

1 **TITLE**

2 Biominalisation plasticity and environmental heterogeneity predict geographic resilience  
3 patterns of foundation species to future change

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5 **Running title**

6 Shell plasticity predicts resilience

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24 **ABSTRACT**

25 Although geographic patterns of species' sensitivity to environmental changes are defined by  
26 interacting multiple stressors, little is known about compensatory processes shaping regional  
27 differences in organismal vulnerability. Here, we examine large-scale spatial variations in  
28 biomineralisation under heterogeneous environmental gradients of temperature, salinity, and  
29 food availability across a 30° latitudinal range (3,334 km), to test whether plasticity in calcareous  
30 shell production and composition, from juveniles to large adults, mediates geographic patterns of  
31 resilience to climate change in critical foundation species, the mussels *Mytilus edulis* and *M.*  
32 *trossulus*. We find shell calcification decreased towards high latitude, with mussels producing  
33 thinner shells with a higher organic content in polar than temperate regions. Salinity was the best  
34 predictor of within-region differences in mussel shell deposition, mineral and organic  
35 composition. In polar, subpolar, and Baltic low-salinity environments, mussels produced thin  
36 shells with a thicker external organic layer (periostracum), and an increased proportion of calcite  
37 (prismatic layer, as opposed to aragonite) and organic matrix, providing potentially higher  
38 resistance against dissolution in more corrosive waters. Conversely, in temperate, higher-salinity  
39 regimes, thicker, more calcified shells with a higher aragonite (nacreous layer) proportion were  
40 deposited, which suggests enhanced protection under increased predation pressure. Interacting  
41 effects of salinity and food availability on mussel shell composition predict the deposition of a  
42 thicker periostracum and organic-enriched prismatic layer under forecasted future environmental  
43 conditions, suggesting a capacity for increased protection of high-latitude populations from  
44 ocean acidification. These findings support biomineralisation plasticity as a potentially  
45 advantageous compensatory mechanisms conferring *Mytilus* species a protective capacity for  
46 quantitative and qualitative trade-offs in shell deposition as a response to regional alterations of

47 abiotic and biotic conditions in future environments. Our work illustrates that compensatory  
48 mechanisms, driving plastic responses to the spatial structure of multiple stressors, can define  
49 geographic patterns of unanticipated species resilience to global environmental change.

50

51 **Keywords**

52 Climate change, *Mytilus*, calcification, biomineralisation, resistance, ocean acidification,  
53 compensatory mechanisms, multiple stressors

54 **INTRODUCTION**

55 Unprecedented global environmental changes are driving scientists towards increased efforts to  
56 investigate the mechanisms underlying geographic variation in biotic responses to future  
57 environmental conditions (Nagelkerken & Connell, 2015; Urban et al., 2016). However, our  
58 ability to predict changes to species and ecosystems in response to climate change remains  
59 limited (Kroeker, Kordas, & Harley, 2017). Current projections are severely constrained by  
60 heterogeneous patterns of ocean warming and acidification (Gattuso et al., 2015), multiple  
61 stressors (Breitburg et al., 2015), and compensatory processes (Cross, Harper, & Peck, 2019;  
62 Ghedini, Russell, & Connell, 2015; Leung, Russell, & Connell, 2017), as well as predictive  
63 models which often exclude important biological mechanisms (Urban et al., 2016). Therefore, a  
64 better mechanistic understanding of environmental sources and processes mediating species'  
65 responses to disturbances is critical for building the theoretical baseline necessary to forecast the  
66 combined effects of multiple emerging stressors (Kroeker et al., 2017; Urban et al., 2016).

67 Advances in macroecology suggest that permanent environmental mosaics, defined by spatial  
68 overlaps of non-monotonic environmental gradients (Kroeker et al., 2016), as well as regional  
69 adaption or acclimatisation (Calosi et al., 2017; Peck, 2018; Vargas et al., 2017), dictate  
70 geographic variations in species performance and sensitivity to disturbances in marine  
71 ecosystems. Key to these works is that responses vary among populations and taxa (Calosi et al.,  
72 2017; Kroeker et al., 2013; Telesca et al., 2018), which often play disproportionately strong roles  
73 in structuring benthic communities (Ashton, Morley, Barnes, Clark, & Peck, 2017). Thus,  
74 species-specific biological processes driving organismal variability likely shape differential  
75 regional responses of foundation species to co-occurring multiple drivers. This can establish

76 spatial patterns of unexpected susceptibility of marine communities to future environmental  
77 conditions.

78 Species producing calcium carbonate ( $\text{CaCO}_3$ ) shells and skeletons are possibly experiencing the  
79 strongest impacts of rapid environmental changes (Kroeker et al., 2013). Knowledge of their  
80 sensitivity is derived largely from short- to long-term studies on model organisms (Kroeker et  
81 al., 2013; Nagelkerken & Connell, 2015), while complex variations under multiple stressors in  
82 natural environments have rarely been investigated (Ashton et al., 2017; Kroeker et al., 2016;  
83 Peck et al., 2015; Watson, Morley, & Peck, 2017). Therefore, inferences made from  
84 experimental studies may not necessarily translate to complex marine ecosystems (Connell et al.,  
85 2017; Vargas et al., 2017). Indeed, species-specific responses to habitat alterations (Kroeker et  
86 al., 2013), on top of mixed outcomes of environmental interactions (Crain, Kroeker, & Halpern,  
87 2008), make future ecosystem predictions extremely challenging (Kroeker et al., 2017). This  
88 leaves open the question: do differences in biological processes, shaping regional variations of  
89 calcifiers' responses to interacting environmental stressors, define geographic patterns of  
90 unanticipated species sensitivity or resilience to global environmental change?

91 A body of research has focused on responses of marine calcifiers to altered water chemistry  
92 (Kroeker et al., 2013; Nagelkerken & Connell, 2015), but studies have rarely considered changes  
93 in biogeochemical cycles strongly mediating biological responses to disturbance (Gattuso et al.,  
94 2015). Among those, a marked intensification of the global water cycle in response to warming  
95 (+4% for +0.5 °C) has been documented over recent decades through changes in ocean salinity  
96 (Durack, Wijffels, & Matear, 2012). Salinity is a major ecological factor dictating distribution  
97 and survival of aquatic organisms, ecosystem functioning (Solan & Whiteley, 2016), as well as  
98 conditions for biomineralisation (Thomsen, Haynert, Wegner, & Melzner, 2015). Indeed, salinity

99 correlates positively with the availability of calcification substrates [bicarbonate ( $\text{HCO}_3^-$ ) and  
100 calcium ( $\text{Ca}^{2+}$ ) ion concentrations] and, therefore, seawater  $\text{CaCO}_3$  saturation state ( $\Omega_{\text{CaCO}_3}$ )  
101 (Thomsen, Ramesh, Sanders, Bleich, & Melzner, 2018). Saturation states of the two main  $\text{CaCO}_3$   
102 polymorphs [calcite ( $\Omega_{\text{calc}}$ ) and aragonite ( $\Omega_{\text{arag}}$ )] control calcification kinetics, driving net  
103 deposition or dissolution of  $\text{CaCO}_3$  structures (Ries, Ghazaleh, Connolly, Westfield, & Castillo,  
104 2016; Sanders, Schmittmann, Nascimento-Schulze, & Melzner, 2018; Thomsen et al., 2018).  
105 Multidecadal studies have revealed a global salinity pattern following the “rich-get-richer”  
106 mechanism, where salty ocean regions (compared to the global mean) are getting saltier (mid-  
107 latitudes), whereas low salinity regions are getting fresher (tropical convergence zones and polar  
108 regions) (Durack et al., 2012). In a future 2 - 3 °C warmer world, a substantial 16 - 24%  
109 intensification of the global water cycle is predicted to occur, making salinity gradients much  
110 sharper (Durack et al., 2012). This may affect deposition rates of biogenic  $\text{CaCO}_3$  through  
111 altered calcification costs. However, the emergent ecological effects of changing salinity on  
112 calcifying species are largely unknown.

