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A biomolecular archaeological approach to 'Nordic grog'

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The combined archaeological, biomolecular, and archaeobotanical evidence from four sites in Denmark (Nandrup, Kostræde, and Juellinge) and Sweden (Havor on the island of Gotland) provide key reference points for reconstructing 'Nordic grog' from ca. 1500 BC to the first century AD. In general, Nordic peoples preferred a hybrid beverage or 'grog,' in which many ingredients were fermented together, including locally available honey, local fruit (e.g., bog cranberry, and lingonberry) and cereals (wheat, rye, and/or barley), and sometimes grape wine imported from farther south in Europe. Local herbs/spices, such as bog myrtle, yarrow and juniper, and birch tree resin rounded out the concoction and provide the earliest chemical attestations for their use in Nordic fermented beverages. The aggregate ingredients probably served medicinal purposes, as well as contributing special flavors and aromas. They continued to be important ingredients for many kinds of beverages throughout medieval times and up to the present.

The importation of grape wine from southern or central Europe as early as ca. 1100 BC, again chemically attested here for the first time, is of considerable cultural significance. It demonstrates the social and ceremonial prestige attached to wine, especially when it was served up as 'Nordic grog' in special wine-sets imported from the south. It also points to an active trading network across Europe as early as the Bronze Age in which amber might have been the principle good exchanged for wine. The presence of pine resin in the beverages likely derives from the imported wine, added as a preservative for its long journey northward.

Keywords: ancient medicine; beer; botanicals; biomolecular archaeology; mead; Scandinavia; wine

A simple question can be posed: What fermented beverages were drunk by the Bronze and Iron Age peoples of the northernmost habitable regions of Europe – referred to as 'Proxima Thule' by the Greeks? Until the advent of modern chemical, archaeological, and archaeobotanical techniques, the answer to this question was perforce based on later Greek and Roman descriptions of the 'barbarians' inhabiting the plains and river valleys north of the Alps (Nelson 2005). If classical writers were to be believed, binge drinking was the rule here by the late Iron Age. For example, according to Varro in his *Chronology*, the Gauls (a general Latin term for the Celtic people living in Europe at the time) were repulsed from the gates of Rome by a surprise attack when they lay in a drunken stupor after torching the city ca. 390 BC. Beer in particular was singled out for its foul smell by Dionysius of Halicarnassus in the late first century BC, who facetiously claimed that the Celtic brew was made from barley rotted in water (*Roman Antiquities* 13.11.1). Diodorus Siculus (*Bibliotheca historica* 5.28.3) drove home the point earlier in the century when he wrote that the rude farmers and mountain folk of the north drank their alcoholic beverages neat (i.e., undiluted)

through tubes or with their moustaches serving as filters, obviously anathema to any cultivated Roman or Greek.

Sometimes, the literary sources provide greater and less contemptuous detail of the ingredients that went into a northern fermented beverage. Diodorus Siculus (5.26.2–3) also noted that Celtic beer included 'the washings of honeycombs, probably mead,' and imported wine. The latter beverage was a mark of the civilized human, at least in Greece probably for thousands of years, in central Italy by at least 700 BC, and in southern France by at least 500 BC (McGovern 2003/2007, pp. 296–298; McGovern 2009/2010, Ch. 5; McGovern *et al.* 2013). Fermented beverages made from honey, the most concentrated simple sugar source in nature, stood equally high on the list of high status drinks in this world and the next, as implied by the famous Greek *kykeon* of the Homeric epics and the Eleusinian Mysteries of the Hellenistic and Roman eras. If northern peoples were importing grape wine from the south and using honey in their fermented beverages, even if it were admixed with 'rotten barley water,' then at least they were partaking of the most prestigious southern beverages.

To gain a fuller and more accurate picture of the composition of these northern beverages before written records, our research group carried out a biomolecular archaeological study of well-preserved residues within vessels from tombs and hoards in Denmark and Sweden, extending over approximately a millennium and a half, from ca. 1500 BC to the first century AD (see Figure 1). In some instances, the archaeobotanical and palynological evidence, which is also presented here, expands upon and/or correlates with the chemical data and interpretations.

Archaeological samples chosen for analysis

Nandrup

The earliest sample examined was recovered from a tumulus (mound) tomb at Nandrup on the island of Mors in Jutland in northwest Denmark (Aner *et al*. 2001: Ke 5282, p. 144; Jensen 2002, pp. 299–300; Koch 2003). It dates to Montelius' Period II of the Nordic Bronze Age (ca. 1500–1300 BC). A single male warrior had been buried with his weapons and a jar, whose residue was the focus of our chemical investigation. The provision of both a bronze sword and a dagger, whose hafts and pommels are nearly identical in their geometric designs of interlocked spirals, roundels, and hatched horizontal lines, implies that the warrior belonged to an elevated social class (Randsborg 1974).

The Nandrup tomb can be compared to a contemporaneous grave at Bregninge in northwestern Zealand in which a male warrior had been buried in an oak coffin with a massively hafted bronze sword, battle-ax, and a jar which contained honey (likely mead) according to a palynological analysis by J. Troels-Smith in an unpublished document in the National Museum of Denmark (see Nielsen, 1977, 1978,

1988, Koch 2003, pp. 128–129). The very well-preserved cist grave at Ashgrove in Scotland of a slightly earlier period (ca. 1700 BC) provides another excellent parallel. A single male warrior with a well-fashioned dagger was buried with a pottery jar (beaker), which again most likely contained honey mead according to the palynological analysis (Dickson 1978; detailed below).

The rounded jar (Figure 2A) from the Nandrup tomb, with a wide mouth and slight vertical lug handles, had been placed at the foot of the man's body. An amber 'button' was recovered from inside the jar, which Koch (2003, p. 128) plausibly proposed was the knob for a lid made of an organic material, such as wood, that disintegrated.

The interior of the jar from its base up to mid-body was covered with a dark residue. Two small pieces (Danish National Museum [NM], sample M 2337), approximately 6 mm on a side (total of 0.25 g), were examined microscopically. They were relatively homogeneous in appearance with different colorations on the exterior (black) and in section (brownish).

Kostræde

A hoard of artifacts from a pit at Kostræde in southern Zealand (Thrane 1966), southwest of Copenhagen, included the only strainer (Figure 2B) yet recovered from the Late Bronze Age (Periods IV–VI, ca. 1100–500 BC) in Denmark. The bronze strainer was quite large, with a diameter of 30.7 cm and a reconstructed height (allowing for the missing base that probably was rounded off) of about 25 cm, comparable to a contemporaneous example of similar size and technological design from the site of Tiszavasvari in eastern Hungary (Thrane 1966). The

Figure 1.　Map of southern Scandinavia, showing archaeological sites investigated.
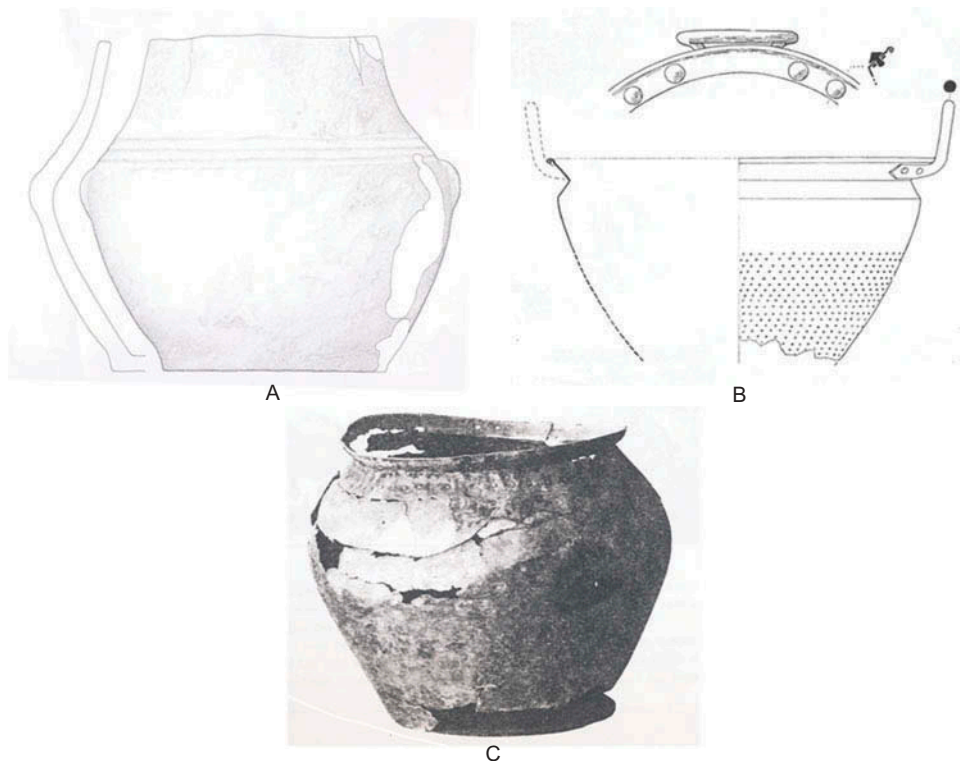
Figure 2.   Artifacts with residues analyzed: Nandrup jar (A), Kostræde strainer (B), and Juellinge bucket (C). (Drawings and photograph, respectively, after Aner *et al*. 2001, pl. 70:5282, mid-body diam. 31 cm [ca. 1:3]; Thrane 1975, fig. 88, rim diam. 30.3 cm [ca. 1:3]; and Müller 1911: fig. 12, mid-body diam. 30.3 cm [ca. 1:3])

strainer was found together with a set of female belt ornaments and a pair of spiral gold earrings.

