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# Pottery Lipids Demonstrate the Marine Product Tribute in Early Historic Japan

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## Abstract

In the 7<sup>th</sup> century CE, Japan adopted a new bureaucratic system called *Ritsuryo*. This system was partly supported by the taxation or tributes of local products ranging from agricultural, marine or even dairy products from rural areas to the capital in Nara. However, the reconstruction of these tributes is mainly based on the written record and little material evidence has been secured to discuss further how and where these tribute items were produced. Here, we focus on a bonito sauce recorded under the name of *Nikatsuo* (boiled bonito) and *Katsuoirori* (bonito broth), the major tribute product from the Pacific coast. We analysed lipids extracted from large-sized ceramic cooking vessels known as *Nabe* excavated from archaeological sites in the Suruga Bay area, that were thought to have been used to produce bonito products on an industrial scale. Clear evidence of marine resource processing is confirmed in these vessels through the presence of chemical compounds related to aquatic organisms and stable carbon isotope values of individual fatty acids, indicating large-scale bonito products processing at these sites. The results newly point to the possibility that such large-scale production and tribute of bonito products may have been practised as early as the Asuka period, earlier than the Nara period as previously recognised.

## Keywords

Organic residue analysis, Asuka period, Nara period, *Ritsuryo* bureaucratic system, Fish sauce

## Introduction

During the 7<sup>th</sup> and 8<sup>th</sup> centuries CE, a centralised state government was first established in Japan. It adhered to the Chinese (Sui and Tang Dynasties) bureaucratic system of *Ritsuryo*, superseding traditional rules and customs of ancient societies. This state was characterised by the balance of advanced, international bureaucracy and traditional, indigenous social structure, both of which were inextricably linked and heavily influenced one another (Baba 2019).

This characteristic can be seen in the taxation system. For instance, although a "*chou*" in the *Ritsuryo* system is a tax, foodstuffs are enumerated in addition to the original monetary items. This was likely to secure ritual rights that formed the core of traditional rule (Otsu 1999). The tribute of tax items was often accompanied by a wooden tablet (Figure 1). Many wooden tablets have been excavated from places where tributes were paid, namely the Fujiwara and Heijo Capitals in Nara, located in the middle of Japan. The inscriptions on these tablets gave an approximate idea of the details of the tribute payments (e.g. agricultural, marine, dairy products etc.) and their source (e.g. place names).

Among them, the item that clearly shows the characteristics of a tribute is processed bonito. Bonito (*Katsuwonus pelamis* L.), a large pelagic fish, has been exploited across the Pacific for at least forty thousand years (O'Connor, Ono and Clarkson 2011; Ono 2017: 84) and has been an important food source for humans throughout the Holocene (Keneko 1965). When processed, the fish provides a fundamental umami taste in Japanese cuisine, as it contains inosinic acid (Kurihara 2015; Okamoto, Tayama, and Furuta 2020). While *katsuo-bushi* (dried fermented bonito) is known to have been produced during the Edo period of the 17<sup>th</sup> to 19<sup>th</sup> centuries CE (Ehara and Higashiyotsuyanagi eds. 2011), its prototypes production and its economic and culinary significance in earlier periods are less well understood.

While bonito is widely caught in the Pacific Ocean, sources of bonito were clearly concentrated in the *Suruga* and *Izu* provinces that is today's Shizuoka prefecture (Figure 2). It is even said that *Izu* Province was established as an administrative division specifically for bonito tribute, ensuring a stable supply of bonito for the state (Nito 1996a), although other products such as rice, coins, seaweeds and mandarins were also known as tributes base on the excavated wooden tablets (Figure 1). This suggests a reorganisation of the production system by the state and the assignment of roles to different regions. However, direct evidence for bonito products production during the 7<sup>th</sup> and 8<sup>th</sup> centuries is lacking. Bonito requires thorough processing to be preserved for long periods, enabling transportation from source to destination of tribute payment and subsequent use. According to the historical legal documents and wooden tablets, there are three methods of bonito processing for tribute: *Arakatsuo* (dried salted bonito), *Nikatsuo* (dried boiled bonito), and *Katsuoirori* (condensed bonito broth). Whatever the process used, the latter two requires an extensive production system including the supply of fuel and specialised wooden and ceramic equipment

The bonito production system is not simply dependent on the natural environment and traditional local livelihoods but on state strategy. Accordingly, detecting archaeological evidence of the production of boiled bonito in *Suruga* and *Izu* provinces implies the *Ritsuryo* system was in operation and therefore key information regarding the formation process of the ancient state system in Japan (Nito 1996a) that has been discussed based solely on written records. Previous studies have paid attention to a distinctive type of ceramic cooking vessel known as *Nabe* excavated from coastal Suruga Bay (Figure 2). This type of pottery is found only in the coastal areas of Shizuoka Prefecture and adjacent areas, with more than 600 individual pottery excavated from 60 sites. These are typically of 15 to 20 litre volume, which is much larger than other normal boiling vessels, typically 8 to 10 litre volume in this region.

These vessels have long been suggested to have been used to cook bonito products listed on wooden tablets in the Nara period, given that they are frequently found in dwellings and possess cooking traces (Segawa 1980). In particular, large-scale settlement sites, such as Fujiibara and Miyukicho that are analysed in this study, may have had a role as fish processing centres in a provincial government office that produced bonito products to transport to the Heijo Capital (Hashiguchi 1987, Okamoto 1989, Nito 1996b, Numazu City ed. 2005, Kameya 2011). In addition to their large size, the metal ornament and inkstone excavated at the Miyukicho site and the large-sized storage houses documented at the Nakahara site support the character of these sites as governmental. The *Nabe*-shaped pottery was produced from the late 7<sup>th</sup> to the 10<sup>th</sup> centuries, suggesting a close relationship with the start of the *Ritsuryo* taxation system (Fujimura 2021 & 2023; Fujimura and Kozaki 2023). The area in which the pottery is mostly concentrated is the *Izu* and *Suruga* Provinces, matching the area of bonito tribute to the Heijo capital (Hashiguchi 1987).

Segawa and Koike (1990) analysed fatty acids and sterols from *Nabe*-shaped vessels, noting that they may have been used for processing migratory fish. However, their identification of the fatty acids was made by comparison with a modern specimen analysed by Gas Chromatography - Flame Ionisation Detection (GC-FID). As this technique does not allow direct identification of compounds,

and the correspondence between retention times of compounds in the archaeological and modern samples were not reported, it is not possible to verify their results. In addition, This work reported free fatty acids C<sub>20:4</sub> (eicosatetraenoic acid) and C<sub>22:6</sub> (docosahexaenoic acid) in their archaeological samples, yet these compounds are unlikely to be preserved in archaeological organic residues as they are readily oxidised in the burial environment (Evershed et al. 1992).

Organic residue analysis of Japanese pottery has witnessed dramatic improvements in recent decades (Craig et al. 2013; Lucquin, Gibbs, et al. 2016; Lucquin et al. 2018), enabling us for example to robustly determine the isotopic range of individual fatty acids from marine fish oils in archaeological ceramics. Harnessing recent advances in GC-MS approaches for the identification of aquatic resources (Evershed 2008; Cramp & Evershed 2014; Lucquin, Colonese et al 2016) combined with compound-specific stable carbon isotope analysis, here we applied these contemporary methodologies to *Nabe*-shaped boiling vessels to test the hypothesis that they were used to boil marine fish such as bonito. This is also a new application to Japanese archaeology in that it sheds new light on historical periods and foodstuff trade that have not been studied before. While it is not possible to distinguish fish species solely based on lipid residues, detecting marine oils from these pottery with clear contexts would certainly reinforce this hypothesis.