113 Atlantic blue mussels, *Mytilus edulis* and *M. trossulus*, are important bed-forming foundation  
114 species throughout the eulittoral ecosystems of the northern hemisphere, and represent valuable  
115 resources for aquaculture (192,000 t produced in 2016 worth 325 million USD) (FAO, 2017).  
116 Growing awareness of the consequences of climate change on biodiversity and the industry that  
117 *Mytilus* species support has stimulated a number of studies to estimate their response potential to  
118 changing ocean conditions (Telesca et al., 2018; Thomsen et al., 2017; Thyrring, Blicher,  
119 Sørensen, Wegeberg, & Sejr, 2017).

120 Calcareous shells perform a range of vital functions including structural support and protection  
121 against predators. Because shell integrity determines survival, shell traits are subject to strong

122 selection pressure with functional success or failure a fundamental evolutionary driver. Blue  
123 mussel shell consists of three layers (Fig. 1a,b): (1) the outer organic periostracum, and the  
124 calcified (2) anvil-type fibrous prismatic and (3) nacreous layers. The periostracum is made of  
125 sclerotised (quinone-tanned) proteins. This layer provides a protected environment for the  
126 deposition of calcareous components, and protects shells from corrosive, acidic waters as well as  
127 predatory and endolithic borers (Harper, 1997). The fibrous prismatic and nacreous layers are  
128 composed of CaCO<sub>3</sub> crystals of different mineral forms, calcite and aragonite respectively, and  
129 inter-crystalline biomineral organic matrix (Checa, Pina, Osuna-Mascaró, Rodríguez-Navarro, &  
130 Harper, 2014). These calcareous layers are characterised by different microstructures (Fig. S1)  
131 and more (i.e. aragonite) or less (i.e. calcite and organics) soluble components (Harper, 2000;  
132 Mucci, 1983), the combination of which determines specific chemical and mechanical shell  
133 protection characteristics (Barthelat, Rim, & Espinosa, 2009; Currey & Taylor, 1974; Fitzer et  
134 al., 2015). Differences in energetic costs of making shell components (Palmer, 1992; Watson et  
135 al., 2017), combined with future alterations in environmental gradients (Gattuso et al., 2015) and  
136 water carbonate chemistry (Thomsen, et al., 2015; Thomsen et al., 2018), will likely influence  
137 variations in shell production and composition, shaping regional patterns of shell resilience to  
138 abiotic and biotic alterations.

139 *Mytilus* spp. growth, biomineralisation and fitness are linked to multiple drivers, including water  
140 temperature, salinity and food supply [chlorophyll-*a* (Chl-*a*) concentration] (Sanders et al., 2018;  
141 Thomsen, Casties, Pansch, Körtzinger, & Melzner, 2013). As is the case for all, but in the  
142 context of this study, in the North Atlantic and Arctic Oceans these key environmental factors  
143 vary heterogeneously with latitude (Fig. 1c,d), encompassing a range of conditions predicted  
144 under different future climate change scenarios (Kirtman et al., 2013). Here, we hypothesise that

145 plasticity in shell biomineralisation, driving spatial variations in shell production, mineral and  
146 organic composition, i) shape regional responses of *Mytilus* species to interacting environmental  
147 drivers, and ii) define geographic patterns of blue mussel vulnerability in the face of global  
148 environmental changes.

149 Despite projected alterations of salinity (Durack et al., 2012; Gattuso et al., 2015) and, therefore,  
150 water carbonate chemistry ( $\Omega_{\text{CaCO}_3}$ ) and calcification costs (Sanders et al., 2018; Thomsen et al.,  
151 2018), salinity gradients have been overlooked in large-scale predictive models for marine  
152 calcifiers. This knowledge is essential to forecast whether environmental changes affect shell  
153 variability and functional capability, especially in ecologically important foundation species such  
154 as *M. edulis* and *M. trossulus*. These factors are crucial for understanding species susceptibility  
155 to other rapidly emergent stressors, such as warming, acidification, and altered species  
156 interactions (Kroeker et al., 2017).

157 In this study, we examine the relationships between variations in *Mytilus* spp. shell  
158 biomineralisation, from juveniles to large adults, and interactive environmental gradients of  
159 temperature, salinity and Chl-*a* concentration in 17 populations spanning a latitudinal range of  
160 30° (3,334 km) across the Atlantic-European and Arctic coastline (Fig. 1c,d). In particular, we  
161 tested for a latitudinal effect on blue mussel shell calcification that we hypothesise will show a  
162 general decrease from temperate to polar regions. We also identified environmental sources of  
163 within-region variations in shell production and composition, to test whether salinity affects shell  
164 biomineralisation during growth, suggesting changes of shell structure, mechanical and chemical  
165 properties. Finally, we modelled spatial trends in shell deposition with environmental gradients,  
166 to test whether plasticity in shell biomineralisation shapes regional responses of *Mytilus* species  
167 to interacting stressors, defining geographic patterns of sensitivity to future changes.



168 **MATERIALS AND METHODS**

169 ***Mytilus* collection**

170 We sampled individuals from 17 *Mytilus* (*M. edulis* and *M. trossulus*) populations along the  
171 North Atlantic, Arctic, and Baltic Sea coastlines from four distinctive climatic regions (warm-  
172 temperate, cold-temperate, subpolar and polar) covering a latitudinal range of 30° (a distance of  
173 3,334 km), from Western European (Brest, North-West France, 48°N) to Northern Greenlandic  
174 (Qaanaaq, North-West Greenland 78°N) coastlines (Fig. 1c). During December 2014 -  
175 September 2015, mussels of various size classes for each site (shell length 26 - 81 mm) were  
176 sampled from the eulittoral zone on rocky shores for a total of 424 individuals (Table S1). At  
177 each site, specimens were collected from the lower limit of the intertidal zone (0 - 0.5 m above  
178 the zero tidal level) on rocky substratum to allow for comparisons (in terms of local conditions)  
179 between Atlantic and Baltic mussels, the latter experiencing short and irregular periods of air  
180 exposure during the year. For each specimen, shell length was measured with digital calipers  
181 (0.01 mm precision) and used as a within-population (collection site) proxy for age (Seed &  
182 Richardson, 1990).

183 We analysed *Mytilus* populations for which the genetic structure was known, with particular  
184 focus on species identity and hybrid status (*M. edulis* × *M. trossulus*). Genetic studies have  
185 revealed various episodes of extensive introgression of *M. edulis* alleles in *M. trossulus*  
186 populations and pronounced hybridisation patterns, especially in the Baltic Sea (Riginos &  
187 Cunningham, 2005; Stuckas, Stoof, Quesada, & Tiedemann, 2009), resulting in the absence of  
188 “pure” *M. trossulus* populations at the North Atlantic and Baltic Sea scales. *Mytilus* shells used  
189 were either from individuals already evaluated in genetic investigations or mussels obtained

190 from sites routinely used in regional monitoring programs that provided information on genetic  
191 status (Table S1). Areas where the Mediterranean mussel, *Mytilus galloprovincialis*, was known  
192 to be present were avoided. We did, however, sample a few sites (3) with very low levels of *M.*  
193 *edulis* × *M. galloprovincialis* hybridisation. Here, mussels from the *M. edulis* species-complex  
194 were analysed together because: i) they share the same shell microstructure (Fig. S1), ii) they  
195 inhabit a wide range of similar habitats in the eulittoral zone, iii) their pervasive hybridisation  
196 patterns (with differential introgression) excluding pure *M. trossulus* populations at the spatial  
197 scale analysed, and iv) the documented smaller contribution of genetic status than environmental  
198 heterogeneity on the variability of *Mytilus* shell traits across large geographic scales (Krapivka et  
199 al., 2007; Telesca et al., 2018).

200

## 201 **Mussel shell preparation**

202 We set left shell valves in polyester resin (Kleer-Set FF, MetPrep, Coventry, U.K.) blocks.  
203 Embedded specimens were sliced longitudinally along their axis of maximum growth (Fig. 1a)  
204 using a diamond saw and then progressively polished with silicon carbide paper (grit size: P800 -  
205 P2500) and diamond paste (grading: 9 - 1 µm). Photographs of polished sections (Fig. 1b) were  
206 acquired with a stereo-microscope (Leica M165 C equipped with a Leica DFC295 HD camera,  
207 Leica, Wetzlar, Germany) and shell thickness was measured using the Fiji software (v1.51w).  
208 Since larger individuals had undergone evident environmental abrasion or dissolution which  
209 removed the periostracum and prismatic layer closer to the umbo, we estimated the thickness of  
210 the whole-shell, prismatic and nacreous layers at the midpoint along the shell cross-section.  
211 Periostracum thickness was measured at the posterior edge where it attaches to the external side

212 of the prismatic layer, to estimate the fully formed organic layer that was unaffected by decay or  
213 abrasion (Harper, 1997).