A small piece (NM, A4614), approximately 6 mm on a side (total of 0.25 g), was removed from one of the perforations of the strainer, evidently a residue from filtering a liquid. The piece was relatively homogeneous with a brownish cross-section and some surface mottling.

Juellinge

The tombs of four women, dating to the Early Roman Iron Age (ca. 200 BC), were found at the Danish site of Juellinge on the island of Lolland, south of Zealand (Müller 1911). Tomb 1, a wood coffin that held the body of a 30-year-old female, was exceptionally rich in finds. Besides cosmetic items and jewelry, the woman held a long-handled bronze strainer-cup, part of a standard imported Roman wine-set, in her right hand. In addition to two Roman-style incised glass beakers, a large bronze 'bucket' (Latin, *situla*) (Figure 2C), also part of the wine-set, was placed at her head, and a whole sheep was laid at her feet outside the coffin.

Inside the bucket was a long-handled ladle, another part of the wine-set. A dark, relatively homogeneous residue had collected on bucket's interior base. A small piece (NM, M18823), approximately 6 mm on a side (total of 0.25 g), was provided for analysis.

Havor

At Havor on the Swedish island of Gotland in the Baltic Sea, a hoard of a filigreed and granulated gold torque, a pair of bronze bells, and an imported Roman bronze wine-set of the Early Roman Iron Age (first century AD) was found buried against the inner face of the wall of a ring fort (Nylén 1962; Nylén *et al*. 2005). The wine-set was comprised of a 'bucket' (situla) (Figure 3), inside of which were three single-handled 'sauce-pans' or mixing/drinking cups, which were nested one inside the another, and a long-handled strainer-cup and ladle inside one another.

The interior of the strainer-cup had a built-up residue in and around its holes. A piece about a centimeter square and 3 mm thick was made available for analysis (see Isaksson 2005: fig. 1a–b). The residue was reddish-brown with dispersed greenish areas.

Archaeobotanical evidence

The botanical, especially palynological, evidence from excavated vessels (particularly 'beakers' and processing vats) in Scotland has proven very illuminating in defining a 'Nordic grog' (McGovern 2009/2010, Ch. 5), going back as early as the fourth millennium BC and continuing down to the second millennium when our earliest sample from Nandrup contributes data for Scandinavia.

Figure 3. Roman drinking-set, comprised of a bucket (*situla*), a ladle, and several 'sauce pans' or drinking cups, from a hoard under the floor of a settlement at Havor (Sweden) in the southern part of the island of Gotland in the Baltic Sea. (Photograph courtesy of E. Nylén and Statens Historiska Museum, Stockholm)

Very consistent results were obtained from the palynological analyses of the Scottish residues from regions throughout the country, including the western islands, and as far north as the Orkneys (Dickson 1978, Wickham-Jones 1990, Dineley 2004, Koch 2003). Honey, deriving mostly from the flowers of the small-leaved lime tree (*Tilia cordata*) and meadowsweet (*Filipendula vulgaris*) or heather (*Calluna vulgaris*), were attested in all the samples. Several vessels also contained cereal pollen. No fruit remains were reported, but since fruits carry minimal pollen compared to the amount and variety that is incorporated into honey as nectar, their presence cannot be excluded. The meadowsweet pollen could have originated from intentionally adding this herb to the beverage. Although some of the pollen evidence is open to interpretation, a good case can be made that the cups and large vessels originally contained a fermented beverage – whether it were a mead, sweetened ale, or a more complex Nordic grog with added herbs.

The palynological analyses of the residues from the Ashgrove and Bregninge warrior burials, with their evident similarity to the Nandrup tomb (above), should be especially noted as a preamble to our Scandinavian findings. The Bregninge jar yielded pollen from lime tree, meadowsweet, white clover (*Trifolium repens*), knotgrass (*Polygonum*), and flowers of the Compositae flower family. A profusion of lime tree, meadowsweet, heather, ribwort plantain (*Plantago lanceolatus*), and thyme (*Thymus*)/mint (*Mentha*) pollen had similarly infused the sphagnum moss and leaves covering the Ashgrove man's upper torso and arms. In both instances, the most convincing explanation was that the jars originally held a diluted honey beverage, which had spilled out onto the moss in the case of the Ashgrove beaker and which had evaporated inside the Bregninge jar. When honey is watered down to about 30%, natural osmophilic yeast in the honey become active and readily converts it to mead.

Nandrup

J. Iversen (Broholm and Hald 1939) made a very similar palynological finding to those at Bregninge and Ashgrove when he examined the residue inside the Nandrup jar, later confirmed by J. Troels-Smith in his unpublished document in the National Museum of Denmark. The residue was dominated by lime tree and meadowsweet pollen, with some white clover pollen, which are best explained again as deriving from a honey product. This was most likely mead, since the residue had the appearance of an evaporated liquid and was confined to the lower part of the vessel.

Juellinge

Our next botanical point-of-reference is the Juellinge residue, dated to ca. 200 BC. Based on his microscopic examination of the residue inside the situla, archaeobotanist B. Gram (1911) identified remnants of barley (Hordeum) grains, a fruit that was most likely bog cranberry (*Oxycoccus palustris*) which also accounted for numerous calcium citrate and malate crystals, lesser amounts of lingonberry or cowberry (*Vaccinium vitis-idaea*), filaments of bog myrtle (*Myrica gale*), and yeast cells. He concluded that the situla had originally contained a mixed beverage of barley beer and a fruit wine made of bog cranberries and lingonberries. Since he did not carry out a palynological analysis, it is not known whether a fermented, diluted honey/mead was also part of the beverage.

Unfortunately, archaeobotanical and/or palynological analyses of the temporally intervening sample from Kostræde, as well as the latest sample in our series from Havor, are yet to be done.

Chemical results

Ancient organic compounds were identified by a combination of chemical techniques: Fourier-transform infrared spectrometry (FT-IR), gas chromatography-mass spectrometry (GC-MS) following extraction, ultra-HPLC tandem mass spectrometry (LC/MS/MS) following extraction, and headspace solid phase microextraction (SPME) coupled to GC-MS (see Supplementary Information).

From earliest to latest, we obtained the following chemical results for our samples:

Nandrup

(1) The FT-IR spectrum of the sample has characteristic but small hydrocarbon peaks in the 2900 cm^{-1} region, indicative of organics, but it is mainly dominated by inorganic iron oxide peaks.

(2) The GC-MS results (Figure 4 and Tables 1 , 2) show odd-numbered n-alkanes C_{27} and C_{29} and even-numbered fatty acids from C_{24} to C_{30}, both characteristic of beeswax. However, the absence of beeswax esters, the C_{25} and C_{31} n-alkanes and the C_{32} and C_{34} fatty acids, together with only moderately intense C_{27} alkane and C_{24} fatty acid peaks, argue against beeswax. Possibly, the results are explained by differential degradation of beeswax compounds that were more susceptible to oxidation (especially wax esters) or were present in lesser amounts. Epicuticular plant wax can be ruled out, because plant sterols and internal diones are absent (see Juellinge, below).

(3) The presence of pristane and phytane, branched long-chain paraffins, are typical petroleum biomarkers (Eglinton and Calvin 1967).

(4) The isopropyl esters of the C_{12}, C_{14}, and C_{16} fatty acids, which have been reported in other ancient liquid containers (e.g., Corinthian 'plastic' vases: Biers *et al*. 1994, pp. 42, 50, 54, 55; a Minoan bowl: Beck *et al*. 2007, pp. 58–59), are likely

volatile contaminants of human skin and/or saliva (Middleditch 1989).

(5) Manoyl oxide, a diterpenoid ether, has been reported (Biers *et al*. 1994, p. 24, with references) to occur in several Pinaceae and Cupressaceae family conifers, including juniper. The lack of evidence for other characteristic terpenes of these tree resins, however, implies that the manoyl oxide is most likely a contaminant.

The Nandrup sample was not analyzed by LC/MS/MS for tartaric acid/tartrate nor by SPME for volatile compounds.

Kostræde

(1) The FT-IR spectrum of the sample shows a mixture of inorganic (iron oxide and a 6-coordinate copper(II) complex) and organic constituents. The latter includes prominent hydrocarbon peaks in the 2900 cm^{-1} region. A doublet at 1730 and 1710 cm^{-1}, combined with a hydroxyl stretch absorption at 1455 cm^{-1}, can be attributed
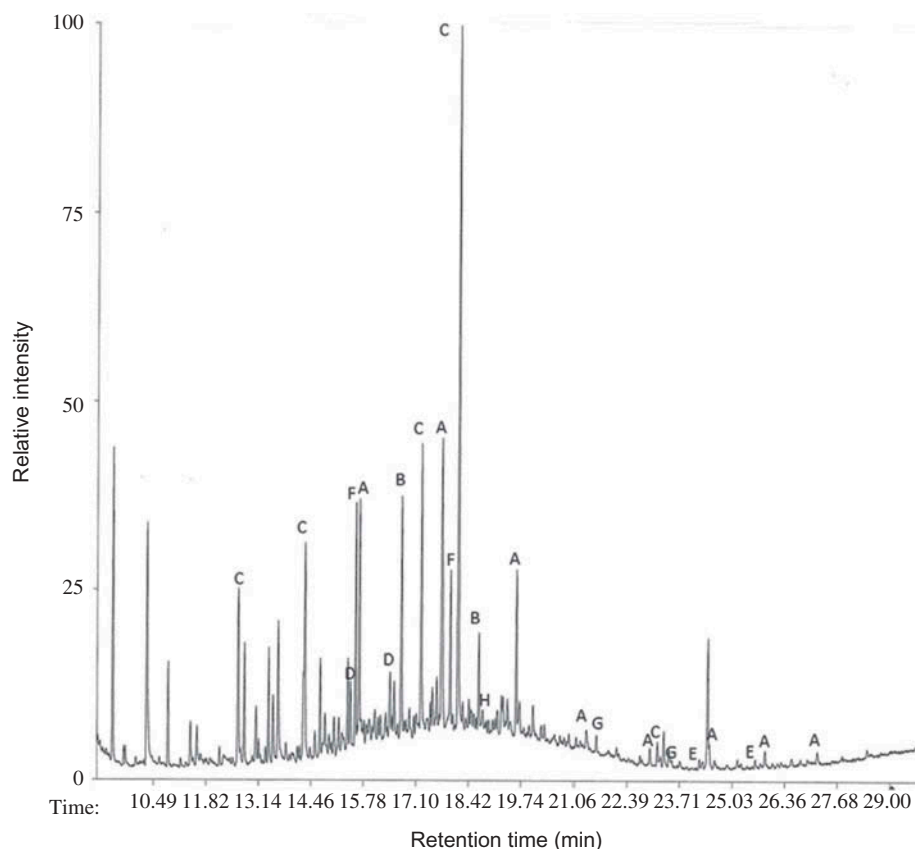

