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Ancient lipids document continuity in the use of early hunter–gatherer pottery through 9,000 years of Japanese prehistory

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The earliest pots in the world are from East Asia and date to the Late Pleistocene. However, ceramic vessels were only produced in large numbers during the warmer and more stable climatic conditions of the Holocene. It has long been assumed that the expansion of pottery was linked with increased sedentism and exploitation of new resources that became available with the ameliorated climate, but this hypothesis has never been tested. Through chemical analysis of their contents, we herein investigate the use of pottery across an exceptionally long 9,000-y sequence from the Jōmon site of Torihama in western Japan, intermittently occupied from the Late Pleistocene to the mid-Holocene. Molecular and isotopic analyses of lipids from 143 vessels provides clear evidence that pottery across this sequence was predominantly used for cooking marine and freshwater resources, with evidence for diversification in the range of aquatic products processed during the Holocene. Conversely, there is little indication that ruminant animals or plants were processed in pottery, although it is evident from the faunal and macrobotanical remains that these foods were heavily exploited. Supported by other residue analysis data from Japan, our results show that the link between pottery and fishing was established in the Late Paleolithic and lasted well into the Holocene, despite environmental and socio-economic change. Cooking aquatic products in pottery represents an enduring social aspect of East Asian hunter–gatherers, a tradition based on a dependable technology for exploiting a sustainable resource in an uncertain and changing world.

archaeology | ceramic | residue analysis | isotope | plant microfossil

The emergence and development of pottery remains one of the most important research questions in archaeology. Once linked exclusively to the development of farming and settled village life, it is now known that the origins of pottery are instead bound-up in a complex process of innovation that ultimately extends back as far as 20,000 y to groups of East Asian hunter–gatherers living during the Late Pleistocene (1–3). One of the earliest and best-studied centers for the innovation and development of ceramic containers is the Japanese archipelago. Pottery was invented here around 16,000 y ago and remained an important part of hunter–gatherer life (Jōmon culture) until the transition to rice cultivation approximately 2,800 y ago. The earliest pots in Japan, produced during the "Incipient" Jōmon phase (16,000–11,500 cal B.P.), were small and found only in low numbers per site. It is thought they had a very limited range of uses, possibly for the occasional small-scale and highly labor-intensive preparation of "exotic" or "prestige" foods (4). Pottery only began to flourish, however, with the steady warming of global temperatures from around 11,500 y ago, and production increased exponentially throughout the early Holocene (11,500–7,000 cal B.P.), with the ameliorated climate (5). These changing climate conditions resulted in ecological shifts in forest vegetation (6) and salt-water inundation of the rich coastal plains that surround the Japanese archipelago. Because it was produced in much greater quantities during the Holocene, it has been hypothesized that pottery may have facilitated new strategies for the processing, storage, and serving of a wider array of increasingly abundant foodstuffs, such as plant foods and shellfish (7). The enhanced production of ceramics has also been linked to increased sedentism, population growth, and perhaps also to the dispersal of pottery technology westwards across Northern Eurasia and northwards toward Alaska (8).

Despite such speculation, it is not known how East Asian hunter–gatherers adapted their pottery to accommodate changing environmental conditions and resource availability, or indeed whether other noneconomic drivers for the uptake of pottery were also in play. Because animal and plant remains are generally very poorly preserved in this region, particularly during the late glacial period, direct determination of the use of pottery is critical to answering this question. We have recently shown that lipids can be reliably characterized in Late Pleistocene ceramic vessels to reveal their original contents (9). We suggested that the primary use of pottery during this period was for processing aquatic resources, albeit based on a limited number of samples from two Incipient Jōmon sites. Beyond this study, little is known about how hunter–gatherer pottery use may have changed or diversified in the Holocene.

Here, we report molecular and isotopic analyses of the contents of pottery vessels from the archaeological site of Torihama, located

Significance

Pottery has had a central role in human society for many millennia, but the reasons for the emergence and spread of this technology are poorly understood. First invented by groups of hunter–gatherers living in East Asia during the last glacial period, production only began to flourish with rising global temperatures in the Holocene, but the reasons for its uptake and spread are unknown. Through chemical analysis of their contents, we herein provide, to our knowledge, the first direct evidence of pottery use across this climatic transition. Contrary to expectations, ceramic vessels had a remarkably consistent use, predominantly for processing aquatic resources, indicating that cultural rather than environmental factors were most important for their widespread uptake.