214

## 215 **Organic content analyses**

216 We performed thermogravimetric analyses (TGA) to estimate the weight proportion (wt%) of  
217 organic matrix within the prismatic layer. A random subsample of 20 *Mytilus edulis* specimens  
218 were selected from four populations (sites 1, 11, 15, 16) to explore differences in shell organic  
219 content under temperate and polar regimes. We removed the periostracum by sanding, and tiles  
220 of prismatic layer ( $8 \times 5$  mm,  $n = 20 \times 4$  sites) were cut along the posteroventral shell margin.  
221 Tiles were cleaned, air-dried and then finely ground. We tested ten milligrams of this powdered  
222 shell with a thermogravimetric analyser (TGA Q500, TA Instruments, New Castle, DE, U.S.A.).  
223 Samples were subjected to constant heating from  $\sim 25$  °C to 700 °C at a linear rate of  $10$  °C  $\text{min}^{-1}$   
224 under a dynamic nitrogen atmosphere and weight changes were recorded (Supporting Document  
225 S1). We estimated the wt% of organic matter within the shell microstructure as the proportion of  
226 weight loss during the thermal treatment between 150 °C and 550 °C (Fig. S2). The TGA  
227 method was used in preference to the traditional muffle furnace approach to explore changes in  
228 organic matrix within a specific shell layer (intra- and inter-crystalline organics). Although  
229 traditional approaches are still used to measure variation of organic content at the whole-shell  
230 level (i.e. estimates of total organics including organic matrix, shell ligament and organics-rich  
231 layers, such as the periostracum and myostracum) (Sanders et al., 2018), TGA represents a more  
232 accurate and widely used method to provide an unbiased estimate (i.e. no influence of residual  
233 intracrystalline water) of the wt% of organic matrix at a single microstructure level in molluscs

234 (Checa, Macías-Sánchez, Harper, & Cartwright, 2016; Zaremba, Morse, Mann, Hansma, &  
235 Stucky, 1998). In a cross calibration experiment where samples of blue mussel shells were  
236 analysed both in a muffle furnace and by TGA ( $n = 5 \times 4$  sites), results obtained were not  
237 significantly different (Fig. S3).

238

### 239 **Environmental characterisation**

240 We selected three key environmental drivers based on their known influence on mussel growth  
241 and calcification, their level of collinearity across the geographic scale investigated, and the  
242 forecasted major ocean alterations under climate change (Kirtman et al., 2013; Sanders et al.,  
243 2018; Thomsen, et al., 2013). For each site measurements of sea surface temperature, salinity,  
244 and Chl-*a* concentration, the latter being used as a proxy for food supply (Thomsen, et al., 2013),  
245 were generated using the Copernicus Marine Environment Monitoring Service (CMEMS)  
246 (<http://marine.copernicus.eu/>). These climate datasets are composed of high-resolution physical  
247 and biogeochemical assimilated (iterative integration of new observational information and  
248 model forecasts over time) daily data (Supporting Document S2). To provide a first order mean  
249 approximation of the average water conditions prevailing during the periods of mussel growth  
250 and shell deposition (Carter & Seed, 1998; Thomsen, et al., 2010) across the life-span of both  
251 young and adult sampled specimens from different age classes (between two and six years old),  
252 we expressed parameters as mean May - October values averaged over the 6-year period 2009 -  
253 2014 (daily observation,  $n = 2,191$  per parameter) and used these as input variables (Fig. 1d;  
254 Table S2).

255 Direct environmental monitoring for each site was not feasible due to the number and geographic  
256 range (> 3,300 km latitudinal span) of the mussel populations analysed, the absence of direct  
257 records for many of the sites used, and the temporal resolution (daily data over six years)  
258 required to provide an average estimate for the growth conditions of young and adult specimens.  
259 For this large-scale study, assimilated data presented potential advantages compared to  
260 traditional measurements due to their spatial and temporal extent, repetition over time, advanced  
261 calibration and validation (i.e. high correlation with discrete field measurements) (IOCCG, 2014;  
262 Telesca et al., 2018; Thomas et al., 2011).

263

## 264 **Statistical analysis**

265 Generalised linear (mixed) models, GL(M)Ms, were used to explain shell thickness and  
266 composition, from juveniles to large adults, with respect to latitude and environmental drivers,  
267 and to compare between the individual shell layers. GLMMs were applied i) to account for the  
268 hierarchical structure of the dataset consisting of multiple specimens ( $n = 24 - 26$  replicates)  
269 from each collection site, ii) to control for variations (“noise”) among sampling units (collections  
270 sites) due to local habitats, and iii) to generalise our results to Atlantic *Mytilus* populations  
271 beyond the study sample (Bolker, 2015; Zuur, Ieno, Walker, Saveliev, & Smith, 2009).

272 We carried out data exploration following the protocol of Zuur et al. (2010). Initial inspection  
273 revealed no outliers. Pairwise scatterplots and variance inflation factors (VIFs) were calculated  
274 to check for collinearity between input variables. VIF values < 2 indicated an acceptable degree  
275 of correlation among covariates to be included within the same model. We applied residual  
276 regression to uncouple the unique from the shared contribution of temperature and Chl-*a*

277 concentration to the response (Graham, 2003). This allowed us to account for the existing causal  
278 link between these two parameters and to avoid inferential problems from modelling non-  
279 independent covariates without losing explanatory power (Graham, 2003). To directly compare  
280 model estimates (effect size metrics) from predictors on different measurement scales, to  
281 estimate biologically meaningful intercepts, and to interpret main effects when interactions are  
282 present, we standardised all the input variables (environmental parameters and shell length)  
283 (Grueber, Nakagawa, Laws, & Jamieson, 2011; Schielzeth, 2010). For standardisation, we  
284 subtracted the sample mean from the variable values and divided them by the sample standard  
285 deviation [ $z_i = (x_i - \bar{x})/\sigma_x$ ] (Schielzeth, 2010).

286 We used separate GLMMs to explore patterns of whole-shell and periostracum thickness with  
287 latitude, environmental conditions, and shell length (size) ( $n = 424$ ). A different approach was  
288 used to investigate relationships between calcareous layers and latitudinal gradients. Prismatic  
289 and nacreous layer thickness were analysed within the same GLMM ( $n = 424 \times 2$  layers) to i)  
290 estimate their variation and covariation across latitudes, ii) predict simultaneously common and  
291 divergent environmental effects on both layers, and iii) reduce the probability of type I error. To  
292 model the relationship between layer thickness and latitude we used a GLMM with a normal  
293 distribution with latitude (continuous), shell layer (categorical, two levels: prismatic and  
294 nacreous) and their interaction as fixed covariates. To model shell thickness as a function of the  
295 environmental predictors we used a GLMM with a normal distribution (Equation 1). In the initial  
296 model, fixed continuous covariates were standardised temperature, salinity and Chl-*a* in addition  
297 to shell layer (categorical, two levels) and their two-way interactions. Shell length (continuous)  
298 was included in both models to control for possible effects of within-population size variation on  
299 layer thickness. To incorporate the dependency among observations for a specific layer from the

300 same collection site, we used site as a random intercept. Preliminary inspection of models'  
301 residuals showed heteroscedasticity in most models. The use of different continuous probability  
302 distributions (i.e. gamma and inverse Gaussian) and link functions did not stabilise the variance,  
303 therefore a ln-transformation of the response was required. Response variables did not require  
304 further transformations.

305 The proportion (wt%) of organic matrix in the prismatic layer ( $n = 80$ ) was modelled with GLMs  
306 as a function of collection site (categorical, four levels) and prismatic thickness (continuous) to  
307 test for differences between polar and temperate regions and association with shell thickness.  
308 The response variable was coded as a value from 0 to 1; therefore, we used a GLM with a beta  
309 distribution and a logistic link function. Pair-wise contrasts with a standard Bonferroni correction  
310 (alpha of 0.0125) were then used to test for differences in wt% among sites within and between  
311 climatic regions.