Figure 4.   GC-MS chromatogram: Nandrup sample: (A) fatty acids (methyl esters); (B) fatty acids (isopropyl esters); (C) phthalate plasticizers; (D) pristane/phytane; (E) C_{27} and C_{29} alkanes; (F) antioxidants in plastic; (G) pine resin family; and (H) manoyl oxide.

Table 1. Chemical compounds identified by GC-MS for Scandinavian samples.

Sample identification	Nandrup	Kostraede	Juellinge	Havor
Pine (Pinaceae) resin diterpenoids				
Dehydroabietic acid	−	+	−	Trace
7-Oxo-dehydroabietic acid	−	+	−	−
Retene	−	Low	−	−
Manoyl oxide	+	−	−	−
Birch (Betulaceae) resin triterpenoids				
Betulin	−	+	−	+
Betulinic acid	−	+	−	+
3-Oxoallobetulane	−	+	−	+
Lupeol	−	+	−	+
Lupenone	−	+	−	+
Lup-2,20(29)-diene	−	+	−	+
Suberic dioic acids				
C_{16}	−	+	−	+
C_{18}	−	+	−	+
C_{20}	−	+	−	+
Plant sterols				
Sitosterol	−	−	+	−
Ergostanol	−	−	+	−
Stigmastanol	−	−	+	−
Stigmasta-3,5 diene-7-one	−	−	+	−
Wax esters of C_{16} fatty acid and C_{24},C_{26},C_{28} alcohols				
C_{16}/C_{24}	−	+	−	−
C_{16}/C_{26}	−	+	−	−
C_{16}/C_{28}	−	+	−	−
n-Alkanes				
C_{25}	−	+	+	−
C_{27}	+	+	+	−
C_{29}	+	+	+	−
C_{31}	−	+	+	−
Epicuticular plant waxes				
Hentriacontane-14,16-dione	−	−	+	−
25-Hydroxy-hentriacontane-14,16-dione	−	−	+	−
Fatty Acids				
Saturated	Up to C_{30}	C_{24} dominant	Higher C-nos. dom.	Up to C_{24}
C_{14}	+	+	+	+
C_{16} (Palmitic)	+	+	+	+
C_{18} (Stearic)	+	+	+	+
C_{20}	+	+	+	+
C_{22}	+	+	+	+
C_{24}	+	Dom.	+	+
C_{26}	+	+	+	−
C_{28}	+	+	+	−
C_{30}	+	+	+	−
C_{32}	−	+	+	−
C_{34}	−	+	+	−
Palmitic/Stearic ratio	>1	~1	>>1	>1
Oleic (C_{18})	−	+	Dom.	+
Azelaic (C_9)	−	+	Dom.	+
Isopropyl esters				
C_{14}	+	−	−	−
C_{16}	+	−	−	−
Petroleum biomarkers				
Pristane	+	−	−	−
Phytane	+	−	−	−

Note: + = Present; − = absent; dom. = dominant; C-nos. = carbon numbers.

Table 2. Retention times for compounds identified by GC-MS.

Retention time (min)	Compound name	Retention time (min)	Compound name
11.15	Heptanedioic acid, dimethyl ester	24.49	Tetracosanoic acid, methyl ester
12.46	Octanedioic acid, dimethyl ester	25.20	Pentacosanoic acid, methyl ester
13.65	Nonanedioic acid, dimethyl ester	25.45	Docosanedioic acid, dimethyl ester
14.83	Decanedioic acid, dimethyl ester	25.61	n-Nonacosane
14.57	Isopropyl laurate	25.90	Hexacosanoic acid, methyl ester
15.44	Pristane	26.55	Heptacosanoic acid, methyl ester
15.66	Tetradecanoic acid, methyl ester	26.93	n-Hentriacontane
16.55	Phytane	27.02	Lup-2,20(29)-diene
16.72	Isopropyl myristate	27.20	Octacosanoic acid, methyl ester
17.78	Hexadecanoic acid, methyl ester	28.04	Ergostanol
18.77	Manoyl oxide	28.45	Triacontanoic acid, methyl ester
19.44	Oleic acid, methyl ester	28.54	Sitosterol
19.65	Octadecanoic acid, methyl ester	28.61	Stigmastanol
20.60	Retene	28.78	14,16-Hentriacontanedione
20.76	Hexadecanedioic acid, dimethyl ester	28.97	Lupenone
21.42	Eicosanoic acid, methyl ester	29.13	Stigmasta-3,5 diene-7-one
21.65	Dehydroabietic acid, methyl ester	29.14	Lupeol
22.42	Octadecanedioic acid, dimethyl ester	29.68	Dotriacontanoic acid, methyl ester
22.99	Docosanoic acid, methyl ester	30.13	25-Hydroxy-14,16-hentriacontanedione
23.14	15-Hydroxydehydroabietic acid, methyl ester	30.58	3-Oxoallobetulane
23.53	7-Oxodehydroabietic acid, methyl ester	31.18	Tetratriacontanoic fatty acid, methyl ester
23.75	Tricosanoic acid, methyl ester	31.69	Betulin
23.98	Eicosanedioic acid, dimethyl ester	36.50	Palmitic acid, tetracosyl ester
24.21	n-Heptacosane	40.52	Palmitic acid, hexacosyl ester

to birch tree resin (see below); its strong absorptions mask any contribution by tartaric acid/tartrate, which is characterized by a doublet at 1740 cm^{-1} (major peak)/1720 cm^{-1} (shoulder) and hydroxyl stretch absorption in the 1450–1430 cm^{-1} region.

(2) Tartaric acid/tartrate is positively identified by LC/MS/MS (Figure 7).

(3) Beeswax was unquestionably present in this sample, based on the GC-MS identification of C_{25}–C_{31} n-alkanes, even-numbered C_{24}–C_{30} fatty acids with C_{24} dominant, and wax esters of the C_{16} fatty acid (palmitic) with C_{24}, C_{26}, and C_{28} alcohols (Figure 5 and Tables 1, 2).

(4) Pine resin is substantiated by dehydroabietic acid and 7-oxo-dehydroabietic acid.

(5) Birch tree resin is extremely well attested by characteristic triterpenoids of the lupeol and betulin families (betulin, betulinic acid, 3-oxoal-lobetulane, lupeol, and lupenone) and suberic C_{16}, C_{18}, and C_{20} dioic acids (Regert 2004 – compare Figure 5 and Tables 1, 2). Lup-2,20 (29)-diene is a marker compound for heat-treated resin (Modugno and Ribechini 2009).

(6) Azelaic acid is most likely a derivative of oleic acid, also attested in the sample, since the latter is readily cleaved at its C_9 double bond and oxidized to the dicarboxylic azelaic acid. Oleic acid is widely distributed in the plant and animal

kingdoms. It should be noted, however, that azelaic acid also occurs naturally in wheat, rye, and barley, which are highly appropriate ingredients for a Nordic grog.

(7) Volatile compounds are identified by SPME (Table 3) and their natural product sources are determined by standard bioinformatics searches of the available chemical literature (McGovern et al. 2009). Several compounds of the hydro-carbon, alcohol, ester, aldehyde, and lactone classes are consistent with grape wine (Bakker and Clarke 2011). However, benzaldehyde, 2-ethyl-1-hexanol, and nonanal could be con-taminants (see Supplementary Information). Any ancient ethanol would have been metabo-lized by microorganisms.

(8) Juniper (*Juniperus communis*) extract, usually added as the cone (fruit), is a probable additive to the liquid filtered through the strainer. It is attested by fenchol, terpineol, and junipene (see Juellinge, below, for better attestation of juniper and herbs/spices in general).

(9) The herb bog myrtle or sweet gale, again as a probable additive, is indicated by fenchol, γ-cadinene, copaene, and other more common compounds.

