Quantitative salinity reconstruction of the Baltic Sea during the Mid-Holocene

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Keywords:	salinity , Baltic Sea, Holocene, multi-proxy

Quantitative salinity reconstructions of the Baltic Sea during the Mid-Holocene

We have reconstructed the past coastal environment of the Baltic Sea from c. 7300 to 3500 cal. a BP, with the focus on sea surface salinity (SSS). To quantitatively determine the SSS, two methods were employed, including measurements of $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in mollusk shells (SSS_{Sr}) and process length variations from dinoflagellate cyst *Operculodinium centrocarpum* (SSS_{pl}). The SSS_{Sr} was c. 6-7 between 6800 and 6400 cal. a BP, similar to modern conditions. Between 6000 and 3900 cal. a BP, SSS_{Sr} was consistently higher, ranging between c. 9-13. Microfossils that are sensitive to salinity variations, such as *Radiosperma corbiform* and *Spiniferites* spp., support the SSS_{Sr} estimate. In comparison with the SSS_{Sr} , the SSS_{pl} values were consistently higher, with an average of c. 14. We suggest that the use of a linear regression model, between the process length and salinity variations, caused the salinity overestimation. A multi-proxy approach, including microfossil, organic carbon content, C/N ratios, and grain size analysis allowed for a division of the study period into three zones, representing different environment settings caused by eustatic sea level, isostatic land movement and climate changes.

Keywords: salinity, Baltic Sea, Holocene, multi-proxy

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Although the Baltic Sea is one of the largest brackish systems in the world today, its salinity varied considerably during the Holocene epoch. During the Yoldia Sea stage between 11700 and 10700 cal. a BP, saline water entered the Baltic Basin through a narrow strait located in the central Swedish lowland. This resulted in a short brackish period between 11300 and 11100 cal. a BP (Andrn *et al.* 2000; Andrn *et al.* 2011). Continuous land uplift prevented further intrusion of sea water, resulting in decreasing salinity and a freshwater stage, the Ancylus Lake phase, which prevailed between 10700 to 10200 cal. a BP. With the eustatic sea level continuously rising, sea water again entered the Baltic Basin, this time through the Danish straits. As a consequence, brackish conditions developed in the Baltic Sea, and can be divided into three stages, the Early Littorina Sea (10200-8500 cal. a BP), the Littorina Sea (8500-3000 cal. a BP) and the Post (Late) Littorina Sea (3000 cal. a BP to present) (Conley *et al.* 2009).

The Littorina Sea phase is characterized by large salinity variations, which strongly affect biogeochemical conditions in the Baltic Sea. Conley *et al.* (2009) demonstrate that intrusion of more saline water during the Littorina Sea potentially caused greater stratification in the water column and restricted the ventilation of the bottom water. Together with the warm and dry climate conditions during the Holocene thermal maximum (HTM), laminated sediment can be observed in sediment cores from the Gotland Basin between c. 8000 and 4000 cal. a BP, indicating an oxygen-depleted bottom water environment (Zilln *et al.* 2008; Renssen *et al.* 2009). However, salinity reconstructions of the Littorina Sea from various studies exhibit considerable differences in terms of pattern and absolute value (Gustafsson & Westman 2002; Emeis *et al.* 2003; Widerlund & Andersson 2011). As a result, the role of salinity variations on the formation of hypoxic bottom conditions in the Baltic is still debatable. Besides, salinity reconstructions in the Baltic Sea have so far mainly been based on qualitative observations with considerable uncertainty in the chronology.

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Two methods of quantitative salinity calibrations in the Baltic Sea have been developed in recent years. Mertens *et al.* (2011) analysed surface sediment samples along a Baltic Sea-Kattegat-Skagerrak gradient and measured the process length of dinoflagellate cysts *Operculodinium centrocarpum* (Wall and Dale 1968). The authors determined a significant linear relationship between sea surface salinity (SSS) and the process length of *O. centrocarpum* ($R^2 = 0.8$, $n = 71$). Salinity reconstruction based on the linear function was applied from a late Holocene sediment core at Limfjord which resulted in satisfactory result (Mertens *et al.* 2011). A similar approach is applied by Sildever *et al.* (2015) and a linear relationship is also observed between process length of *O. centrocarpum* and sea surface salinity ($R^2 = 0.86$, $n = 7$), from surface samples collected in a Baltic Sea-Kattegat-Skagerrak transect. The second salinity reconstruction method is based on the strontium isotope ratio ($^{87}\text{Sr}/^{86}\text{Sr}$) measured in carbonate shell remains. The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio found in biogenic carbonates corresponds to the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of the surrounding water, which is also a function of the ambient salinity (Depaolo & Ingram 1985). This method has the potential to reconstruct the salinity of the water where the mollusks lived (Andersson *et al.* 1992; Widerlund & Andersson 2006). In the brackish Baltic Sea the water is a mixture between sea water and river water and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the Baltic Sea water are significantly higher than in sea water. Because of this, the Sr concentration and isotopic composition in Baltic Sea water can be characterized as a general two-component conservative mixing system between sea water and river water (Andersson *et al.* 1992). If the end-member values of the river and sea water can be determined and the $^{87}\text{Sr}/^{86}\text{Sr}$ composition in carbonate shells measured, salinity can be reconstructed (Widerlund & Andersson 2006).