312 Models were optimised by first selecting the random structure and then the optimal fixed  
313 component. The principal tools for model comparison were the corrected Akaike Information  
314 Criterion (AICc) and bootstrapped likelihood ratio tests. Random terms were selected on prior  
315 knowledge of the dependency structure of the dataset. Visual inspection of residual patterns  
316 indicated violation of homogeneity in most cases. This required the use of variance structures  
317 (generalised least squares) allowing the residual spread to vary with respect to shell layer. The  
318 fixed component was optimised by rejecting only non-significant interaction terms that  
319 minimised the AICc value. For all model comparisons, variation of AICc between the optimal  
320 (lowest AICc value) and competing models were greater than 8, and fixed-effect estimates were  
321 nearly identical, indicating that competing models were very unlikely to be superior (Burnham &  
322 Anderson, 2002). The proportion of variance explained by the models was quantified with

323 conditional or pseudo determination coefficients ( $cR^2$  or  $\text{pseudo}R^2$ ) (Nakagawa, Johnson, &  
324 Schielzeth, 2017). We used variograms to assess the absence of spatial autocorrelation. Final  
325 models were validated by inspection of standardised residual patterns to verify GLMM  
326 assumptions of normality, homogeneity and independence. We used optimal models fitted on  
327 standardised input variables (same measurement scale) to estimate the mean effect sizes of  
328 environmental drivers on the response (Schielzeth, 2010). Ninety-five per cent confidence  
329 intervals (95% CIs) for the regression parameters were generated using bias-corrected parametric  
330 bootstrap methods (10,000 iterations). 95% CIs were used for statistical inference due to  
331 estimation of approximated significance values ( $p$ -value) in mixed-modelling. If the confidence  
332 intervals did not overlap zero, then the effect was considered significant. Conditional modes and  
333 variances of the random effect were calculated for each GLMM to inspect differences in  
334 collection site-level effect on the variation of individual layer thickness after accounting for the  
335 effect of environmental covariates and shell size (fixed component). All data exploration and  
336 statistical modelling were performed in R (v3.4.1) (for packages see Table S3).

337 A principal component analysis (PCA), with a singular value decomposition method, was  
338 performed on shell traits (i.e. thickness of prismatic layer, nacreous layer, and periostracum) to  
339 observe variations in shell composition among individuals from different climatic regions. The  
340 PCA was used to create new independent variables, the principal components (PCs), resulting  
341 from the linear combinations of shell traits (Table S4), and to observe how these changed  
342 together among populations.

343



## 344 **RESULTS**

### 345 **Latitudinal patterns of shell deposition**

346 GLMMs indicated a general decrease of *Mytilus* whole-shell thickness with increasing latitude  
347 (95% CI = -0.36 to -0.01,  $cR^2 = 0.81$ ) (Fig. 2, S4). We detected a significant negative  
348 relationship between the prismatic and nacreous layers thickness and latitude (95% CI = -0.258  
349 to -0.068,  $cR^2 = 0.71$ ; Fig. 2, S4), while no variation in periostracum thickness (95% CI = -0.14  
350 to 0.07,  $cR^2 = 0.81$ ; Fig. 2) was detected. No significant change in the relative thickness of  
351 prismatic and nacreous layers was observed across the sampled latitudinal range (latitude  $\times$  layer  
352 interaction, 95% CI = -0.24 to 0.15; Table S5). Shell length was positively correlated with  
353 thickness in all layers indicating thickening during growth (Fig. 2; Table S5).

354 Prismatic layers were characterised by a significantly higher wt% of organic content (lower  
355 proportion of  $CaCO_3$ ) in mussel shells from polar than temperate regions, indicating decreased  
356 shell calcification at high latitudes (Fig. 3a, S3). Polar shells [sites 15, 16; mean (SD) = 1.8 wt%  
357 (0.31)] were characterised by an average of 29% more organic content compared to temperate  
358 mussels [sites 1, 11; mean (SD) = 1.4 wt% (0.16)]. The wt% of organics was negatively  
359 correlated with prismatic thickness (Fig. 3b), indicating a lower proportion of  $CaCO_3$  and  
360 thinner, less calcified, shells at polar latitudes.

361

### 362 **Environmental influence on shell production and composition**

363 We identified significant trends in shell thickness with environmental gradients depending on the  
364 shell measurement considered (Fig. 2, S5; Table 1). Whole-shell thickness was positively related

365 to temperature, salinity and shell length, but there was no influence of Chl-*a* ( $cR^2 = 0.93$ ; Fig. 2).  
 366 Salinity had an effect on shell thickness that was 3.4 and 2.1 times larger than temperature and  
 367 length, respectively (Fig. 2; Table 1).

368 Prismatic and nacreous layer thicknesses were analysed within the same GLMM. After model  
 369 selection, fixed continuous covariates of the optimal model, Equation (1), were standardised  
 370 *temperature*, *salinity*, *Chl-a*, shell *length* in addition to shell *layer* (categorical, two levels:  
 371 prismatic and nacreous) and the *salinity*  $\times$  *layer*, *length*  $\times$  *layer* interactions. The random  
 372 component was collection *site* used as a random intercept. The model was of the form:

$$373 \ln(Thickness_{ijk}) \sim N(\mu_{ijk}; \sigma_j^2)$$

$$374 \mu_{ijk} = Temperature_{ik} + Salinity_{ik} + Chl-a_{ik} + Length_{ik} + Layer_j$$

$$375 + Salinity_{ik} \times Layer_j + Length_{ik} \times Layer_j + Site_{ij}$$

$$376 Site_{ij} \sim N(0; \sigma_{Site}^2)$$

377 (Equation 1)

378 where  $Thickness_{ijk}$  is the  $k$ th thickness observation from layer  $j$  ( $j = \text{prismatic, nacreous}$ ) and site  $i$   
 379 ( $i = 1, \dots, 17$ ).  $Site_{ij}$  is the random intercept for layer  $j$ , which is assumed to be normally  
 380 distributed with expectation 0 and variance  $\sigma_{Site}^2$ .

381 Sea surface temperature, salinity and shell length all successfully predicted ( $cR^2 = 0.93$ )  
 382 variations in the thickness of prismatic and nacreous layers, while no influence of Chl-*a* on either  
 383 layer was detected (Table 1). The mean effect size of salinity on the response was twice as large  
 384 as the effect of shell length, while it was 2.9 and 4.7 times larger than the effect of temperature  
 385 on the prismatic and nacreous layers, respectively (Equation 2; Fig. 2). This indicates salinity

386 had a stronger contribution to predicting shell structure than the effects of temperature, Chl-*a*,  
387 and shell length combined (Fig. 4).

388  $\mu_{ijk} =$

389 
$$\begin{cases} 5.907 + 0.138 \times \text{Temperature} + 0.396 \times \text{Salinity} + 0.028 \times \text{Chl-}a + 0.197 \times \text{Length} & \text{Prismatic} \\ 5.853 + 0.138 \times \text{Temperature} + 0.654 \times \text{Salinity} + 0.028 \times \text{Chl-}a + 0.308 \times \text{Length} & \text{Nacreous} \end{cases}$$

390 (Equation 2)

391 Interactions between shell layer and both salinity and shell length (Equation 2) indicate  
392 deposition of proportionally thicker prismatic layers (higher proportion of calcite) under low  
393 salinities and proportionally thicker nacreous layers (higher proportion of aragonite) under  
394 higher salinities across the entire range of shell lengths (Fig. 4). No change in the relative  
395 thickness of prismatic and nacreous layers with water temperature was detected (Table S6).

396

### 397 **Periostracum variability**

398 Models of periostracum thickness revealed significant exponential relationships with Chl-*a* and  
399 shell length ( $cR^2 = 0.81$ ) (Table 1). Length had a mean effect that was three times larger than  
400 Chl-*a* (Fig. 2), showing a rapid thickening of the periostracum during shell growth. The  
401 interactions between shell length and both salinity and temperature indicate that the effects of  
402 these variables on the periostracum were interdependent. At low salinities, the higher values of  
403 shell length had a greater positive effect on periostracum thickness, while the reverse was true  
404 for higher temperatures which had a marginal effect only on thickening rates (Fig. 5a, S6). This  
405 suggests that periostracum thickening during shell growth was faster in fresher waters than in  
406 relatively saltier conditions.