(10) The natural source for the high level of eucalyp-tol (cineole) is uncertain; this compound is found in mugwort and other wormwood species,
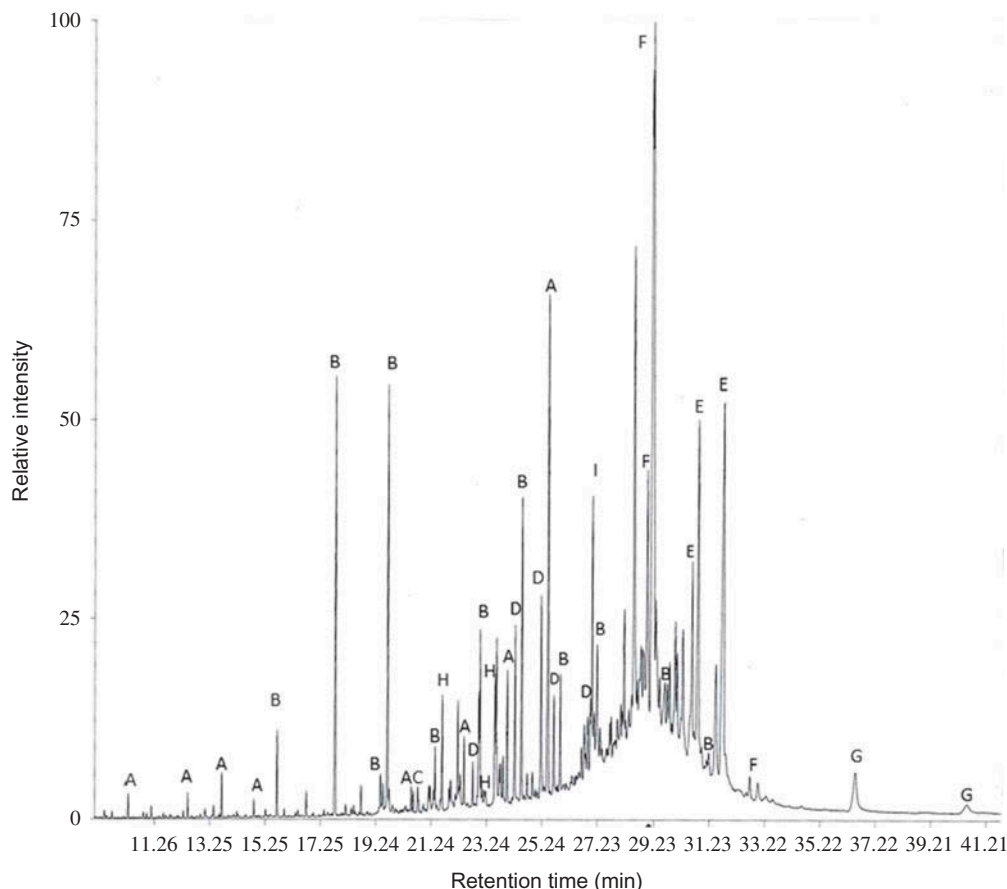
Figure 5.   GC-MS chromatogram: Kostræde sample: (A) dioic acids; (B) fatty acids (saturated and unsaturated); (C) retene; (D) odd-numbered n-alkanes from C_{25} to C_{31}; (E) betulin birch resin family; (F) lupeol birch resin family; (G) wax esters of C_{16} (palmitic) fatty acid and C_{26} and C_{28} alcohols; (H) pine resin family; and (I) lup-2,20(29)-diene (heat-treated birch resin marker).

cranberry, and rosemary, which are known to have been additives to ancient European fermented beverages (e.g., Juan-Tresserras 1998, Stika 1996, McGovern *et al.* 2013).

Juellinge

(1) The FT-IR spectrum of the sample shows a mixture of inorganic and organic constituents, which are ill-defined.

(2) The LC/MS/MS data (Figure 7) are borderline positive for tartaric acid/tartrate.

(3) An epicuticular plant wax, which is common on the surfaces of fruits such as bog cranberry and lingonberry, is well attested by GC-MS (Figure 6 and Tables 1, 2), based on the presence of the plant sterols (sitosterol, ergostanol, stigmastanol, and stigmasta-3,5 diene-7-one) and large amounts of internal plant diones (hentriacontane-14,16-dione) and its associated hydroxyl compound (Evershed *et al.* 1991). A very high palmitic to stearic fatty acid ratio is also typical of plant products. Consequently and in contrast to the Nandrup sample (above), very similar peak intensity patterns of key odd-numbered n-alkanes and even-numbered fatty acids, coupled with the absence of wax esters and the C_{32} and C_{34} fatty acids, are better explained as deriving from epicuticular wax, rather than degraded beeswax.

(4) Azelaic acid is again most likely a derivative of oleic acid (see Kostræde, above), but a natural source from wheat, rye, or barley cannot be ruled out.

(5) Volatile compounds are very well represented by SPME (Table 3), including probable grape wine, juniper, and bog myrtle compounds, as already pointed out for Kostræde (above).

(6) Additionally, pine resin (Pinaceae) is attested by fenchone, terpineol, and possibly γ-cadinene; D-limonene and camphor might also derive from pine resin, but could be contaminants.

Table 3.   Chemical compounds identified by SPME for Scandinavian samples.

Retention time (min)	Compound name	Relative intensity (%)			Probable source
		Kostræde	Juellinge	Havor	
2.64	1-Butanol, 3-methyl-	–	2.52	–	1
2.67	1-Butanol, 2-methyl-	–	1.02	–	1
3.01	Toluene	–	–	0.20	?
3.37	Hexanal	0.40	0.39	0.25	?
4.93	Heptanal	0.54	–	0.41	1
6.08	Benzaldehyde	3.86	5.24	2.05	1
6.20	Heptanol	0.97	–	0.77	?
6.37	Hexanoic acid	2.04	1.52	–	?
6.43	Phenol	1.05	17.02	–	?
6.56	5-Hepten-2-one, 6-methyl-	0.52	–	–	?
6.87	Octanal	0.80	0.50	0.48	1/6?
7.31	Benzene, 1,2,3-trimethyl-	–	–	0.41	6
7.39	1-Hexanol, 2-ethyl-	4.41	1.46	1.60	?
7.43	D-Limonene	–	0.54	–	1,2,3,5
7.51	Eucalyptol (cineole)	0.54	–	0.26	3
7.54	Benzyl alcohol	–	7.54	–	1
7.75	Salicylaldehyde	–	7.75	–	?
7.93	p-Cresol	–	7.93	–	?
8.09	Cyclohexene	–	8.09	–	?
8.24	Cyclooctane	–	8.24	–	?
8.25	1-Octanol	2.69	–	1.68	1
8.71	Fenchone	0.52	–	–	2,3 (as fenchol),5
8.71	2-Nonanone	–	0.64	0.27	?
8.96	Nonanal	1.13	0.88	0.54	1
9.06	Thujone	–	0.33	–	4
9.64	Pentanedioic acid, dimethyl ester	–	0.16	–	?
9.89	Camphor	–	0.82	–	2,3,4,5
10.05	Menhone	–	0.33	–	?
10.23	2,4,6-Octatriene, 2,6-dimethyl-	–	0.25	–	2
10.31	Octanoic acid	3.87	1.77	0.61	?
10.33	1-Nonanol	–	–	0.44	?
10.42	Menthol	1.82	0.84	0.49	?
10.6	Benzenemethanol, α,α,4-trimethyl	–	0.18	–	?
10.68	Naphthalene	2.36	3.78	0.72	?
10.78	2-Decanone	–	0.95	–	?
10.8	Terpineol	0.98	–	–	1,2,5
10.86	Octanoic acid, ethyl ester	0.51	–	–	1
10.89	Methyl salicylate	0.80	0.56	–	?
11.23	l-Verbenone	–	0.14	–	2
11.51	Benzothiazole	0.52	0.65	1.15	?
12.25	Nonanoic acid	2.13	0.57	0.72	?
12.78	2-Undecanone	–	0.59	–	?
12.82	Nonanoic acid, ethyl ester	0.39	0.48	–	1
12.88	Naphthalene, 2-methyl-	0.75	0.48	–	6
13.04	Undecanal	–	0.29	–	?
13.21	Naphthalene, 1-methyl-	0.53	0.28	–	6
14.09	Decanoic acid	0.46	–	–	?
14.15	γ-Nonalactone	0.52	0.34	–	1
14.27	1-Undecanol	–	0.16	–	?
14.49	Biphenyl	–	0.16	–	?
14.69	2-Dodecanone	–	0.11	–	?
14.76	Tetradecane	0.50	–	–	?
14.97	Naphthalene, 1,6-dimethyl-	0.98	–	–	6
15.07	Junipene	0.86	–	–	2
15.24	Naphthalene, 2,3-dimethyl-	–	0.10	–	1/6?
15.24	Naphthalene, 2,7-dimethyl-	0.46	–	–	1/6?
15.3	Naphthalene, 2,6-dimethyl-	0.40	0.19	–	1/6?
16.11	1-Hexadecanol	–	0.25	–	?
16.35	α-Cucumene	–	0.17	–	?
16.77	Pentadecanal	–	0.14	–	?
16.97	Dibenzofuran	0.74	0.28	–	6

Table 3.   (*Continued*).