In this study, we reconstruct surface salinity within the Littorina Sea phase using both methods: process length variations of *O. centrocarpum* and $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in shell remains. As far as we know, this is the first down-core salinity reconstruction based on these two

74 methods in the Baltic Sea. The results from the two methods are subsequently compared with
75 each other and then with previous salinity reconstructions. Furthermore, we use a range of
76 different proxy variables such as organic carbon content (C_{org}), C/N ratios, microfossil
77 assemblages and grain size analyses to further reconstruct environment changes over our
78 study period.

79

80 **Modern Baltic salinity regime**

81 As a result of the Baltic Sea-Kattegat-Skagerrak estuarine system, Baltic Sea salinity is
82 governed by the relative amount of freshwater input (runoff and precipitation) and the sea
83 water intruding through the Danish Straits. Limited water exchange with the open ocean and
84 considerable freshwater input ($16100 \text{ m}^3 \text{ s}^{-1}$) led to an average salinity of about 7.4 in the
85 Baltic Sea over the 20th century (Janssen *et al.* 1999). Freshwater input generally causes a
86 higher water level in the Baltic Sea than the in the Kattegat, resulting in an outflow of less
87 saline water from the Baltic Sea. The inflow of more saline water maintains the salt balance in
88 the Baltic Sea. Lehmann *et al.* (2002) demonstrate that the volume exchange through the
89 Danish Straits is correlated with the sea level pressure difference between Oslo in Norway
90 and Szczecin in Poland.

91 Saline inflows intruding through the Danish Straits drive the vertical circulation in the
92 Baltic Sea. The more saline water is first spilled over the shallow sills and then flows into the
93 Arkona Basin and Bornholm Basin (Fig. 1). During events of Major Baltic Inflows, which
94 occur mainly in the winter season, deep basins further away from the Danish Straits such as
95 the Gotland Basin can be replenished (Matthäus & Franck 1992). These events are more
96 likely to occur after strong easterly winds lasting several weeks followed by strong westerly
97 winds of similar duration (Lass & Matthäus 1996).

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99 **Hydrographic setting of the study site**

100 Our coring site KYR in the south-western Baltic Sea is located close (c. 3 km) to the

101 environmental monitoring station KAARV4 (Fig.1). The surface salinity from KAARV4 was

102 compared with the environmental monitoring stations BY5 and BY10, which are located in

103 the open Baltic (Fig. 1). The hydrographic time series from these stations were obtained from

104 the Swedish Oceanographic Data Center (SHARK) at the Swedish Meteorological and

105 Hydrological Institute (SMHI). Average surface salinity, calculated using data covering the

106 near-surface layer (down to 20 m depth), was 7.1, 7.4 and 7.1 at station KAARV4, BY5 and

107 BY10, respectively, over 1998-2011 (Fig. 1). Spearman correlations of the salinity data

108 between KAARV4 and BY5 ($r = 0.82$, $p < 0.01$), and between KAARV4 and BY10 ($r = 0.56$,

109 $p < 0.01$) indicate that the salinity of station KAARV4 is determined by the open Baltic Sea. T-

110 test of KAARV4 and BY10 salinity data resulted in an equal mean salinity between stations

111 KAARV4 and BY10, i.e. no significant difference in average surface water salinity between

112 KAARV4 and BY10. We are therefore confident that the salinity at our study site is

113 representative of sea surface water salinity over the southern Baltic Sea.

114

115 **Material and methods**

116 **Sediment core sampling**

117 Sediment cores were collected during a research cruise with *R/V Ocean Surveyor* in August,

118 2011. The coring site KYR (56 °7'53"N, 15 °32'55"E) is located 2 km SE of the city of

119 Karlskrona, SE Sweden, with 10 m water depth (Fig. 1). Four short cores, KYR10A, B, F and

120 J of c. 40 cm long each, were collected using a Gemini corer. One piston core (KYR10L) of

121 520 cm long was also retrieved. The piston core was split lengthwise into two equal halves,
122 described and photographed on board. The lithology was characterized by homogeneous
123 clayed gyttja of brown color. Homogeneity of the sediment core was also confirmed by X-ray
124 image. The short cores and one half of the piston core were subsampled every one centimeter
125 at the Dept. of Geology, Lund University. The samples were all freeze-dried and archived
126 thereafter. The other half of the piston core is stored in a cold room (+4°C), at Lund
127 University, Sweden.

128

129 **Chronology**

130 AMS ^{14}C dating was applied to establish the chronology, with twelve shell remains dated
131 from the piston core KYR10L and one from a short core KYR10B at Lund University
132 Radiocarbon Dating Laboratory (Table 1). To determine the reservoir age in our study, six
133 samples of *Macoma balthica* shell were analysed for $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ measurements at the
134 University of Amsterdam (Table 2). Estimated reservoir ages were subsequently determined,
135 using the relationship between $\delta^{18}\text{O}$ in *Macoma balthica* shells and reservoir age in the Baltic
136 region (Lougheed *et al.* 2013). After the reservoir age was subtracted from the dating results,
137 an age model of the sediment sequence was established with Oxcal 4.2 (Ramsey 2009).