## Material and methods

### *Sampling ceramic powder*

A total of 39 *Nabe*-shape vessel fragments, from six large-scale settlement sites in the Suruga Bay area, were sampled by drilling out two grams of ceramic powder for each, using sterilised drill bits, after removing the surface that had potential contaminants before sampling. The *Nabe*-shaped pottery tends to be found near the cooking hearth in dwelling pits, therefore an intensive cooking function is assumed. All selected sample pottery sherds were excavated from within the dwelling pits.

In particular, it has been pointed out that the Nakahara, Fujiibara and Miyukicho sites may have a higher degree of specialization than other settlements sites based on the size of dwelling houses in these sites and additional storage buildings (Fujimura and Kozaki 2023). Notably, these sites are divided into two typo-chronological phases; 1) 7<sup>th</sup> century CE sites such as Nakahara and Sawahigashi A, and 2) 8-9<sup>th</sup> centuries CE sites such as Miyukicho, Fujiibara, Sanshinden and Kashiwabara. While the latter period has wooden tablet record of bonito tribute from this area, the former still lacks written evidence of such practice.

These sherds were excavated in the 1970s-2000s (Numazu City Board of Education 1978, 1998, 2016; Fuji City Board of Education 1997, 2000, 2002), washed with tap water, and then stored in plastic containers. Site locations are shown in Figure 2 and typical vessel forms are shown in Figure 3. As shown in the Figure 4, taking an example of the Nakahara site, large-scale dwelling houses are distributed in these sites and the *Nabe*-shaped vessels were excavated from such structures. The site and sample summaries are presented in Table 1.

### *Modern fish sauce sample*

To compare with the archaeological samples, modern product of *Katsuoirori* was analysed using the same protocol but in a separate batch. This product is produced by Kanesa Katsubushi Shoten (Kanesa Bonito Shop) located in Tago, Nishiizu-cho, Kamo-gun, Shizuoka Prefecture. The bonito used in this product is caught from nearby sea. To make *Katsuoirori*, approximately 150 kg of bonito's head and backbone are carefully simmered in a large pot to avoid burning and skimming the scum off the top. The broth is gradually filtered during the simmering process to remove excess bones and other

materials. The resulting bonito broth is then further simmered until it becomes a highly viscous concentrated liquid. Five mg of this product was sampled for the analysis.

#### *Lipid residue extraction*

Acidified methanol extraction was performed following established methods (Craig et al. 2013; Correa-Ascencio et al. 2014). Approximately 4 mL of methanol (CH<sub>3</sub>OH) was added to clean glass vials containing around 1 g ( $\pm$  5 %) of ceramic powder and 10  $\mu$ L of a *n*-tetratriacontane internal standard (1  $\mu$ g  $\mu$ L<sup>-1</sup>) before being placed in an ultrasonic bath (25 °C, 15 minutes). After taking out from the bath, approximately 800  $\mu$ L of sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) was added to each vial before they were placed in a heating block set at 70 °C for 4 hours. After cooling the vials to room temperature, they were centrifuged (4000 rpm, 5 minutes). The resulting supernatant was then transferred to another clean vials. Approximately 2 mL of hexane (C<sub>6</sub>H<sub>14</sub>) was added to the second vials, mixed using a vortex mixer, and allowed to stand for static separation. The supernatant layer of hexane, which contained the extracted lipids, was transferred to a third vial. The process of adding, mixing, and transferring hexane was repeated two more times. The extract was gently dried under a gentle stream of nitrogen. The dried extract was transferred into a GC vial containing 10  $\mu$ L of another internal standard *n*-hexatriacontane (1  $\mu$ g  $\mu$ L<sup>-1</sup>) in two steps (first: 90  $\mu$ L, second: 100  $\mu$ L) using hexane, to produce a final extract suspended in 200  $\mu$ L of hexane. One blank sample was included in each batch to identify any potential contamination that may have occurred during the extraction process, as well as a standard sample with known isotopic values of fatty acids to correct values obtained by GC-c-IRMS.

#### *GC-MS (Gas Chromatography - Mass Spectrometry)*

Extracts were analysed by gas chromatography – mass spectrometry using a Shimadzu GCMS-QP2010 Ultra (Shimadzu, Kyoto, Japan). The inlet temperature was set to 300 °C and 1  $\mu$ L sample was introduced to the GC-MS using splitless injection. Ultra ALLOY-5 (Frontier Laboratories Ltd., Japan: 30 m  $\times$  0.25 mm, film thickness 0.25  $\mu$ m) column was used. The oven temperature was set to 50 °C for 2 min, then raised to 325 °C, at 10 °C/min, and held for 12 min. The *m/z* scan range was between 50 and 800, with a total acquisition time of 41.5 min. The ion source temperature was set to 230 °C and ionization voltage to 70 eV. Helium was the carrier gas, with a flow rate of 3 ml/min.

Extracts were also analysed in selected ion monitoring (SIM) mode for the detection of aquatic biomarkers (Evershed 2008; Lucquin, Gibbs, et al. 2016). The inlet temperature was set to 300 °C and 1  $\mu$ L sample was introduced to the GC-MS using splitless injection. DB-23 (J&W Scientific, Folsom, CA, USA: 60 m  $\times$  0.25 mm, film thickness 0.25  $\mu$ m) column was used. The oven temperature was set to 50 °C for 2 min, raised to 160 °C at 0.5 °C/min, then raised to 250 °C at 20 °C/min, and held for 13.5 min with a total acquisition time of 75.0 min. Selected ions were classified into four groups: *m/z* 74, 87, 213, and 270; 74, 88, 101, and 312; 74, 101, 171, and 326; and 74, 105, 262, 290, 318, and 346 for the detection of 4,8,12-trimethyltridecanoic acid (TMTD); pristanic acid; phytanic acid; and  $\omega$ -(*o*-alkylphenyl) alkanolic acids of carbon length C<sub>16</sub> to C<sub>22</sub> respectively (Lucquin et al. 2018). The ion source was set to 230 °C and ionization voltage to 70 eV. Helium was used as the carrier gas, with a flow rate of 4 ml/min.

#### *GC-c-IRMS (Gas Chromatography-combustion-Isotope Ratio Mass Spectrometry)*

GC-c-IRMS measurements of fatty acid methyl esters C<sub>16:0</sub> and C<sub>18:0</sub> were undertaken using a Agilent 7890B series Gas Chromatograph (Agilent Technologies, Cheadle, Cheshire, UK) linked to Isoprime

100 (Isoprime, Cheadle, UK) via the Isoprime GC5 interface (Isoprime Cheadle, UK). For the GC, 1  $\mu\text{L}$  of sample (in hexane) was injected at 300 °C by splitless mode. DB-5MS UI column (PN 122-5562UI; 60 m  $\times$  250  $\mu\text{m}$   $\times$  0.25  $\mu\text{m}$ ; J&W Scientific technologies, Folsom, CA, USA) was used and the carrier gas (ultra-high purity grade helium) set at constant flow of 2 mL/min. The temperature started at 50 °C (for 0.5 min), then increased by 25 °C min<sup>-1</sup> up to 175 °C, then by 8 °C min<sup>-1</sup> up to 325 °C. The final temperature was maintained for 20 min. The column flow was directed through the GC5 furnace tube (CuO) maintained at 850 °C to oxidise all the carbon species to CO<sub>2</sub>. 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 <sup>13</sup>C / <sup>12</sup>C ratio of each peak. Computation was made with IonVantage and IonOS softwares (Isoprime, Cheadle, UK). Results 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  $\pm$  S.D. values of these were  $-29.87 \pm 0.24\text{‰}$  and  $-23.47 \pm 0.04\text{‰}$  for the methyl ester of C<sub>16:0</sub> (reported mean value vs. VPDB  $-29.90 \pm 0.03\text{‰}$ ) and C<sub>18:0</sub> (reported mean value vs. VPDB  $-23.24 \pm 0.01\text{‰}$ ), respectively.