Retention time (min)	Compound name	Relative intensity (%)			Probable source
		Kostræde	Juellinge	Havor	
17.21	Azulene, 4,6,8-trimethyl-	0.38	–	–	?
17.48	Naphthalene, 1,2,3,4-tetrahydro-1,4,6-trimethyl	–	0.23	–	?
18.25	Hexadecane	0.69	–	–	?
18.48	Tetradecanal	–	0.16	–	?
18.89	Naphthalene, 1,2,3,4,4a,7-hexahydro-1,6-dimethyl-4-(1-methylethyl)-	–	0.27	–	5?
19.11	γ-Cadinene	–	0.48	–	2,3,4,5?
19.17	Copaene	–	0.16	–	2,3,4
19.32	Naphthalene, 1,2,3,4,4a,5,6,8a-octahydro-4a, 8-dimethyl-2-(1-methylethylidene)-,(4aR-trans)-	–	0.30	–	?
19.64	Cadalin (Cadalene)	–	1.92	–	?
19.86	Heptadecane	0.64	0.14	–	?
21.26	Phenanthrene	1.08	0.30	–	1/6?
21.26	Anthracene	–	–	0.17	1/6?
23.81	1,2-Benzenedicarboxylic acid, butyl 2-ethylhexyl ester	–	–	–	6
		–	–	–	
24.06	Octacosane	–	5.71	–	?
24.15	Eicosane	–	4.11	–	?

Notes: 1 = Grape wine (*Vitis vinifera*) constituent.
2 = Juniper (*Cupressaceae juniperus*) constituent.
3 = Bog myrtle (*Myrica gale*) constituent.
4 = Yarrow (*Achillea millefolium*) constituent.
5 = Pine (Pinaceae) resin constituent.
6 = Probable modern or ancient contaminant.
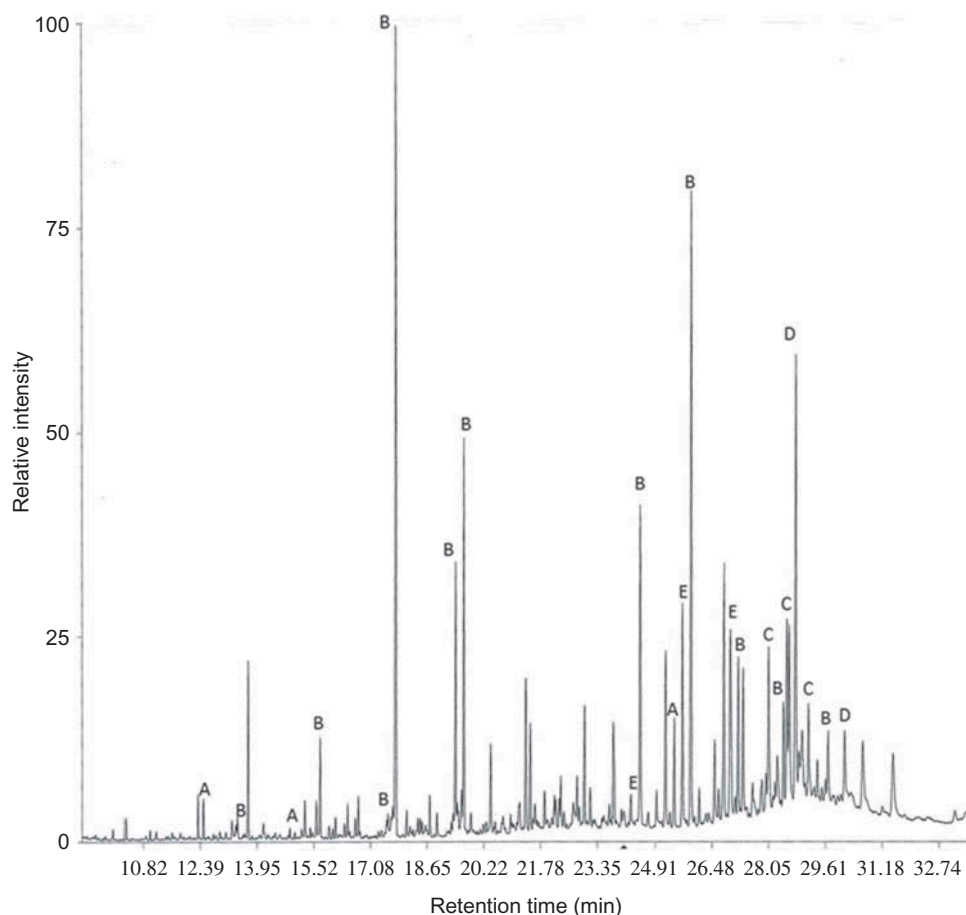
Figure 6.   GC-MS chromatogram: Juellinge sample: (A) dioic acids; (B) fatty acids (saturated and unsaturated); (C) plant sterols; (D) epicuticular wax: hentriacontane-14,16-dione and 25-hydroxy-hentriacontane-14,16-dione; and (E) odd-numbered n-alkanes from C_{25} to C_{31}.
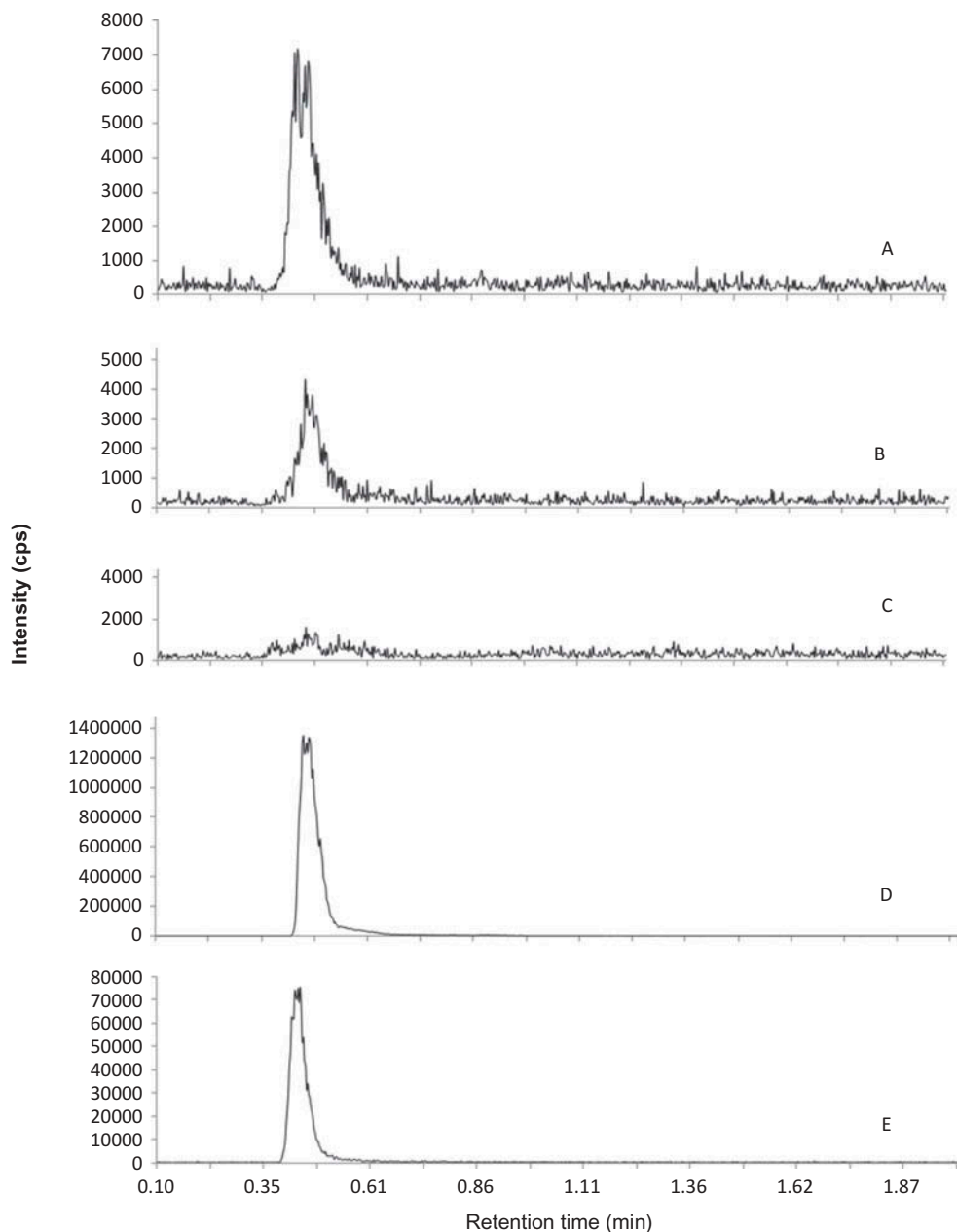
Figure 7. MRM LC/MS/MS traces of L-tartaric acid corresponding to *m/z* 149→87 molecular ion fragmentation for Kostræde (A), Juellinge (B), and Havor (C) samples, compared with standard solutions of L-tartaric acid (D) and modern generic red wine (E).

(7) The herb yarrow (*Achillea millefolium*) is indicated by copaene, γ-cadinene, and camphor.

(8) The presence of both terpineol and benzyl alcohol is consistent with bog cranberry, but weakly indicated.

Havor

(1) The absorptions in the FT-IR spectrum of the sample can all be ascribed to organic groups.

(2) Tartaric acid/tartrate is positively identified by LC/MS/MS (Figure 7). A Feigl spot test (Feigl 1966; see Supplementary Information) was also positive.

(3) According to GC-MS (Figure 8 and Tables 1, 2), the same suite of birch tree resin compounds are present in this sample as in the Kostræde sample (above). A trace of dehydroabietic acid is likely due to pine resin.