138

139 **Microfossil analysis**

140 Organic-walled microfossils were processed following the guidelines from Rochon *et al.*
141 (1999) and Pospelova *et al.* (2010). Prior to chemical treatments, two tablets of calibrated
142 *Lycopodium clavatum* spores were added into a known amount of sediment. Samples were
143 first treated with 10% hydrochloric acid (HCl) and then 48% hydrofluoric acid (HF). The

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144 samples were neutralized through rinsing with distilled water. To concentrate dinoflagellate
145 cysts and the other microfossils, the residuals after treatment were rinsed through a 125 µm
146 sieve and a 20 µm sieve. The residues between 20 and 125 µm were then sonicated gently for
147 up to 60 seconds before sieving with 20 µm again. Organic-walled microfossils were
148 identified with 60x based on Rochon et al. (1999) and Price and Pospelova (2011). The
149 concentrations of the microfossils were calculated and expressed as amount of individuals per
150 gram (Benninghoff 1962).

151 For foraminiferal analysis, 1 gram of dry sediment was wet sieved with 100 µm and 45 µm
152 sieves. To enhance the disintegration of sediment aggregates, sodium diphosphate (Na₂P₂O₇)
153 was used when sieving. The residues > 45 µm were analysed using a binocular stereoscopic
154 zoom microscope (Nikon SMZ 1500).

155
156 **Salinity reconstruction based on *Operculodinium centrocarpum***

157 Digital images of *O. centrocarpum* were taken with an Olympus SC30 camera mounted on an
158 Olympus BX53 microscope. Process lengths were measured with cellSens® software
159 designed for the camera, following the method described by Mertens *et al.* (2011). The three
160 longest processes were measured for each *O.centrocarpum* cysts. Thirty cysts were measured
161 at each sample depth except for samples with few occurring cysts (average = 29, n = 22). The
162 average process length and its standard deviation from each sample were calculated. A sea
163 surface salinity reconstruction based on the process length of *O.centrocarpum* (SSS_{pl}) was
164 calculated using the function by Mertens *et al.* (2011), which is expressed as
165 $SSS_{pl}=3.16 \times \text{average process length}-0.84$.

$^{87}\text{Sr}/^{86}\text{Sr}$ measurements and salinity reconstruction

Pretreatment of shell remains for the $^{87}\text{Sr}/^{86}\text{Sr}$ analyses followed Widerlund and Andersson (2006). The shell samples were leached with 1M acetic acid followed by 2.5M HCl acid. Between 10 and 40% of the shells were lost after the acid treatment. Scanning electron microscope images revealed that the shells were well preserved, without signs of post-depositional mineralization (Fig. S1). The remaining sample was dissolved in 2.5 M HCl and Sr was separated following Widerlund and Andersson (2006). Approximately 200-400 ng of Sr from the purified sample was loaded and mixed with tantalum activator on a single rhenium filament. The $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic composition was determined at the Swedish Museum of Natural History in Stockholm using a Thermal ionization mass spectrometer (TIMS) and a Thermo Scientific TRITON. The external precision of $^{87}\text{Sr}/^{86}\text{Sr}$ measurements for the NBS 987 standard (0.710217 ± 16) was ± 22 ppm (2 SD, $n = 12$).

The two-component mixing model reported by Widerlund and Andersson (2006) was used to calculate the paleosalinity based on the $^{87}\text{Sr}/^{86}\text{Sr}$ measurements in the fossil shells. The sea water end-member for Sr concentration, $C_{\text{Sr}}^{\text{SW}}$, and Sr isotopic composition values used in the calculations are from Widerlund and Andersson (2011): $C_{\text{Sr}}^{\text{SW}} = 7.73 \text{ mg L}^{-1}$ and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio = 0.709157. For the river water end-member, which represents a single-river effective end member for the Baltic Proper, a concentration of Sr in river water ($C_{\text{Sr}}^{\text{RW}}$) = 0.085 mg L^{-1} and the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio = 0.713607 were used. The sea surface salinity reconstructed with $^{87}\text{Sr}/^{86}\text{Sr}$ measurements is expressed as SSS_{Sr} (Table 3).

Organic carbon and C/N ratio

A total of 38 samples with c.10 cm interval were subsampled for organic carbon (C_{org}) and carbon nitrogen ratio (C/N) measurements. The capsule method by Verardo *et al.* (1990) was employed, which involved acidification of 6-7 mg sediment within a silver (Ag) capsule. The samples were then moistened and 10% HCl acid was added in steps of 10 μ l, 20 μ l, 30 μ l and 50 μ l. The samples were dried on a hotplate at 50 °C for 1 hour. The Ag capsules were then wrapped with the tin (Sn) capsules for better combustion. The samples were analysed with a Costech ECS 4010 element analyzer at the Dept. of Geology, Lund University.

Grain size analysis

A total of 37 samples were analysed for grain size from sediment core Kyr10L, following the method described by van Hengstum *et al.* (2007). In order to obtain enough material for the analyses, several neighboring 1 cm samples were combined. To remove the organic matter, 15 ml of 33% H_2O_2 was added. The samples were then heated on a hot plate until reaction ceased, and allowed to cool down to 40 °C. Then 15 ml of 10% HCl was added to remove the carbonate material. Samples were subsequently diluted, centrifuged and decanted until a neutral pH was reached. To further remove biogenic silica, samples were boiled in 100 ml of 8% NaOH until reaction ceased. Sample were further diluted and centrifuged until neutral pH was reached. Sand grains (> 63 μ m) were separated by sieving and the mass fraction of sand in the freeze-dried sample was calculated. Grain size less than 63 μ m were analysed with the laser diffraction method with SediGraph III at the Dept. of Geology, Lund University. Three groups, <1 μ m (clay), 1-8 μ m (fine silt) and 8-63 μ m (medium-coarse silt) were classified and their corresponding mass distributions were calculated automatically.

