

**$\delta^{18}\text{O}$ -inferred salinity from *Littorina littorea* (L.) gastropods in a Danish shell midden at the Mesolithic-Neolithic transition**

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**Abstract**

Norsminde Fjord has received extensive geoarchaeological investigation, hosting one of the classic Stone Age shell midden sites in Denmark, and one of the best examples of the widespread oyster decline at the Mesolithic-Neolithic transition. Here intra-shell  $\delta^{18}\text{O}$  (and  $\delta^{13}\text{C}$ ) analyses from the common periwinkle, *Littorina littorea* (L.) are used to infer inter-annual environmental changes at the Mesolithic-Neolithic transition (four from each period). This study utilises a modern  $\delta^{18}\text{O}$  *L. littorea*-salinity training set previously developed for the Limfjord, Denmark to quantify winter salinity.  $\delta^{18}\text{O}$  values range between +1.6 and +4.0‰ in the Late Mesolithic and -6.3‰ to +2.0 in the Early Neolithic. Using maximum  $\delta^{18}\text{O}$  values, winter salinity at the known temperature of growth cessation in *L. littorea* (i.e.  $+3.7 \pm 1^\circ\text{C}$ ) for the first annual cycle of each shell ranges between 25.5–26.8 psu (s.d. 0.56) for the Late Mesolithic, with an average salinity of 26.1 psu. Early Neolithic shells range between 19.4 to 28.2 psu (s.d. 4.59) with an average salinity of 23.7 psu. No statistically significant change in salinity occurs between the Late Mesolithic and Early Neolithic. This result supports recent diatom/mollusc-based inferences that salinity was not the sole cause of the oyster decline,

though some evidence is presented here for more variable seasonal salinity conditions in the Early Neolithic, which (along sedimentary change and temperature deterioration) might have increased stress on oyster populations in some years. It is recommended here that for robust palaeoenvironmental inferences, where possible, multiple specimens should be used from the same time period in conjunction with multiproxy data.

**Keywords:** *Littorina littorea*, oxygen isotope, salinity, Denmark, shell midden, oyster decline, Mesolithic-Neolithic transition, coastal, paleoenvironmental

## Introduction

Since their discovery in Denmark in the mid–nineteenth century (Steenstrup et al., 1851), shell middens (termed kitchen middens or “køkkenmøddinger”, e.g. Fig. 1C), formed in connection with human shell–fishing, have proved one of the best archaeological deposits for studying patterns of human subsistence through time. Extensive excavation of many Danish stratified shell middens have produced a detailed temporal record of changes in human practices and resource exploitation (Andersen, 1989; Andersen, 2007; Andersen, 2008a; Andersen, 2008b), which along with isotopic analysis of bone collagen from humans and domesticated animals (e.g. Fischer et al., 2007; Tauber, 1981) demonstrates that marine resources have played a central role in the subsistence of cultures and societies in Denmark over large parts of the Middle- and Late-Mesolithic (Andersen, 2007; Fischer et al., 2007). At the Mesolithic–Neolithic transition, there is a shift in diet from a largely marine to a predominately terrestrial sourced diet (Fischer et al., 2007), coincidental with a marked change in molluscan faunal composition in many Stone Age shell middens; i.e. from predominately high salinity and warmer-habitat demanding oysters (*Ostrea edulis* L.) in the Mesolithic midden sections to lower salinity and temperature tolerant cockles (*Cerastoderma edule* L.) in the Neolithic sections (Andersen, 2007) and refs therein; Fig. 1D).

Environmental hypotheses (e.g. temperature/salinity decline, increased sediment accumulation; Rowley-Conwy, 1984; Andersen, 2007; Lewis et al., 2016) have been put forward to explain this widespread faunal shift in Danish shell middens, but to date the role played by intra–annual/seasonal climate change remains unknown and untested. This is due to the almost total lack of seasonal/intra–annual resolution data concerning salinity or temperature change available from coastal Denmark over the Holocene in the direct vicinity of important archaeological settings; in fact only one quantitative record of Holocene temperature change (i.e. pollen–inferred January and July temperature (i.e. Brown et al., 2012) exists for the entire country. One potential method of inferring seasonal/intra–annual

temperature and salinity change is to analyse  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  profiles along the direction of growth in molluscs present within the shell middens (and collected from the adjacent fjord by humans) (Burman and Schmitz, 2005; Andrus, 2011; Leng and Lewis, 2016).

Here we investigate seasonal/intra-annual climate variability (particularly salinity and temperature) through  $\delta^{18}\text{O}$  (and  $\delta^{13}\text{C}$ ) data from periwinkles (*Littorina littorea* L.) collected by humans and deposited in the Norsminde shell midden (Fig. 1). Eight periwinkles were analysed from stratigraphic midden layers, either side of the culturally important Mesolithic/Neolithic transition and broadly synchronous with the oyster decline (both dated to ca. 5900 cal. yrs BP; Andersen, 2007) detected in the shell midden. To test repeatability, four shells were analysed from each of two layers near the Mesolithic-Neolithic Transition, one layer (L7; Fig 1D) from the Late Mesolithic (LM) when oysters were abundant and the other layer (L4; Fig 1D) from the Early Neolithic (EN) when the shell midden is dominated by cockles (and blue mussels, *Mytilus edulis* L.).

We also attempt to quantify winter salinity from the  $\delta^{18}\text{O}$  isotope data using the previously determined isotope-temperature-salinity relationship (or transfer function) for *L. littorea* (Burman and Schmitz, 2005) established for the Limfjord (inland coastal waters of North Jutland, Denmark). This transfer function was previously applied to two subfossil periwinkles from the Mesolithic Danish Ertebølle *locus classicus* shell midden (Burman and Schmitz, 2005), and to Eemian specimens from the English channel and the Swedish Kattegat to compare sea surface temperatures and salinity during the Eemian with present day (Burman and Pässe, 2008). The Danish Ertebølle study, though based on two shell, gave some indication that within the Mesolithic Ertebølle period (ca. 7,400–5,900 cal. yrs BP), both salinity and sea surface temperature were higher in the central Limfjord than present day (Burman and Schmitz, 2005).

Building on this previous work, here we aim to determine if growth series data from *L. littorea* specimens is repeatable throughout individual shell midden layers. This will better enable us to assess the potential of this technique for inferring qualitative (and potentially quantitative) temporal changes in salinity (and/or temperature) and seasonality directly from stratigraphic shell middens over the entire shell midden accumulation (i.e. occupation) phase. This is critical for robustly testing if changes in faunal composition within shell middens correspond with environmental/climatic change (e.g. Fig 1D), and subsequently whether clear links can be determined between environmental (including climate) change, shell fish availability and human subsistence trends (i.e. environmental hypotheses of cultural change). Furthermore, to fully understand human-environment interactions through time, the role played by

seasonal environmental/climate change must be considered in geoarchaeological investigation.

## Methods and materials

Norsminde Fjord (56 °01'018"N, 10 °14'049"E) is a small estuarine system (~6 km long, 250 m wide, surface area of ~1.68 km<sup>2</sup>, max. depth 2 m, average depth 0.6 m) situated in a glacial meltwater erosion basin, along the east coast of Jutland about 20 km south of the city of Århus (Fig. 1). It is predominately fed by one major inflow (Odder Å) that drains a catchment area of ~101 km<sup>2</sup> (Nielsen et al., 1995). Odder Å (fed by several tributaries) is responsible for draining 85 % of the catchment before entering the fjord in the south–west at its innermost section (Fig. 1B). This creates a salinity gradient within the fjord with salinities of ~0 psu freshwater occurring at the mouth of the Rævs Å, gradually increasing up to ~24 psu at the Kattegat entrance (with most of the fjord around ~18–20 psu). At present Norsminde Fjord has no regular temperature or salinity stratification regime. Recent annual monitoring of Norsminde Fjord at a sampling site in the northern area of the fjord (Fig. 1B) showed that temperature varies by ~20°C over the year (range 0.5 to 20.0°C in 1989/90) with the maximum occurring in July and the minimum in February (Therkildsen and Lomstein, 1994). In the same year salinity varied between 15.0 to 26.8 psu.