(4) Azelaic acid is again most likely a derivative of oleic acid (see Kostræde, above), but a natural source from wheat, rye, or barley cannot be ruled out.
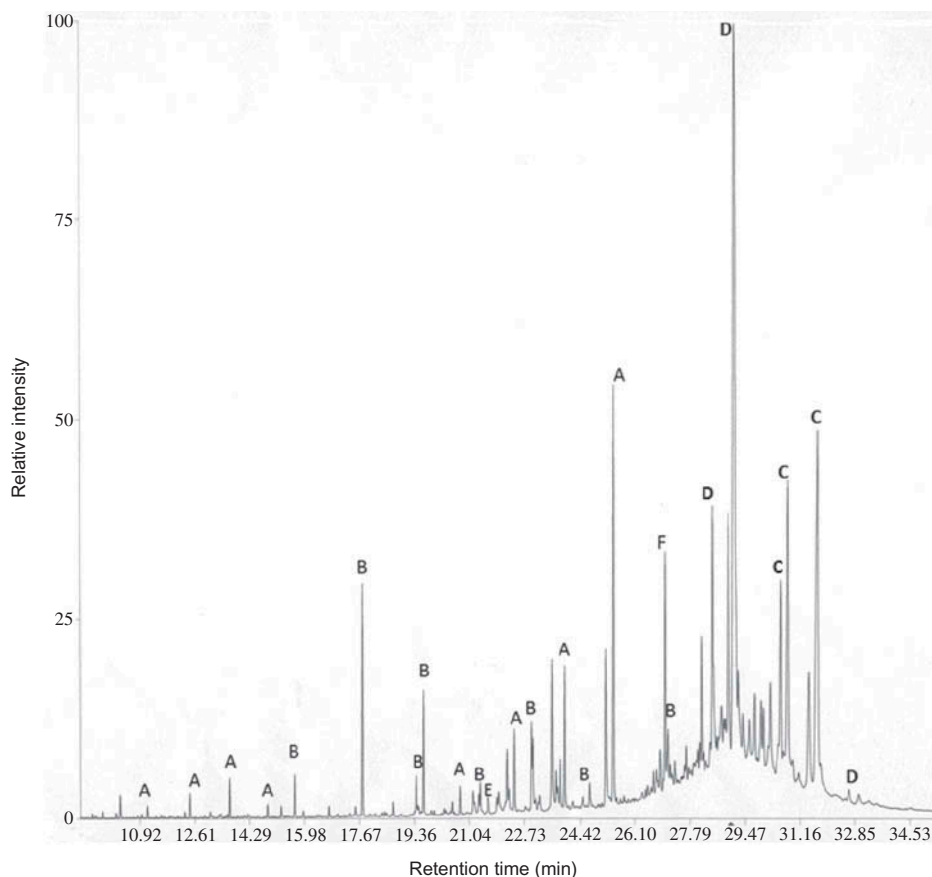
Figure 8.   GC-MS chromatogram: Havor sample: (A) dioic acids; (B) fatty acids (saturated and unsaturated); (C) betulin birch resin family; (D) lupeol birch resin family; (E) pine resin family; and (F) lup-2,20(29)-diene (heat-treated birch resin marker).

(5) The high palmitic to stearic fatty acid ratio is typical of plant products. No cholesterol, characteristic of animal fats, was detected.

(6) Probable grape wine compounds are attested by SPME (Table 3), but herbal and pine resin compounds were absent.

(7) The natural source for the high level of eucalyptol (cineole) is uncertain; this compound is found in mugwort and other wormwood species, cranberry, and rosemary, which are known to have been additives to ancient European fermented beverages (see above).

Discussion and conclusions

The combined archaeological, biomolecular, and archaeobotanical evidence from four sites in Denmark and Sweden, stretched out in time over a millennium and a half (ca. 1500 BC to the first century AD), presents a coherent picture of how Nordic peoples conceived of and made their fermented beverages. In general, they preferred a hybrid beverage or 'grog,' in which various ingredients were most likely fermented together, including locally available honey, locally grown fruit (e.g., bog cranberry and lingonberry) and cereals (wheat, rye, and/or barley), and sometimes grape wine imported from farther south in Europe. Single batch fermentation, rather than separate fermentation of each beverage with subsequent mixing, best accounts for the uniformity of the beverage over time. At some stage in the process, locally available juniper, birch, and possibly pine (unless it had arrived via the grape wine from southern or central Europe), and/or herbs, especially bog myrtle and yarrow, might be mixed in. More of the fermentables might also have been added as sweeteners or flavorants after fermentation or when the beverage was served.

Nandrup

Our earliest sample from Nandrup might appear to be the most anomalous 'Nordic grog' in our corpus, because the residue inside the jar has thus far been shown to only comprise honey. This conclusion is based on well-attested palynological data, which are consistent with, but not

proven, by the chemical data. The latter evidence thus far rules out any fruit being added. Herbal additives are also moot until a volatile organic analysis is carried out. Thus, the princely warrior in the Nandrup tomb, on current knowledge, most likely took an unadulterated mead to his death.

The case can be made that mead was the drink of elite northerners for millennia, whether in the Bronze Age (Koch 2003) or during Viking times (Husberg 1994). Honey is the most concentrated simple sugar source in nature, composed of 60–80% simple sugars, and procuring it in the wild (before hives were introduced, possibly as late as the first century AD) was difficult. As a relatively rare, expensive commodity, honey and its fermented beverage, mead, were likely the prerogative of the rich and powerful. The relatively high alcoholic content of mead of about 10–12% (alcohol by volume), like grape wine, contributed to its appeal. Other native fruits, grains, and natural products were lower in sugar and yielded lower alcoholic beverages. Such beverages might be enjoyed for their own sake or considered appropriate for the afterlife, as shown by barley beer in a beaker of the Single-Grave Culture, dated around 2600 BC, in a warrior burial at Refshøjgård on Jutland (Klassen 2005).

While mead might have been at the top of the Nordic drink pyramid, hybrid beverages were not far behind. The burial of a young woman in an oak coffin under a mound at Egtved in Jutland (Thomsen 1929), not far from Nandrup and of the same time period, is sufficient proof for this assertion. The woman's amazingly well-preserved attire, especially an open woolen skirt in which strings would have dangled from her hips, likely marked her as a priestess or ceremonial dancer. A large bronze belt disk at her midriff displayed interlocking spirals, which was a well-known entoptic symbol of the Nordic sun-god (Kaul 1998, 2005). Bronze Age female figurines from other Danish sites wear similar dress and jewelry, and are sometimes shown dancing with hands on their hips, arching their backs in acrobatic positions, displaying their breasts, and holding vessels. Dancers, amid spiral designs, are also shown on Scandinavian rock carvings executing backward flips, line or chain dancing, and doing the ancient equivalent of a 'jig' (conveniently illustrated in Koch 2003, see especially figs. 12.16, 12.17, and 12.18 [no. 15]).

Most importantly for the purposes of this study, a birch-bark container had been placed at the foot of the Egtved woman's coffin. The archaeobotanists B. Gram (Thomsen 1929, p. 20) and J. Troel-Smith (Christensen, Jensen 1991, p. 16) examined the container's contents and identified the remains of bog cranberries and cowberries, wheat grains, bog myrtle filaments, and pollen from the lime tree, meadowsweet, and white clover derived from honey. It was concluded that the Egtved woman likely belonged to the upper class. As such, she took to her

death a special mixed fermented drink of mead, barley beer, and fruit, which might also have led to more inspired dancing during life. The bog myrtle would have given the brew a special flavor; it is still a popular additive to Scandinavian aquavits. Intriguingly, a flowering yarrow had been placed between the upper and lower halves of the coffin.

Kostræde

Our first probable evidence of imported wine in Nordic grog comes from the next sample in the chronological sequence, Kostræde, dated ca. 1100–500 BC. The chemical evidence was definitive for tartaric acid/tartrate (from the Eurasian grape, which did not grow in Scandinavia during this period), honey (likely as mead, which could more easily be filtered as a liquid than viscous honey), and birch and pine tree resins. Additives to the beverage probably included juniper and bog myrtle. Only very tentative chemical evidence was obtained for a cereal ingredient in the beverage. Archaeobotanical investigation of the Kostræde residue, yet to be carried out, might well shed light on whether a cereal, such as wheat, barley and/ or rye, was also added to the beverage, making it a true hybrid beer/wine/mead.

The pine resin additive in the Kostræde beverage is not unexpected, since this tree exudate had long been used as a mastic, sealant, medicinal agent, etc., but in the case at hand, its antioxidative, preservative properties are likely the most important. Humans had discovered by the Neolithic period in the Near East that pine and other conifer resins helped to prevent grape wine from going to vinegar. The practice, along with viniculture itself, later spread to Europe. The *retsina* of modern Greece is the only modern carryover of this widespread practice of antiquity. Any wine that made the long trip to Scandinavia from southern or central Europe, such as that likely in the Kostræde beverage, might well have been protected by pine resin. Or it could have been added to the Nordic grog after it arrived.

The presence of the Eurasian grape in the mixed beverage is best explained as having been imported as wine from the south. The strainer itself was most likely made in central Europe (viz., Hungary: Thrane 1975), which might have been producing its own wine by this time or, if not, were in contact with regions farther to the south or east which were. Wine is an excellent candidate in exchange for amber in particular, since large deposits of the petrified resin occur along Baltic shores including Zealand in Denmark where Kostræde is located. The amber trade routes had already been in operation for centuries (Kaul 2013; also see Mukherjee *et al*. 2008 *re* the chemical identification of Baltic amber in a Bronze Age royal tomb in Syria).

Birch tree resin as an additive in any beverage would be intriguing, but its occurrence in the Kostræde beverage is so far the earliest chemically attested instance of such. Its use for a variety of other purposes – especially as a mastic to hold haftings onto weapons and tools and as a sealant – can be cited from northern and central Europe since at least Neolithic times. That the resin was considered fit for human consumption and might well go into a beverage is implied by the finding of lumps of it with human teeth impressions in Neolithic Swiss lake settlements and recently at a Neolithic site in Finland (Scientists find ancient chewing gum in Finland 2013). Apparently, it had been chewed as gum; since some of compounds in birch tree resin have anaesthetic and antibiotic effects, the goal might have been to alleviate pain or protect against tooth decay.