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in Wakasa Bay, Fukui Prefecture, Japan (Fig. 1). The earliest Jōmon pots (Incipient Jōmon) available from this site date from approximately 14,000 B.P., but crucially the pottery sequence continues through the Younger Dryas chronozone into the Holocene (Initial Jōmon, 11,500 B.P.) and Early Jōmon period (ca. 7,000 B.P.), when a shell mound was formed at the site. Exceptionally for East Asia, organic artifacts and ecofacts are preserved in the waterlogged deposits of Torihama, providing a rare opportunity to examine pottery use against changes in the fauna and flora that were exploited, as well as other material culture associated with their procurement and processing. It has been tentatively proposed that the site was initially a seasonal (summer/autumn) hunting and fishing station in the Late Pleistocene but became occupied for longer periods of time during the Early Holocene, with greater emphasis on plant and freshwater shellfish collection (10–13). Despite sea-level rise at the Pleistocene/Holocene transition, marine foods were most likely available throughout the sequence as the distance to the open sea remained reachable because of the steep coastal morphology (Fig. 1) (14).

To investigate changes in the pottery use at this site, three complementary methods were deployed, using well-established protocols (*Methods*), to 143 vessels from all nine stratigraphic phases at Torihama (Table S1). First, lipids were extracted from ceramics and adhering charred surface deposits (foodcrusts) and their structural and carbon isotope characteristics determined using gas chromatography-mass spectrometry (GC-MS) and GC-combustion-isotope ratio MS (GC-c-IRMS), respectively. Second, charred deposits were directly analyzed by elemental analysis-isotope ratio mass spectrometry (EA-IRMS) to determine their bulk carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) stable isotope values. Finally, plant microfossils (starch and phytoliths) were extracted and counted in charred deposits adhering to interior and, where possible, exterior surfaces.

Results

Diagnostic compounds ("biomarkers") for aquatic foods were identified by GC-MS in a large proportion of samples analyzed (Fig. 2*A* and Table 1), regardless of period (Incipient, Initial, or Early) or subphase (Fig. 2*D* and Table S1). In total, over 50% of the samples analyzed that produced an interpretable residue contained isoprenoid alkanoic acids and long-chain (C_{18}-C_{20}) ω-(*o*-alkylphenyl) alkanoic acids (APAAs) (Fig. 2*A*), satisfying the full molecular criteria for aquatic products in archaeological pottery (15). Such APAAs are only formed from protracted or repeated heating of polyunsaturated fatty acids in aquatic oils, and therefore must be derived from primary use of the pot (16). The high proportion of APAAs recovered is remarkable given the antiquity of the vessels and must represent only a minimum estimate for the presence of aquatic products, and that APAAs are not easily formed and are susceptible to degradation and loss because of their low abundance. A greater number of samples had partial sets of biomarkers consisting of C_{18} APAAs and at least one isoprenoid (Fig. 2*A* and Table 1), and many more had lipid profiles consisting of medium- and long-chain saturated (C_{14}-C_{24}), monounsaturated ($C_{16:1}$-$C_{22:1}$), and dicarboxylic (C_7-C_{13}) fatty acids that are typical of degraded fish and aquatic mammal oils (Table S1). In addition, the relative frequencies of two naturally occurring diastereomers of phytanic acid were assessed (3*S*,7*R*,11*R*,15-phytanic and 3*R*,7*R*,11*R*,15-phytanic acid). Phytanic acid is an isoprenoid acid only present in high abundance in the tissues of ruminant, freshwater, and marine animals, with a predominance (i.e., >60%) of the *SRR*-isomer in aquatic species (17, 18). Phytanic acid was observed in 89% of the archaeological samples analyzed, and in the majority of cases (approximately 90%) the *SRR*-isomer was most abundant, consistent with an aquatic rather than ruminant source (Fig. 2*B*). However, a slight decrease in the contribution of *SRR* was observed through time (Fig. 2*B*), which may indicate a broadening of pottery use.

