

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

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18
19 **Abstract**

20
21 Norsminde Fjord has received extensive geoarchaeological investigation, hosting one of the
22 classic Stone Age shell midden sites in Denmark, and one of the best examples of the
23 widespread oyster decline at the Mesolithic-Neolithic transition. Here intra-shell $\delta^{18}\text{O}$ (and
24 $\delta^{13}\text{C}$) analyses from the common periwinkle, *Littorina littorea* (L.) are used to infer inter-
25 annual environmental changes at the Mesolithic-Neolithic transition (four from each period).
26 This study utilises a modern $\delta^{18}\text{O}$ *L. littorea*-salinity training set previously developed for the
27 Limfjord, Denmark to quantify winter salinity. $\delta^{18}\text{O}$ values range between +1.6 and +4.0‰ in
28 the Late Mesolithic and -6.3‰ to +2.0 in the Early Neolithic. Using maximum $\delta^{18}\text{O}$ values,
29 winter salinity at the known temperature of growth cessation in *L. littorea* (i.e. $+3.7\pm 1^\circ\text{C}$) for
30 the first annual cycle of each shell ranges between 25.5–26.8 psu (s.d. 0.56) for the Late
31 Mesolithic, with an average salinity of 26.1 psu. Early Neolithic shells range between 19.4 to
32 28.2 psu (s.d. 4.59) with an average salinity of 23.7 psu. No statistically significant change in
33 salinity occurs between the Late Mesolithic and Early Neolithic. This result supports recent
34 diatom/mollusc-based inferences that salinity was not the sole cause of the oyster decline,

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

40

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

43

44 **Introduction**

45 Since their discovery in Denmark in the mid–nineteenth century (Steenstrup et al., 1851),
46 shell middens (termed kitchen middens or “køkkenmøddinger”, e.g. Fig. 1C), formed in
47 connection with human shell–fishing, have proved one of the best archaeological deposits
48 for studying patterns of human subsistence through time. Extensive excavation of many
49 Danish stratified shell middens have produced a detailed temporal record of changes in
50 human practices and resource exploitation (Andersen, 1989; Andersen, 2007; Andersen,
51 2008a; Andersen, 2008b), which along with isotopic analysis of bone collagen from humans
52 and domesticated animals (e.g. Fischer et al., 2007; Tauber, 1981) demonstrates that
53 marine resources have played a central role in the subsistence of cultures and societies in
54 Denmark over large parts of the Middle- and Late-Mesolithic (Andersen, 2007; Fischer et al.,
55 2007). At the Mesolithic–Neolithic transition, there is a shift in diet from a largely marine to a
56 predominately terrestrial sourced diet (Fischer et al., 2007), coincidental with a marked
57 change in molluscan faunal composition in many Stone Age shell middens; i.e. from
58 predominately high salinity and warmer-habitat demanding oysters (*Ostrea edulis* L.) in the
59 Mesolithic midden sections to lower salinity and temperature tolerant cockles (*Cerastoderma*
60 *edule* L.) in the Neolithic sections (Andersen, 2007) and refs therein; Fig. 1D).

61

62 Environmental hypotheses (e.g. temperature/salinity decline, increased sediment
63 accumulation; Rowley-Conwy, 1984; Andersen, 2007; Lewis et al., 2016) have been put
64 forward to explain this widespread faunal shift in Danish shell middens, but to date the role
65 played by intra–annual/seasonal climate change remains unknown and untested. This is due
66 to the almost total lack of seasonal/intra–annual resolution data concerning salinity or
67 temperature change available from coastal Denmark over the Holocene in the direct vicinity
68 of important archaeological settings; in fact only one quantitative record of Holocene
69 temperature change (i.e. pollen–inferred January and July temperature (i.e. Brown et al.,
70 2012) exists for the entire country. One potential method of inferring seasonal/intra–annual

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

74

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

85

86 We also attempt to quantify winter salinity from the $\delta^{18}\text{O}$ isotope data using the previously
87 determined isotope-temperature-salinity relationship (or transfer function) for *L. littorea*
88 (Burman and Schmitz, 2005) established for the Limfjord (inland coastal waters of North
89 Jutland, Denmark). This transfer function was previously applied to two subfossil periwinkles
90 from the Mesolithic Danish Ertebølle *locus classicus* shell midden (Burman and Schmitz,
91 2005), and to Eemian specimens from the English channel and the Swedish Kattegat to
92 compare sea surface temperatures and salinity during the Eemian with present day (Burman
93 and Pässe, 2008). The Danish Ertebølle study, though based on two shell, gave some
94 indication that within the Mesolithic Ertebølle period (ca. 7,400–5,900 cal. yrs BP), both
95 salinity and sea surface temperature were higher in the central Limfjord than present day
96 (Burman and Schmitz, 2005).