407

### 408 **Among-site shell variation**

409 GLMMs showed no difference in collection site-level effects (conditional modes) on each  
410 thickness measurement (Fig. 5b). Conditional modes indicated that environmental factors and  
411 shell size accounted for most of the among-site shell variations. This suggested no residual effect  
412 of species identity or hybridisation (or other potentially influential factors) on the thickness of  
413 individual shell layers at different sites after accounting for the effects of environmental  
414 conditions and shell size.

415 A PCA on shell traits indicated marked differences in shell composition among sites from  
416 different climatic regions (Fig. S7). PC1 captured most of the shell variation among individual  
417 (74.7%) indicating differences in shell composition due to the wide range of size classes (shell  
418 length) available. PC2 (16.9%) indicating formation of shells with thicker periostracum in low-  
419 salinity environments (polar and Baltic region). PC3 (8.36%) captured heterogeneous within-  
420 region variations in prismatic and nacreous layers deposition, supporting no change in the  
421 relative deposition of calcareous shell components with latitude.

422

## 423 **DISCUSSION**

424 Our results demonstrate that plasticity in shell biomineralisation in *Mytilus* species shapes  
425 regional differences in shell production and composition as a response to the spatial structure of  
426 environmental conditions. An understanding of the biological processes driving differences in  
427 responses of species among regions to multiple interacting stressors is crucial for improving

428 predictive accuracy and informing more realistic projections of species and ecosystem resilience  
429 to climate change (Urban et al., 2016). Heterogeneous population-level responses from different  
430 climates act as a natural laboratory for investigating potential effects of future change. These  
431 differing responses suggest salinity is the best predictor of within-region variations in *Mytilus*  
432 shell production, mineral (prismatic and nacreous layers) and organic (periostracum)  
433 composition during growth. Spatial variations and trade-offs in shell biomineralisation suggest  
434 geographic differences in chemical and mechanical protection, shaping spatial patterns of  
435 resistance of these foundation species to global environmental changes.

436 Decreasing shell calcification (increasing organic content and thinner shells) towards high  
437 latitudes (Fig. 2, 3) supports documented patterns of skeletal production and estimated costs  
438 (Watson et al., 2017, 2012). Two explanatory paradigms exist for decreased skeletal size at  
439 higher latitudes: i) increased calcification costs due to poleward decrease in  $\Omega_{\text{CaCO}_3}$  and reduced  
440 ectotherms metabolic rate (Watson et al., 2017, 2012) and ii) reduced predation pressure of  
441 durophagous (shell crushing) and drilling predators (e.g. crabs, dog whelks and seabirds)  
442 (Aronson et al., 2007; Harper & Peck, 2016). Given the higher production cost of shell organics  
443 than  $\text{CaCO}_3$  deposition (Palmer, 1992; Sanders et al., 2018; Watson et al., 2017) and problematic  
444 protein production at polar temperatures (Peck, 2016, 2018), we might expect a reduced  
445 proportion of organic matrix. Moreover, decreasing predation pressure should result in thinner  
446 shells (Freeman, 2007; Sherker, Ellrich, & Scrosati, 2017) of the same composition irrespective  
447 of geographic area. However, the wt% of organic matrix was higher at Arctic latitudes. This  
448 could suggest either a marked increase in the cost of calcification in polar regions (Watson et al.,  
449 2017), altering significantly the relative costs of organics and  $\text{CaCO}_3$  production (Sanders et al.,  
450 2018), or seawater  $\Omega_{\text{CaCO}_3}$  below one ( $\Omega \leq 1$ ) due to low temperatures and salinity

451 thermodynamically favouring net dissolution of CaCO<sub>3</sub> structures (Ries et al., 2016; Thomsen et  
452 al., 2018). In either case, these effects would result in decreased shell calcification at high  
453 latitudes. Increased proportions of insoluble organic matrix, which protects the calcified shell  
454 components from dissolution (Harper, 2000), and deposition of thinner shells suggest a trade-off  
455 between potential resilience to dissolving conditions and increased vulnerability to predators.  
456 This may have adaptive beneficial effects on mussels in more corrosive, polar and subpolar  
457 waters where predation pressure is low.

458 For over 60 years, temperature and shell size have been considered primary drivers of biogenic  
459 CaCO<sub>3</sub> mineralogy across latitudes, dictating the formation of predominantly aragonitic  
460 structures in temperate regions and increased calcite precipitation in cold climates (Carter &  
461 Seed, 1998; Lowenstam, 1954; Ramajo, Rodriguez-Navarro, Duarte, Lardies, & Lagos, 2015).  
462 Although our study partly corroborates previous findings, we observed no significant change in  
463 the relative deposition of calcite (prismatic layer) and aragonite (nacreous layer) with latitude or  
464 temperature. But we demonstrate that salinity had the strongest effect on shell production and  
465 composition in Atlantic *Mytilus* (Fig. 2), supporting the strong influence of salinity on water  
466 carbonate chemistry ( $\Omega_{\text{CaCO}_3}$ ) and calcification costs (Sanders et al., 2018; Thomsen, et al., 2015;  
467 Thomsen et al., 2018).

468 The interaction between shell layer, salinity and shell size (Equation 2) indicates changes in shell  
469 production (quantity) and composition (quality) in *Mytilus* spp. across different salinities (Fig.  
470 4). Shifts in shell structure from juveniles to large adults lead to the formation of thinner,  
471 prismatic-dominated shells in brackish waters and thicker, nacre-dominated structures under  
472 marine conditions (Fig. 4b,c). Observed variations in predominant shell mineralogy with salinity  
473 regime, suggest changes most likely driven by altered seawater carbonate saturation state. Low

474 temperatures and salinities in polar and subpolar regions, relative to temperate areas, would lead  
475 to lower  $\Omega_{\text{CaCO}_3}$  and favour net dissolution of the less stable of the two main forms of  $\text{CaCO}_3$ ,  
476 the aragonite (Mucci, 1983; Ries et al., 2016; Thomsen, et al., 2015), with formation of thinner,  
477 calcite-dominated shells. Conversely, in temperate Atlantic regions, warmer and saltier waters  
478 (higher  $\Omega_{\text{CaCO}_3}$ ) are less likely to constrain  $\text{CaCO}_3$  production and deposition of thicker shells.  
479 Very low salinities in the Baltic Sea, compared to the mean oceanic salinity, correlate with  
480 limiting concentrations of calcification substrates (Thomsen et al., 2018) and lead to extended  
481 period of aragonite undersaturation ( $\Omega_{\text{arag}} < 1$ ), imposing kinetic constraints on calcification  
482 (Tyrrell, Schneider, Charalampopoulou, & Riebesell, 2008). This will likely increase energetic  
483 costs of calcification (Sanders et al., 2018; Thomsen et al., 2018) and favour net dissolution of  
484 aragonite over calcite structures with formation of thinner shells characterised by higher  
485 proportions of calcite (Melzner et al., 2011). No difference in site-level effects on individual  
486 layers was found, suggesting modelled shell variations are independent of species identity and  
487 hybridisation (Fig. 5b). This supports the relatively smaller contribution of genetic status than  
488 environmental heterogeneity on the variability of *Mytilus* shell traits across large-geographic  
489 scales suggested by Krapivka et al. (2007) and Telesca et al. (2018). Observed response patterns  
490 suggest a strong potential for qualitative and quantitative shell adjustments in blue mussels to  
491 produce the most appropriate shell structure for specific environmental conditions.

492 Under current scenarios, plasticity in shell biomineralisation could represent an advantageous  
493 compensatory mechanism for *Mytilus* species when facing different water chemistries and  
494 predation levels. In fact, at high-latitudes and in the Baltic region, where durophagous predators  
495 are rare or absent (Aronson et al., 2007; Harper & Peck, 2016; Kautsky, Johannesson, &  
496 Tedengren, 1990; Reimer & Harms-Ringdahl, 2001) and the water is more likely to constrain

497 CaCO<sub>3</sub> deposition ( $\Omega_{\text{CaCO}_3} \leq 1$ ) (Watson et al., 2017), mussels are characterised by thinner,  
498 prismatic-dominated (calcitic) shells enriched in organic matrix, providing a generally higher  
499 protection from dissolution (Harper, 2000; Mucci, 1983). Conversely, at mid-latitudes, where  
500 durophagous predators are more abundant (Harper & Peck, 2016; MacArthur, 1972) and  
501  $\Omega_{\text{CaCO}_3}$  is generally higher (Watson et al., 2017), mussels display thicker, nacre-dominated  
502 (aragonitic) shells suggesting higher mechanical resistance to predation (Barthelat et al., 2009;  
503 Lowen, Innes, & Thompson, 2013; Sherker et al., 2017).