Norsminde Fjord has been subject to extensive archaeological research (e.g. Andersen, 1976; Andersen, 1983; Andersen, 1989; Bailey and Milner, 2008; Andersen and Malmros, 1965; Gabrielsen, 1953) and is now one of the most important Stone Age regions in Denmark, with its shorelines hosting a whole series of Mesolithic and Neolithic aged coastal settlements (e.g. kitchen middens; Flynderhage, Norslund, Kalvø; Gabrielsen, 1953; Andersen and Malmros, 1965; Andersen, 1983; Fig 1D) and individual archaeological finds (e.g. flint tools, various axes, bones/antlers). Of interest here, the characteristic kitchen midden ("køkkenmødding"; Fig. 1C) spanning the Mesolithic–Neolithic transition was unearthed during the latest major excavation (1972–1989; Andersen, 1989) and clearly documents the importance of the fjord and marine environment to Stone Age cultures and societies. The *L. littorea* shells analysed in this study were retrieved during this excavation period (Andersen, 1989). The mollusc fauna of this shell midden is diverse, though it is generally dominated by *Ostrea edulis* (European flat oyster), *Cerastoderma edule* (common cockle), *Mytilus edulis* (blue mussel) and *Littorina littorea* (common periwinkle), the abundances of which change markedly throughout the stratigraphic layers (Bailey and Milner, 2008; Fig. 1D). For this study, only well-preserved shells were selected for analysis, i.e. they had a continuous section from the apex to outer margin, with no visible signs of

erosion. All shells selected were relatively large (i.e. diameter >1.7cm, height >2.2cm), avoiding juvenile or very young specimens. Shell details are provided in Table 1.

The common or edible periwinkle (*L. littorea*) is common in the upper shore region down to the shallow sublittoral, particularly favouring rocky shores, but is also abundant on soft substrates (such as in estuaries and fjords) and algal biofilms (Fretter and Graham, 1980; Petersen, 2004). The species is omnivorous, feeding on small invertebrates such as barnacle larvae, as well as macroalgae, microalgae and germlings; Watson and Norton, 1985, Wilhelmsen and Reise, 1994). In Denmark *L. littorea* is present today on all coasts (apart from exposed sandy beaches) as far south east as Bornholm in the Baltic Sea (Petersen, 2004; Fretter and Graham, 1980). Since the Littorina transgression in Denmark/Kattegat (ca. 9,600–8,000 cal. yrs BP; Petersen, 1981; Bendixen et al., 2015; Christiansen et al., 1993; Bennike et al., 2000; Bennike et al., 2004), *L. littorea* has been continually present (and often abundant) throughout the Danish waters and the Baltic Sea (commonly as part of the *Mytilus* epifauna; Petersen, 2004; Petersen et al., 2005), extending as far as Estonia up until ca. 3,000 BP (Petersen, 2004). It has remained present north of Bornholm throughout the late Holocene up until present day.

*Littorina littorea* has a broad temperature and salinity tolerance making it useful for isotope-based archaeological studies, rarely disappearing completely from the record in Danish shell deposits north of the Belt Sea's and Øresund. The lower range of salinity tolerance of *L. littorea* is unknown, but it can thrive in salinities >11.5–14 psu through to fully marine conditions (~35 psu), and can survive for short periods of time in salinities <14 psu, though relatively inactive (Todd, 1964; Rosenberg and Rosenberg, 1972). The lower temperature limit of *L. littorea* is below zero (perhaps as low as –13°C), through to ~+35°C (Davenport and Davenport, 2005). A previous stable isotope study of *L. littorea* shells show that seasonal/annual (and longer-term) changes in the temperature or salinity of the ambient water are reflected in the isotopic composition of the shell (principles outlined in Fig. 2), making this species a potentially valuable recorder of past environmental/climatic conditions (Andreasson et al., 1999).

Whilst reproduction can occur most of the year in some environments, in more temperate climates such as the UK and Denmark, it is common only in late winter/spring (Fretter and Graham, 1980). The larval stage usually lasts between 2–6 weeks (being temperature-dependent) and can result in dispersal distances of over 10 km (Fish, 1972; Fish, 1979). Naturally, *L. littorea* reaches maturation in 1–3 years (Williams, 1964; Fish, 1972; Fretter and Graham, 1980) and can live for over 9 years (Heller, 1990), though specimens found in archaeological deposits are often much younger (here, collected at 2–3 years old based on

the number of  $\delta^{18}\text{O}$  cycles apparent in each shell; Fig. 3A, Table 1). Growth rates are generally highest in the early years due to an ontogenetic decrease in growth for *L. littorea* (Williams, 1964; Andreasson et al., 1999; Ekaratne and Crisp, 1984).

## **Analytical methods**

All eight periwinkle shells were cleaned thoroughly by brief submersion in (5%) sodium hypochlorite, followed by rinsing and scrubbing of the outer surface with deionised water to remove any extraneous material. After drying, each shell was successively drilled along the direction of growth (from the apex to the aperture) using a microdrill fitted with a 0.3 mm diameter drill bit. A sampling resolution of  $\sim 0.3$  mm (amounting to 54–83 samples per shell; 542 samples in total) was obtained, with care taken to drill only the outer prismatic calcium carbonate layer (Leng and Lewis, 2016). Approximately 50–100 micrograms of carbonate were used for isotope analysis using an Isoprime dual inlet mass spectrometer plus Multiprep device. Samples are loaded into glass vials and sealed with septa. The automated system evacuates vials and delivers anhydrous phosphoric acid to the carbonate at  $90^\circ\text{C}$ . The evolved  $\text{CO}_2$  is cryogenically cleaned and passed to the mass spectrometer. Isotope values ( $\delta^{13}\text{C}$ ,  $\delta^{18}\text{O}$ ) are reported as per mille (‰) deviations of the isotopic ratios ( $R^{13}\text{C}/^{12}\text{C}$ ,  $^{18}\text{O}/^{16}\text{O}$ ) calculated to the VPDB scale using a within-run laboratory standard calibrated against NBS-19:  $\delta = (R_{\text{sample}}/R_{\text{standard}} - 1) \times 1000$ . Craig correction is also applied to account for  $^{17}\text{O}$ . Analytical reproducibility of the standard calcite (KCM) is  $< 0.1\text{‰}$  for  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$ .

## **Quantification of salinity and temperature**

Using the above mentioned Limfjord training set (Fig. 1A) quantifying relationships between  $\delta^{18}\text{O}_{\text{water}}$  and  $\delta^{18}\text{O}_{\text{shell}}$  in modern *L. littorea* specimens over a salinity/temperature gradient,  $\delta^{18}\text{O}$  from subfossil *L. littorea* shells can be used to quantify winter salinity and summer temperature changes with accuracy over Holocene timescales. This is based on the assumption that the annual range for variation in the shell  $\delta^{18}\text{O}$  values is largely determined by the seasonal change in water temperature and that the seasonal salinity changes are small in comparison between April and December across the Limfjord, averaging  $3.37\text{‰}$  (range 3 to  $3.6\text{‰}$ ; Burman and Schmitz, 2005). Burman and Schmitz (2005) found a relationship of  $+0.25\text{‰}$   $\delta^{18}\text{O}$  change per +1 unit of salinity (psu) and  $-0.22\text{‰}$   $\delta^{18}\text{O}$  change per  $+1^\circ\text{C}$  in modern specimens, and argue that this relationship has not changed over the Holocene. Based on the linear relationship for  $\delta^{18}\text{O}$  of the shell calcite versus temperature, Burman and Schmitz (2005) determined that shell growth ceases around  $3.7^\circ\text{C}$  for the modern Limfjord *L. littorea* specimens. As the  $\delta^{18}\text{O}$  record reflects largely the combined