Bulk carbon ($\delta^{13}C$), nitrogen ($\delta^{15}N$) stable isotopes, and atomic carbon-to-nitrogen ratios (C:N) were obtained from the charred deposits adhering to 70 vessel interiors to assess the origin of the bulk organic matter in these residues. Although this approach has been widely applied to study the use of East Asian pottery (19, 20), it offers only crude resolution of contents because of uncertainties in the isotope end-points of different foodstuffs and because of diagenetic alteration (21). The $\delta^{15}N$ values of 75% of charred deposits analyzed ranged between 8.8‰ and 13.4‰ (Fig. 2*C*), regardless of period. A similar

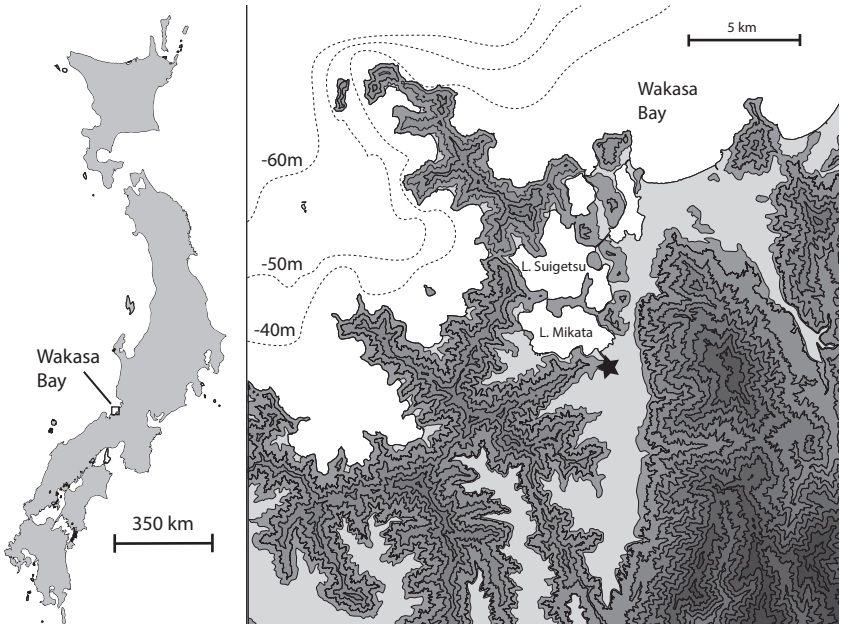

Fig. 1. Location of the Torihama site (starred) on Wakasa Bay. Dotted contours indicate the bathymetry relative to present mean sea level. The coastline corresponding to the Incipient period was between approximately 65–50 m below present-day sea level (14). Topography is shaded by 50-m increments. Image courtesy of Hideaki Kojima (Wakasa Mikata Jōmon Museum).