97

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

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

110

111 **Methods and materials**

112 Norsminde Fjord (56 °01'018"N, 10 °14'049"E) is a small estuarine system (~6 km long, 250
113 m wide, surface area of ~1.68 km², max. depth 2 m, average depth 0.6 m) situated in a
114 glacial meltwater erosion basin, along the east coast of Jutland about 20 km south of the city
115 of Århus (Fig. 1). It is predominately fed by one major inflow (Odder Å) that drains a
116 catchment area of ~101 km² (Nielsen et al., 1995). Odder Å (fed by several tributaries) is
117 responsible for draining 85 % of the catchment before entering the fjord in the south–west at
118 its innermost section (Fig. 1B). This creates a salinity gradient within the fjord with salinities
119 of ~0 psu freshwater occurring at the mouth of the Rævs Å, gradually increasing up to ~24
120 psu at the Kattegat entrance (with most of the fjord around ~18–20 psu). At present
121 Norsminde Fjord has no regular temperature or salinity stratification regime. Recent annual
122 monitoring of Norsminde Fjord at a sampling site in the northern area of the fjord (Fig. 1B)
123 showed that temperature varies by ~20°C over the year (range 0.5 to 20.0°C in 1989/90)
124 with the maximum occurring in July and the minimum in February (Therkildsen and
125 Lomstein, 1994). In the same year salinity varied between 15.0 to 26.8 psu.

126

127 Norsminde Fjord has been subject to extensive archaeological research (e.g. Andersen,
128 1976; Andersen, 1983; Andersen, 1989; Bailey and Milner, 2008; Andersen and Malmros,
129 1965; Gabrielsen, 1953) and is now one of the most important Stone Age regions in
130 Denmark, with its shorelines hosting a whole series of Mesolithic and Neolithic aged coastal
131 settlements (e.g. kitchen middens; Flynderhage, Norslund, Kalvø; Gabrielsen, 1953;
132 Andersen and Malmros, 1965; Andersen, 1983; Fig 1D) and individual archaeological finds
133 (e.g. flint tools, various axes, bones/antlers). Of interest here, the characteristic kitchen
134 midden ("køkkenmødding"; Fig. 1C) spanning the Mesolithic–Neolithic transition was
135 unearthed during the latest major excavation (1972–1989; Andersen, 1989) and clearly
136 documents the importance of the fjord and marine environment to Stone Age cultures and
137 societies. The *L. littorea* shells analysed in this study were retrieved during this excavation
138 period (Andersen, 1989). The mollusc fauna of this shell midden is diverse, though it is
139 generally dominated by *Ostrea edulis* (European flat oyster), *Cerastoderma edule* (common
140 cockle), *Mytilus edulis* (blue mussel) and *Littorina littorea* (common periwinkle), the
141 abundances of which change markedly throughout the stratigraphic layers (Bailey and
142 Milner, 2008; Fig. 1D). For this study, only well-preserved shells were selected for analysis,
143 i.e. they had a continuous section from the apex to outer margin, with no visible signs of

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

146

147 The common or edible periwinkle (*L. littorea*) is common in the upper shore region down to
148 the shallow sublittoral, particularly favouring rocky shores, but is also abundant on soft
149 substrates (such as in estuaries and fjords) and algal biofilms (Fretter and Graham, 1980;
150 Petersen, 2004). The species is omnivorous, feeding on small invertebrates such as
151 barnacle larvae, as well as macroalgae, microalgae and germlings; Watson and Norton,
152 1985, Wilhelmsen and Reise, 1994). In Denmark *L. littorea* is present today on all coasts
153 (apart from exposed sandy beaches) as far south east as Bornholm in the Baltic Sea
154 (Petersen, 2004; Fretter and Graham, 1980). Since the Littorina transgression in
155 Denmark/Kattegat (ca. 9,600–8,000 cal. yrs BP; Petersen, 1981; Bendixen et al., 2015;
156 Christiansen et al., 1993; Bennike et al., 2000; Bennike et al., 2004), *L. littorea* has been
157 continually present (and often abundant) throughout the Danish waters and the Baltic Sea
158 (commonly as part of the *Mytilus* epifauna; Petersen, 2004; Petersen et al., 2005), extending
159 as far as Estonia up until ca. 3,000 BP (Petersen, 2004). It has remained present north of
160 Bornholm throughout the late Holocene up until present day.

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

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

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

183

184 **Analytical methods**

185 All eight periwinkle shells were cleaned thoroughly by brief submersion in (5%) sodium
186 hypochlorite, followed by rinsing and scrubbing of the outer surface with deionised water to
187 remove any extraneous material. After drying, each shell was successively drilled along the
188 direction of growth (from the apex to the aperture) using a microdrill fitted with a 0.3 mm
189 diameter drill bit. A sampling resolution of ~ 0.3 mm (amounting to 54–83 samples per shell;
190 542 samples in total) was obtained, with care taken to drill only the outer prismatic calcium
191 carbonate layer (Leng and Lewis, 2016). Approximately 50–100 micrograms of carbonate
192 were used for isotope analysis using an Isoprime dual inlet mass spectrometer plus
193 Multiprep device. Samples are loaded into glass vials and sealed with septa. The automated
194 system evacuates vials and delivers anhydrous phosphoric acid to the carbonate at 90°C .
195 The evolved CO_2 is cryogenically cleaned and passed to the mass spectrometer. Isotope
196 values ($\delta^{13}\text{C}$, $\delta^{18}\text{O}$) are reported as per mille (‰) deviations of the isotopic ratios ($R^{13}\text{C}/^{12}\text{C}$,
197 $^{18}\text{O}/^{16}\text{O}$) calculated to the VPDB scale using a within-run laboratory standard calibrated
198 against NBS-19: $\delta = (R_{\text{sample}}/R_{\text{standard}} - 1) * 1000$. Craig correction is also applied to account for
199 ^{17}O . Analytical reproducibility of the standard calcite (KCM) is $< 0.1\text{‰}$ for $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$.