effects of salinity and temperature, then if one of these parameters is known the other can be inferred with high accuracy (within  $\sim 1\text{-}2^\circ\text{C}$  or  $1\text{-}2\text{psu}$ ; Burman and Schmitz, 2005), on the assumption that the modern relationship holds true for the sub-fossil shells (method for conversion outlined in Supplementary Material). Here, as the winter temperature is known at the point that growth ceases (i.e.  $3.7\pm 1^\circ\text{C}$ ), salinity at this time can be calculated. Taking the first clear winter maximum  $\delta^{18}\text{O}_{\text{shell}}$  in the sub-fossil shells as the point at which growth ceases (see Fig. 3A), we used the modern relationship for *L. littorea* determined by Burman and Schmitz (2005) to convert each shell  $\delta^{18}\text{O}$  (VPDB) winter value into  $\delta^{18}\text{O}_{\text{water}}$  (VSMOW) (Fig. S1) at the point of growth cessation (which would be at  $3.7\pm 1^\circ\text{C}$ ). We then converted each  $\delta^{18}\text{O}$  (VSMOW) value into a salinity estimate using the modern relationship between  $\delta^{18}\text{O}_{\text{water}}$  (VSMOW) and salinity (i.e.  $0.25\text{‰}$  per salinity unit) (Fig. S2).

In the sub-fossil specimens from the Norsminde shell midden, where annual cycles are clear (see Fig. 3A) the highest  $\delta^{18}\text{O}$  value is used to determine the point at which growth stops and subsequently the marker for  $3.7^\circ\text{C}$ . This is supported by regional temperature reconstructions which suggest that over the study period average winter temperature were below  $3.7^\circ\text{C}$  (Antonsson and Seppä, 2007; Seppä et al., 2009; Brown et al., 2012) and in most shells the highest  $\delta^{18}\text{O}$  values are often followed by a relatively rapid decrease in  $\delta^{18}\text{O}$ . This follows the methodology used by Burman and Schmitz (2005) and is verified by the modern *L. littorea* specimens. Andreasson et al. (1999) highlight that only the  $\delta^{18}\text{O}$  cycle closest to the apex should be used for estimation of annual ranges of surface-water temperature due to the cessation or decrease in growth related to ontogeny and spawning after 2-3 years. In contrast to the Limfjord sites sampled by Burman and Schmitz (2005), Norsminde Fjord currently exhibits relatively large annual differences in salinity ( $>10\text{psu}$ ; Therkildsen and Lomstein, 1994). This means that summer maximum temperature cannot be calculated using the methods employed by Burman and Schmitz (2005) (in the absence of high-quality independent palaeo-temperature curve, as an unknown portion of the change in the annual  $\delta^{18}\text{O}$  will be due to salinity change (resulting from variable incorporation of freshwater into the fjord between wetter and drier months and potentially evaporation during warmer summers).

## Results

The individual shell profiles for  $\delta^{18}\text{O}$  are provided in Figure 3A and shell metrics (range, minimum, maximum, standard deviation, and winter salinity inferences) are provided in Table 1 and Figure 3B. Whilst the sample population is low ( $n = 8$ ) for a season of collection study, these periwinkles appear to have been collected predominately in the

summer/autumn for the Mesolithic shells and the autumn/winter for the Neolithic shells (Table 1).  $\delta^{13}\text{C}$  values range between  $-2.3\text{‰}$  to  $+1.6\text{‰}$  (VPBD) in the Late Mesolithic and  $-9.0\text{‰}$  to  $+3.2\text{‰}$  in the early Neolithic shells, the predominantly high values suggesting that shell carbon is sourced from marine dissolved inorganic carbon. Due to the focus of this study on changing  $\delta^{18}\text{O}$  derived salinity/temperature signals from *L. littorea* shells, no further consideration of the  $\delta^{13}\text{C}$  data is provided here (see Supplementary Material for more details).

## **$\delta^{18}\text{O}$ and winter salinity estimates**

$\delta^{18}\text{O}$  values range between  $-6.3\text{‰}$  and  $+2\text{‰}$  (VPBD), with generally a similar overall distribution of values in both the Mesolithic and Neolithic shells ( $-4\text{‰}$  to  $+2\text{‰}$ ), but with the very lowest values (below  $-4\text{‰}$ ) occurring only in the Early Neolithic shell samples. All shells show at least one or more complete annual cycle (see Fig. 3A, Table 1), though in some cases annual cycles are difficult to determine confidently (labelled in Fig. 3A). The highest  $\delta^{18}\text{O}$  values (lowest temperatures) tend to be followed by a sharp decrease in  $\delta^{18}\text{O}$  values, marking the start of the winter growth cessation, then growth resuming in the warmer months in these specimens.

Winter salinity inferences for the Mesolithic shells range between 25.5–26.8 psu (standard deviation of 0.56 psu) with an average salinity of 26.1 psu. The average winter salinity inference for the Neolithic is slightly lower (i.e. 23.7 psu) than for the Mesolithic, but Neolithic values are generally more variable (standard deviation of 4.59), ranging from 19.4 to 28.2 psu. The salinity calculations are shown in the Supplementary Material (see Fig. S1 and S2). No significant change is apparent between Mesolithic and Neolithic average winter salinity (t-test with unequal variances:  $p > 0.05$ ).

## **Discussion**

### **$\delta^{18}\text{O}$**

The cyclical variability displayed by the sub-fossil *L. littorea* shells from Norsminde primarily reflects the annual changes in water temperature and salinity of the fjord and growth patterns of the individual shells (cf. Andreasson et al., 1999; Burman and Schmitz, 2005; Fig. 3A). Within a shell, winters are represented by highest  $\delta^{18}\text{O}$  values and therefore changes are largely driven by temperature due to the inverse temperature- $\delta^{18}\text{O}_{\text{shell}}$  relationship in Danish coastal waters ( $-0.22\text{‰}$  per  $^{\circ}\text{C}$ ; Burman and Schmitz, 2005). This results in  $\delta^{18}\text{O}$  minima during the summer, due to the associated large temperature increase, which outweighs annual salinity changes (e.g.  $20\text{ }^{\circ}\text{C}$  annual temperature fluctuation from January



to July, compared to ~12 psu salinity change in 1989/90; Therkildsen and Lomstein, 1994). However, salinity changes will have an impact on the  $\delta^{18}\text{O}_{\text{shell}}$  content (+0.25‰  $\delta^{18}\text{O}$  change per +1 psu; Burman and Schmitz (2005)) over the year primarily due to changing freshwater input (Burman and Schmitz, 2005), though evaporation might contribute during particular warm/dry periods, e.g. during the Holocene thermal maximum (HTM).

A positive salinity- $\delta^{18}\text{O}_{\text{shell}}$  relationship exists in Danish coastal waters (0.25‰ per psu; Burman and Schmitz, 2005), driven by highest levels of precipitation input in winter (modern precipitation for South Scandinavia is ~-8 to -11‰ (VSMOW) yielding lower  $\delta^{18}\text{O}$  than adjacent coastal/marine waters ~-4‰ (VSMOW); Frohlich et al., 1988; IAEA, 2001; Burgman et al., 1987; LeGrande and Schmidt, 2006). Whilst it is possible that some of the winter precipitation is locked up in snow and ice and released later in the season, we believe that in the late Mesolithic period, at the peak of the HTM, there was likely to have been less snow/ice cover effects (Brown et al., 2012). As indicated above, these effects mean that absolute annual temperature information cannot be extracted from the  $\delta^{18}\text{O}_{\text{shell}}$  data. The only time that relative temperature and salinity effects can be separated and a quantitative inference produced is for winter salinity, at a time when the temperature is known: i.e. here at 3.7°C, the point that growth ceases in the *L. littorea* shells (based on modern specimens; Burman and Schmitz, 2005). The (winter) salinity within Norsminde fjord is driven by the relative mixing of freshwater and seawater, with high salinity water input from the Kattegat and freshwater from the catchment delivered predominately by fluvial inputs (Fig. 1B). Salinity in the fjord was higher in the Late Mesolithic/Early Neolithic than present day (Lewis et al., 2016), due to higher sea-levels (Berglund et al., 2005; Christensen, 2001) and consequently a wider and more open with the Kattegat. This is also supported by the salinity inferences presented here (Table 1; Fig. 4E) and discussed in more detail below.