## Results

Interpretable quantities of lipids ( $> 5\mu\text{g g}^{-1}$ , Craig et al. 2013) were obtained from 27 (69%) of the 39 samples analysed. However, it was difficult to find any significant difference in lipid concentrations between different periods and sites. Although the Kashiwabara site showed relatively higher concentration, it has only 2 samples thus it is difficult to assess its statistical significance (Figure 6).

A typical chromatogram obtained by GC-MS is shown in Figure 7. Compounds that are likely modern contaminants, including phthalates and fatty alcohols (Whelton et al. 2021), were identified in several samples. They are presumed to derive from events prior to this analysis such as during the curation and storage as the blank control samples did not contain these contaminants. Some degree of contamination is perhaps unavoidable, given that these ceramics were wrapped and stored in plastic materials. Nevertheless, the extent is not serious enough to affect the interpretation.

Low abundances of C<sub>18</sub>  $\omega$ -(*o*-alkylphenyl)alkanoic acids (C<sub>18</sub>-APAAs) were identified in samples from Fujiibara ( $n = 3$ ) and Miyukicho ( $n = 2$ ). These compounds are formed through the heating of unsaturated fatty acids (Hansel et al. 2004; Evershed et al. 2008). Ratios of E and H C<sub>18</sub>-APAA isomers have been shown to vary between animal, leafy plant, and other plant products, potentially enabling broad distinction between residue types (Bondetti et al. 2020; Shoda et al. 2021). In addition, it was suggested that the APAA C<sub>20</sub>/C<sub>18</sub> ratio could be used to distinguish aquatic sources from terrestrial products with values above 0.06 associated with aquatic products (Bondetti et al. 2020). In the samples from Miyukicho, sample MYC001 yielded an APAA C<sub>20</sub>/C<sub>18</sub> ratio of 0.12, thereby meeting this criterion. Also, the E/H value of the same sample (4.7) and of sample MYC007 (3.6) are consistent with the interpretation of aquatic resources being the main source of these residues. On the other hand, the abundance of E and H isomers in the Fujiibara samples were too low for the quantification.

The relative abundances of iso- and anteiso-branched chain C<sub>15</sub> (C<sub>15</sub>ivstot = i15:0/ (i15:0 + a15:0)) and C<sub>17</sub> (C<sub>17</sub>ivstot = i17:0/ (i17:0 + a17:0)) fatty acids were calculated for each extract in order to assess the contribution of aquatic resources to residues (Demirci et al. 2021). The values (C<sub>15</sub>ivstot =  $0.58 \pm 0.14$ ; C<sub>17</sub>ivstot =  $0.59 \pm 0.15$ ) obtained in this study are similar to those reported for archaeological organic residues that comprise aquatic biomarkers and modern fish; C<sub>15</sub>ivstot = 0.61 and mean C<sub>17</sub>ivstot = 0.53 (Demirci et al. 2021) and C<sub>15</sub>ivstot =  $0.59 \pm 0.1$  and C<sub>17</sub>ivstot =  $0.59 \pm 0.5$  (Hauff and Vetter 2010). However, it is undeniable that the reference range corresponding to the

exhaustive resource is not known, and there is insufficient data to use this value as the basis for resource identification.

Isoprenoid fatty acids, including phytanic, pristanic, and 4,8,12-trimethyltridecanoic acid (4,8,12-TMTD), are employed as biomarkers for aquatic products in the investigation of archaeological organic residues when they are accompanied by APAA C<sub>18</sub>, C<sub>20</sub> and sometimes C<sub>22</sub> (Evershed 2008; Lucquin et al. 2016). While phytanic acid may derive from both aquatic and terrestrial ruminant resources, calculating the relative abundance of SRR and RRR phytanic acid isomers can enable a certain degree of distinction between sources (Lucquin et al. 2016). The SRR/RRR ratios of the analysed samples are clearly lower than values typically associated with both ruminant and aquatic sources. Indeed, there is a significant difference between our results and those of aquatic reference materials, requiring further research (t-test,  $p < 0.05$ ). Nevertheless, 4,8,12-TMTD is recognised to only occur in the aquatic environment (Cramp & Evershed 2014) and to be useful to determine whether the samples originated from marine organisms. A total of 27 (69%) of samples comprised 4,8,12-TMTD, demonstrating that aquatic products were highly likely processed in these vessels regardless of their chronological phase. Yet, as the full criteria of the aquatic biomarkers are not obtained from these samples, interpretation must be done with caution.

Stable carbon isotope ratios of palmitic and stearic acids were measured by GC-c-IRMS for each sample. All samples, including the modern one, demonstrated substantial enrichment (-25.5 to -20.9‰ for  $\delta^{13}\text{C}_{16:0}$  and -24.3 to -22.0‰ for  $\delta^{13}\text{C}_{18:0}$ ) and correspond to the reference range of marine resources (Figure 8). There is little difference between the 7<sup>th</sup> century and the 8<sup>th</sup>-9<sup>th</sup> century samples, indicating continuity of function.

Figure 9 shows the partial chromatogram of the extract from the modern bonito broth, obtained by the scanning mode of GC-MS analysis. Omega-3 fatty acids such as EPA (eicosapentaenoic acid), which is generally found in aquatic organisms was identified. However, APAAs were not detected, while isoprenoid fatty acids, such as phytanic, pristanic acids, and 4,8,12-TMTD were detected by the SIM mode. The phytanic acid SRR ratio was found to be significantly lower than both the values obtained in the present study and the values of reference samples (Figure 10). Stable carbon isotope ratio of palmitic and stearic acids, on the other hand, plotted within a range close to that of marine resources (Figure 8).

## Discussion and Conclusions

### *Cooking marine fish in pottery*

The results demonstrate the potential processing of marine resources, in at least 69% of vessels analysed, through the detection of 4,8,12-TMTD (Cramp and Evershed 2014), the relative abundance of iso- and anteiso- branched chain fatty acids of C<sub>15</sub> and C<sub>17</sub>, and observation of extensive  $\delta^{13}\text{C}$  enrichment among individual fatty acids, corresponding to modern marine references (Lucquin et al. 2018). Furthermore, the presence of C<sub>18</sub>-APAAs in five samples clearly indicates that these vessels were heated, most likely for cooking, given the excavated contexts of these pottery found nearby cooking hearths. Despite the limited numbers of samples in which these compounds were identified, APAAs C<sub>20</sub>/C<sub>18</sub> ratios and E/H values of C<sub>18</sub> APAAs support the interpretation of aquatic oil being the main source of these residues. Importantly, this tendency was observed in both chronological periods, highlighting the continuity of marine product processing using this vessel type.

It is difficult to determine the proportion of bonito caught at the time as archaeozoological findings are limited in this area due to the unfavourable soil conditions for bone preservation. Nevertheless, the marine topography of Suruga Bay provides ideal conditions for large migratory fish to approach the shore, as the topography of the seabed in the vicinity stirs up warm currents. Indeed, in

modern times, large-scale fishing for migratory fish such as bonito and tuna has been extremely active in this area (Numazu History and Folklore Museum 2006). Furthermore, iron fish hooks and large clay net sinkers have been excavated from Nakahara, indicating that fishing certainly took place in the gulf nearby the studied area.