200 **Quantification of salinity and temperature**

201 Using the above mentioned Limfjord training set (Fig. 1A) quantifying relationships between
202 $\delta^{18}\text{O}_{\text{water}}$ and $\delta^{18}\text{O}_{\text{shell}}$ in modern *L. littorea* specimens over a salinity/temperature gradient,
203 $\delta^{18}\text{O}$ from subfossil *L. littorea* shells can be used to quantify winter salinity and summer
204 temperature changes with accuracy over Holocene timescales. This is based on the
205 assumption that the annual range for variation in the shell $\delta^{18}\text{O}$ values is largely determined
206 by the seasonal change in water temperature and that the seasonal salinity changes are
207 small in comparison between April and December across the Limfjord, averaging 3.37‰
208 (range 3 to 3.6‰ ; Burman and Schmitz, 2005). Burman and Schmitz (2005) found a
209 relationship of $+0.25\text{‰}$ $\delta^{18}\text{O}$ change per +1 unit of salinity (psu) and -0.22‰ $\delta^{18}\text{O}$ change
210 per $+1^\circ\text{C}$ in modern specimens, and argue that this relationship has not changed over the
211 Holocene. Based on the linear relationship for $\delta^{18}\text{O}$ of the shell calcite versus temperature,
212 Burman and Schmitz (2005) determined that shell growth ceases around 3.7°C for the
213 modern Limfjord *L. littorea* specimens. As the $\delta^{18}\text{O}$ record reflects largely the combined

214 effects of salinity and temperature, then if one of these parameters is known the other can
215 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
216 assumption that the modern relationship holds true for the sub-fossil shells (method for
217 conversion outlined in Supplementary Material). Here, as the winter temperature is known at
218 the point that growth ceases (i.e. $3.7\pm 1^\circ\text{C}$), salinity at this time can be calculated. Taking the
219 first clear winter maximum $\delta^{18}\text{O}_{\text{shell}}$ in the sub-fossil shells as the point at which growth
220 ceases (see Fig. 3A), we used the modern relationship for *L. littorea* determined by Burman
221 and Schmitz (2005) to convert each shell $\delta^{18}\text{O}$ (VPDB) winter value into $\delta^{18}\text{O}_{\text{water}}$ (VSMOW)
222 (Fig. S1) at the point of growth cessation (which would be at $3.7\pm 1^\circ\text{C}$). We then converted
223 each $\delta^{18}\text{O}$ (VSMOW) value into a salinity estimate using the modern relationship between
224 $\delta^{18}\text{O}_{\text{water}}$ (VSMOW) and salinity (i.e. 0.25‰ per salinity unit) (Fig. S2).

225

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

244

245 **Results**

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

250 summer/autumn for the Mesolithic shells and the autumn/winter for the Neolithic shells
251 (Table 1). $\delta^{13}\text{C}$ values range between -2.3‰ to $+1.6\text{‰}$ (VPBD) in the Late Mesolithic and $-$
252 9.0‰ to $+3.2\text{‰}$ in the early Neolithic shells, the predominantly high values suggesting that
253 shell carbon is sourced from marine dissolved inorganic carbon. Due to the focus of this
254 study on changing $\delta^{18}\text{O}$ derived salinity/temperature signals from *L. littorea* shells, no further
255 consideration of the $\delta^{13}\text{C}$ data is provided here (see Supplementary Material for more
256 details).

257 $\delta^{18}\text{O}$ and winter salinity estimates

258 $\delta^{18}\text{O}$ values range between -6.3‰ and $+2\text{‰}$ (VPBD), with generally a similar overall
259 distribution of values in both the Mesolithic and Neolithic shells (-4‰ to $+2\text{‰}$), but with the
260 very lowest values (below -4‰) occurring only in the Early Neolithic shell samples. All shells
261 show at least one or more complete annual cycle (see Fig. 3A, Table 1), though in some
262 cases annual cycles are difficult to determine confidently (labelled in Fig. 3A). The highest
263 $\delta^{18}\text{O}$ values (lowest temperatures) tend to be followed by a sharp decrease in $\delta^{18}\text{O}$ values,
264 marking the start of the winter growth cessation, then growth resuming in the warmer months
265 in these specimens.