Raw birch sap, which flows in quantity from the trees in Spring, has a sugar content of 0.5–2% by weight. It can be concentrated down to much higher concentrations by heating, which appears to have been the case for the samples from Kostræde and Havor (below), based on the marker compound, lup-2,20(29)-diene. As such, it had considerable potential as a principal ingredient, or at least a fermentable adjunct, in Nordic grog. The use of lesser amounts for medicinal purposes and flavor enhancement is also possible. The common man's drink in Russia today, *kvass* (leaven), perpetuates this ancient tradition; in addition to leavened rye, wheat, or barley bread, which produce a mildly (1–1.5%) alcoholic drink, birch sap and various fruits are sometimes thrown into the brew to increase the sugar and resulting alcohol content.

Juniper might also be viewed as a curious additive to any beverage. Its unique flavor and aroma profiles, however, would have been incentives for its use. The spice was recognized for its medicinal value and flavor in Roman times (e.g., Galen, *De Alimentorum Facultatibus* [On the Powers of Foods], bk. 2) and probably much earlier. Even the invention of the popular, juniper-infused distilled beverage, gin, is attributed to a seventeenth century AD Dutch physician. More traditional Nordic beverages with juniper can be cited (also see Madej *et al.* forthcoming), including Finnish *sahti*, which like *kvass*, is made from barley, rye, wheat, and/or oats, often prebaked. On Gotland where the Havor sample (below) was obtained, the traditional beverage, *Gotlandsdryka*, is a barley beer spiced with juniper extract. Added sugar, often as honey, makes for a more powerful brew.

Bog myrtle as an additive to the Nordic grog was already known by the time of Kostræde, since it had been identified in the Egtved beverage, based on archaeobotanical evidence. Its combination with yarrow, however, is its first attestation by chemical methods. These two herbs are of special importance to European beer brewing in general. They constitute two of the principal herbs in medieval 'gruit,' which was used as the bittering agent for beers and other fermented beverages until it was displaced by hops in the sixteenth century AD throughout Europe (Unger 2007).

Juellinge

The evident association between women and the Nordic grog, which we have already seen illustrated at Egtved and Kostræde (above), is highlighted once again at Juellinge more than a millennium later during the Early Roman Iron Age (ca. 200 BC). Archaeobotanical and biomolecular analyses concur in the reconstruction of a mixed fermented fruit beverage of bog cranberry, lingonberry, and likely grape wine. The archaeobotanical investigation contributed additional information to the biomolecular investigation, viz., that barley was another main ingredient, and that the drink had clearly been fermented as shown by the yeast cells. The biomolecular investigation, in turn, provided new information that beeswax and thus honey were probably absent from the beverage. It also revealed that pine resin and juniper, together with yarrow and bog myrtle, contributed special flavors and aromas to the wine/beer concoction.

The Kostræde and Juellinge beverages are similar in other important respects: they both were made with imported, resinated wine, and both were spiced with bog myrtle and juniper. They differed in that the Kostræde drink was combined with honey mead and birch tree resin, whereas the Juellinge drink was a composite of barley beer and fruit wines made not just from grape but also native lingonberry and possibly bog cranberry.

Juellinge illustrates the pattern of importing special wine-sets in early Roman times that spreads out to encompass the rest of Denmark, southern Sweden (Scandia), Gotland, and parts of Norway and Finland in the centuries to follow. Increasingly, Roman and Greek wine-sets of large situlae, ladles, strainers, and drinking cups and horns, often of previous metals (gold and silver) and glass, were imported northwards (Hansen 1987). Indeed, the wine-set became a fixture of upper-class Scandinavian tombs, placed behind the head of the deceased. At Hoby on Lolland, not far from Juellinge and also belonging to early Roman times, a male chieftain was laid out in all his finery, which included the jug, ladle, and situla of a wine-set, a drinking horn, and two silver drinking cups with scenes from the *Iliad* (Johansen 1923). A wine-strainer, in particular, was a key item of any respectable woman's drinking repertoire for serving herself and her mate, as attested for the woman in grave 1949–2 at the wealthy, third century AD community of Himlingøje on Zealand (Hansen 1995, pp. 152–158).

The finding of the richest Bronze Age tomb thus far in Scandinavia at Kivik (Kristiansen and Larsson 2005), in the vicinity of Simris and contemporaneous with Nandrup, implies that connections between northern and southern

Europe had been in the making for over a millennium. The Mycenaean motifs (horse and chariot, boat, weaponry, sun-symbol of a cross within a circle, etc.) on the stone slabs of the burial cist at Kivik support this hypothesis in an extraordinary way, especially in light of the rich amber deposits on the nearby shores. Baltic amber is plentiful in Mycenaean tombs and palaces, and must have been exchanged for goods or items of commensurate value in the eyes of the northerners. The exchange mechanism of ideas and goods between northern and southern Europe, both direct and indirect, was complex at this time (Kaul 2013); it became more regularized in later periods.

Drinking-sets and wine might well have been two of the principal exchange commodities. Precedents for providing a drinking-set in the afterlife can be found as early as 1200 BC in the Urnfield culture of central Europe, when bronze buckets, handled cups, and strainers, like the one from Kostræde, were common items in burials. Imported drinking-bowls from central Europe, which had probably been inspired by Mycenaean prototypes, also made their way into bogs. Large 'cauldrons' or buckets were sometimes mounted on 'sun'-wheels, like those from Ystad in Scandia, Milavec in Bohemia, and Skallerup on the island of Zealand in Denmark (Rausing 1997). The later introduction of Greek and Roman drinking-sets into the continent elaborated upon this tradition, which eventually encompassed almost all of Europe.

What motivations lay behind these developments? Whether we call it elite emulation or conspicuous consumption, the rulers of Proxima Thule had much to gain by exchanging amber and other natural resources for imported goods from the south, beginning as early as the Bronze Age and reaching a crescendo by Roman times. They enhanced their status in the eyes of their subjects and the gods when they could provide them with grand feasts and offerings served up in quintessential drinking and serving vessels, which exhibited the consummate skill of the artisan. The northern rulers further set themselves apart, as much as any host of a Greek *symposion* or Roman *convivium*, when they served a more potent and distinctive version of Nordic grog, which might contain more valuable honey or imported wine than usual. The competition between Celtic chiefs to purchase only the best food and drink for their feasts and ceremonies was so intense that J. M. de Navarro (1928, p. 435) claimed that Celtic art owed its existence to Celtic thirst.

Havor

The Havor situla fits the pattern of importing both grape wine and the wine-set to go with it in Roman times. The wine was attested by multiple lines of evidence, including a Feigl spot test and, most definitively, LC/MS/MS. Since our earlier collaborator on this study, Sven Isaksson (2005), did not have access to these data when he published his analyses, he did not report the presence of tartaric acid/tartrate. There is agreement on the presence of birch tree resin compounds, which dominates the FT-IR spectrum and which, as we have already seen, laced the mixed beverage at Kostræde (above). Herb or spice additives are unlikely, based on the absence of SPME evidence. Traces of diterpenoid compounds of pine resin, which were not reported by Isaksson, show that the imported wine was probably resinated. No beeswax compounds from honey were detected, and lacking an archaeobotanical analysis, other ingredients, such as a cereal, cannot be determined. In short, the available evidence, including a very high palmitic to stearic fatty acid ratio and the absence of cholesterol (cf. Isaksson 2005), are consistent with the situla having originally contained an exclusively plant-derived product, viz., a resinated wine with added birch tree resin. It is highly probable that the 'dregs' of this beverage, which contained an elevated level of birch tree resin, was filtered through the strainer-cup, and better explain the built-up residue in its pore holes rather than its use as a censor (*pace* Isaksson 2005).

Far from having been the 'barbarians' that we might imagine them to have been from classical sources, the northern inhabitants of Proxima Thule exhibit the same innovation in making fermented beverages from a variety of natural sources that humans around the globe have shown since we came 'out of Africa.' They were not averse to adopting the accoutrements of southern or central Europeans in drinking their preferred beverages out of imported vessels of grand proportions, artistic finesse, and ostentatious materials. They were also not averse to importing and drinking the southern beverage of preference, grape wine, even if it were sometimes mixed with northern honey, fruits, herbs, and resins. To begin with, wine was a trickle, thus far first chemically attested at Kostræde (ca. 1100–500 BC). But its importation likely picked up momentum, as suggested by the results reported here from Juellinge and Havor and as will likely be filled out by future analyses, both earlier and covering the intervening time periods.

Once accepted as an alternative fermented beverage and fruit source, grape wine eventually took the northerners by storm, as a flood of the beverage was exported by amphoras and barrels up and down the principal river arteries of the continent, whose forerunners were the amber trade routes. When the Celts themselves had established a native wine making industry in southern France (McGovern *et al.* 2013), the process was inevitable.

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Supporting Information

FT-IR Analyses and Database Searches

The FT-IR data were obtained on Thermo Nicolet spectrometers, both by diffuse-reflectance on samples mixed with potassium bromide (KBr) or on neat (unmixed) samples run using a diamond cell. The primary FT-IR data are not presented here, because of limitations of space. Moreover, for the purpose of this paper, the pertinent compounds are much more exactly characterized by GC-MS, SPME, and LC/MS/MS.

