred cereal and pulse seeds. *STAR: Science & Technology of Archaeological Research* 1(1):1–8.
39. Fraser RA, et al. (2013) Assessing natural variation and the effects of charring, burial and pre-treatment on the stable carbon and nitrogen isotope values of archaeobotanical cereals and pulses. *J Archaeol Sci* 40(12):4754–4766.
40. Yoshida K, et al. (2013) Dating and stable isotope analysis of charred residues on the Incipient Jomon pottery (Japan). *Radiocarbon* 55(2–3):1322–1333.
41. Kunikita D, et al. (2013) Dating Charred Remains on Pottery and Analyzing Food Habits in the Early Neolithic Period in Northeast Asia. *Radiocarbon* 55(2–3):1334–1340.
42. Ishida Y, et al. (2001) Archeological evidence of Pacific salmon distribution in northern Japan and implications for future global warming. *Prog Oceanogr* 49(1):539–550.
43. Ishida Y, Yamada A, Nagasawa K (2016) Future Climate-Related Changes in Fish Species Composition Including Chum Salmon (*Oncorhynchus keta*) in Northern Japanese Waters, Inferred from Archaeological Evidence. *North Pacific Anadromous Fish Commission Bulletin* 6: 243–258.
44. Lambeck K, Rouby H, Purcell A, Sun Y, Sambridge M (2014) Sea level and global ice volumes from the Last Glacial Maximum to the Holocene. *Proc Natl Acad Sci USA* 111(43):15296–15303.
45. O'Connor S, Ono R, Clarkson C (2011) Pelagic fishing at 42,000 years before the present and the maritime skills of modern humans. *Science* 334(6059):1117–1121.