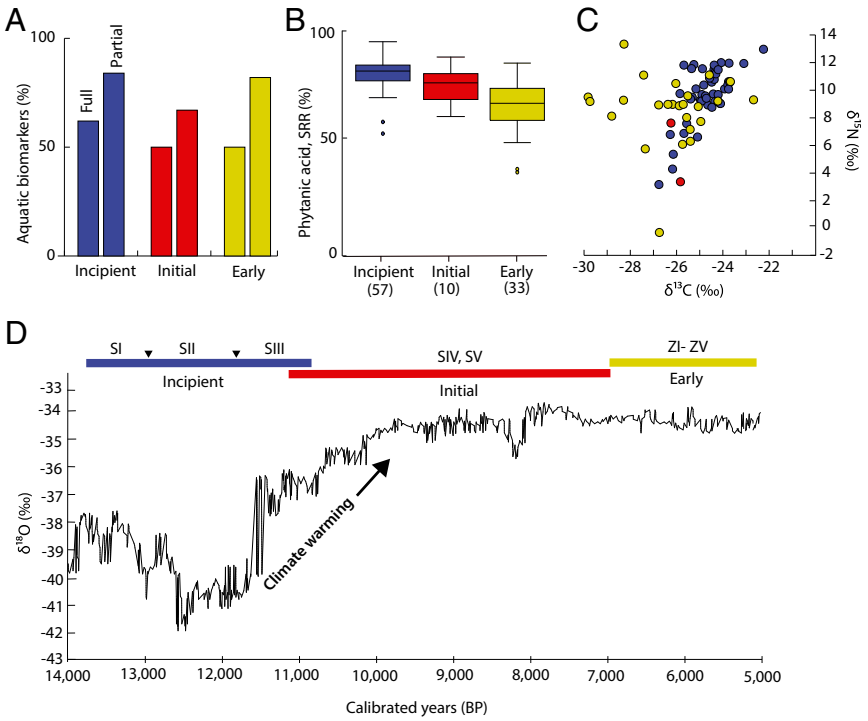

Fig. 2. Correspondence between the phases of pottery at Torihama and their molecular and isotopic characteristics. (*A*) Proportion of pots with an interpretable lipid residue (>5 μg/g^{-1}) containing full (C_{18}-C_{20} APAAs and at least one isoprenoid fatty acid) or partial sets (C_{18} APAAs and at least one isoprenoid fatty acid) of aquatic biomarkers. (*B*) Boxplot showing the range in the contribution of the *SRR* diastereomers of phytanic acid with number of observations indicated for each period. (*C*) Plot of bulk isotope values obtained from analysis of charred deposits. (*D*) Showing the pottery sequence at Torihama based on available radiocarbon dates (Table S3) against the Greenland ice-core oxygen isotope record, with lower $\delta^{18}O$ values generally corresponding to lower temperatures.

range has been previously observed in charred ceramic deposits from different coastal archaeological sites associated with the exploitation of fish and marine mammals (22, 23), and is more consistent with reference tissues from aquatic organisms than terrestrial animals or plants (24), although mixing of the latter cannot be ruled out or accurately quantified using this approach. We note, however, that over 90% of the samples have atomic C:N ratios less than 12 (median = 9.7) (Table S1), which characterize protein rich foods rather than starchy plants (20). Charred deposits more depleted in ^{15}N (i.e., below 6‰) and consistent with terrestrial plant and animal foods were only observed in the minority of cases (approximately 8% of observations). In one charred deposit (T309), associated with an Early Jōmon vessel, we observed a bulk ^{15}N value of −0.3‰ and a C:N ratio of 21.5. Coupled with the absence of aquatic lipid biomarkers, this result provides the only convincing example of plant processing in pottery from Torihama (19).

Carbon isotope ratios ($\delta^{13}C$) of charred deposits showed a greater difference between periods, with Early Jōmon pots much more variable (Fig. 2*C*). This parameter is mainly controlled by the carbon source. Marine carbon is more enriched in ^{13}C compared with terrestrial and freshwater sources. In this case, the samples most depleted in ^{13}C were some of the most enriched in ^{15}N (Fig. 2*C*) and also had relatively low atomic C:N ratios (Table

Table 1. Table summarizing organic residue analysis results by period in relation to the relative frequency of pottery and description of stone artifacts, plant, and faunal remains

Period associates dates (×10^3 cal B.P.)	Samples analyzed	Samples yielding lipids (%)*	Samples yielding aquatic biomarkers (%) (with partial set)	Relative pottery abundance to stone artifact†	Stone artifact assemblage‡	Species representation (aquatic to terrestrial, a/t)§
Incipient 13.8–10.9	64	94	62 (84%)	0.7	Projectiles, net sinkers, grinding stones	Freshwater, marine, terrestrial (a/t = 1.8)
Initial 11.1–8.0	15	80	50 (67%)	0.4	Projectiles, net sinkers and grinding stones	(N/A)
Early 7.9–5.1	80	84	50 (82%)	2.0	Net sinkers, grinding stones, projectiles	Marine, freshwater, terrestrial (a/t = 3.6).

*Greater than 0.2 μg·mg^{-1}.
†Pottery weights for the area excavated were estimated from analysis of a partial sample (12 m^2) with layers S1 to Z1. It is expressed relative to the amount of stone artifact.
‡Order of frequency.
§a/t: Species representation for aquatic versus terrestrial animals in terms of minimum number of individuals.