504 Despite projected global changes in salinity gradients (Durack et al., 2012), *Mytilus* species show  
505 a strong capacity for compensatory responses in shell production to mitigate the emergent  
506 negative effects of changing water chemistry. In fact, the interacting effects of salinity, shell  
507 length, and a minor influence of temperature on the periostracum (Fig. 5a, S6), which represents  
508 a strong chemical barrier to shell dissolution in molluscs (Harper, 1997; Peck, Tarling, Manno,  
509 Harper, & Tynan, 2016; Tunnicliffe et al., 2009), indicated deposition of thicker periostraca  
510 under decreasing salinities. This likely increases the durability of periostracum to environmental  
511 abrasion during aging and better mediates impacts of ocean acidification.

512 Although populations in high-latitude ecosystems will experience globally the most rapid  
513 acidification (Gattuso et al., 2015), decreasing salinity (lower  $\Omega_{\text{CaCO}_3}$ ) predicts deposition of  
514 thinner shells with an increased proportion of organic-enriched, prismatic layers and thicker  
515 periostraca, potentially increasing shell resistance to future more corrosive conditions.

516 Conversely, in temperate areas, increasing salinity (higher  $\Omega_{\text{CaCO}_3}$ ) predicts deposition of thicker  
517 shells with relatively thicker nacreous layers, favouring mechanical protection from higher  
518 predation pressure in warmer climates (Freeman, 2007; Harper & Peck, 2016; Lowen et al.,



519 2013; Sherker et al., 2017). However, forecasted changes in the thickness of periostracum with  
520 salinity depend on shell size and would be more evident in larger or faster growing individuals  
521 (length > 48 mm) (Fig. 5a).

522 In Greenland, where the rate of melting of the ice sheet has doubled in the last decade (Kjeldsen  
523 et al., 2015), lower salinities during summer (< 20 psu) (Sejr et al., 2017), decreasing  $\Omega_{\text{CaCO}_3}$  and  
524 increasing primary productivity (food supply) in coastal areas (Meire et al., 2017), predict  
525 formation of thicker periostraca and proportionally thicker organic-enriched calcitic layers.  
526 These shell adjustments in Arctic *Mytilus* spp. could represent compensatory responses for a  
527 potentially increased resilience to future water conditions favouring shell dissolution at the price  
528 of decreased protection from predators. In contrast, in the Baltic Sea, the projected decrease in  
529 salinity (up to 45% reduction in the north-eastern and central Baltic) (Gräwe, Friedland, &  
530 Burchard, 2013), combined with the considerable physiological osmotic stress (salinity from 22  
531 psu to 3 psu), would be particularly critical for mussels inhabiting already unfavourable  
532 conditions for calcification (i.e. limiting  $[\text{Ca}^{2+}]$  and aragonite undersaturated seawater) (Sanders  
533 et al., 2018; Thomsen et al., 2018). Moreover, the reduced shell size of Baltic *Mytilus* does not  
534 predict formation of thicker, durable periostraca, which could further increase vulnerability to  
535 dissolution. Impacts of changing salinity on *Mytilus*, which contributes up to 90% of the Baltic  
536 benthic invertebrate biomass (Kautsky et al., 1990), could have large-scale implications for  
537 coastal communities in the near future (Johannesson, Smolarz, Grahn, & André, 2011).

538 *Mytilus* species have a marked shell plasticity and thick periostracum compared to other  
539 calcifiers that often compete with it for space (e.g. barnacles and spirorbid polychaetes).  
540 Biomineralisation plasticity may act as a mechanism conferring *Mytilus* species a protective  
541 capacity for quantitative and qualitative trade-offs in shell deposition to produce the most

542 appropriate shell structure for a specific set of abiotic (i.e. CaCO<sub>3</sub> water chemistry and sources)  
543 and biotic (i.e. predation pressure) conditions. This potential mechanism could represent a major  
544 factor for keystone calcifiers, not only molluscs, to maintain their ecological role and functions  
545 in rapidly changing oceans. Moreover, the periostracum provides a strong defence against shell  
546 dissolution and allows mytilids to survive in oligohaline waters (~5 psu) and extremely acidified  
547 conditions (e.g. hydrothermal vents) (Harper, 1997; Tunnicliffe et al., 2009). These factors may  
548 shift the ecological balance and community structure in favour of species with a greater response  
549 potential and stronger resistance to corrosive conditions, such as mussels, when ocean waters  
550 become fresher and more acidic in future decades.

551 As hypothesised, plasticity in shell biomineralisation shapes regional differences in *Mytilus* shell  
552 responses to interacting environmental conditions and drives spatial variations of chemical and  
553 mechanical shell protection, dictating geographic patterns of Atlantic *Mytilus* sensitivity to future  
554 environmental change. Overall, mussel shell calcification decreased towards high latitudes, with  
555 salinity being the best predictor of within-region variations in shell production, mineral and  
556 organic composition. Quantitative and qualitative differences in shell deposition among regions  
557 indicate compensatory trade-offs in shell components suggesting the potential for a higher  
558 resistance against dissolution for mussels in polar, low-salinity environments, and an enhanced  
559 mechanical protection from predators in temperate, higher-salinity regions. The strong response  
560 potential of blue mussel shell periostracum suggests a potentially increased resilience to ocean  
561 acidification in polar and sub-polar *Mytilus*, and a higher sensitivity of Baltic populations under  
562 future environmental conditions.

563 Our findings indicates that a better understanding of key biological processes mediating species'  
564 response to habitat alterations will be essential for identifying vulnerability and informing

565 conservation practices, especially for species having both high climate sensitivity and key  
566 ecological roles in shaping marine communities. This knowledge underpins our ability to predict  
567 accurately and reduce the damaging effect of climate change on future biodiversity under any  
568 range of scenarios (Urban et al., 2016). Our study has important implications because it explores  
569 the links between i) the mechanisms of biological variation, as biomineralisation plasticity, ii)  
570 species' responses to the spatial co-occurrence of multiple environmental drivers, and iii)  
571 potential regional differences in resilience of calcifying species to habitat change. This  
572 understanding is of critical importance for making realistic projections of emergent ecological  
573 effects of global environmental changes, such as altered salinity regimes, and to improve our  
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576

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589

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800

801 **TABLES**802 **Table 1. Environmental GLMMs summary.**

803 Estimated statistics and bootstrapped 95% CIs for regression parameters are reported for the  
 804 modelled relationships between thickness of the various shell layers and whole-shell against  
 805 standardised covariates. For the summary of model in Equation (1), estimates for group means,  
 806 slopes, and standard errors are reported separately for the prismatic and nacreous layers (Table  
 807 S6). (Parameters' significance is determined when the 95% CI does not include zero).