Here, short-term variations in the winter  $\delta^{18}\text{O}$  maximum (i.e. year to year variation in isotope values within shells and between shells from the same layer at the 3.7°C growth cease) are interpreted as salinity change, linked to climate (i.e. freshwater input vs marine water), rather than collection location and movement of *L. littorea* specimens along the salinity gradient within Norsminde Fjord (see below). Whilst some evidence exists for a fall in sea-level near the Late Mesolithic/Early Neolithic boundary (Berglund et al., 2005; Christensen, 1995), shorter term (annual to sub-decadal) variations in sea-level are unlikely to be of large enough magnitude to effect marine water exchange and fjord salinity alone. It is also important to note that there is substantial fluctuation of summer minimum  $\delta^{18}\text{O}$  values within shells, suggesting that summer salinity and/or temperature fluctuated from year to year over the study period, though as these signals cannot be separated without a high-quality independent salinity or temperature record, we focus here on winter salinity.

Change in average winter salinity is minimal (and not significant) between shells from the Late Mesolithic and those from the Early Neolithic, though there is some difference in the range and variability (as shown by the standard deviation of all  $\delta^{18}\text{O}$  values) in shells from the Early Neolithic (Fig. 3A and 4E). Given the salinity gradient within the fjord today (and likely during the late Mesolithic/Early Neolithic), this could be explained by several possible mechanisms including a change in collection location, movement/displacement of *L. littorea* shells or climate/sea level variability, the latter affecting inputs of fresh- and saline- water into the fjord and consequently salinity. A change in the collection location of some of the Early Neolithic shells is possible (e.g. L4-1 and L4-2 give lower winter salinity estimates; Table 1), though we deem this unlikely to be the controlling factor as *L. littorea* was harvested in much smaller quantities than oysters or cockles, and therefore unlikely to be placed under such resource pressure. Though not systematically measured (as for oysters and cockles; Bailey and Milner, 2008) no report of any change in average size of *L. littorea* shells has been reported. Hence it is likely that they were available in sufficient supply and most likely collected at the most convenient (nearby) location.

Movement of the individual *L. littorea* shells from high to low salinity parts of the shore is also possible, but unlikely. *Littorina littorea* is capable of some movement, and able to move up and down the shore, often tracking the tide at slow speeds ( $\sim 2 \text{ cm min}^{-1}$ ) to maintain immersion, feeding and optimum salinity (Newell, 1958). However, in terms of moving from higher to lower salinity areas within the fjord, this seems unlikely, as individuals tend to crawl down the shore with the receding tide and then return to the same location as the tide comes in, often leaving U-shaped trails in sandy/muddy sediment (Newell, 1958). Wind and wave action could redistribute individuals, though in a relatively sheltered shore with a small tidal range ( $< 1 \text{ m}$ ) and generally low energy system, living individuals are unlikely to be re-distributed long distances.

The most likely explanation for these varying trends is fluctuation in annual climatic conditions. The Early Neolithic was a time of changing climate, with cooling temperatures (Seppä et al., 2009; Antonsson and Seppä, 2007; Brown et al., 2012) and increased precipitation or surface wetness (Seppä et al., 2005) as the Holocene thermal maximum began to wane. Layers in shell middens cover relatively coarse time periods (perhaps up to 200 years in Norsminde; Andersen, 1989; Bailey and Milner, 2008), so the lifespans of individual molluscs (here, 2-3 years) within a layer are unlikely to overlap. Variable summer/winters (e.g. warmer or colder and wetter or drier years) were likely recorded in some specimens and not others, which highlights a danger of using single shell analyses which may be skewed by atypical conditions. Furthermore, in a transitional climate period, fluctuating temperature and rainfall (leading to salinity variations) between extremes might

be expected, hence explaining the larger range and lower  $\delta^{18}\text{O}$  winter values (and lower salinity) in two of the four Early Neolithic shells. Sea-level might also have contributed to the freshwater-marine water balance within the fjord (and consequently  $\delta^{18}\text{O}$  content of the water and *L. littorea* shells), with higher sea-levels likely to increase the volume of high-salinity marine water entering the fjord. The regional sea-level curves (Fig. 4D) suggest some potential decline between the Late Mesolithic and Early Neolithic (particularly at Blekinge, though more muted and fluctuating at the closer site Vedbæk; Fig. 4D) but the DI-salinity from Norsminde Fjord suggests that this did not manifest in long-term changes to average salinity of the Fjord (Fig. 4E). However, shorter-term atmospheric/climate driven sea-level variability (e.g. Andersson, 2002; Woolf et al., 2003) combined with variable freshwater input might have affected salinity within the fjord on annual to sub-decadal timescales.

When converted to salinity (at the  $3.7\pm 1^\circ\text{C}$  temperature induced growth cessation) using the modern training set (after Burman and Schmitz, 2005), the  $\delta^{18}\text{O}$  data shows a larger range in winter salinities in the Early Neolithic than in the Late Mesolithic (Fig 4E; Table 1). A shift towards wetter conditions is widely observed in the Early Neolithic (Snowball et al., 2004; Seppä et al., 2005) and potentially increased the number of extreme wet winters. This would in turn increase the input of freshwater into fjord systems such as Norsminde Fjord and could account for the occasional low salinities recorded in the *L. littorea* shells. It is important also to acknowledge that there is some contradictory evidence suggesting that drier conditions might have prevailed in the Early Neolithic (e.g. Lake Bliden; Olsen et al., 2010) and that precipitation/surface wetness likely exhibits regional variability. Although no data are available specifically for the Norsminde Fjord catchment, short-term/annual winter (and/or summer) extremes are possible under any climate regime and highlights the need for independent multiproxy data and high-quality local and regional records.

### **Comparison with regional parameters**

During the late Stone Age (ca. 8,000-3,700 cal. yrs BP), both sea-level (e.g. Berglund et al., 2005; Christensen, 2001; Fig. 4D) and salinity (e.g. Petersen, 2004; Lewis et al., 2016; Weckström et al., 2017; Fig 4E) were higher than present day in Denmark and western areas of the Baltic Sea. The winter (minimum) salinities presented here (from the *L. littorea* shells) also imply higher salinity than present day within Norsminde Fjord, though the  $\delta^{18}\text{O}$  based estimates generally suggest higher salinity compared to the diatom record (Lewis et al., 2016) (Fig. 4E). This offset between the DI-salinity and *L. littorea* winter minimum salinity is likely explained by DI-salinity inferences not being season-specific, averaged over several years and incorporating diatoms from a spatially wider area of the fjord (i.e. lower

salinity inferred from the assemblage), in addition to standard error in both methodologies. Furthermore, it is likely that the periwinkles were originally collected in close proximity to the shell midden, which is located much closer to the mouth of Norsminde Fjord than the sediment coring site, and therefore subject to higher salinities (Fig. 1B)