S1). These values are consistent with freshwater fish and may therefore indicate a change from marine species in the Incipient period to a mixture of freshwater, brackish and marine by the Early Jōmon. This shift corresponds to the establishment of the shell midden and may indicate a broadening of the aquatic species targeted at this juncture. However, the remnant macronutrient composition of the residue also influences $\delta^{13}C$ values, with lipids depleted in ^{13}C compared with proteins and carbohydrates (21); hence, our interpretation may be confounded by preferential loss of these different compound classes. To circumvent this problem, the stable carbon isotope ratio of two medium chain-length saturated alkanoic acids ($C_{16:0}$ and $C_{18:0}$) were determined individually using GC-c-IRMS in 52 charred deposits and 58 sherds (Fig. 3). These values were compared with references from authentic modern reference fats and extracted lipids from skeletal tissues of known species from the site (Fig. 3 and Table S2).

The GC-c-IRMS data confirm a predominant marine aquatic source in the Incipient Jōmon period pottery, which could be either open marine species, salmonids, or a mixture of these (Fig. 3*A*). The *n*-alkanoic acids extracted from the Holocene age samples (Fig. 3 *B* and *C*) are more variable and are consistent with reference values from freshwater fish/molluscs and nonruminants, such as wild boar (Fig. 3 and Table S2), although marine-derived residues are also represented. The correspondence between relatively low lipid $\delta^{13}C$ values and aquatic biomarkers strongly supports evidence for the processing of freshwater products. Interestingly, this freshwater signal is only observed in the Early Jōmon pottery (Fig. 3*C*), despite the fact that freshwater fish, particularly carp (Cyprinidae), are found throughout the sequence and the site is situated near freshwater and brackish lakes (Fig. 1). It is hard to determine whether freshwater fish or freshwater molluscs were processed in pottery at this time. The nitrogen isotope values of charred residues from Early Jōmon pottery with aquatic biomarkers are more consistent with reference values from higher trophic level fish than the freshwater molluscs (24, 32) that mainly comprise the midden, although a contribution from the latter cannot be completely ruled out or easily distinguished using lipid residue analysis.

A small number of vessels without aquatic biomarkers and with alkanoic acid $\delta^{13}C$ values consistent with both ruminant and nonruminant terrestrial animals are observed in all periods. Of the Initial Jōmon vessels analyzed, those without charred deposit showed an absence of aquatic biomarkers, whereas all charred surface deposits from this period produced the full range of these (Table S1). Moreover, this difference is supported by the stable carbon isotope signature of *n*-alkanoic acids, which are more enriched in the samples with charred surface deposits. These data may point to deliberate separation of terrestrial and aquatic resources and more dedicated pottery use during the Initial Jōmon period. There was no such correspondence between vessel use and the presence or absence of charred deposits during the other periods; here, aquatic biomarkers were readily formed in both surface deposits and within the vessel wall, although vessels without foodcrusts were rare in our sample. Nevertheless, despite some variation and possible ambiguity because of mixing, we conclude that aquatic foods were a dominant feature of pottery use in all periods; in only the minority of cases (<10%) can we rule out vessels used for this purpose.

None of the samples had very depleted ^{13}C *n*-alkanoic acids values, consistent with reference values from acorns obtained from Japanese forests (Fig. 3), despite the abundance of acorn macroremains in the Torihama deposits. Similarly, the atomic C:N values are not linearly correlated with $\delta^{13}C$ (Pearson's $R = -0.21$, $P = 0.0754$), as would be expected if ^{13}C-depleted starchy plant foods were making a significant contribution. The distributions of lipids observed in the pottery are also inconsistent with plant oils or waxes, although plant sterols and terpenes were occasionally observed at low abundance (Table S1). It is possible that low amounts of plant-derived lipids may have been masked by lipid-rich animal products. To investigate further, plant starch granules and phytoliths (silica bodies) were extracted from interior and exterior charred deposits from 15 Incipient and 6 Early Jōmon vessels using established protocols optimized for pottery residues (33, 34). In each case the number of starch granules (<1 count per mg^{-1}) and phytoliths (<10 count per mg^{-1}) were very low and significantly indistinguishable from exterior surface deposits [Kruskal–Wallis, χ^2(phytoliths) = 0.0385, $P = 0.8444$; χ^2(starch) = 1.6662, $P = 0.1968$] (Table S4). Although starch granules deteriorate during cooking, low counts of more thermally durable phytoliths were recorded in the same samples, supporting the proposition that an absence of plant remains was not a result of poor preservation. Although plant processing remains a possibility, as lack of evidence is always difficult to interpret, we argue that given: (*i*) the optimal organic conditions for preservation at Torihama, (*ii*) the fact that plant microfossils have been readily extracted from other examples of hunter–gatherer pottery (33, 34), (*iii*) the elevated bulk $\delta^{15}N$ values, (*iv*) the low atomic C:N ratios, and (*v*) the overwhelming molecular evidence for lipids derived from aquatic animals, the absence of significant plant processing in pottery from Torihama can be reasonably concluded.