#### *The interpretation of SRR ratios*

SRR/RRR ratios of phytanic acids have been effectively used for distinguishing ruminants from aquatic resources in extracted lipid residues (Lucquin, Colonese, et al. 2016; Shoda et al. 2017). However, SRR/RRR ratios obtained in this study are generally lower than contemporary aquatic samples, which is inconsistent with other evidence for marine resource processing such as  $\delta^{13}\text{C}$  enrichment among individual fatty acids. Notably, Lucquin et al. (2016) only analysed SRR/RRR ratios among four different groups of aquatic species, namely, *Babylonia* sp. (marine gastropod), *Globicephala* sp. (Pilot whale), *Venerupis philippinarum* (marine bivalve), *Salmonidae* sp. (salmon). None of these species are migratory fish, such as bonito, which could be a potential reason for this contradiction. Interestingly, although it is limited to only one measurement, the analysis of the modern *Katsuoirori* in this study showed significantly lower values compared to the reference presented by the study (Lucquin et al. 2016). Currently it is difficult to interpret this low ratio because it is assumed that the origin of phytanic acids is marine phytoplankton, which should also be at the base of the aquatic foodchain for migratory species such as bonito. Further research is necessary to assess phytanic acid concentrations and isomeric ratios among different fish species.

#### *Bonito production under the ancient tribute system*

*Nabe*-shaped pottery is thought to have been highly specialised, serving a specific purpose, as their size and common archaeological context being excavated near cooking structures in large-size settlement sites. Additionally, the large-scale production system and governmental nature of Fujiibara and Miyukicho sites suggest that their products were closely linked to state administration. Written records suggest that the sources of bonito tribute to the Heijo Capital in the Nara period were clearly concentrated in the *Suruga* and *Izu* Provinces and the *Nabe*-shaped pottery was likely used for tribute bonito products.

Although it is difficult to identify whether the detected marine lipids derive from either bonito or other fish species, the clear evidence of marine resource processing in this context strongly indicate that these sites and vessels were involved in the administrative production of boiled bonito or/and bonito broth. This is the first indication from the material cultural side to illuminate how and where the tribute items were produced during the initial development stage of ancient Japanese taxation system. Significantly, our findings newly illuminated that this tradition dates back to the 7<sup>th</sup> century Asuka period.

Lastly, until now, pottery lipid residue analyses were limited to prehistoric materials at least in East Asia. This study applies for the first time this approach to the pottery from historic periods, highlighting the usefulness of this method for the interpretation of historic activities that are often predominantly understood through written records. Further application of this technique to detect other tribute products such as different kinds of agricultural products from different regions, will enable us to reconstruct the ancient state bureaucratic systems through the investigation of material culture and biomolecules bequeathed to them.

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### References Cited

Baba, Hajime.

2019 Explore the Heijo Capital. *Ancient capital*: 93–155, eds. Yoshimura, Takehiko., Yoshikawa, Shinji and Kawajiri Akio. Tokyo: Iwanami Shoten, Publishers (in Japanese).

Bondetti, Manon., Erin Scott, Blandine Courel, Alexandre Lucquin, Shinya Shoda, Jasmine Lundy, Catalina Labra-Odde, Léa Drieu and Oliver E. Craig

2021 Investigating the formation and diagnostic value of  $\omega$ -(o-alkylphenyl)alkanoic acids in ancient pottery. *Archaeometry* 63(3): 594–608.

Correa-Ascencio, Marisol. and Richard P. Evershed

2014 High throughput screening of organic residues in archaeological potsherds using direct acidified methanol extraction. *Analytical Methods* 6 (5): 1330–1340.

Corr, Lorna T., Michael P. Richards, Susan Jim, Stanley H. Ambrose, Alexander Mackie, Owen Beattie and Richard P. Evershed

2008 Probing dietary change of the Kwäday Dän Ts' inč̓ individual, an ancient glacier body from British Columbia: I. Complementary use of marine lipid biomarker and carbon isotope signatures as novel indicators of a marine diet. *Journal of Archaeological Science* 35(8): 2102–2110.

Craig, Oliver E., Hayley Saul, Alexandre Lucquin, Yastami Nishida, Karine Taché, Leon Clarke, Anu J. Thompson, David. T. Alftoft, Junzo Uchiyama, Mayumi Ajimoto, Kevin Gibbs, Sven Isaksson, Carl P. Heron and Peter Jordan

2013 Earliest evidence for the use of pottery. *Nature* 496(7445): 351–354. doi: 10.1038/nature12109

Cramp, Lucy and Richard P. Evershed

2014 Reconstructing Aquatic Resource Exploitation in Human Prehistory Using Lipid Biomarkers and Stable Isotopes. *Treatise on geochemistry: archaeology and anthropology*: 319–339.

Demirci, Özge., Alexandre Lucquin, Canan Çakırlar, Oliver E. Craig and Daan C.M. Raemaekers

2021 Lipid residue analysis on Swifterbant pottery (c. 5000–3800 cal BC) in the Lower Rhine-Meuse area (the Netherlands) and its implications for human-animal interactions in relation to the Neolithisation process. *Journal of Archaeological Science* 36: 102812.

Evershed, Richard P., Carl Heron, Stephanie Charters and L. John Goad

1992 The Survival of Food Residues: New Methods of Analysis, Interpretation & Application. *In Proceedings of the British Academy* 77: 187–208.

Evershed, Richard P., Mark S. Copley, Luke Dickson and Fabricio A. Hansel  
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.

Fuji City Education Board  
1983 *Excavation Survey Report of the Sanshinden Site*. Shizuoka: Fuji City Education Board (in Japanese).

Fuji City Education Board  
1997 *Excavation Report of the Sawahigashi A Site, Zone 5*. Shizuoka: Fuji City Education Board (in Japanese).

Fuji City Education Board  
2000 *Excavation Report of the Sanshinden Site, Zone D*. Shizuoka: Fuji City Education Board (in Japanese).

Fuji City Education Board  
2002 *Excavation Report of Archaeological Sites within Fuji City - Fiscal Years 2010 and 2011*. Shizuoka: Fuji City Education Board (in Japanese).

Fujimura, Sho.  
2021 Structure and evolution of ancient settlements in Suruga Province Fujigun. *Structure and Evolution of Ancient Settlement* 1(1): 109–133 (in Japanese).

Fujimura, Sho.  
2023 Kofun construction and regional development in the Suruga and Izu regions: Focus on the 6th and 7th centuries. *Kofun Construction and Regional Development*: 17–32 (in Japanese).

Fujimura, Sho. and Susumu Kosaki  
2023 Characteristics and distribution of Nabe-shaped pottery in the Suruga and Izu regions in ancient times. *Symposium: Archaeology of Ancient Bonito Part I(resume)* (in Japanese).

Hansel, Fabricio A., Mark S. Copley, Luiz Augusto dos Santos Madureira and Richard P. Evershed  
2004 Thermally produced  $\omega$ -(o-alkylphenyl)alkanoic acids provide evidence for the processing of marine products in archaeological pottery vessels. *Tetrahedron Letters* 45(14): 2999–3002.

Hauff, Simone. and Walter Vetter  
2010 Quantification of branched chain fatty acids in polar and neutral lipids of cheese and fish samples. *Journal of Agricultural and Food Chemistry* 58(2): 707–712.

Heron, Carl., Shinya Shoda, Adrià Breu Barcons, Janusz Czebreszuk, Yvette Eley, Marise Gorton, Wiebke Kirleis, Jutta Kneisel, Alexandre Lucquin, Johannes Müller, Yastami Nishida, Joon-ho Son and Oliver E. Craig

2016 First molecular and isotopic evidence of millet processing in prehistoric pottery vessels. *Scientific Reports* 6(1): 38767.

Horiuchi, Akiko., Yoshiki Miyata, Nobuhiko Kamijo, Lucy Cramp and Richard P. Evershed

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.

Kametani, Hiroaki.

2011 *Studies on ancient wooden plates and local communities*: 197-360, Tokyo: Azekura-syobo (in Japanese).