212 Results

213 Chronology

214 Twelve AMS ^{14}C dates from sediment core KYR10L (Table 1) indicate an age between c.
 215 3500 to 7300 cal. a BP, from core depth 100 to 520 cm (Fig. 2). Results of $\delta^{18}\text{O}$
 216 measurements from *Macoma balthica* give an estimated reservoir age of 260-300 years, with
 217 an average of 270 years (Table 2). A 270-years reservoir age was subsequently subtracted
 218 from the ^{14}C ages before the age-depth model was established. A reservoir age of 206 years
 219 was determined from the sample in short core KYR10B, indicating a reduced reservoir age in
 220 recent samples compared with the older ones from mid-Holocene. The sedimentation rate is
 221 more or less constant, about 1.1 mm/ year. One sample from short core KYR10B was also
 222 dated, giving a rather recent age (Table 1).

224 Salinity reconstructions

225 Three mollusks samples with ages between 6400 and 6800 cal. a BP were analysed for
 226 $^{87}\text{Sr}/^{86}\text{Sr}$ measurements, resulting in SSS_{Sr} (sea surface salinity based on Sr isotope
 227 measurement) of c. 6-7, similar to the modern salinity (Fig. 3). Shell remains for $^{87}\text{Sr}/^{86}\text{Sr}$
 228 measurements are more frequently found between 3900-6000 cal. a BP and absent between
 229 6000-6400 cal. a BP. SSS_{Sr} fluctuated between c. 9 to 13 between 3900-6000 cal. a BP.
 230 Modern samples suggest SSS_{Sr} are in the range of 6-8, covering the current salinity (c. 7) in
 231 the region. SSS_{pl} (sea surface salinity based on process length) values were generally higher
 232 than SSS_{Sr} , with also larger uncertainties. Average SSS_{pl} values were stable and generally
 233 ranged between 13-15 from 3500 to 7300 cal. a BP, except from 5000 to 5300 cal. a BP,
 234 during which the highest SSS_{pl} values (16-18) were observed. The lowest SSS_{pl} was 13,
 235 observed in a sample from the top of the piston core KYR10L.

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237 Microfossil analysis, C_{org}, C/N and grain size

238 Dinoflagellate species observed from our study include *Operculodinium centrocarpum* (Wall
239 & Dale 1968), *Pyxidiniopsis psilata* (Head 1994), *Spiniferites* spp. (Wall & Dale 1968),
240 *Lingulodinium machaerophorum* (Deflandre & Cookson, 1955) and *Ataxiodinium choane*
241 (Reid 1974) (Fig. 3). It is noted *P. psilata* was classified as *O. centrocarpum* with ‘zero’
242 process due to eco-phenotypic reason in some studies (Mertens *et al.* 2011; Willumsen *et al.*
243 2013). On average more than 90% of dinoflagellate cyst assemblage was comprised of *O.*
244 *centrocarpum*. The dinoflagellate cysts assemblage was dominated by autotrophic species
245 whereas only one heterotrophic species *A. choane* was sparsely recorded. The observed
246 tintinnids lorica included *Tintinnopsis* sp and *Stenosemella* sp, and they were grouped
247 together. The marine-to-brackish algae *Radiosperma corbiferum* (Meunier 1910) is sensitive
248 to salinity variations in the Baltic Sea and only regularly recorded at lower core depth.
249 Calcareous benthic foraminifers were only observed from two neighboring samples,
250 corresponding to c. 5800 cal. a BP. C_{org} values ranged between 10 to 12%, with the lowest
251 values observed at 6500 cal. a BP. The C/N ratios ranged from 7 to 8, with slightly higher
252 value 6600-7300 cal. a BP. Grain sizes distribution (< 63 µm) was generally stable,
253 dominated by clay sized particles. The sand content was low, generally less than 1%. Three
254 zones were visually drawn based on the multi-proxy analysis (Fig. 3).

255

256 Zone I 7300-6400 cal. a BP

257 The *O. centrocarpum* concentration continuously increased since 7300 cal. a BP and peaked
258 at 6900 cal. a BP with more than 300k cysts/g. A subsequent decrease to c. 150k cyst/g at
259 6400 cal. a BP was observed. The concentration of *L. machaerophorum* was stable at 6k
260 cyst/g and decreased to almost no finds at 6400 cal. a BP. *Spiniferites* spp. demonstrated a
261 slightly increasing trend, reaching 7k cyst/g around 6500 cal. a BP. The concentrations of *P.*

262 *psilata* and *A. choane* were low in this zone. Tintinnids lorica were sparse, with the highest
263 concentrations observed between 7000 and 7300 cal. a BP, c. 10k individuals/g.
264 Concentrations of *Radiosperma corbiferum* were high, at c. 4-8k individuals/g. There were no
265 foraminifera observed in this zone. C_{org} values ranged between 10 and 11%, and demonstrated
266 a decreasing trend from c. 7300 to 6500 cal. a BP. The lowest values ~10% was recorded at c.
267 6600 cal. a BP. C/N values (>7.5) were rather consistent between 6400-7300 cal. a BP. The
268 minerogenic mass composition ($<63 \mu m$) of the sediment was dominated by clay (~50%),
269 followed by fine silt (25%) and medium-coarse silt (25%). The sand content ranged between
270 0.6% and 1.2%, with the highest concentrations recorded at 6800 cal. a BP.

