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Chemical analyses of organic residues in archaeological pottery from Arbon Bleiche 3, Switzerland – evidence for dairying in the late Neolithic

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Abstract

Fatty acids distribution and stable isotope ratios (bulk $\delta^{13}C$, $\delta^{15}N$ and $\delta^{13}C$ of individual fatty acids) of organic residues from 30 potsherds have been used to get further insights into the diet at the Late Neolithic (3384–3370 BC) site of Arbon Bleiche 3, Switzerland. The results are compared with modern equivalents of animal and vegetable fats, which may have been consumed in a mixed ecology community having agrarian, breeding, shepherd, gathering, hunting, and fishing activities. The used combined chemical and isotopic approach provides valuable information to complement archaeological indirect evidence about the dietary trends obtained from the analysis of faunal and plant remains. The small variations of the $\delta^{13}C$ and $\delta^{15}N$ values within the range expected for degraded animal and plant tissues, is consistent with the archaeological evidence of animals, whose subsistence was mainly based on C_3 plants. The overall fatty acid composition and the stable carbon isotopic compositions of palmitic, stearic and oleic acids of the organic residues indicate that the studied Arbon Bleiche 3 sherds contain fat residues of plant and animal origin, most likely ruminant (bovine and ovine). In several vessels the presence of milk residues provides direct evidence for dairying during the late Neolithic in central Europe.

Keywords: Organic residue; Fatty acids; Potsherds; Gas chromatography-mass spectrometry; GC-MS; Stable carbon and nitrogen isotope ratios; Compound specific isotope analysis; CSIA; Neolithic; Dairying; Switzerland

1. Introduction

1.1. Archaeological residues

The Neolithic settlement of Arbon Bleiche 3 is situated on the southeastern shore of Lake Constance, Canton Thurgau, Switzerland (Fig. 1). From 1993 to 1995, rescue excavations of the site conducted by the Cantonal Archaeological Service found 27 houses, all built between 3384 and 3370 BC (dendrochronological dating), in the excavated area of about 1100 m^2 [34]. The cultural layer lies in the waterlogged zone, where the humidity helped the preservation of the archaeological organic material [30]. In the frame of a long-term project, with the aim to understand the economy and particularly food procurement strategies, preparation and consumption of Neolithic lake shore settlers in central Europe, a multidisciplinary approach, applying botanical, zoological and chemical methodologies to the material recovered from Arbon Bleiche 3 have been adopted [30]. Arbon Bleiche 3 is therefore the most

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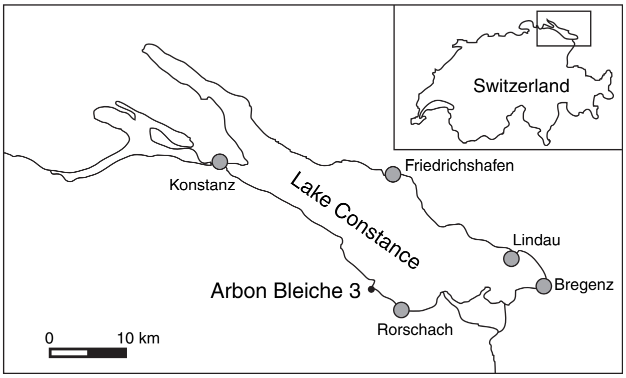

Fig. 1. Location of Arbon Bleiche 3, Lake Constance, Switzerland (adapted from [2]).