	Estimate	SE	95% CI	<i>t</i> -value	<i>p</i> -value (approximate)
<b>Whole-shell*</b>					
(Intercept)	6.617	0.051	<b>6.517; 6.717</b>	128.71	< <b>0.0001</b>
Temperature	0.156	0.054	<b>0.014; 0.240</b>	2.89	<b>0.013</b>
Salinity	0.525	0.060	<b>0.411; 0.672</b>	8.69	< <b>0.0001</b>
Chl- <i>a</i>	0.074	0.054	-0.042; 0.216	1.37	0.20
Length	0.248	0.037	<b>0.181; 0.327</b>	6.44	< <b>0.0001</b>
<b>Prismatic (Pr) &amp; nacreous (Na)†</b>					
(Intercept)Layer(Pr)	5.907	0.031	<b>5.775; 6.038</b>	188.31	< <b>0.0001</b>
(Intercept)Layer(Na)	5.853	0.083	<b>5.715; 5.990</b>	70.81	< <b>0.0001</b>
Temperature	0.138	0.033	<b>0.016; 0.263</b>	4.17	<b>0.0008</b>
Chl- <i>a</i>	0.028	0.033	-0.084; 0.141	0.86	0.40
Salinity × Layer(Pr)	0.396	0.039	<b>0.262; 0.529</b>	10.22	< <b>0.0001</b>
Salinity × Layer(Na)	0.654	0.093	<b>0.501; 0.811</b>	7.07	< <b>0.0001</b>
Length × Layer(Pr)	0.197	0.031	<b>0.094; 0.295</b>	6.39	< <b>0.0001</b>
Length × Layer(Na)	0.308	0.065	<b>0.196; 0.419</b>	4.74	< <b>0.0001</b>
<b>Periostracum‡</b>					
(Intercept)	3.500	0.048	<b>3.406; 3.596</b>	71.03	< <b>0.0001</b>
Temperature	0.049	0.043	-0.036; 0.134	1.12	0.28
Salinity	-0.009	0.061	-0.131; 0.111	-0.14	0.89

Chl- <i>a</i>	0.147	0.038	<b>0.071; 0.221</b>	3.88	<b>0.0020</b>
Length	0.439	0.041	<b>0.357; 0.522</b>	10.25	<b>&lt;0.0001</b>
Temperature × Length	-0.064	0.035	-0.135; 0.006	-1.77	0.082
Salinity × Length	-0.151	0.061	<b>-0.271; -0.029</b>	-2.38	<b>0.020</b>

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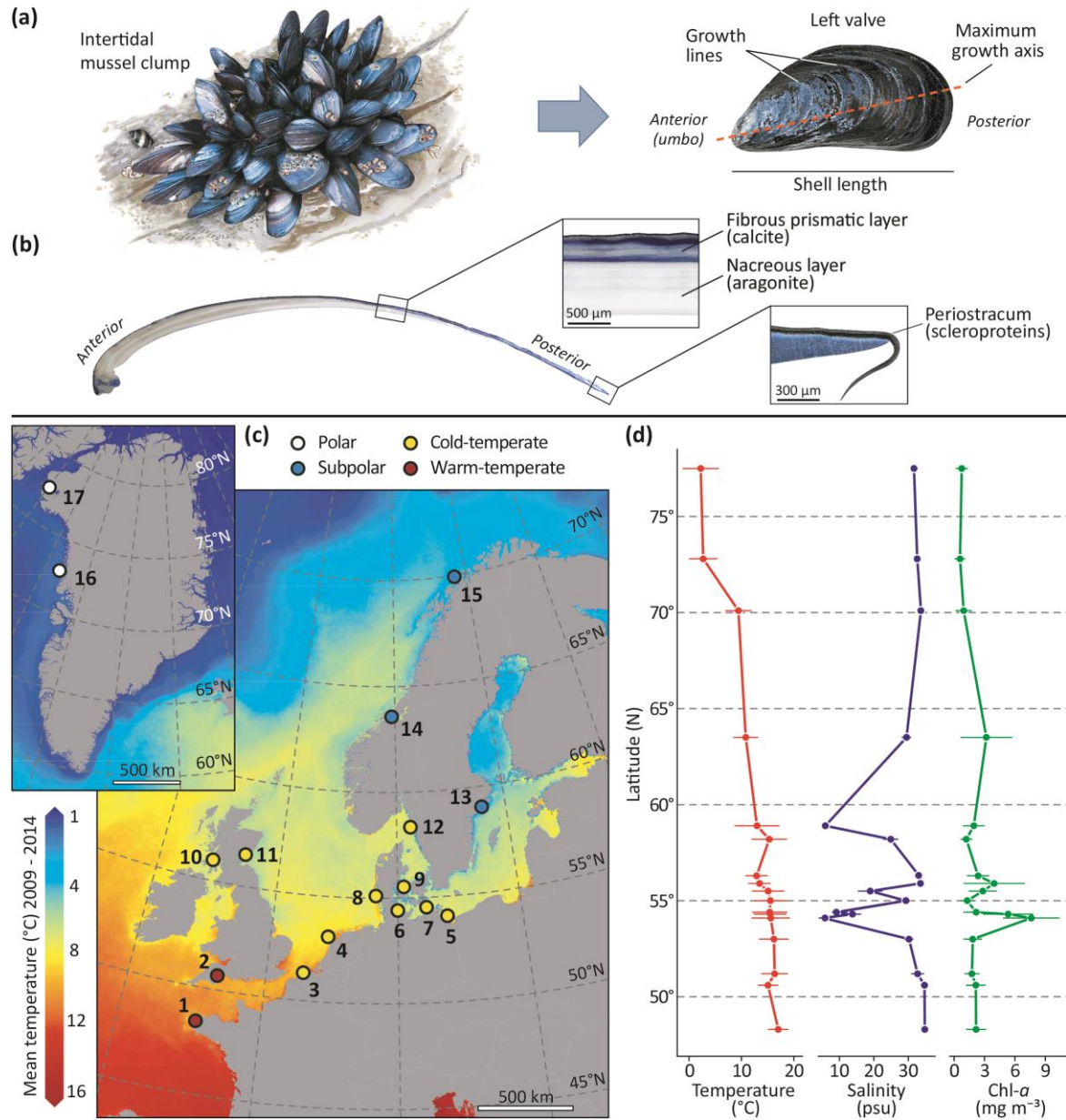
808 \* Whole-shell, the random intercept was normally distributed with mean of 0 and variance  
809 0.209<sup>2</sup>.

810 † Prismatic and nacreous layers, the random intercepts were normally distributed with mean 0,  
811 and variances 0.123<sup>2</sup> and 0.310<sup>2</sup>, respectively.

812 ‡ Periostracum, the random intercept was normally distributed with mean 0 and variance 0.130<sup>2</sup>.

813





815

816 **Figure 1.** *Mytilus* spp. shell, collection sites and environmental heterogeneity. **(a)** *Mytilus* shell

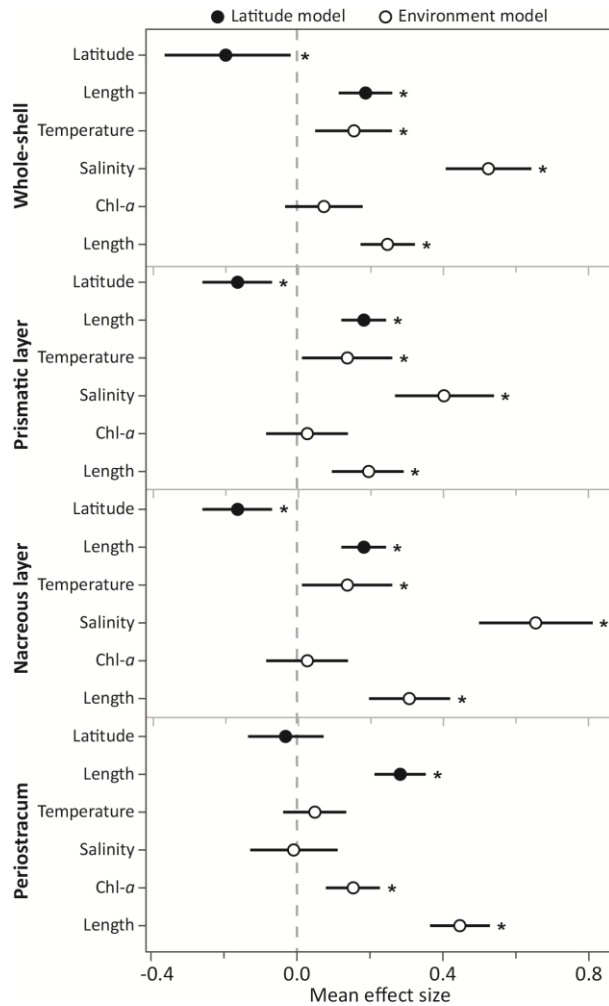
817 valve morphology and dimensions. **(b)** Anteroposterior cross-section of shell valve along the

818 axis of maximum growth (from umbo to posterior commissure, dashed line) showing internal

819 structure and composition of individual mineral (prismatic and nacreous) and organic

820 (periostracum) shell layers. **(c)** Thermal map of North-East Atlantic and Arctic surface waters

821 from the CMEMS (<http://marine.copernicus.eu/>) biogeochemical datasets showing locations at  
822 different climatic regions (open circles) where *Mytilus* specimens were collected from across the  
823 Eastern European and Greenlandic coastlines (from 48°N to 78°N): (1) Brest, France, (2)  
824 Exmouth, England, (3) Oostende, Belgium, (4) Texel, Netherlands, (5) Usedom, (6) Kiel, (7)  
825 Ahrenshoop, (8) Sylt, all Germany, (9) Kerteminde, Denmark, (10) Tarbet, Kintyre, Scotland,  
826 (11) St. Andrews, Scotland, (12) Kristineberg, Sweden, (13) Nynäshamn, Sweden, (14)  
827 Trondhiem, Norway, (15) Tromsø, Norway, (16) Upernavik, Greenland and (17) Qaanaaq,  
828 Greenland. Map created with ArcMap 10.5 (ArcGIS software by Esri, <http://esri.com>),  
829 background image courtesy of OpenStreetMap (<http://www.openstreetmap.org>). **(d)**  
830 Heterogeneous latitudinal gradients for sea surface temperature, salinity, and Chl-*a* concentration  
831 across the study regions. Mean values (May - October, filled circles) and SD (horizontal solid  
832 lines) for the 6-year period 2009 - 2014 were estimated from CMEMS datasets.