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272 **Zone II 6400-3900 cal. a BP**

273 Concentration of *O. centrocarpum* generally decreased from 6400 to 3900 cal. a BP,
274 especially between 5500 and 3900 cal. a BP. Concentrations of both *Spiniferites* spp. and *L.*
275 *machaerophorum* were generally higher in this zone than in Zone I. High concentrations of
276 these species were found especially from 5800 to 5000 cal. a BP. The concentration of *P.*
277 *psilata* was low, except between 5600 and 4000 cal. a BP. A peak of *P. psilata* abundance at
278 5000 cal. a BP (100k cyst/g) was recorded. The tintinnids lorica concentrations were high,
279 especially between 5000-6400 cal. a BP. The peak abundance of tintinnids lorica was
280 recorded at 5800 cal. a BP, with 20k/g. The concentration of *R. corbiferum* was much lower
281 than in Zone I, with less than 2k individuals/g. Calcareous benthic foraminifera, *Ammonia*
282 *beccarii*, were encountered around 5800 cal. a BP from two samples. The C_{org} values (c. 12%)
283 were relatively high between 4400 and 4200 cal. a BP. The C/N ratios were stable (c. 7.5) and
284 were generally lower than in Zone I. Clay content showed a slight shift from below 50% in
285 Zone I to c. 55% in this zone. Medium-coarse silt was lower than in Zone I, with most

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286 samples below 20%. The sand content was consistently below 1%, with slightly higher value
287 at 5000-4400 cal. a BP.

289 **Zone III 3900-3500 cal. a BP**

290 The dinoflagellate cyst species, tintinnids lorica and *R. corbiferum* concentrations decreased
291 to their lowest concentrations in Zone III compared with previous zones (Fig. 3). The C_{org}
292 values decreased slightly from 12.5 to 11%. The C/N ratios were consistently below 7.5 in
293 this zone. The clay fraction decreased slightly to 50%, whereas silt and sand fractions
294 increased slightly.

296 **Discussion**

297 **Mid-Holocene sea surface salinity of the Baltic Sea**

298 Our salinity reconstruction from ⁸⁷Sr/⁸⁶Sr isotope measurements demonstrates that the lowest
299 salinity, about 6 to 7, occurred in Zone I in the time span between 6400 to 6800 cal. a BP.
300 This estimate is supported by the high abundance of *Radiosperma corbiferum*, of which the
301 highest concentrations were recorded in the central Baltic when the summer surface salinity is
302 around 6-7 (Gundersen 1988; Sorrel *et al.* 2006). SSS_{Sr} ranged between 9 and 13 from 6000
303 to 3900 cal. a BP in Zone II. The high SSS_{Sr} values correspond well with the generally high
304 *Spiniferites* spp. concentrations. Sildever *et al.* (2015) demonstrate that *Spiniferites* spp.
305 abundance from surface sediment in the Baltic Sea, Kattegat and Skagerrak region is
306 positively correlated with salinity (R²=0.91, n=5). Therefore, the higher abundance of
307 *Spiniferites* spp. in Zone II, compared with Zone I, further confirms the higher SSS_{Sr} estimate.
308 SSS_{Sr} increased c. 4 units from 6400 to 6000 cal. a BP. Such temporal sea surface salinity
309 increases have also been observed by Emeis *et al.* (2003) based on δ¹³C_{TOC} measurements

from a sediment core in the Gotland Basin. We note that the timing coincides with the deceleration of the global rise in sea levels (Peltier & Fairbanks 2006; Billeaud *et al.* 2009). It is likely that as the global sea level rise decelerated significantly around 6400 cal. a BP, the stratification between the bottom and upper parts of the water column became weaker due to the decrease in frequent saline water inflows into the Baltic Sea. A less stratified water column could result in a rapid salinity increase at 6400-6000 cal. a BP. Alternatively, the relatively low salinity between 6800 and 6400 cal. a BP could be caused by increased run-off due to a more humid climate, which was recorded as a brief event in southern Sweden around 6700 cal. a BP (Hammarlund *et al.* 2003). The findings of calcareous benthic foraminifera, although very rare, at c.5800 cal. a BP also indicate a potential salinity increase in Zone II. The climate conditions in Zone II were generally warm and dry, favoring the sea water inflow through the Danish Straits (Emeis *et al.* 2003; Hammarlund *et al.* 2003). The high SSS_{Sr} in Zone II was also facilitated by the large inlet areas, including the Öresund and Darss sills, at that time (Gustafsson & Westman 2002).

The absence of mollusk shell remains makes it impossible to obtain SSS_{Sr} after 4000 cal. a BP. However, the SSS_{Sr} was 11 at 4000 cal. a BP, implying a salinity decrease until the present. A climate shift to wetter and colder conditions after 4000 cal. a BP, which was observed in the Scandinavia region, might be one cause of the salinity decrease (Hammarlund *et al.* 2003; Krossa *et al.* 2014).