Kaneko, Horomasa.

1965 Jomon shellmiddens and bonito fishing. *Fukushima Kouko* 6: 15–25 (in Japanese).

Kosaki, Susumu.

2023 Aspects of ancient monuments in Numazu. *Ancient society created by the Kano and Fuji rivers: the original landscape of Numazu and Fuji*: 9–16 (in Japanese).

Kurihara, Kenzo.

2015 Umami the Fifth Basic Taste: History of Studies on Receptor Mechanisms and Role as a Food Flavor. *BioMed Research International* 2015: 189402.

Lucquin, Alexandre., Kevin Gibbs, Junzo Uchiyama, Hayley Saul, Mayumi Ajimoto, Yvette Eley, Anita Radini, Carl P. Heron, Shinya Shoda, Yastami Nishida, Jasmine Lundy., Peter Jordan, Sven Isaksson and Oliver E. Craig

2016 Ancient lipids document continuity in the use of early hunter-gatherer pottery through 9,000 years of Japanese prehistory. *Proceedings of the National Academy of Sciences of the United States of America* 113(15): 3991–3996.

Lucquin, Alexandre., André C. Colonese, Thomas F.G. Farrell and Oliver E. Craig

2016 Utilising Phytanic Acid Diastereomers for the Characterisation of Archaeological Lipid Residues in Pottery Samples. *Tetrahedron Letters* 57(6): 703–707.

Lucquin, Alexandre., Harry K. Robson, Yvette Eley, Shinya Shoda, Dessislava Veltcheva, Kevin Gibbs, Carl Heron, Sven Isaksson, Yastami Nishida, Yasuhiro Taniguchi, Shōta Nakajima, Kenichi Kobayashi, Peter Jordan, Simon Kaner and Oliver E. Craig

2018 The impact of environmental change on the use of early pottery by East Asian hunter-gatherers. *Proceedings of the National Academy of Sciences of the United States of America* 115(31): 7931–7936.

Miyata, Yoshiki., Akiko Horiuchi, Hideki Takada and Toshio Nakamura

2015 Evaluation of sea mammals as marine resource by lipid analysis in pottery excavated from Mawaki Archaeological Site, Ishikawa, Japan. *In Proceedings of the Japan Society for Scientific Studies on Cultural Properties (JSSSCP) Conference 2015*: 40–41.

Nito, Atsushi.

1996a. A Study on the Establishment and Peculiarities of the Izu Province in Ancient Japan. *Historical Research of Shizuoka Prefecture* 12: 1–24 (in Japanese).

Nito, Atsushi.

1996b. A Study on the Bonito Tribute in the Suruga and Izu Provinces. *Research on the History of Traffic Facilities in the Tokaido*: 59–102 (in Japanese).

Numazu History and Folklore Museum

2006 *The History of Fishing Tools: Fishing Methods and Tools in the Innermost Part of Suruga Bay*. Shizuoka: Bunkodo Printing Co., Ltd (in Japanese).

Nara National Research Institute for Cultural Properties

2008 *Heijo Palace Excavation Report* 22: 22. Nara: Nara National Research Institute for Cultural Properties (in Japanese).

Numazu City Board of Education

1978 *Excavation Report of the Fujiibara Site Part I: Remains*. Shizuoka: Numazu City Board of Education (in Japanese).

Numazu City Education Board

1998 *Excavation Report of the Miyukicho Site: Artifacts (Pottery)*. Shizuoka: Numazu City Board of Education (in Japanese).

Numazu City Education Board

2016 *Excavation Report of the Nakahara Site*. Shizuoka: Numazu City Board of Education (in Japanese).

Numazu City

2005 *The History of Numazu City: Complete History I: The Prehistoric, Ancient and Medieval Periods*. ed. Numazu City History Compilation Committee and Numazu City Municipal Board of Education, Tokyo: Tosho Printing Co., Ltd (in Japanese).

O'Connor, Sue., Rintaro Ono and Chris Clarkson

2011 Pelagic fishing at 42,000 years before the present and the maritime skills of modern humans. *Science* 334(6059): 1117–1121.

Okamoto, Noriyuki.

1989 History of the production of Nikatsuo in the Ritsuryo period: with a focus on Suruga Province. *The 10th Anniversary Issue of the Yamanashi Prefecture Archaeological Society*: 391–407 (in Japanese).

Okamoto, Yoko., Kenji Tayama and Ayumi Furuta

2020 Sensory Comparisons between Handmade and Instant Bonito-Kelp Stocks in Japanese Dishes. *International Journal of Gastronomy and Food Science* 21: 100233.

Ono Rintaro.

2017. *Human History of Ocean*. Tokyo: Yuzankaku (in Japanese).

Otsu Toru.

1999. *Imperial system of ancient*. Tokyo: Iwanami Shoten, Publishers (in Japanese).

Segawa, Yuichiro.

1980. Large bowls from Fujiibara site: Changes in Nabe (boiling pot)-shaped pottery in the Ritsuryo Period. *Numazu City Museum of History and Folklore Bulletin* 4: 51–102 (in Japanese).

Segawa, Yuichiro. and Hiroko Koike

1980. A Note on Nikatsuo (boiled bonito) and Nabe-shaped Pottery. *Numazu Museum Bulletin* 14: 1–19 (in Japanese).

Shoda, Shinya., Takayuki Shinzato, Miho Suzuki, Hiroto Takamiya, Helen M. Talbot and Oliver E. Craig

2021 Reconstruction of Diet and Natural Resource Use at the North Edge of the Kaizuka Culture by Pottery Lipid Residue Analysis. *Scientific studies on cultural properties* 83: 55–76 (in Japanese).

Shoda, Shinya., Alexandre Lucquin, Jae-ho Ahn, Chul-joo Hwang and Oliver E. Craig

2017 Pottery Use by Early Holocene Hunter-Gatherers of the Korean Peninsula Closely Linked with the Exploitation of Marine Resources. *Quaternary Science Reviews*.  
<https://www.sciencedirect.com/science/article/pii/S0277379117301154>.

Taché, Karine., Yitzchak Jaffe, Oliver E. Craig, Alexandre Lucquin, Jing Zhou, Hui Wang, Shengpeng Jiang, Edward Standall and Rowan K. Flad

2021 What do “barbarians” eat? Integrating ceramic use-wear and residue analysis in the study of food and society at the margins of Bronze Age China. *PloS One* 16(4): e0250819.

Whelton, Helen L., Simon Hammann, Lucy J.E. Cramp, Julie Dunne, Mélanie Roffet-Salque and Richard P. Evershed

2021 A Call for Caution in the Analysis of Lipids and Other Small Biomolecules from Archaeological Contexts. *Journal of Archaeological Science* 132: 105397.



Figure 1. 1: A wooden tablet attached to the container of broth from dried bonito that was transported from the Suruga province, dated circa 735 CE, excavated from the Heijo Capital site. It is written that the volume of the broth is about 0.9 litres. The length is about 10 cm which is small for a cargo tag in this period but typical for the tags of condensed bonito broth (<https://mokkanko.nabunken.go.jp/ja/6AFIUO14000141>). 2: A wooden tablet attached to the tributed boiled bonito that was transported from the Suruga province, dated 760 CE, excavated from Eastsouth district of the Heijo Palae Site. The length is about 20cm (<https://mokkanko.nabunken.go.jp/ja/6AAICJ63000062>) 3: A wooden tablet attached to the tributed mandarins that was transported from the Suruga province, dated 770 CE, excavated from the Zoshushi (Ministry of Brewery) district of Heijo Palace Site. The length is about 20cm (<https://mokkanko.nabunken.go.jp/ja/6AACVV12000001>).