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

273 Discussion

274 $\delta^{18}\text{O}$

275 The cyclical variability displayed by the sub-fossil *L. littorea* shells from Norsminde primarily
276 reflects the annual changes in water temperature and salinity of the fjord and growth
277 patterns of the individual shells (cf. Andreasson et al., 1999; Burman and Schmitz, 2005; Fig.
278 3A). Within a shell, winters are represented by highest $\delta^{18}\text{O}$ values and therefore changes
279 are largely driven by temperature due to the inverse temperature- $\delta^{18}\text{O}_{\text{shell}}$ relationship in
280 Danish coastal waters (-0.22‰ per $^{\circ}\text{C}$; Burman and Schmitz, 2005). This results in $\delta^{18}\text{O}$
281 minima during the summer, due to the associated large temperature increase, which
282 outweighs annual salinity changes (e.g. $20\text{ }^{\circ}\text{C}$ annual temperature fluctuation from January

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

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

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

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

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

344 The most likely explanation for these varying trends is fluctuation in annual climatic
345 conditions. The Early Neolithic was a time of changing climate, with cooling temperatures
346 (Seppä et al., 2009; Antonsson and Seppä, 2007; Brown et al., 2012) and increased
347 precipitation or surface wetness (Seppä et al., 2005) as the Holocene thermal maximum
348 began to wane. Layers in shell middens cover relatively coarse time periods (perhaps up to
349 200 years in Norsminde; Andersen, 1989; Bailey and Milner, 2008), so the lifespans of
350 individual molluscs (here, 2-3 years) within a layer are unlikely to overlap. Variable
351 summer/winters (e.g. warmer or colder and wetter or drier years) were likely recorded in
352 some specimens and not others, which highlights a danger of using single shell analyses
353 which may be skewed by atypical conditions. Furthermore, in a transitional climate period,
354 fluctuating temperature and rainfall (leading to salinity variations) between extremes might

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

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

380 **Comparison with regional parameters**

381 During the late Stone Age (ca. 8,000-3,700 cal. yrs BP), both sea-level (e.g. Berglund et al.,
382 2005; Christensen, 2001; Fig. 4D) and salinity (e.g. Petersen, 2004; Lewis et al., 2016;
383 Weckström et al., 2017; Fig 4E) were higher than present day in Denmark and western
384 areas of the Baltic Sea. The winter (minimum) salinities presented here (from the *L. littorea*
385 shells) also imply higher salinity than present day within Norsminde Fjord, though the $\delta^{18}\text{O}$
386 based estimates generally suggest higher salinity compared to the diatom record (Lewis et
387 al., 2016) (Fig. 4E). This offset between the DI-salinity and *L. littorea* winter minimum salinity
388 is likely explained by DI-salinity inferences not being season-specific, averaged over
389 several years and incorporating diatoms from a spatially wider area of the fjord (i.e. lower

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

394 Burman and Schmitz (2005) originally used the methodology employed here to reconstruct
395 changes in two sub-fossil shells from the Ertebølle shell midden (with these two shells dated
396 to the Late Mesolithic). The data presented here suggest lower salinity in Norsminde Fjord
397 (26.1 psu) than at Ertebølle (i.e. 31 psu) during the Late Mesolithic. This is expected due to
398 where Ertebølle is in the Limfjord, and in closer proximity to the high salinity water of the
399 North Sea (Fig. 1A). Modern-day salinity at Ertebølle is ~26 psu compared to an average of
400 ~18-20 psu at Norsminde Fjord, though it is important to highlight that a different land–sea
401 configuration (e.g. Kysing Fjord; Fig. 1B) existed in both the Limfjord and Norsminde Fjord at
402 that time due to isostatic uplift and higher sea level (Andersen, 1989; Christensen, 2001).
403 During the mid-Holocene high sea-level stand, an opening to the Skagerrak and North Sea
404 in the north has been inferred (Andersen, 1995; Andersen, 2007; Petersen et al., 2005),
405 closer to the Ertebølle site than the modern-day entrance in the west (Fig. 1A). Also, during
406 the mid–Holocene (ca. 7000–4000 cal. yrs BP), Norsminde Fjord was significantly larger
407 incorporating the Kysing palaeo-fjord (~10 km long, 2.7–3 km wide and ~500 m at the
408 Kattegat mouth, Andersen, 1989; Fig. 1B). A wider connection between Norsminde Fjord
409 and the Kattegat under higher sea-levels would have improved exchange of marine water
410 (and sediment) resulting in higher fjord salinity and lower sedimentation rates (Lewis et al.,
411 2016). This created optimal conditions for a variety of marine molluscs such as oysters, an
412 important component of Stone Age shell middens, particularly in the Late Mesolithic layers
413 (Andersen, 2007; Bailey and Milner, 2008). However, this connection started to narrow
414 sometime in the late Mesolithic (though exact date remains unknown), due to longshore
415 transport of sediment and subsequent development of beach ridges (Andersen, 1989).