FT-IR spectra were searched for "matches" against large databases of relevant natural products and processed organic materials, synthetic compounds, modern wine samples, and "ancient wine reference samples." The latter were residues from ancient vessels which likely originally contained wine, based on strong archaeological criteria or exterior inscriptions which recorded their contents.

GC-MS Extractions and Analyses

For the liquid-injection GC-MS analyses, the Nandrup, Kostræde, and Havor samples were pulverized and dissolved in a 2:1 mixture of chloroform and methanol, which was heated for 30-45 min at 60°C. The solubles were concentrated down by centrifugation and evaporation to dryness, followed by methylation with Alltech II Me-Prep. As an experiment, the pulverized Juellinge sample was put directly into Methyl Prep reagent and heated for 1 hr at 60°C. After filtering through a cotton-filled pipette, a heavier liquid had separated out; it was solubilized by adding dimethylformamide to the filtrate.

One microliter samples were injected splitless onto a 30 m x 250 μ x 0.25 μ film thickness HP-5MS column (5% phenyl methyl siloxane) of an Agilent HP 6890 GC, run at a 1.5 ml/min flow rate. An HP 5973 mass selective detector was used with the injector port at 325°C. The oven temperature was held at 50°C for 2 min, then programmed to increase at 10°C/min to 325°C where it was held for 10.5 min for a total run time of 40 min. The transfer line to the mass spectrometer was at 300°C. Compound identification was made by retention time and mass spectrum using NIST 05.

LC/MS/MS Extractions and Analyses

Approximately 10 mg samples were mixed in 5 mL of 1-2.8% ammonium hydroxide in water/methanol (80:20, v/v), stirred overnight, and ultrasonicated for 1 hr. Ammonium hydroxide enhances dissolution of tartaric acid in basic solution, so that the latter can be detected as the negative ion and its fragments. All aqueous extracts/suspensions were concentrated by evaporating the

methanol and/or reducing the water content, followed by filtration through a 0.45 μm Nylon Acrodisc filter.

It should also be noted that short retention times are typical for ultrahigh performance LC methods, and present no problem in separating tartaric acid from other compounds that elute at later retention times. More importantly, our identification techniques relied on multiple factors, including retention times and accurate mass measurements that enable the unambiguous identification of tartaric acid.

SPME Analyses

Using freshly powdered samples, the headspace SPME analyses were carried out on an Agilent HP 6890 GC with a 5973 mass selective detector, equipped with an HP-5MS column (30 m × 250 μ × 0.25 μ film thickness) and Gerstel MPS2 Multipurpose Autosampler with a divinylbenzene/carboxen/polydimethylsiloxane 50/30 μm fiber. Fifty milligrams of sample were suspended in 1 mL of deionized water, to which 0.5 g of NaCl was added. The fiber was exposed to the headspace of the saline suspension at 70°C for 10 min, followed by 3 min desorption and splitless injection into the GC-MS at 250°C. To identify possible carryover compounds or contaminants, blank control samples, consisting of only the aqueous saline solutions, were run between the analyzed samples. The mass spectrometer was operated in the scan mode from 40 to 400 atomic mass units. The oven was heated for 29 min from 50°C to 250°C at 7°C/min, and a constant pressure flow rate of 1.2 mL/min was maintained on the column. The compounds were identified by matching scores of 80 or above to those in the NIST 05 and 08 mass spectral libraries (comprising more than 160,000 compounds).

Some of the compounds which were detected (Table 3), especially low-boiling compounds up to hexenal, might derive either from ancient and/or modern "background contaminants" due to groundwater percolation or sample handling (e.g., plasticizers and anti-oxidants from plastic, including compounds in the phthalate family).

SPME is of great utility in biomolecular archaeological studies. It requires only milligram quantities of valuable archaeological samples, and analyses can be performed rapidly, at lower detection limits, in an aqueous saline solution without prior extraction in an organic solvent.

Feigl Test

A Feigl (1966) chemical spot test for tartaric acid/tartrate, with microgram sensitivity, was used for only the Havor sample. A methanol extract showed a dark green fluorescence when irradiated by UV light; malic acid, one of the few cross-interfering compounds, gives a non-fluorescent

greenish solution. The sample was tested together with blanks and solutions of the acids at low concentrations.

Tartaric Acid as the Principal Grape Biomarker in the Near East and Mediterranean.

Barnard *et al.* (2011) recently claimed that malvidin is a better biomarker than tartaric acid/tartrate for identifying the Eurasian grape and its products in the Near East and Mediterranean region, including Italy. However, a recent, very thorough bioinformatics search confirms the long-established and general reliability of Singleton's data (1995), viz., that the concentration of tartaric acid in grape (4000 mg/L) is twenty times that of malvidin (200 mg/L), as a conservative estimate. Natural sources for malvidin, as might be expected for a pigment, are also much more broadly distributed than plants with tartaric acid. They include pomegranate (*Punica granatum*), carrot (*Daucus carota*), apple (*Malus domestica*), whortleberry/ bilberry (*Vaccinium myrtillus*), red clover (*Trifolium pratense*), and crocus (*Crocus sativa*).

Barnard *et al.* (2011) also incorrectly state that Middle Eastern hawthorn fruit has high amounts of tartaric acid. While the tartaric acid concentrations in two Chinese hawthorn species (*Crataegus pinnatifida* and *C. cuneata*) do exceed those of grape (McGovern *et al.* 2004), the chemistries of different species of the same genus in different regions of the world can vary enormously. Unless trade relations can be established by archaeological evidence between diverse regions at the time under consideration, other plants with high tartaric acid–e.g., tamarind from the Indian sub-continent, hawthorn fruit and star fruit from east Asia, or yellow plum from the New World– are irrelevant. For the period of this paper, ca. 1500 B.C. to the early 1st c. A.D. in Europe, no archaeobotanical evidence exists for these non-native plants.

Pomegranate is the only close contender to grape in having relatively large amounts of both tartaric acid and malvidin. Aarabi *et al.* (2008) state that pomegranate has about 600 mg/L of tartaric acid. But this fruit is also inconsequential for this discussion, since no archaeobotanical remains in association with European winemaking installations of the period in view have yet been reported.

Thus, if tartaric acid/tartrate is present in an ancient sample, together with alcohols, esters, aldehydes and terpenoids compounds characteristic of modern grape (as identified by SPME here), then the probability increases for a grape product.

Methodological Approach to Identifying an Ancient Grape Product as Wine

Assuming that tartaric acid/tartrate has been identified in an ancient vessel or utensil, then several other archaeological and enological factors must be assessed, to determine whether the intended product was wine and not another grape product. As argued above, the presence of the Eurasian grape in the mixed beverages is best explained as having been imported as wine from southern Europe. The grapevine did not grow in Scandinavia during this period. The admixture of pine resin in the beverages, a common preservative in ancient wines, supports this interpretation.

In terms of production of the wine in southern Europe, any grape juice there would not have remained non-alcoholic for long in a warm climate, given the slow pressing methods used in antiquity. Grape juice naturally ferments to wine in several days, because yeast (*Saccharomyces cerevisiae*) is always present on some grape skins. These microorganisms thrive in grape juice, which is an ideal medium of water and nutrients for their multiplication, and convert the sugars in the juice into alcohol and carbon dioxide.

Ancient Medicinal Wines and Mixed Fermented Beverages

Chemical analysis opens up a new perspective on ancient pharmacology, even preceding written texts, by providing contemporaneous data on the botanicals added to fermented beverages (McGovern *et al.* 2010). For example, adding a tree resin to wine, to protect against wine disease as well as for medicinal purposes and covering up off-tastes and off-aromas, was a popular and widespread practice throughout the ancient world (McGovern 2009/2010). Later literary references in Pliny the Elder, Strabo, Cato, and others make it abundantly clear that Etruscan wine from central Italy was often mixed with both fresh pine resin and processed pitch to make *vinum picatum* (Latin, "pitched wine") (Hostetter *et al.*, 1994), which left resinous splotches on sidewalls and accumulations on the bases of bronze wine cauldrons at sites throughout Etruscan and Ligurian Italy and Celtic Gaul as early as the fifth century B.C. (Bouloumié 1978). A metal such as bronze did not need to be sealed with tar, as became more customary for pottery amphoras and other containers in later periods. Resinated wines were still being made in the Middle Ages, according to the extensive agricultural and medical compilations based on classical writings, collectively known as the *Geoponica* (see Dalby 2011). The chemical evidence presented here for birch tree resin, juniper, bog myrtle, yarrow, bog cranberry, and lingonberry as adjuncts or additives in the mixed fermented beverages from Scandinavia implies that they were part of an emergent pharmacology there.

Other researchers have begun to report botanical and chemical evidence for herbal concoctions in alcoholic beverages from sites in central and southern Europe. For example, native rosemary, mint, and thyme were added to a

fermented emmer wheat and barley beverage at Genó, near Barcelona in Spain, around 3000 B.C. (Juan-Tresserras 1998). Mugwort (*Artemisia vulgaris* in the wormwood family), also added to some of the early Spanish brews, was hypothesized as an additive, together with carrot, to a dark, sour barley beer (Stika 1996) at the settlement of Hochdorf of the late 5th-early 4th century B.C. This reconstruction was based on the archaeobotanical remains from probable malting and drying ditches, close to the earlier tumulus burial for the Celtic prince honored in death by a cauldron filled with mead (Vorwohl 1985). Significantly, the principal bittering agents in early medieval European beer were bog myrtle, yarrow, meadowsweet, and other herbs (Nelson 2005), which are already being used for this purpose for centuries, even millennia, earlier in Scandinavia.