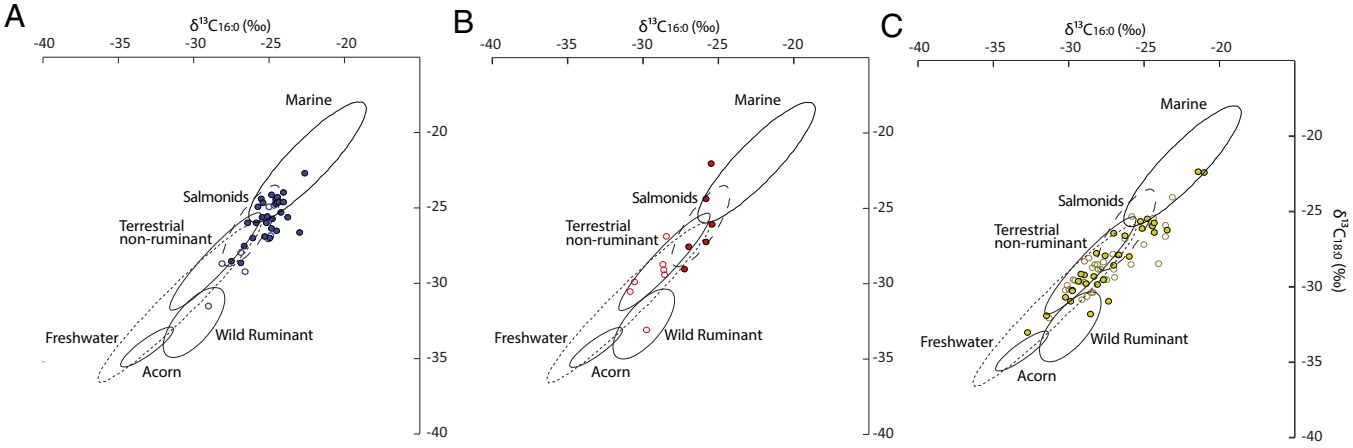

Fig. 3. $\delta^{13}C$ values of $C_{16:0}$ and $C_{18:0}$ *n*-alkanoic acids extracted in three phases of Jōmon pottery from Torihama: (*A*) Incipient Jōmon, (*B*) Initial Jōmon, (*C*) Early Jōmon. The data are compared with reference ranges for authentic reference lipids from modern tissues and archaeological bone (Table S2) (9, 22, 25–31) (66.7% confidence). Samples with the full range of aquatic biomarkers are shown by filled circles.

Discussion

The close and continued use of pottery for processing aquatic resources contrasts with shifts in artifact assemblages and faunal remains at Torihama. The exceptional preservation of organic materials in the waterlogged deposits at this site shows that a range of terrestrial plant and animal species were exploited in all periods, in addition to freshwater and marine species (12). Pollen from nearby Lake Suigetsu indicates that deciduous broadleaf forest was already established in the Late Pleistocene and persisted even during the much cooler conditions that prevailed during the Younger Dryas, corresponding to the Incipient Jōmon phase at Torihama (35). Nevertheless, the resurgence of forests in the early Holocene (Initial Jōmon) must have greatly increased opportunities for hunting terrestrial animals and exploiting nut-bearing trees. Grinding stones for preparing plant foods, projectiles for hunting, as well as acorns, water-chestnut, wild boar, and sika deer are found throughout the sequence. The relative importance of these terrestrial-based activities compared with fishing and shellfish collection is hard to accurately assess. Storage pits filled with acorns and an increase in the number of grinding stones relative to other lithic artifacts characterize the Early Jōmon layers (36) and provide the clearest evidence for economic change driven by an increase in the exploitation of forest products. This change also corresponds to an increase in the abundance of pottery relative to lithic artifacts (Table 1) and the start of significant shellfish exploitation. Despite these changes, a broad subsistence strategy is observed throughout the sequence and clearly contrasts with our evidence for specialization in the use of pottery.