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

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

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

429 For the oyster decline, the change in seasonality might be of importance, with salinity (and
430 $\delta^{18}\text{O}$ values) appearing to exhibit greater variability in the Early Neolithic than in the Late
431 Mesolithic (both a higher range and standard deviation is observed between these layers;
432 Fig. 3B, 4E). Greater seasonal salinity (and temperature) fluctuations might also have
433 contributed to the reduction in oysters, by adding further stress upon accessible oyster
434 populations in addition to increasing sedimentation rates in fjords (Fig. 4C) and a general
435 temperature decline (Fig. 4A; outlined by Lewis et al., 2016). The isotope data suggest that
436 in some years winter salinity fell near or below the minimum level required to support
437 breeding oyster populations (i.e. ~ 23 psu; Jensen and Spärck, 1934; Spärck, 1924; Yonge,
438 1960). Whilst oysters require water temperatures above $\sim 15^\circ\text{C}$ to breed (Jensen and
439 Spärck, 1934; Spärck, 1924; Yonge, 1960), in some years low salinity could have extended
440 into the warmer months (e.g. low salinity winters followed by wet springs/summer) and
441 narrowed the breeding season (in spring/summer). In these years human exploitation with
442 reduced replenishment might have caused decline/stress of oyster populations. In addition,
443 Norsminde Fjord was near the southern limits of oyster habitation in the inner Danish waters
444 (Jensen and Spärck, 1934; Petersen, 2004) even in the late Stone Age (7,400-3,700 cal. yrs
445 BP), when salinity and sea-levels were above present day (Berghlund et al., 2005;
446 Christensen, 2001), perhaps with the result that small fluctuations in salinity might have had
447 severe impacts. This, coupled with changing sedimentation patterns within the fjord (Lewis et
448 al., 2016; Fig. 4C) and climate change, including both temperature decline (Snowball et al.,
449 2004; Antonsson and Seppä, 2007; Brown et al., 2012; Seppä et al., 2009; Fig 4A) and a
450 potential shift towards wetter conditions (Seppä et al., 2005; Fig. 4B), might have put
451 cumulative stress on oyster populations in Danish coastal waters. However, as layer 4 is
452 several hundred years later than the oyster decline, further assessment of seasonal change
453 is needed from earlier layers (i.e. layers 5 and 6; Fig. 1D) of the Norsminde shell midden.

454 **Future implications**

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

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

470 Lastly, we acknowledge that finer resolution drilling and implementation of
471 sclerochronological techniques could have provided further information concerning the
472 timing, duration and rate of growth in the analysed shells (Schöne, 2008; Wingard and
473 Surge, 2017; Schöne and Gillikin, 2013) and should be incorporated in future studies.
474 However, for the sake of this study, we argue that these shells were drilled at sufficient
475 resolution (0.3mm) to capture intra-annual variation as demonstrated by the similar intra-
476 shell isotope profiles to previous studies (e.g. Andreasson et al., 1999; Burman and Schmitz,
477 2005) and further sclerochronological analyses are unlikely to have altered final
478 salinity/seasonality inferences and interpretation.

479 **Conclusions**

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

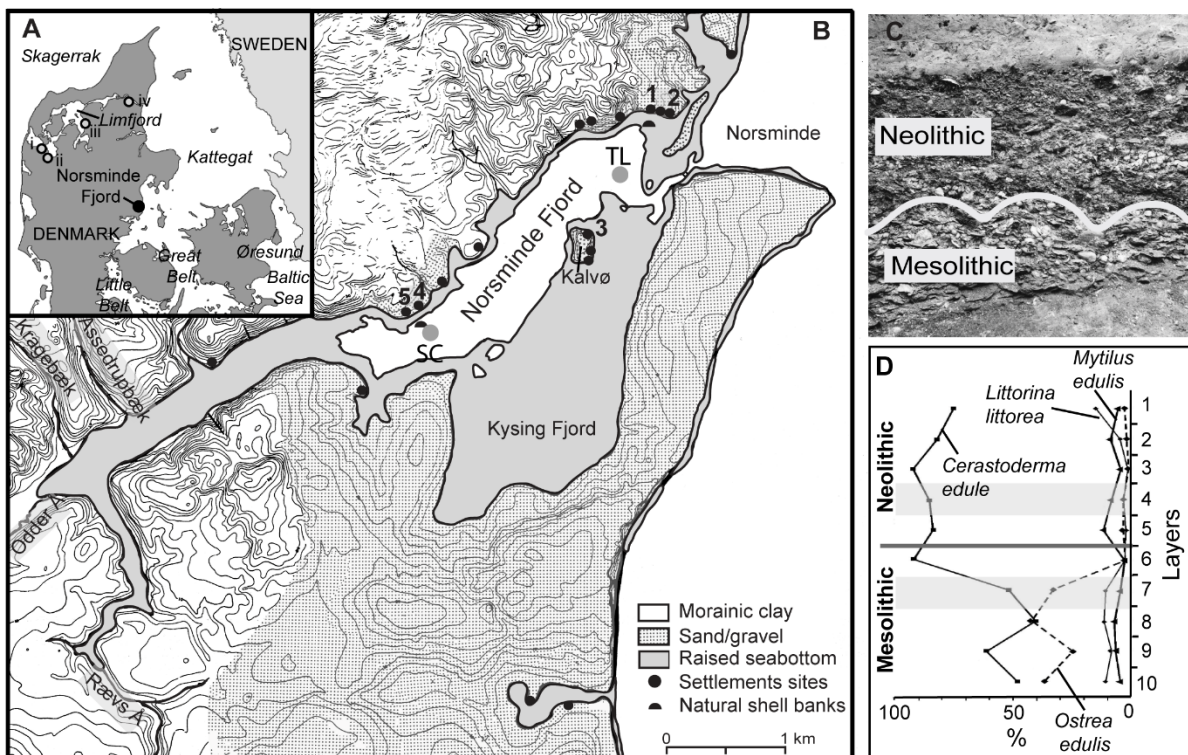
495 not easily disappear from the record entirely under changing environmental conditions. As it
 496 has a more continuous and abundant presence than other common molluscs, it is very
 497 suited for such isotopic or sclerochronological work. However, in order to enhance
 498 geoarchaeological debate,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

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 506 midden. Carol Arrowsmith and Hilary Sloane (from the British Geological Survey) are
 507 thanked for sample preparation and isotope analyses. This project was funded by NERC
 508 facility grant IP-1550-0515 awarded to D.B.Ryves and J.P.Lewis.