Previous SSS_{Sr} estimates in the Baltic Proper, based on the mollusks collected in the raised beaches along the Baltic coast, suggest a salinity of c. 9-13 between 6000 and 3900 cal. a BP (Widerlund & Andersson 2011). Such estimate is similar from our SSS_{Sr} estimate. However, the authors found about one third of the SSS_{Sr} estimates (<7) were too low, which could partly be explained by the unknown environment when the mollusks were deposited. Our SSS_{Sr}

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334 results demonstrate that mollusk shells collected from sediment cores are probably more
335 suitable for the method.

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337 **Uncertainty with SSS_{pl}**

338 The SSS_{pl} results, indicating a salinity of 13-18 between 3500-7300 cal. a BP, were higher
339 than SSS_{sr}. The SSS_{pl} values were also higher than previous salinity estimates (Gustafsson &
340 Westman 2002; Widerlund & Andersson 2011). We attribute these higher values partly to
341 overestimation of the salinity reconstruction from the univariate calibration, in a low salinity
342 (<20) environment (Fig. S2). If only *O. centrocarpum* recorded below a salinity of 20 were
343 taken into account from sediment surface samples analysed by Mertens *et al.* (2011) and
344 Jansson *et al.* (2014), SSS_{pl} reconstruction would result in an average of c. 10 between 7300-
345 3500 cal. a BP (Fig. S3), close to the SSS_{sr} estimates.

346 Jansson *et al.* (2014) apply a multi-variant model to determine the impact of temperature,
347 salinity and nutrient data on the process length of *O. centrocarpum*. The authors conclude that
348 sea surface temperature and sea surface salinity explained 22% and 78%, respectively, of the
349 variance in process length variation. This suggests that using salinity as the only variable to
350 explain the process length variation is not appropriate. Moreover, it is very likely that the *O.*
351 *centrocarpum* has two genetically different groups, one with short processes and the other
352 with long ones. This would result in a highly significant correlation coefficient between
353 salinity and process length. However, it is not appropriate to use the linear function for a
354 salinity calibration. SSS_{pl} estimates based on the calibration function from Sildever *et al.*
355 (2015) result in similar salinity values (Fig. S3). However, we suggest further studies, where
356 field and culture studies are combined, to better understand the relationships between process
357 length of *O. centrocarpum* and environment variables, before using univariate calibrations.

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359 Regional environment of Littorina Sea phase

360 The Baltic Sea changed from the freshwater Ancylus Lake system to the brackish Littorina
361 Sea system about c. 8500 cal. a BP, due to the continuous global sea level rise and the
362 subsidence of the Danish Straits (Sohlenius *et al.* 2001; Rößler *et al.* 2011). The concentration
363 of *O. centrocarpum* peaked at 6800-7000 cal. a BP in Zone I, reflecting the Littorina
364 transgression maximum. This event was also recorded in the Smygen Bay (Fig. 1) on the
365 southeast Swedish coast from 7100 to 6900 cal. a BP, and in the Gotland Basin from 6800 to
366 6600 cal. a BP (Brenner 2005; Yu & Berglund 2007). In our study area, close to Smygen Bay,
367 estimated relative sea level was c.8 m above the present sea level during the transgression
368 maximum (Berglund *et al.* 2005; Pässe & Andersson 2005).

369 The higher C/N values in Zone I probably indicate an increased terrestrial material input
370 due to erosion during the transgression period (Meyers 1994). As terrigenous organic matter
371 has much higher C/N ratios (>20) than the marine-brackish organic matter (<10), C/N ratios
372 in our record (c. 7-8) imply an organic material dominantly from brackish-marine origin
373 (Meyers 1994). During the transgression period in Zone I, bottom water intensity is most
374 likely high, which is supported by relatively higher silt and sand proportions compared with
375 clay. Zone II represents the period after the Littorina transgression maximum during which
376 the regional sea level generally decreased. Small variations in the C_{org}, C/N and grain size
377 distributions indicate a stable sedimentary environment. We note that the main changes in
378 Zone II are associated with the organic-walled microfossil assemblages. The overall high
379 abundance of *L. machaerophorum*, especially between 5000 and 6000 cal. a BP, probably
380 implies a high productivity and salinity environment (Dale 2001; Mertens *et al.* 2009).
381 Tintinnids are heterotrophic organisms and they are abundant in an open marine environment

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382 such as the Atlantic Ocean (Cordeiro *et al.* 1997; Pospelova *et al.* 2010; Price & Pospelova
383 2011). High abundance of tintinnids lorica during 5400-6000 cal. a BP also indicates this
384 period as a nutrient-rich and high salinity period. High abundance of *P. psilata* was recorded
385 4900-5100 cal. a BP, which is observed from the Gotland Basin between 5500 and 5300 cal. a
386 BP (Brenner 2005). Considering the age uncertainties, it is quite likely that the period of
387 increased *P. psilata* abundance is the same event across the Baltic Sea. Such an increase
388 implies a temporal shift towards conditions favoured by *P. psilata*, which needs further
389 investigation. The transition between Zone II and Zone III occurred at c. 3900 cal. a BP,
390 marking the termination of the Littorina Sea. The low abundances of nearly all microfossils in
391 Zone III indicate the transition to the Post Littorina Sea, characterized by a subtle and gradual
392 decrease in sea level, and reduced salinity. The Zone III in our study has a very similar
393 dinoflagellate cyst composition to the dinoflagellate cyst Zone III from Yu and Berglund
394 (2007).