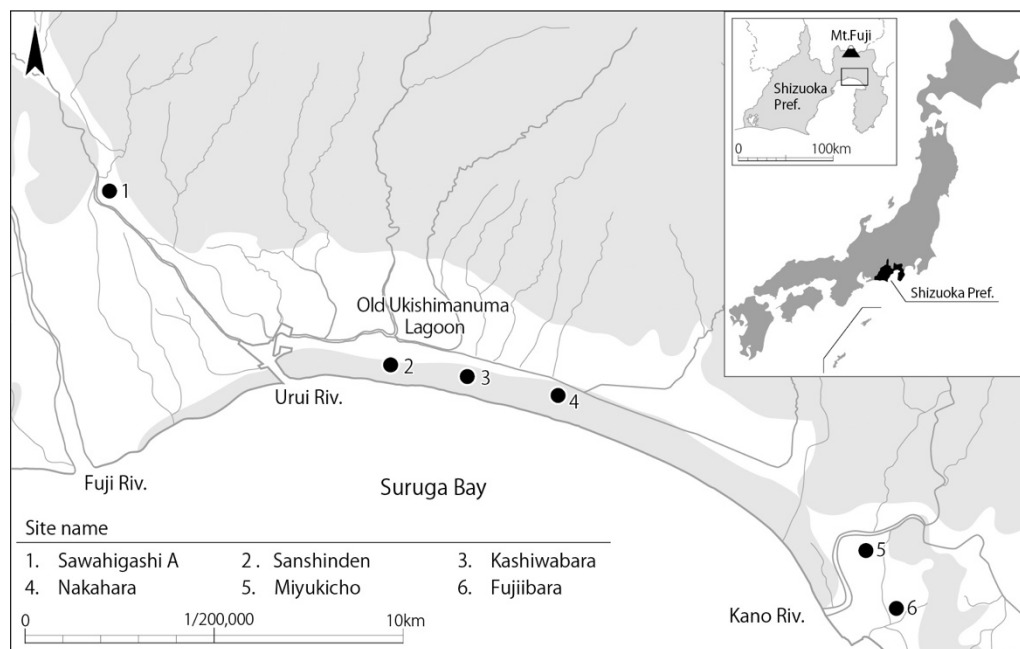


Figure 2. The locations of the investigated sites.

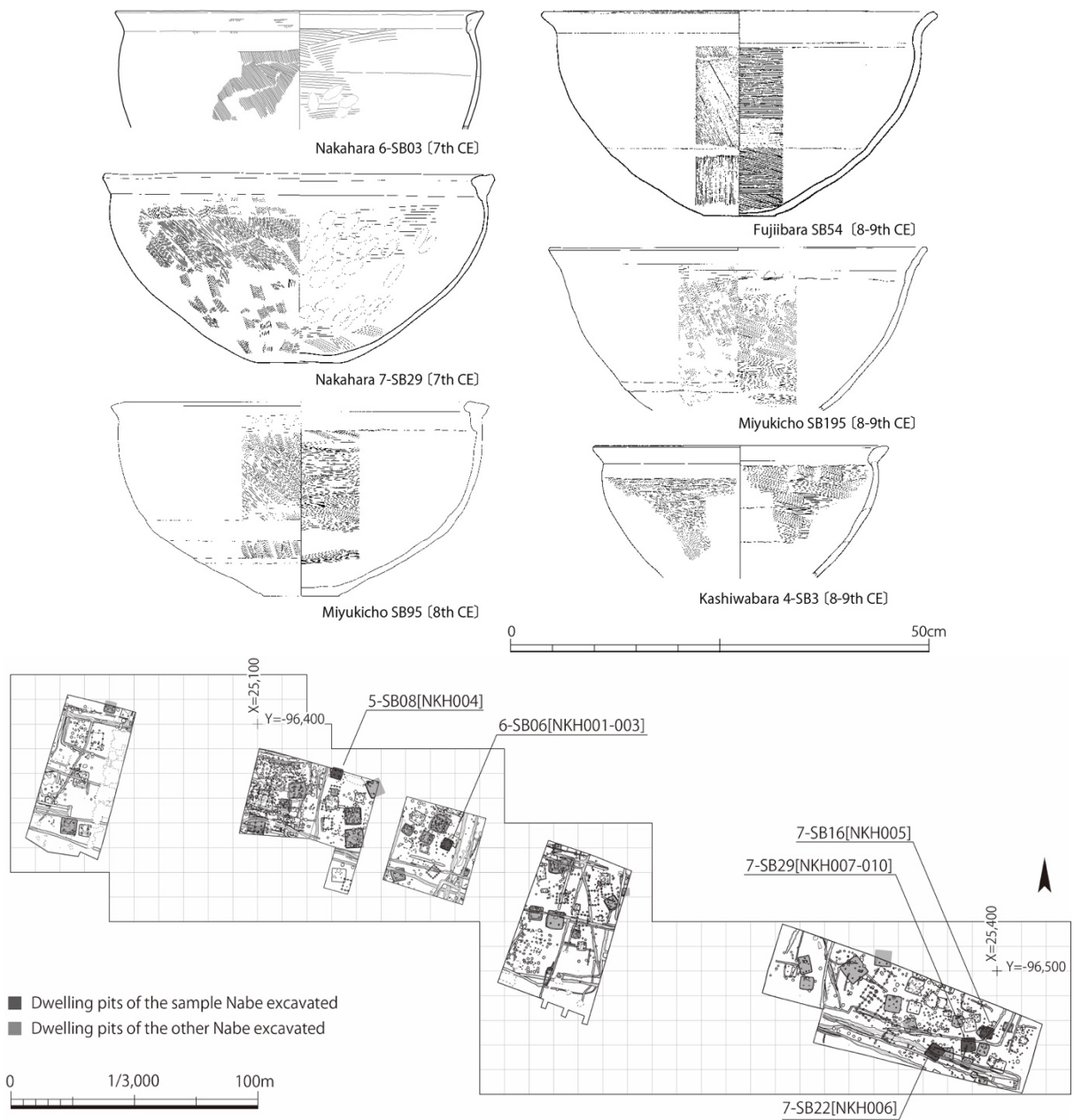


Figure 3. Typical form of Nabe-shaped pottery excavated from coastal sites in the Suruga Bay.



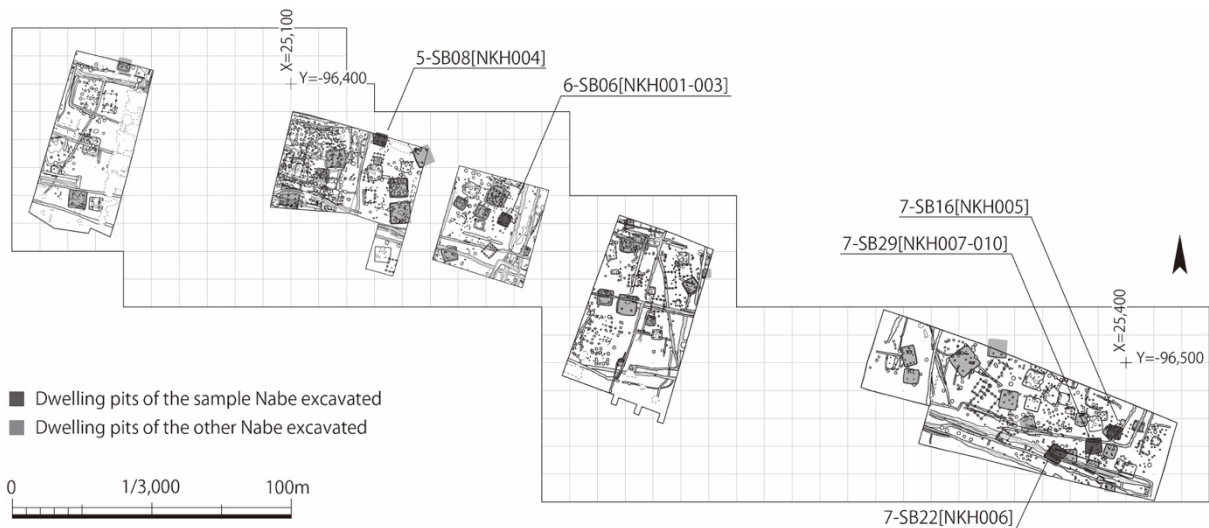


Figure 4. The distribution of building features and finding places of sample pottery in the Nakahara site.