thoroughly investigated Neolithic lake shore settlement in central Europe. Among the cultural remains associated with the plant and animal relicts were abundant fragments of unglazed ceramic vessels [6,5]. Dark brown consolidated organic residues were recognized in the interior of some vessels [5], suggesting that they were used for storage and processing of food. Within these organic crusts rare relics of cereals and fish bones were preserved [5]. Mostly, the relics were too small to be visible with low magnification. A microscopic analysis shows that the organic residues contained regularly cereals and other plant tissues; however, no structures of tissues or bones of other animals were detected [36]. The investigations of faunal and plant remains (mammal and fish bones, teeth, seeds, fruits, twigs, pollen, and phytolites) and artifacts associated with food consumption provided evidence for the presence of domestic cows (*Bos taurus*), pigs (*Sus domesticus*), goats (*Capra hircus*), sheeps (*Ovis aries*), and dogs (*Canis familiaris*), and a wide range of wild animals such as red deer (*Cervus elaphus*), wild boar (*Sus scrofa*), brown bear (*Ursus arctos*), and frogs [10,11,26,27]. A further faunal diet component came from lacustrine resources, including Cyprinidae (e.g. *Rutilus rutilus*), Salmonidae (e.g. *Coregonus* sp.), pike (*Esox lucius*), catfish (*Silurus glanis*), and perch (*Perca fluviatilis*) [28]. Cultivated plants such as cereals (*Triticum div*. sp., *Hordeum* sp.), flax (*Linum usitatissimum*) and opium poppy (*Papaver somniferum*) were important food elements [26,27]. Many wild food plants were gathered in the surroundings of the settlement to complement the crops. These included hazelnut (*Corylus avellana*), strawberries (*Fragaria* sp.), blackberries (*Rubus fruticosus* s.l.), raspberries (*Rubus idaeus* s.l.), crab apple (*Malus sylvestris*), and sloe plum (*Prunus spinosa*). The diet of the domestic animals included silver fir (*Abies alba*), mistletoe (*Viscum album*), blackberry (*Rubus frulicosus* s.l.), alder (*Alnus* sp.) and hazel [1,2,32], suggesting their presence in the settlement mainly during wintertime.

1.2. Fat preservation

The main fatty acids found in plant and animal lipids are straight chain carboxylic acids (abbreviated as $C_{x:y}$, where "x" the number of carbon atoms and "y" the number of double C–C bonds in the chain). The more abundant saturated fatty acids are the lauric ($C_{12:0}$), myristic ($C_{14:0}$), palmitic ($C_{16:0}$), and stearic ($C_{18:0}$) acid, and the unsaturated acids are palmitoleic ($C_{16:1}$), oleic ($C_{18:1}$), linoleic ($C_{18:2}$), and linolenic ($C_{18:3}$). During hydrolytic degradation of the lipids, the fatty acids are released from the triglycerides. The short- and medium-chain fatty acids (e.g., $C_{4:0}$ to $C_{14:0}$) are appreciably more water-soluble and volatile than the long-chain fatty acids. So, degraded fat is identified by high concentrations of palmitic and stearic acid. The survival of lipids in association with many archaeological materials is widely documented [15]. In subsequent studies Evershed et al. [17] have shown that the stable carbon isotopic composition of the main fatty acids preserved in unglazed archaeological pottery appears unaffected by diagenetic alteration during burial. Thus providing, by comparison with modern reference fats, a highly robust criteria to distinguish a range of commodities associated with the use of vessels in the past, including among others, animal fats [19,40], milk [14], plant oils [3], plant leaf waxes [16], and beeswax [18].

The relative consumption of marine versus terrestrial foods or use of C_3- vs. C_4-plants based foods within terrestrial ecosystems can be inferred from stable carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) isotopic composition. The carbon isotope composition of plants and their products are linked to the processes of photosynthetic CO_2 fixation. The most important atmospheric CO_2-fixing reactions are the C_3 and C_4 pathways [41]. C_3 plants (plants adapted to temperate ecosystems, including most vegetables, fruit and wheat) use the Calvin cycle for CO_2 fixation, and the $\delta^{13}C$ values fall into the range −34 to −22‰ [48]. The C_4 plants use the Hatch–Slack cycle, and have lower isotopic fractionation compared to C_3 plants. C_4 plants are plants adapted to hot, arid environments, comprising most plants in the tropics, including millet, maize, sugar cane and savanna grasses, and are relatively enriched in ^{13}C (−16 to −9‰). Small shifts of +1 to +2‰ in $\delta^{13}C$ occur between the muscle tissue or whole body of a consumer and its food source [7,21]. Animals, including humans, who consume a great deal of C_4 plants (e.g., maize) can have $\delta^{13}C$ values close to or higher than −12‰. More negative $\delta^{13}C$ values, lower than −22‰, indicate that the food that the individual has consumed comes mainly from terrestrial C_3 plants environment, as well as from the flesh or milk of animals that also subsisted on only C_3 plants. In the region under consideration in this study, Neolithic terrestrial food chains are based on photosynthesis by C_3 plants. The diet of the domestic and herbivore prey