396 **Conclusions**

397 Sea surface salinity was in the range of 6-7 during 6400-6800 cal. a BP and varied between 9
398 and 13 during 3900-6000 cal. a BP in the southern Baltic, based on the $^{87}\text{Sr}/^{86}\text{Sr}$
399 measurements. The salinity reconstructions based on the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios, measured from
400 mollusks shells, and on the process length from dinoflagellate cysts *Operculodinium*
401 *centrocarpum*, differ from each other. The sea surface salinity estimates based on the *O.*
402 *centrocarpum* process length variations seem to overestimate salinity.

403 By applying a multi-proxy approach, our study has provided additional information on the
404 regional environment changes in the Littorina Sea phase, and three zones can be identified
405 over the study period. Zone I represents the Littorina transgression maximum period,

characterized by a relatively high sea level, greater input of terrestrial material and higher bottom water intensity. Zone II is characterized by high salinity (9-13) in the Littorina Sea phase and higher marine productivity. Zone III represents the starting period of the Post Littorina Sea phase, indicating a shift to the modern Baltic phase with decreased salinity and productivity.

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555 **Figure captions (Fig. 1-3)**

556 *Fig. 1.* Location of the coring site, surface salinity data from three monitoring stations and other sites
 557 referred to in the text. A. Map of the Baltic Sea and its adjacent water bodies. Two monitoring stations,
 558 BY5 and BY10 are marked with filled circles. AB=Arkona Basin, BB=Bornholm Basin, GB=Gotland
 559 Basin. B. Map showing the coring site KYR and the nearby monitoring station KAARV4. C. Time
 560 series (1998-2011) of surface salinity data from the three monitoring stations. Crosses represent the
 561 average salinity of the top 20m water from different stations. Yearly-averaged salinity series are
 562 shown as solid lines.

564 *Fig. 2.* Age-depth curve for the sediment sequence of KYR10L. Dashed lines correspond to the 95%
 565 confidence interval associated with age-depth modelling.

567 *Fig. 3.* Results from salinity reconstructions and multi-proxy analysis. The shaded bar corresponds to
 568 the modern salinity from the station KAARV4. Error bars associated with the SSS_{sr} indicate method
 569 precision of $\pm 5\%$ (Widerlund & Andersson 2011). SSS_{pl} is plotted with $\pm 1SD$ error, associated with
 570 the process length measurements. The mollusk shell (a) was collected from the top of piston core
 571 Kyr10L; mollusk shell (b) was collected from 18cm of short core KYR10B, which was dated to a
 572 modern age (Table 1); sample (c) was collected from the top of KYR10L. Numbers for microfossils

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correspond to the total number from one gram dry sediment. Filled and open circles represent existence and absence of benthic foraminifera respectively.

Supplementary figure captions (Fig. S1-S3)

Fig. S1. Scanning electron microscope images of mollusk shell remains: (A) Sample KYR10L-209, *Macoma balthica* (B) Sample KYR10L-273, *Mytilus edulis* (C) Sample KYR10L-312, *Macoma balthica* (D) Sample KYR10L 379, *Mytilus edulis*

Fig. S2. Correlation between sea surface salinity and process length of *O. centrocarpum* with two salinity ranges. Blue dots are the whole data set with salinity range 5-35 from the Baltic–Kattegat–Skagerrak region used by Jansson *et al.* (2014). Linear fit of the data set by the authors is the black line. The data set can be obtained in supplementary Table 1 by Jansson *et al.* (2014). The red dashed rectangle corresponds to the salinity range 5-20 of the data set, which covers the estimated salinity range over the mid-Holocene. The red solid line corresponds to the linear fit based on salinity values within the range of 5-20. The two different calibrations can lead to significant differences based on the measurement of *O. centrocarpum* process length.

Fig. S3. Comparison of salinity reconstructions based on two different calibration functions. Salinity reconstruction calculated using the function from Mertens *et al.* (2011) is marked in black, and salinity reconstruction using the function by Sildever *et al.* (2015) is marked in red. The error bar indicates $\pm 1SD$.

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Table 1. Details of AMS ¹⁴C dates, δ¹³C and δ¹⁸O results obtained from shell remains in the piston core KYR10L and short core KYR10B

Core name	Core depth (cm)	Lab. code	Material	¹⁴ C date (BP)	Error (±)	δ ¹³ C (‰ V-PDB)	Calibrated age (cal. a BP±σ)
KYR10B	18	LuS 9529	<i>Macoma balthica</i>	136 pMC	0.6	-2.86	-25±2
KYR10L	146	LuS 10712	<i>Macoma balthica</i>	3825	40	4	3841±65
KYR10L	187	LuS 9850	<i>Macoma balthica</i>	4045	50	-	4152±88
KYR10L	209	LuS 9851	<i>Macoma balthica</i>	4225	50	-	4404±82
KYR10L	235	LuS 9852	<i>Macoma balthica</i>	4410	50	-	4680±88
KYR10L	267	LuS 9853	<i>Mytilus edulis</i>	4620	50	-0.26	4939±76
KYR10L	295	LuS 9854	<i>Macoma balthica</i>	4585	50	-	4903±65
KYR10L	323	LuS 9855	<i>Macoma balthica</i>	5065	50	-0.98	5514±66
KYR10L	341	LuS 9856	<i>Macoma balthica</i>	5200	50	-1.19	5670±59
KYR10L	353	LuS 9525	<i>Mytilus edulis</i>	5265	50	1.64	5745±78
KYR10L	369	LuS 9526	<i>Macoma balthica</i>	5370	50	-0.68	5831±62
KYR10L	392	LuS 9527	<i>Mytilus edulis</i>	5455	50	-0.5	5950±76
KYR10L	440	LuS 9528	<i>Macoma balthica</i>	6065	50	0.01	6593±62