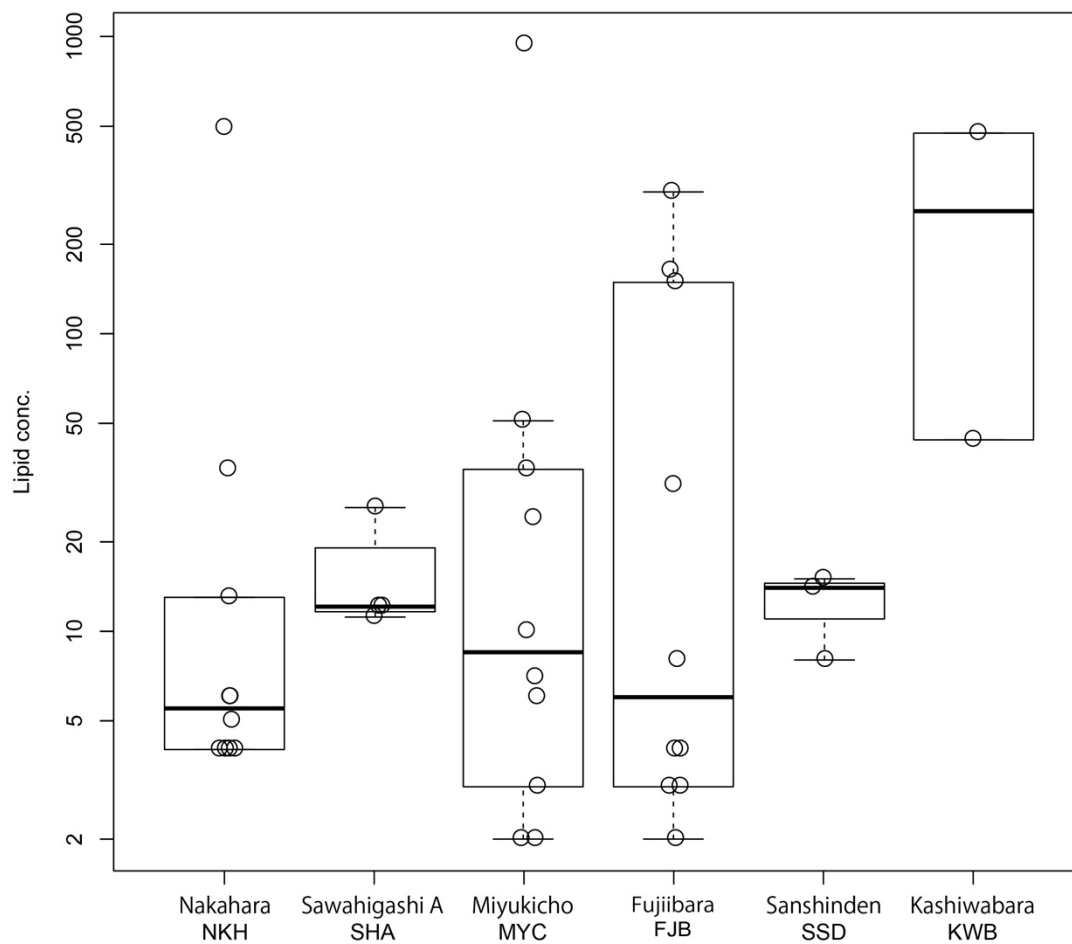


Figure 5. Scattered boxplot of the lipid concentrations in each site analysed. The boxes represent a 68% credible interval while the whiskers represent a 95% credible interval. The horizontal line indicates the mean value.

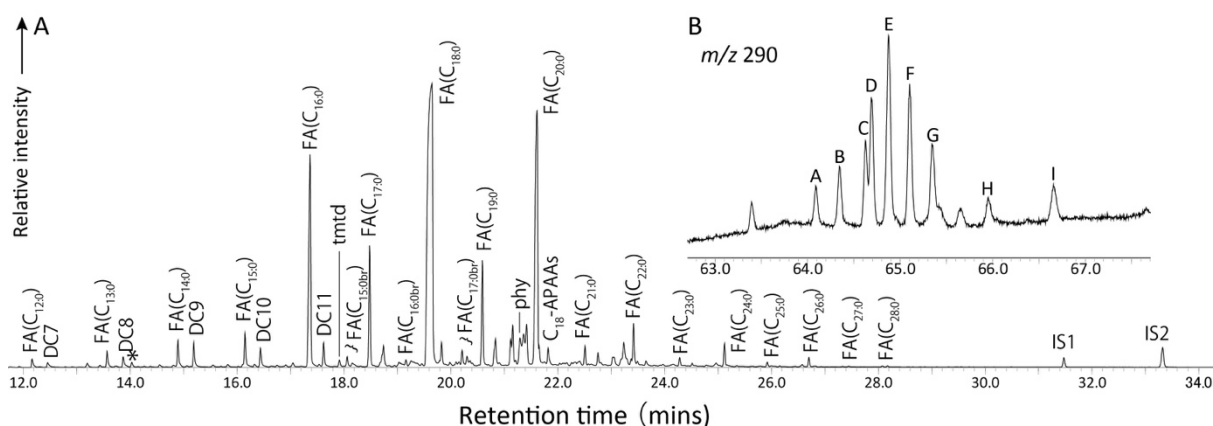


Figure 6. (A) Partial total ion chromatogram of MYC001AE and (B) partial extracted ion chromatogram of  $m/z$  290 obtained during selected ion monitoring of the same sample. FA(C<sub>x:y</sub>) = fatty acids with carbon lengths of x and degree of unsaturation as y. DC<sub>x</sub> = dihydroxy fatty acids with carbon lengths of x. phy = phytanic acid, pri = pristanic acid, tmt = 4,8,12-trimethyltridecanoic acid, and C<sub>18</sub>-APAA = C<sub>18</sub> ω-(o-alkylphenyl)alkanoic acids. IS1 = n-tetratriacontane and IS2 = n-hexatriacontane. \* = modern contaminants.