It has been argued that the sharp increase in the frequency of pottery across the Japanese archipelago at the start of the Holocene (37) was associated with new uses for pottery related to the exploitation of a wider range of food products that became available with climatic amelioration. Within this context, the lack of evidence for plant foods in the majority of vessels analyzed throughout the sequence at Torihama is particularly interesting, because protracted boiling of nuts to remove toxic tannins and saponins is often cited as a major driver for the uptake of early ceramics (19, 37–39). The paucity of fat-rich ruminant products in pottery, which are easily distinguishable by GC-c-IRMS (25), is similarly intriguing. Given their size and abundance, sika deer in particular made a substantial contribution to diet and are found in all phases at Torihama (12), but fat from this source could only be clearly identified in three samples (Fig. 3). Nor does it seem likely, given the relatively high ^{15}N values observed in the charred deposits, that exploitation of molluscs was the main driver for increased pottery production, as has been suggested (40), even though freshwater molluscs were heavily exploited at Torihama. Instead, the direct evidence of pottery use reported here supports the idea that pottery was invented in the late glacial period with the aim of processing a broader range of aquatic products (9) and that it retained this primary function at least until the mid-Holocene. Such functional resilience in the use of pottery in the face of altered environmental conditions, dramatic changes in the scale of manufacture, as well as proliferation in form and design, is remarkable.

The association between fishing and the hunting of aquatic mammals and pottery production may be a broader feature of preagricultural communities. Similarly high $\delta^{15}N$ values have been found in charred deposits on Jōmon pots throughout the Japanese archipelago (9, 19, 20). Lipid residue analysis has shown that marine and freshwater products were frequently processed in pottery produced by Holocene hunter–gatherers from Northeastern North America (22) and the Baltic (23, 41), and in Japan as late as the Final Jōmon phase (1000–400 B.C.) (42). Because the earliest Incipient Jōmon pottery vessels were relatively small, typically 1–2 L (43), and were only produced in low numbers, their effectiveness for substantially increasing aquatic resource production is questionable. Our findings are more consistent with the view that pottery was initially a "prestige technology" with a limited range of uses for special foods for aggrandizing or in competitive feasting (4), particularly during periods of high resource abundance and social aggregation. Practically, pottery may have facilitated the rendering and storage of highly prized aquatic oils during seasonal gluts of fish that occur during short-lived episodes of spawning or migration, in concert with other larger perishable containers, as has been documented historically (44). However, it is interesting that this specialized function did not change substantially as new forms emerged and pottery became more abundant and easier to produce during the Holocene, unless the perceived "value" of aquatic foods also changed through time. A broadening of the types of aquatic resources processed in pottery in the Holocene to encompass freshwater and brackish species provides the only evidence that the tight control governing pottery use was relaxed. Increases in the size and diversity of pottery in the Early Jōmon may well reflect increased ability to obtain surplus fish, to control labor, and an increased demand for fish oil for more elaborate and diverse feasting contexts.

Regardless of the significance or scale of the activity, our study shows that pottery retained its primary function despite substantial warming at the start of the Holocene, increased exploitation of the burgeoning forests, increased sedentism, and the proliferation of artifacts associated with plant processing and fishing. For this to happen, we suggest that pottery production, specifically for the exploitation of aquatic resources, must have been embedded in the social memory of these East Asian foragers for thousands of years, as a cultural norm. This dependable strategy was used by successive generations, perhaps to mitigate against risks associated with environmental change, the adaptation to new forms of subsistence, social transformation, and changes in territorial control. We hypothesize that this same functional driver was at least partly responsible for the long-distance spread of pottery westwards across Eurasia through lacustrine and riverine ecological corridors in the early Holocene (45). However, this needs formal testing.

Methods

We obtained 143 ceramic vessels from nine different stratigraphic phases at Torihama (Table S1). Each phase was dated by the associated pottery typology and independently through radiocarbon dating of associated organic artifacts (Table S3).