Burman and Schmitz (2005) originally used the methodology employed here to reconstruct changes in two sub-fossil shells from the Ertebølle shell midden (with these two shells dated to the Late Mesolithic). The data presented here suggest lower salinity in Norsminde Fjord (26.1 psu) than at Ertebølle (i.e. 31 psu) during the Late Mesolithic. This is expected due to where Ertebølle is in the Limfjord, and in closer proximity to the high salinity water of the North Sea (Fig. 1A). Modern-day salinity at Ertebølle is ~26 psu compared to an average of ~18-20 psu at Norsminde Fjord, though it is important to highlight that a different land-sea configuration (e.g. Kysing Fjord; Fig. 1B) existed in both the Limfjord and Norsminde Fjord at that time due to isostatic uplift and higher sea level (Andersen, 1989; Christensen, 2001). During the mid-Holocene high sea-level stand, an opening to the Skagerrak and North Sea in the north has been inferred (Andersen, 1995; Andersen, 2007; Petersen et al., 2005), closer to the Ertebølle site than the modern-day entrance in the west (Fig. 1A). Also, during the mid-Holocene (ca. 7000–4000 cal. yrs BP), Norsminde Fjord was significantly larger incorporating the Kysing palaeo-fjord (~10 km long, 2.7–3 km wide and ~500 m at the Kattegat mouth, Andersen, 1989; Fig. 1B). A wider connection between Norsminde Fjord and the Kattegat under higher sea-levels would have improved exchange of marine water (and sediment) resulting in higher fjord salinity and lower sedimentation rates (Lewis et al., 2016). This created optimal conditions for a variety of marine molluscs such as oysters, an important component of Stone Age shell middens, particularly in the Late Mesolithic layers (Andersen, 2007; Bailey and Milner, 2008). However, this connection started to narrow sometime in the late Mesolithic (though exact date remains unknown), due to longshore transport of sediment and subsequent development of beach ridges (Andersen, 1989).

#### **Archaeological debate: salinity change at the Mesolithic–Neolithic transition**

This issue of salinity change at the Mesolithic-Neolithic transition and its potential role in the oyster decline is briefly revisited here, using shell  $\delta^{18}\text{O}$  as an independent proxy for salinity change. Lewis et al. (2016) argue that there was no sudden salinity decline or persistently lower salinity in the Danish coastal waters in the Early Neolithic period, and therefore rebuke the hypotheses that declining salinity caused the reduction in oysters present in numerous coastal shell middens across Denmark, including Norsminde (Rowley-Conwy, 1984; Andersen, 2007). The *L. littorea* isotope results presented here support Lewis et al. (2016) as statistical analyses suggest that there is no significant ( $p>0.05$ ) change in winter salinity

between the Late Mesolithic and Early Neolithic (Table 1; Fig. 4E). However, it is important to highlight here that this is only a comparison between two layers of the Norsminde shell midden (with 600-700 years time difference) and higher resolution (and longer-term) changes still need investigation.

For the oyster decline, the change in seasonality might be of importance, with salinity (and  $\delta^{18}\text{O}$  values) appearing to exhibit greater variability in the Early Neolithic than in the Late Mesolithic (both a higher range and standard deviation is observed between these layers; Fig. 3B, 4E). Greater seasonal salinity (and temperature) fluctuations might also have contributed to the reduction in oysters, by adding further stress upon accessible oyster populations in addition to increasing sedimentation rates in fjords (Fig. 4C) and a general temperature decline (Fig. 4A; outlined by Lewis et al., 2016). The isotope data suggest that in some years winter salinity fell near or below the minimum level required to support breeding oyster populations (i.e. ~23 psu; Jensen and Spärck, 1934; Spärck, 1924; Yonge, 1960). Whilst oysters require water temperatures above ~15°C to breed (Jensen and Spärck, 1934; Spärck, 1924; Yonge, 1960), in some years low salinity could have extended into the warmer months (e.g. low salinity winters followed by wet springs/summer) and narrowed the breeding season (in spring/summer). In these years human exploitation with reduced replenishment might have caused decline/stress of oyster populations. In addition, Norsminde Fjord was near the southern limits of oyster habitation in the inner Danish waters (Jensen and Spärck, 1934; Petersen, 2004) even in the late Stone Age (7,400-3,700 cal. yrs BP), when salinity and sea-levels were above present day (Berglund et al., 2005; Christensen, 2001), perhaps with the result that small fluctuations in salinity might have had severe impacts. This, coupled with changing sedimentation patterns within the fjord (Lewis et al., 2016; Fig. 4C) and climate change, including both temperature decline (Snowball et al., 2004; Antonsson and Seppä, 2007; Brown et al., 2012; Seppä et al., 2009; Fig 4A) and a potential shift towards wetter conditions (Seppä et al., 2005; Fig. 4B), might have put cumulative stress on oyster populations in Danish coastal waters. However, as layer 4 is several hundred years later than the oyster decline, further assessment of seasonal change is needed from earlier layers (i.e. layers 5 and 6; Fig. 1D) of the Norsminde shell midden.

#### **Future implications**

This study uses multiple shells from individual layers of a shell midden sequence. The data presented here demonstrate clear inter-layer variability between shells; e.g. Neolithic minimum winter  $\delta^{18}\text{O}$  in the first annual cycle range from -0.77 to +1.98 resulting in salinity inference ranges from 19.4 to 28.2 psu. This expresses the need for care in single shell per layers studies. For example, from this study, while the Late Mesolithic shells produce very

similar inferred salinity, the variability within the Early Neolithic shells demands that at least 3 shells are used to adequately provide a useful mean salinity for this period and give a realistic idea of its variability. Comparing any of the four Late Mesolithic shells with any from the Early Neolithic, winter salinity might be inferred to have fallen (L4-1, L4-2), stayed broadly similar (L4) or risen (OEM) across the Mesolithic-Neolithic transition. Therefore, in order to avoid inferences being potentially skewed by this variability, we suggest that future studies of this type should use multiple specimens per layer if possible. In cases where this is unfeasible, perhaps due to preservation of suitable specimens or time/money constraints, then results should be interpreted with extreme caution and compared against other, independent multiproxy data.

Lastly, we acknowledge that finer resolution drilling and implementation of sclerochronological techniques could have provided further information concerning the timing, duration and rate of growth in the analysed shells (Schöne, 2008; Wingard and Surge, 2017; Schöne and Gillikin, 2013) and should be incorporated in future studies. However, for the sake of this study, we argue that these shells were drilled at sufficient resolution (0.3mm) to capture intra-annual variation as demonstrated by the similar intra-shell isotope profiles to previous studies (e.g. Andreasson et al., 1999; Burman and Schmitz, 2005) and further sclerochronological analyses are unlikely to have altered final salinity/seasonality inferences and interpretation.