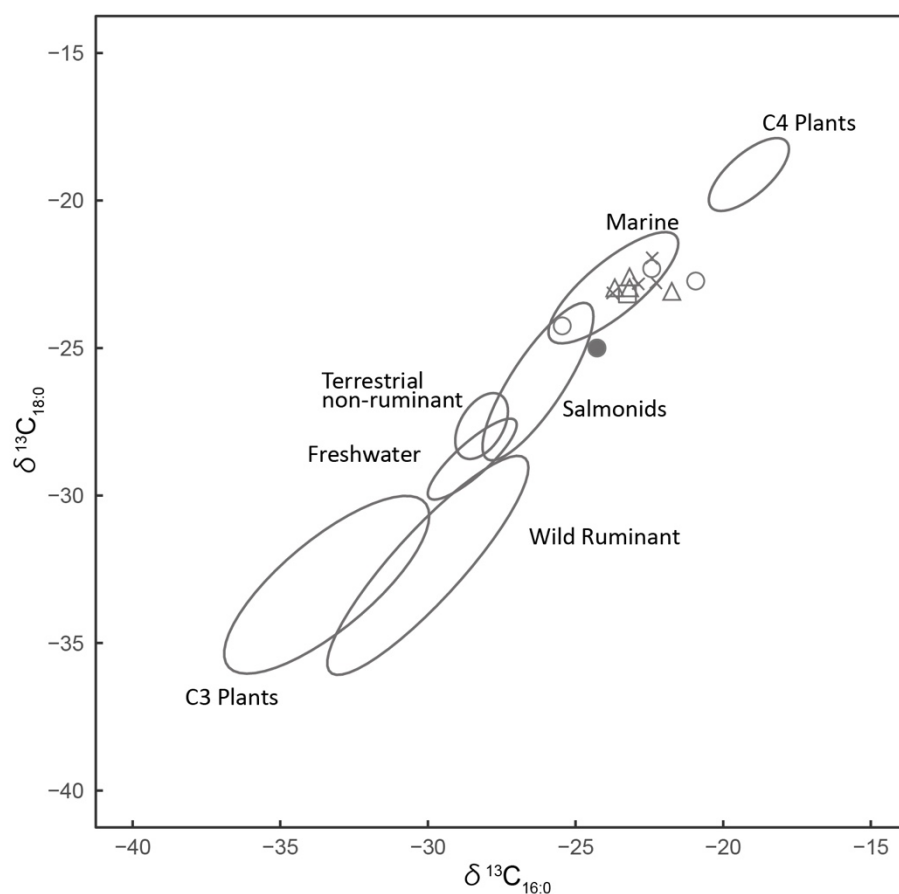


Figure 7. Scattered boxplots of the SRR diastereomer ratio (%) of phytanic acids in archaeological samples and modern references (Lucquin, Colonese et al. 2016).

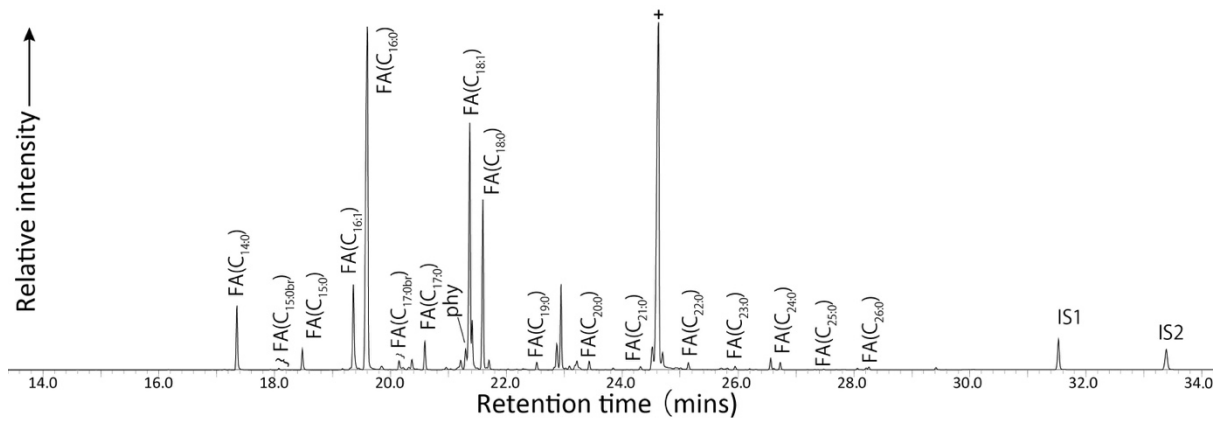


Figure 8. Plot of  $\delta^{13}\text{C}_{16:0}$  against  $\delta^{13}\text{C}_{18:0}$  of archaeological samples with ranges (68 % confidence) of modern authentic reference products (Craig et al., 2013; Horiuchi et al., 2015; Heron et al., 2016; Lucquin et al., 2016; Miyata et al., 2015; Taché et al., 2021). ○: Nakahara (7th century), ×: Miyukicho (8-9th century), △: Fujiibara (8-9th century), □: Kashiwabara (8-9th century), ●: Modern *Katsuoirori* (condensed bonito broth).

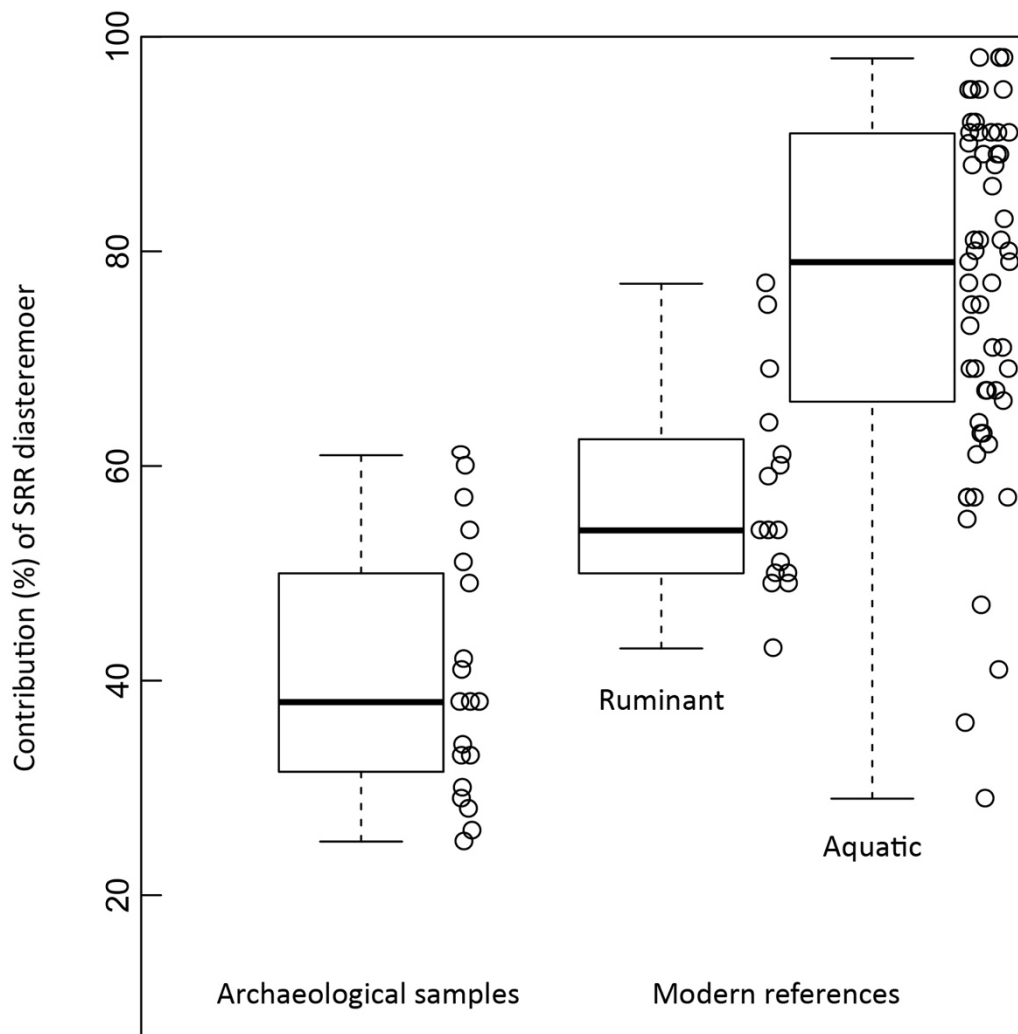


Figure 9. Partial total ion chromatogram of *Katsuoirori*.  $\text{FA}(\text{C}_{x:y})$  = fatty acids with carbon lengths of  $x$  and degree of unsaturation as  $y$ . phy = phytanic acid, IS1: n-tetratriacontane, IS2: n-hexatriacontane, and +: EPA.