509

510 Figures:

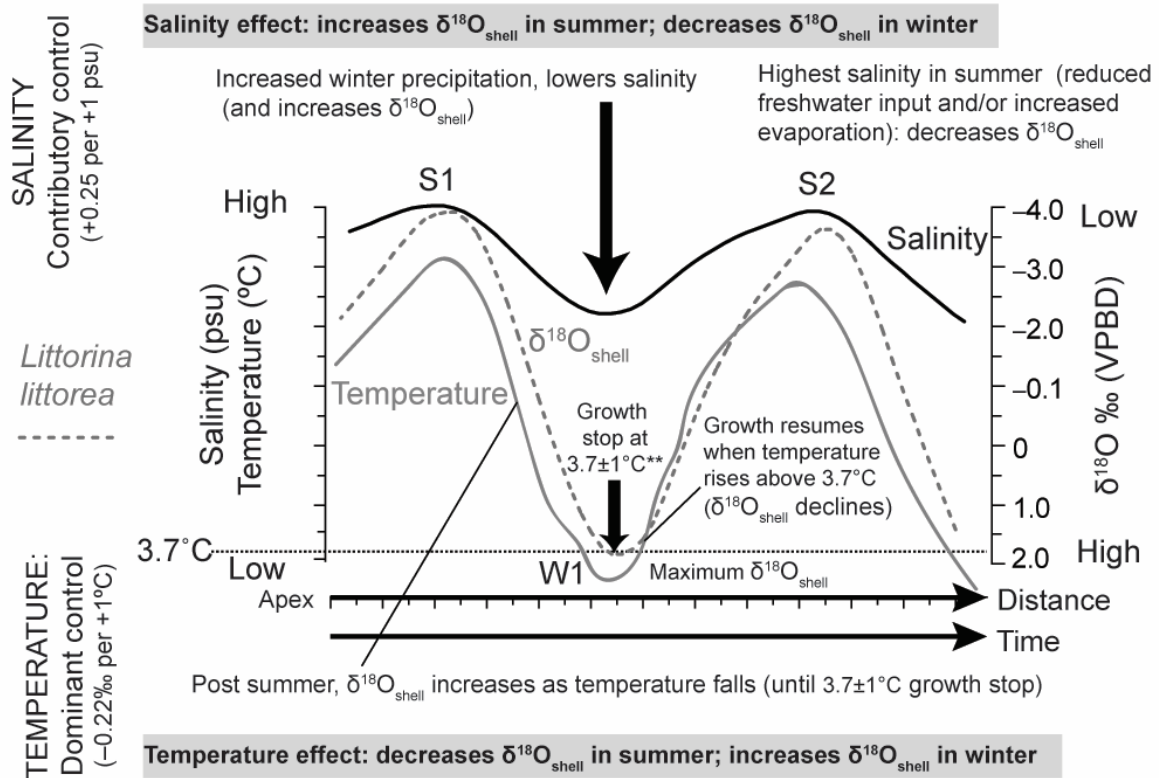


511

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

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

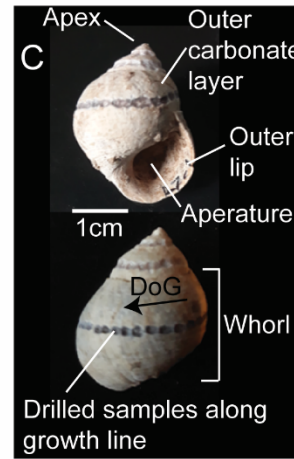
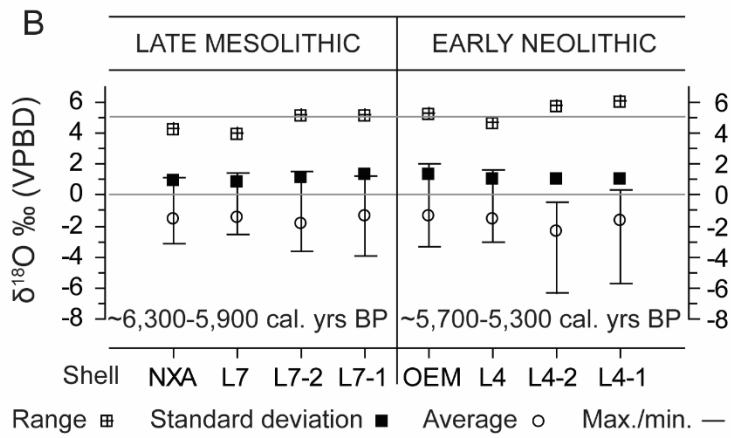
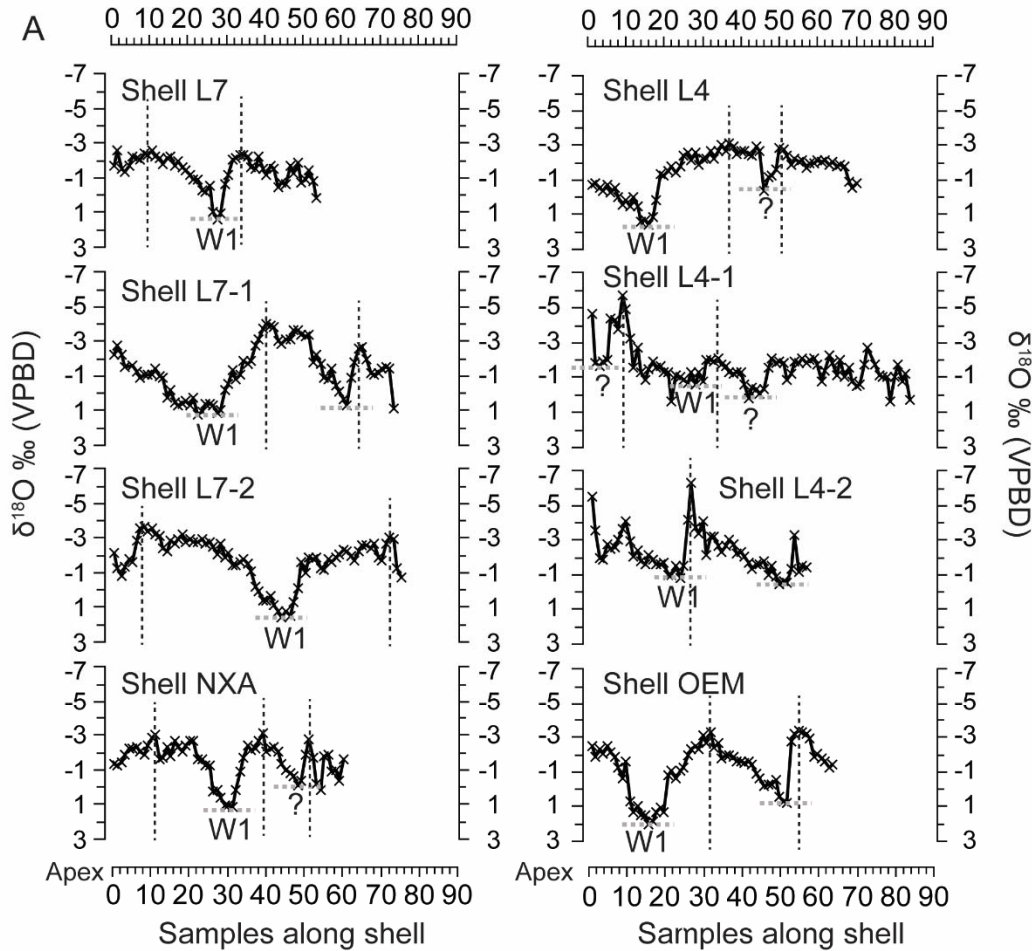
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535

536 **Figure 2.** Theoretical relationship between $\delta^{18}\text{O}$ in a *Littorina littorea* shells in response to
 537 variation in annual seawater temperature and salinity. Temperature exerts the dominant
 538 control on the $\delta^{18}\text{O}_{\text{shell}}$ over an annual cycle as yearly temperature variation exceeds salinity
 539 change (hence why $\delta^{18}\text{O}_{\text{shell}}$ declines during summer. ******At known temperature (i.e. $3.7 \pm 1^\circ\text{C}$
 540 the point at which growth ceases in modern *L. littorea* specimens) salinity can be calculated
 541 using modern day relationship between $\delta^{18}\text{O}_{\text{shell}}$ and temperature/salinity (i.e. transfer
 542 function produced by Burman and Schmitz (2005), which also includes conversion from
 543 $\delta^{18}\text{O}_{\text{water}}$ (measured in VSMOW) to $\delta^{18}\text{O}_{\text{shell}}$ (measured in VPDB; see Supplementary
 544 Material and correction for fractionation effects).