## Conclusions

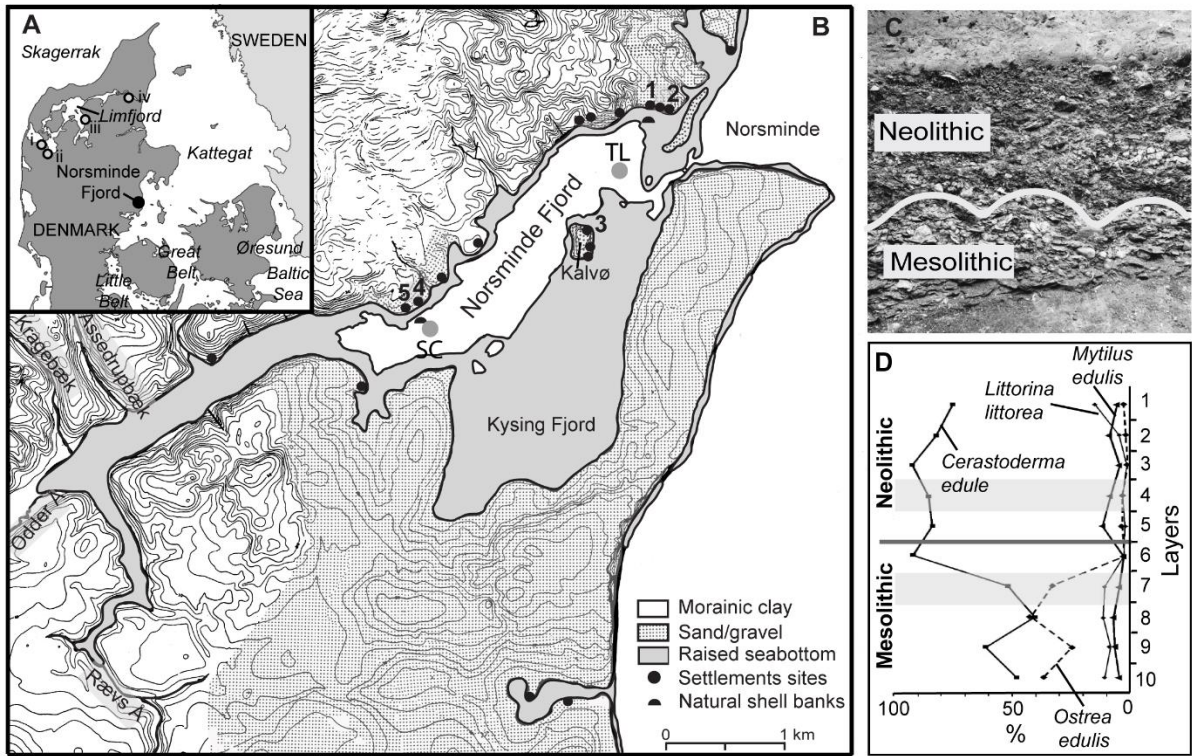
Using the modern relationship between  $\delta^{18}\text{O}_{\text{shell}}$  in *L. littorea* with temperature and salinity, the maximum  $\delta^{18}\text{O}$  values for the first annual  $\delta^{18}\text{O}$  cycle can be used to calculate winter salinity at the known temperature of growth cessation in *L. littorea* (i.e.  $+3.7 \pm 1^\circ\text{C}$ ; as derived from a Limfjord based training set). Here,  $\delta^{18}\text{O}$  –based quantitative reconstruction of salinity from subfossil *L. littorea* shells has provided further insight on the environmental conditions at the Mesolithic-Neolithic transition in Denmark and the widespread oyster decline. The data presented here support the inference that no change in average salinity occurred between the Late Mesolithic and Early Neolithic period, but shows that the Early Neolithic was characterised by a more variable climate (and consequently salinity). High-resolution (seasonal to inter-annual) isotopic analysis of *L. littorea* shells from archaeological deposits (and sedimentary cores) offer great potential in future geoarchaeological studies. Shell geochemistry from the common periwinkle can provide detailed information on seasonal to inter-annual conditions and, with a modern training set, can be used to infer palaeo-salinity quantitatively. Further, as *L. littorea* was commonly collected for subsistence purposes by prehistoric people and is able to tolerate a wide range of temperatures and salinities, it does

not easily disappear from the record entirely under changing environmental conditions. As it has a more continuous and abundant presence than other common molluscs, it is very suited for such isotopic or sclerochronological work. However, in order to enhance geoarchaeological debate, certain criteria must be met for any such target mollusc. These include a knowledge of modern-day ecology and local and regional shell-secretion response to key environmental parameters (i.e. temperature and salinity), clear growth cycles within the sub-fossil shells, the use of multiple shells from individual layers/time periods and wherever possible, other independent multiproxy data for comparison.

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Figures:

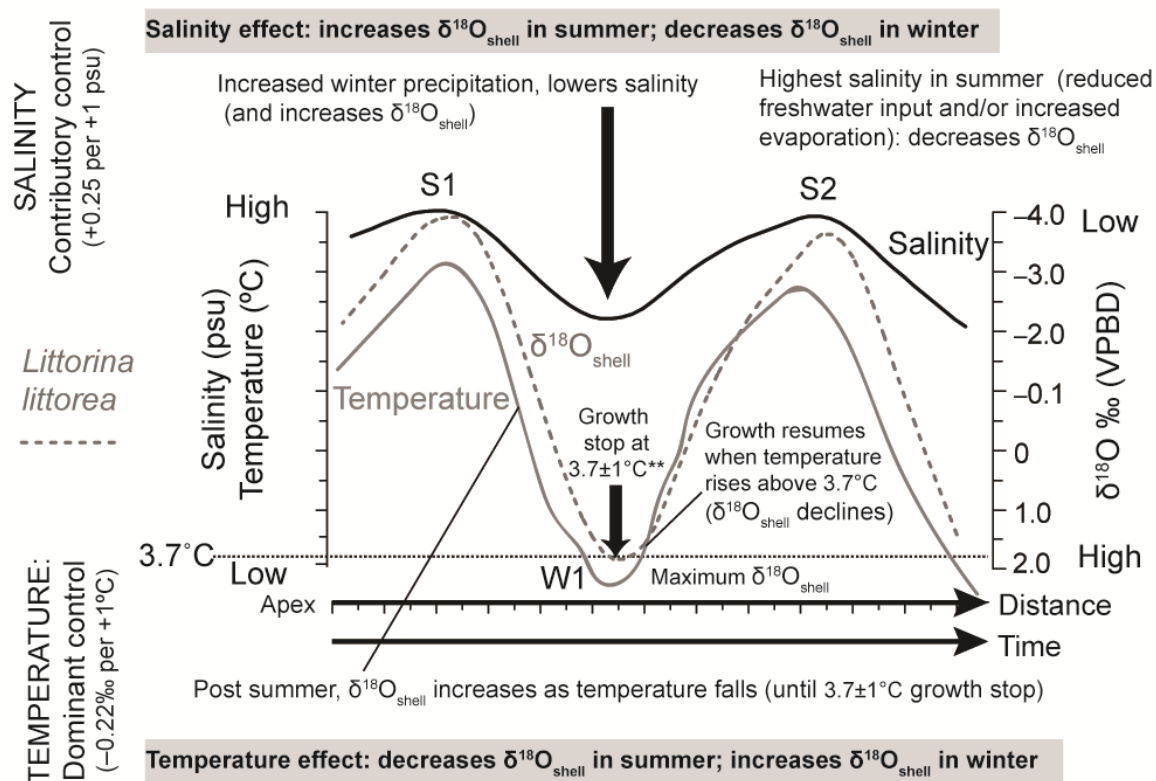


**Figure 1:** Study area and shell midden data. A. Map of Denmark showing location of the study area and the training sites (i-iv) incorporated in the Burman and Schmitz (2005) study