Lipid Analysis of Ceramic Samples. Lipids were extracted and methylated in one step with acidified methanol (46, 47). Briefly, methanol was added to 99 homogenized charred deposits (1 mL to 10–30 mg) and 57 ceramic powders drilled (d. 2–5 mm) from the sherd surface (4 mL to 1 g). The mixture was sonicated for 15 min, and then acidified with concentrated sulphuric acid (200 μL). The acidified suspension was heated in sealed tubes for 4 h at 70 °C and then allowed to cool. The lipids were then extracted with *n*-hexane (3 × 2 mL), and directly analyzed by GC-MS and GC-C-IRMS using standard conditions and protocols (16, 25, 48). Alternatively, lipids from four homogenized charred deposits were extracted by alkali saponification [2 mL of sodium hydroxide (5% [wt/vol] in methanol) for 2 h at 70 °C]. Saponified extracts were cooled, neutral lipids were removed (*n*-hexane, 3 × 2 mL), the extracts were acidified with HCl and the acid fraction was extracted (*n*-hexane, 3 × 2 mL) and methylated using BF_3-methanol complex [14% (wt/vol), 200 μL, 1 h, 70 °C]. For GC-C-IRMS, instrument precision on repeated measurements was ±0.3‰ (SEM) and the accuracy determined from in-house FAME and *n*-alkane isotope standards was ±0.5‰ (SEM). All $\delta^{13}C$ values are expressed in per mille (‰) relative to the Vienna PeeDee Belemnite international standard. Where sufficient sample remained, solvent extraction was also carried out on drilled pottery sherds and crushed surface residues. Samples were sonicated three times with DCM:MeOH (2:1, vol/vol). The extracts were combined and evaporated to dryness under a stream of N_2. Solvent extracted and a selection of acid-methanol–extracted samples were silylated with BSTFA at 70 °C for 1 h, and then evaporated to dryness under a gentle stream of N_2. Derivatised samples were redissolved in *n*-hexane and analyzed directly by GC-MS, as described previously (16, 25, 48).

Lipid Analysis of Faunal Remains. Lipids were extracted and analyzed by GC-c-IRMS from selected faunal remains at Torihama to provide additional comparative reference data (Fig. 3 and Table S2), using previously described procedures (29). Exogenous lipids were first removed with dichloromethane/methanol (2:1 vol/vol; 3 × 2 mL) from each bone sample (~1 g). After each solvent addition, samples were ultrasonicated for 15 min and then centrifuged at 867 × *g* for 10 min and the supernatant was removed. The remaining bone powder was dried completely

under gentle stream of N_2 and then extracted with a selection of modern animal tissues (~30 mg) and experimental cooking vessel (approximately 1 g) using the acid-methanol procedure as outlined above for ceramics. The methylated fatty acids were analyzed by GC-C-IRMS as described above. Modern reference samples were further corrected for the burning of fossil fuels (49) to allow comparison with archaeological data.

Bulk Isotope Analysis. Crushed surface residues (~1 mg) were analyzed by elemental analysis IRMS as previously reported (48). Samples yielding less than 1% N were discarded and instrument precision on repeated measurements was ±0.2‰ (SEM). $\delta^{13}C$, $\delta^{15}N = [(R_{sample}/R_{standard} - 1)] \times 1{,}000$, where $R = {}^{13}C/{}^{12}C$ and ${}^{15}N/{}^{14}N$. All sample measurements are expressed in per mil relative to Vienna PeeDee Belemnite for $\delta^{13}C$ values and air N_2 for $\delta^{15}N$ values.

Plant Microfossil Analyses. Surface residues (approximately 1.5–7 mg) were treated with H_2O_2, 3% (vol/vol), 10 mL, 15–30 min, and manually disaggregated. Samples were then centrifuged (1,000 × *g*, 3 min) and the supernatant reduced to 2 mL. The remaining residues were washed three times with UltraPure water and made up to 1-mL suspensions. This supernatant, containing liberated phytoliths and starches, was added to microscope slides and left to dry at room temperature. Samples were mounted in glycerol before viewing in rotated planes using an inverted polarizing microscope fitted with a digital camera. All silica bodies and starches were counted by scanning the mounted specimen in a grid pattern at a magnification of 400×.

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