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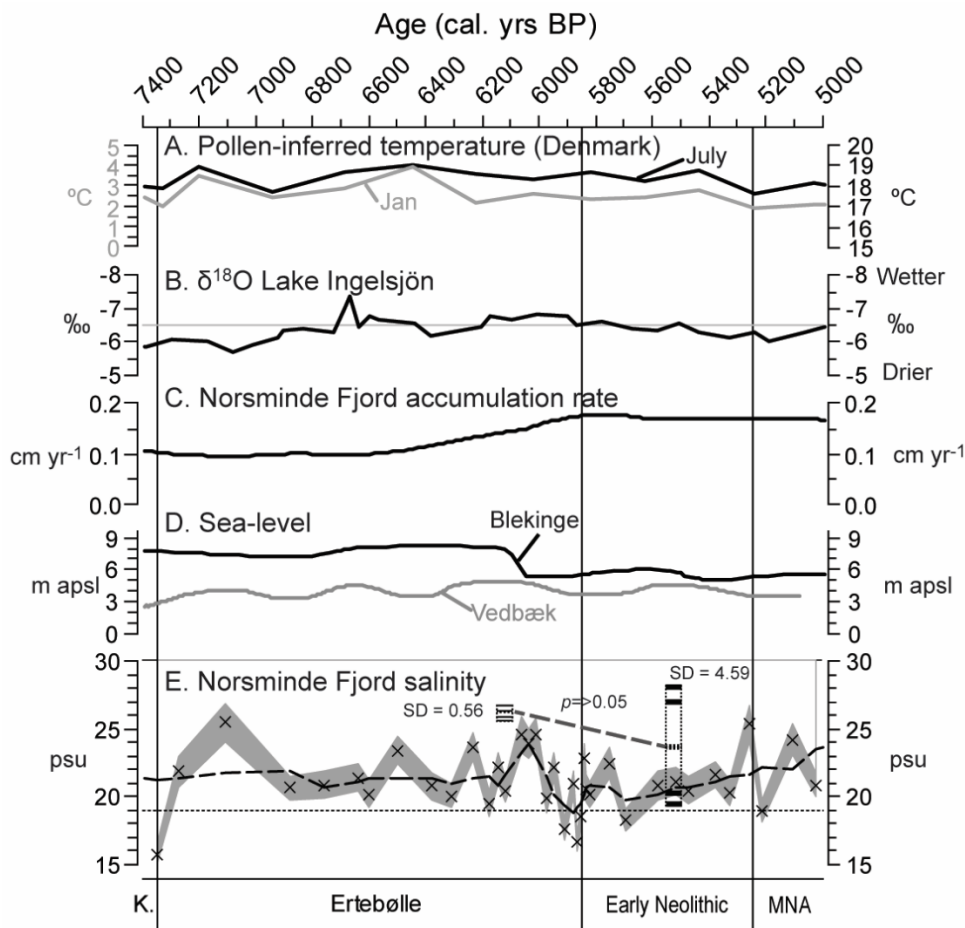


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547 **Figure 3.** A. Temporal (i.e. interannual) profiles of $\delta^{18}\text{O}$ data from eight *Littorina littorea*
 548 shells from the Norsminde shell midden (left panel: Late Mesolithic shells; right panel: Early
 549 Neolithic shells). Isotope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

554 2005). Dubious winter maxima are indicated with a question mark and not included for
 555 quantification of winter salinity. B. $\delta^{18}\text{O}$ shell–isotope metrics (including range, standard
 556 deviation, maximum, minimum and average) and approximate age range of the shells
 557 analysed in this study. C Photos of drilled *L. littorea* shells with key features labelled. DoG =
 558 direction of growth.

559



560

561 **Figure 4.** Comparison of the $\delta^{18}\text{O}$ *Littorina littorea* based quantitative salinity estimates from
 562 the Norsminde shell midden (presented here) with other Norsminde Fjord palaeo data and
 563 regional temperature and sea-level change (7.400-5.000 cal. yrs BP). **A.** Mean January and
 564 July pollen-inferred temperatures from Denmark (Brown et al., 2012). **B.** Oxygen isotope
 565 analysis ($\delta^{18}\text{O}$) of lacustrine carbonates from Lake Ingelsjön showing effective humidity
 566 (Seppä et al., 2005). **C.** Norsminde Fjord sediment accumulation rate over the study period
 567 reconstructed from a ^{14}C dated sediment core (Lewis, 2011; Lewis et al., 2016; P.
 568 Rasmussen and J. Olsen unpublished data; SC in Fig. 1). **D.** Sea-level change from
 569 Vedbæk, Zealand, (Christensen, 2001) and Blekinge, SE Sweden (Berglund et al., 2005). **E.**
 570 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	Max: 1.58; Min: -1.40; Range: 2.98; Average: 0.41; SD: 0.58	Max: 1.15; Min: -3.12; Range: 4.27; Average: -1.48; SD: 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	Max: 1.60; Min: -2.31; Range: 3.92; Average: -0.05; SD: 0.84	Max: 1.24; Min: -3.97; Range: 5.21; Average: -1.29; SD: 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	Max: 1.38; Min: -1.09; Range: 2.47; Average: 0.60; SD: 0.54	Max: 1.57; Min: -3.61; Range: 5.18; Average: -1.74; SD: 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	Max: 1.56; Min: -1.31; Range: 2.86; Average: 0.74; SD: 0.54	Max: 1.39; Min: -2.58; Range: 3.97; Average: -1.38; SD: 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	Max: 2.24; Min: -7.78; Range: 10.03; Average: -0.63; SD: 1.65	Max: 0.37; Min: -5.72; Range: 6.08; Average: -1.59; SD: 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?	Max: 2.00; Min: -8.99; Range: 10.99; Average: -1.36; SD: 2.11	Max: -0.50; Min: -6.26; Range: 5.77; Average: -2.26; SD: 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	Max: 1.23; Min: -0.89; Range: 2.12; Average: 0.15; SD: 0.53	Max: 1.59; Min: -3.08; Range: 4.67; Average: -1.48; SD: 1.19	$r = 0.42$; $r^2 = 0.17$; $p =$	27.1 psu

							0.00033 (p<0.01*)	
EN	OEM	64	2 (2)	Spring	Max: 3.15; Min: -0.79; Range: 3.95; Average: 1.45; SD: 0.72	Max: 1.98; Min: -3.37; Range: 5.35; Average: -1.22; SD: 1.45	r = 0.11; r ² = -0.01; p = 0.40 (p >0.01)	28.2 psu

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

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