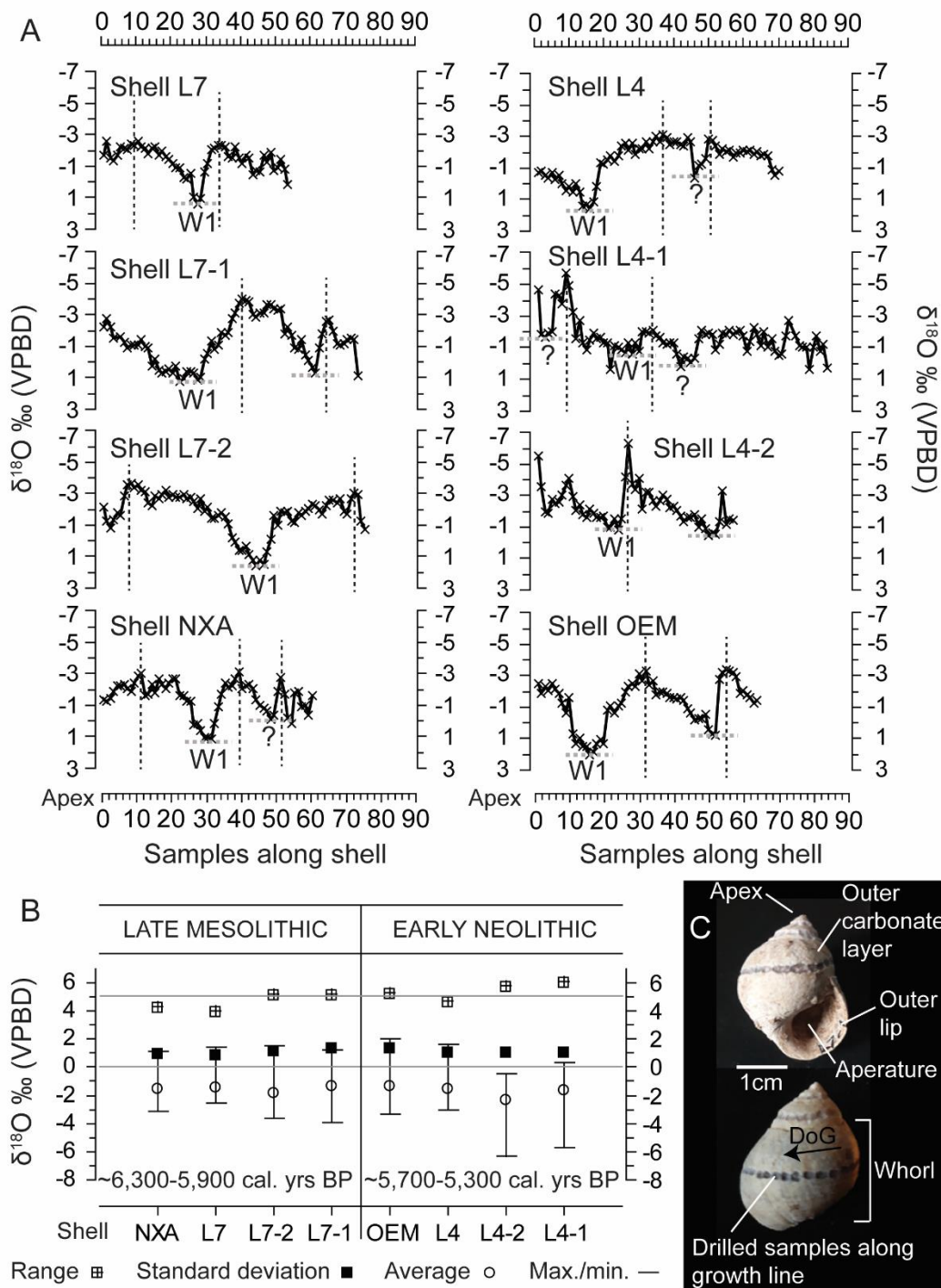
from which the modern-day training set is produced. B. Norsminde Fjord geology, topography and archaeological map (modified from Andersen, 1989). Today only Norsminde Fjord (white area) is sub–merged by marine waters, but at times of higher sea–level (during the late Stone Age) a much larger fjord existed as shown by the extent of the Kysing palaeo–fjord (light grey shading). Kalvø formerly existed as an island in the middle of the Norsminde–Kysing Fjord (Andersen, 1983). TL = location of sampling station in Therkildsen and Lomstein (1994). SC = location of sedimentary core for palaeoenvironmental analyses (P. Rasmussen et al. unpublished data; Lewis, 2011; Lewis et al., 2016; and presented in Fig. 4C, E). Numbered sites refer to key archaeological sites: 1. Norsminde shell midden (or køkkenmødding), 2. Frederiks Odde, 3. Kalvø, 4. Norslund, 5. Flynderhage. C. Photo of a section through the Stone Age Norsminde shell midden, covering the mid to late–Mesolithic Ertebølle period and Early Neolithic Funnel Beaker period (ca. 6,800–5,500 cal. yrs BP), including the much–debated Mesolithic–Neolithic transition (photo courtesy of S.H. Andersen). D. Percentage abundance of key molluscs present in stratigraphic layers of the Stone Age Norsminde shell midden between ca. 6,500–5,200 cal. yrs BP/4,500–3,200 BC (from Bailey and Milner, 2008). Note that *Littorina littorea* is ever-present throughout the study period and the reduction of *Ostrea edulis* in the Late Mesolithic, and concurrent increases in *Cerastoderma edule* and *Mytilus edulis*. The shells used in this study originate from shell midden layer 7 (Late Mesolithic shells L7, L7-1, L7-2 and NXA) and layer 4 (Early Neolithic shells L4, L4-1, L4-2 and OEM); these 2 layers are shaded grey.

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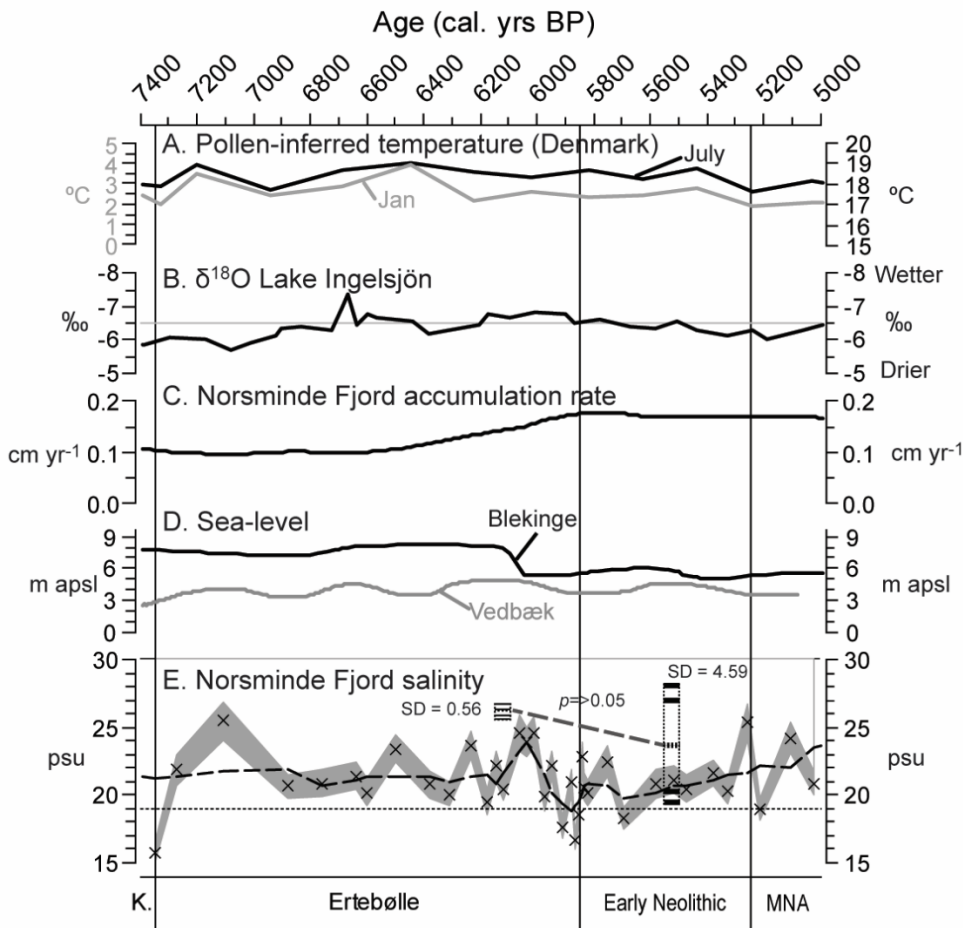


**Figure 2.** Theoretical relationship between  $\delta^{18}\text{O}$  in a *Littorina littorea* shells in response to variation in annual seawater temperature and salinity. Temperature exerts the dominant control on the  $\delta^{18}\text{O}_{\text{shell}}$  over an annual cycle as yearly temperature variation exceeds salinity change (hence why  $\delta^{18}\text{O}_{\text{shell}}$  declines during summer. **\*\***At known temperature (i.e.  $3.7 \pm 1^\circ\text{C}$  the point at which growth ceases in modern *L. littorea* specimens) salinity can be calculated using modern day relationship between  $\delta^{18}\text{O}_{\text{shell}}$  and temperature/salinity (i.e. transfer function produced by Burman and Schmitz (2005), which also includes conversion from  $\delta^{18}\text{O}_{\text{water}}$  (measured in VSMOW) to  $\delta^{18}\text{O}_{\text{shell}}$  (measured in VPDB; see Supplementary Material and correction for fractionation effects).