Table 2. Results of $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ measurements (reproducibility > 0.1%) from core KYR10L and short core KYR10B

Core name	Core depth (cm)	Lab. code	Species	$\delta^{13}\text{C}$ (‰ V-PDB)	$\delta^{18}\text{O}$ (‰ V-PDB)	Estimated reservoir age* (years)
KYR10B	11	HFKYR10B18	<i>Macoma balthica</i>	-0.53	-4.4	206
KYR10L	209	HFKYR10L209	<i>Macoma balthica</i>	-0.16	-2.59	282
KYR10L	295	HFKYR10L295	<i>Macoma balthica</i>	0.02	-2.21	298
KYR10L	311	HFKYR10L311	<i>Macoma balthica</i>	-0.75	-3.08	280
KYR10L	362	HFKYR10L362	<i>Macoma balthica</i>	0.49	-3.10	261
KYR10L	427	HFKYR10L427	<i>Macoma balthica</i>	-0.75	-3.08	261
KYR10L	440	HFKYR10L440	<i>Macoma balthica</i>	0.24	-2.9	269

*The estimated reservoir age (Rt) was calculated based on the function by Loughheed *et al.* (2013) : $Rt = 41.8 \times \delta^{18}\text{O} + 390$

Table 3. Strontium isotope data measured on mollusk shells and calculated salinities

Sediment core	Core depth (cm)	Lab. code	Shell type	Age (cal. a BP)	⁸⁷ Sr/ ⁸⁶ Sr**	SSS _{Sr} ***
Kyr10B	18	Kyr10B-18	<i>Macoma balthica</i>	-25	0.709311 ± 8	7.9(8.3)8.6
Kyr10L	0	Kyr10L-0	<i>Macoma balthica</i>	*	0.709368 ± 8	6.2(6.4)6.6
Kyr10L	157	Kyr10L-157	<i>Mytilus edulis</i>	3967	0.709260 ± 8	10.6(11.1)11.8
Kyr10L	187	Kyr10L-187	<i>Macoma balthica</i>	4225	0.709272 ± 8	9.8(10.2)10.8
Kyr10L	209	Kyr10L-209	<i>Macoma balthica</i>	4441	0.709266 ± 8	10.2(10.7)11.3
Kyr10L	237	Kyr10L-237	<i>Mytilus edulis</i>	4694	0.709245 ± 8	11.7(12.4)13.2
Kyr10L	247	Kyr10L-247	<i>Mytilus edulis</i>	4776	0.709267 ± 8	10.1(10.6)11.2
Kyr10L	263	Kyr10L-263	<i>Mytilus edulis</i>	4910	0.709264 ± 8	10.3(10.8)11.3
Kyr10L	267	Kyr10L-267	<i>Mytilus edulis</i>	4948	0.709281 ± 8	9.3(9.7)10.2
Kyr10L	273	Kyr10L-273	<i>Mytilus edulis</i>	4996	0.709259 ± 8	10.6(11.2)11.8
Kyr10L	286	Kyr10L-286	<i>Macoma balthica</i>	5105	0.709283 ± 8	9.2(9.6)10.1
Kyr10L	312	Kyr10L-312	<i>Macoma balthica</i>	5380	0.709279 ± 8	9.4(9.9)10.4
Kyr10L	333	Kyr10L-333	<i>Macoma balthica</i>	5592	0.709281 ± 8	9.3(9.7)10.3
Kyr10L	341	Kyr10L-341	<i>Macoma balthica</i>	5661	0.709247 ± 8	11.5(12.2)13.0
Kyr10L	355	Kyr10L-355	<i>Macoma edulis</i>	5766	0.709256 ± 8	10.8(11.5)12.1
Kyr10L	379	Kyr10L-379	<i>Mytilus edulis</i>	5955	0.709262 ± 8	10.4(11.0)11.6
Kyr10L	427	Kyr10L-427	<i>Macoma balthica</i>	6440	0.709382 ± 8	5.8(6.0)6.2
Kyr10L	439	Kyr10L-439	<i>Mytilus edulis</i>	6567	0.709355 ± 8	6.5(6.7)6.9
Kyr10L	457	Kyr10L-457	<i>Macoma balthica</i>	6736	0.709349 ± 8	6.7(6.9)7.1

* Sample Kyr10L-0 is an undated shell fragment found at the top of the piston core.

** The reported error represents the external precision in the ⁸⁷Sr/⁸⁶Sr determination (±0.000018 (±2σ) for the NBS 987 standard).

***SSS_{Sr} is calculated from based on Eqs. (3)–(5) in Windlerlund & Andersson (2006). Salinity ranges are calculated based on the ±2σ from ⁸⁷Sr/⁸⁶Sr measurements. Salinity_{Sr} values in parentheses are the SSS_{Sr} used in the text.