**Figure 3.** A. Temporal (i.e. interannual) profiles of  $\delta^{18}\text{O}$  data from eight *Littorina littorea* shells from the Norsminde shell midden (left panel: Late Mesolithic shells; right panel: Early Neolithic shells). Isotope data are plotted against sample number (on x-axis), starting from the apex (i.e. youngest part of the shell = 1; see inset C) and following the direction of growth round the spiral towards to the outer lip. Vertical dotted black lines represent inferred summer  $\delta^{18}\text{O}$  minimum (July/August) and horizontal grey lines represent winter maximum (and the likely temperature induced cessation of growth at  $\sim 3.7^\circ\text{C}$ ; Burman and Schmitz,

2005). Dubious winter maxima are indicated with a question mark and not included for quantification of winter salinity. B.  $\delta^{18}\text{O}$  shell-isotope metrics (including range, standard deviation, maximum, minimum and average) and approximate age range of the shells analysed in this study. C Photos of drilled *L. littorea* shells with key features labelled. DoG = direction of growth.



**Figure 4.** Comparison of the  $\delta^{18}\text{O}$  *Littorina littorea* based quantitative salinity estimates from the Norsminde shell midden (presented here) with other Norsminde Fjord palaeo data and regional temperature and sea-level change (7.400-5.000 cal. yrs BP). **A.** Mean January and July pollen-inferred temperatures from Denmark (Brown et al., 2012). **B.** Oxygen isotope analysis ( $\delta^{18}\text{O}$ ) of lacustrine carbonates from Lake Igelsjön showing effective humidity (Seppä et al., 2005). **C.** Norsminde Fjord sediment accumulation rate over the study period reconstructed from a  $^{14}\text{C}$  dated sediment core (Lewis, 2011; Lewis et al., 2016; P. Rasmussen and J. Olsen unpublished data; SC in Fig. 1). **D.** Sea-level change from Vedbæk, Zealand, (Christensen, 2001) and Blekinge, SE Sweden (Berglund et al., 2005). **E.** Diatom-inferred salinity (x) from Lewis et al. (, 2016) with a 0.1 span Lowess smooth (black

571 dotted line) and quantitative winter minimum salinity estimates from the 4 Late Mesolithic  
572 and 4 Early Neolithic periwinkles. The salinity estimates (calculated from the highest winter  
573  $\delta^{18}\text{O}$  in the cycle nearest the apex) are shown with solid black lines (—) and the average with  
574 dotted lines (···). The grey dashed line between the two averages show salinity change  
575 between the Late Mesolithic and early Neolithic: this minor drop in salinity is not significant  
576 ( $p>0.05$ ). SD = standard deviation of the salinity estimates for the Late Mesolithic and Early  
577 Neolithic shells respectively

Period	Shell	No. of samples	No. of winters (summers)	Season of collection	$\delta^{13}\text{C}$ isotope metrics (See Supplementary Material; Fig. S3)	$\delta^{18}\text{O}$ isotope metrics	$\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ (See Fig. S3)	Winter salinity estimate
Late Mesolithic (LM)	NXA	61	2 (2)	Autumn/ Winter	<b>Max:</b> 1.58; <b>Min:</b> -1.40; <b>Range:</b> 2.98; <b>Average:</b> 0.41; <b>SD:</b> 0.58	<b>Max:</b> 1.15; <b>Min:</b> -3.12; <b>Range:</b> 4.27; <b>Average:</b> -1.48; <b>SD:</b> 1.06	$r = 0.30$ ; $r^2 = 0.09$ ; $p = 0.019$ ( $p > 0.01$ )	25.5 psu
LM	L7-1	74	2 (1)	Winter	<b>Max:</b> 1.60; <b>Min:</b> -2.31; <b>Range:</b> 3.92; <b>Average:</b> -0.05; <b>SD:</b> 0.84	<b>Max:</b> 1.24; <b>Min:</b> -3.97; <b>Range:</b> 5.21; <b>Average:</b> -1.29; <b>SD:</b> 1.46	$r = 0.29$ ; $r^2 = 0.08$ ; $p = 0.012$ ( $p > 0.01$ )	25.8 psu
LM	L7-2	76	1 (2)	Spring	<b>Max:</b> 1.38; <b>Min:</b> -1.09; <b>Range:</b> 2.47; <b>Average:</b> 0.60; <b>SD:</b> 0.54	<b>Max:</b> 1.57; <b>Min:</b> -3.61; <b>Range:</b> 5.18; <b>Average:</b> -1.74; <b>SD:</b> 1.28	$r = 0.50$ ; $r^2 = 0.25$ ; $p = 0.0000045$ ( $p < 0.01^*$ )	26.8 psu
LM	L7	54	2? (3?)	Autumn/ Winter	<b>Max:</b> 1.56; <b>Min:</b> -1.31; <b>Range:</b> 2.86; <b>Average:</b> 0.74; <b>SD:</b> 0.54	<b>Max:</b> 1.39; <b>Min:</b> -2.58; <b>Range:</b> 3.97; <b>Average:</b> -1.38; <b>SD:</b> 0.91	$r = 0.22$ ; $r^2 = 0.05$ ; $p = 0.10$ ( $p > 0.01$ )	26.2 psu
Early Neolithic (EN)	L4-1	83	2+ (2+)	Winter	<b>Max:</b> 2.24; <b>Min:</b> -7.78; <b>Range:</b> 10.03; <b>Average:</b> -0.63; <b>SD:</b> 1.65	<b>Max:</b> 0.37; <b>Min:</b> -5.72; <b>Range:</b> 6.08; <b>Average:</b> -1.59; <b>SD:</b> 1.10	$r = 0.83$ ; $r^2 = 0.69$ ; $p = 4.80\text{E-}22$ ( $p < 0.01^*$ )	20.1 psu
EN	L4-2	60	2 (1+)	Spring?	<b>Max:</b> 2.00; <b>Min:</b> -8.99; <b>Range:</b> 10.99; <b>Average:</b> -1.36; <b>SD:</b> 2.11	<b>Max:</b> -0.50; <b>Min:</b> -6.26; <b>Range:</b> 5.77; <b>Average:</b> -2.26; <b>SD:</b> 1.17	$r = 0.88$ ; $r^2 = 0.77$ ; $p = 3.64\text{E-}19$ ( $p < 0.01^*$ )	19.4 psu
EN	L4	70	2? (2?)	Spring	<b>Max:</b> 1.23; <b>Min:</b> -0.89; <b>Range:</b> 2.12; <b>Average:</b> 0.15; <b>SD:</b> 0.53	<b>Max:</b> 1.59; <b>Min:</b> -3.08; <b>Range:</b> 4.67; <b>Average:</b> -1.48; <b>SD:</b> 1.19	$r = 0.42$ ; $r^2 = 0.17$ ; $p = 0.00033$	27.1 psu

							(p<0.01*)	
EN	OEM	64	2 (2)	Spring	<b>Max:</b> 3.15; <b>Min:</b> -0.79; <b>Range:</b> 3.95; <b>Average:</b> 1.45; <b>SD:</b> 0.72	<b>Max:</b> 1.98; <b>Min:</b> -3.37; <b>Range:</b> 5.35; <b>Average:</b> -1.22; <b>SD:</b> 1.45	<b>r</b> = 0.11; <b>r</b> <sup>2</sup> = -0.01; <b>p</b> = 0.40 (p>0.01)	28.2 psu

**Table 1.** Details of the *Littorina littorea* shells analysed in this study.

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