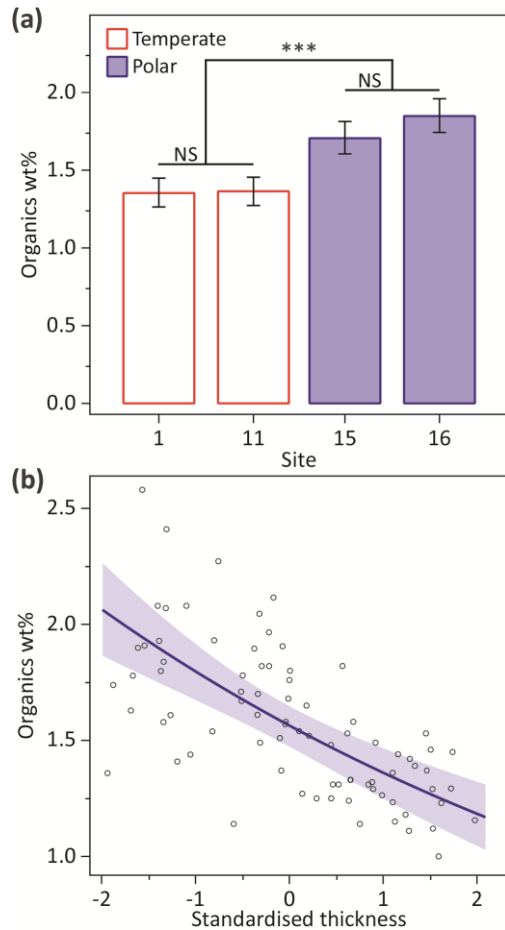


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834

**Figure 2.** Mean effect size of predictors on *Mytilus* shell measurements. Effect sizes were estimated from individual latitudinal (filled circles) and environmental (open circles) GLMMs. Mean effect sizes and direction of impacts of latitude, shell length, sea surface temperature, salinity, and Chl-*a* concentration on layer In-thickness ( $\mu\text{m}$ ) measurements are reported for the whole-shell, prismatic layer, nacreous layer, and periostracum. Significance of regression parameters is identified when the bootstrapped 95% CI (error bars) does not cross zero (\* denotes a significant difference from zero).

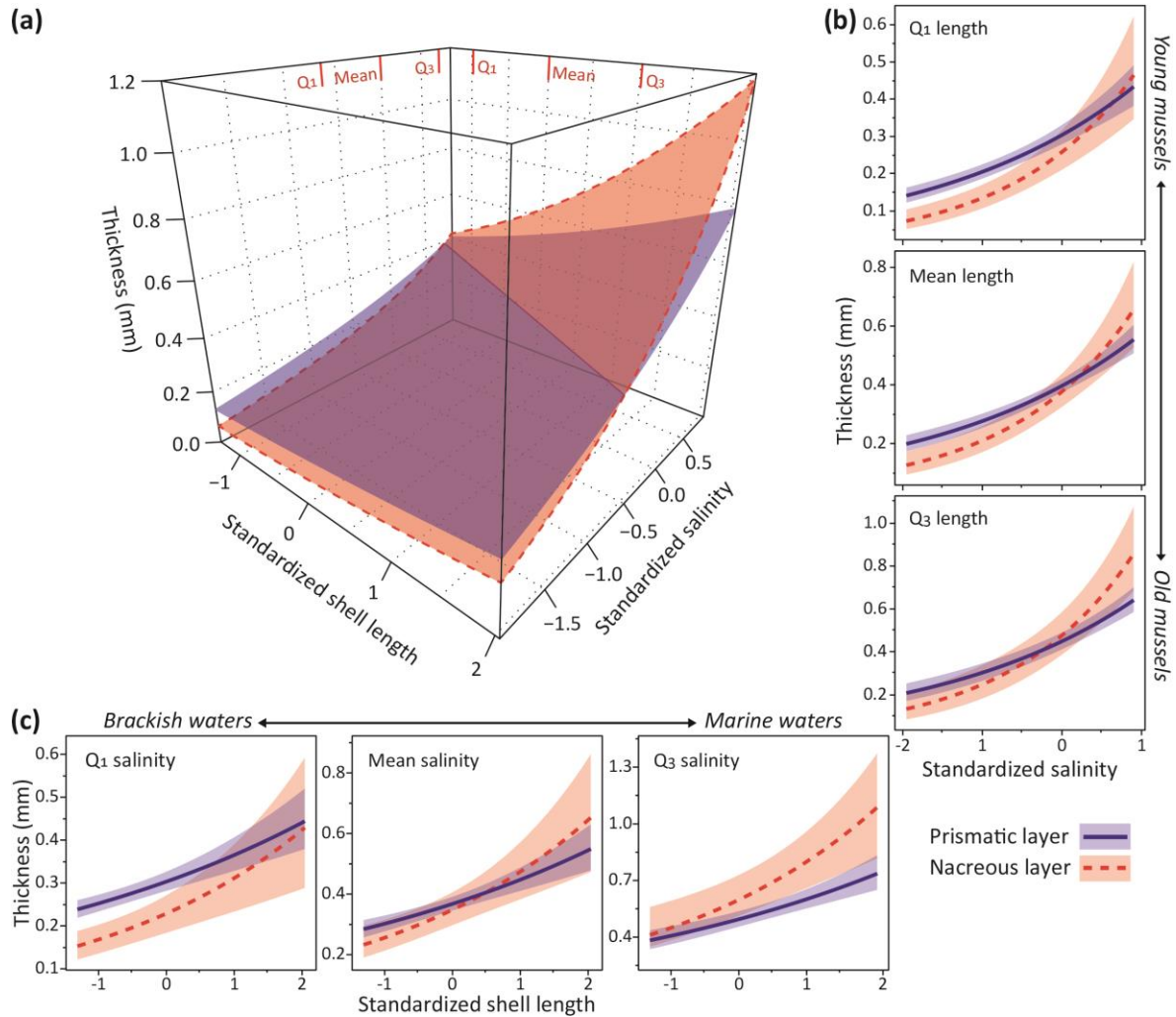
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842

843 **Figure 3.** Latitudinal patterns of shell organic content and calcification. **(a)** Variations in organic  
 844 content within prismatic layers among shells from temperate (sites 1, 11; open bars) and polar  
 845 (sites 15, 16; solid bars) climates. Pair-wise contrasts indicated significantly higher proportions  
 846 of organics in high-latitude than low-latitude specimens [mean difference = 0.44%;  $z = 8.27$ ,  $p <$   
 847  $0.0001$  (\*\*\*)],  $\text{pseudoR}^2 = 0.49$ ,  $n = 80$ ], in addition to non-significant differences (NS) among  
 848 temperate (mean difference = 0.002%;  $z = 0.12$ ,  $p = 0.91$ ) and polar (mean difference = 0.13%,  $z$   
 849  $= 1.86$ ,  $p = 0.063$ ) populations. Error bars indicate 95% CIs. **(b)** Relationship between the wt%  
 850 of organics and standardised thickness of the prismatic [mean (SD) = 529  $\mu\text{m}$  (174)] (sites 1, 7,  
 851 10 and 11), indicating a negative association between layer thickness and calcification level ( $z =$

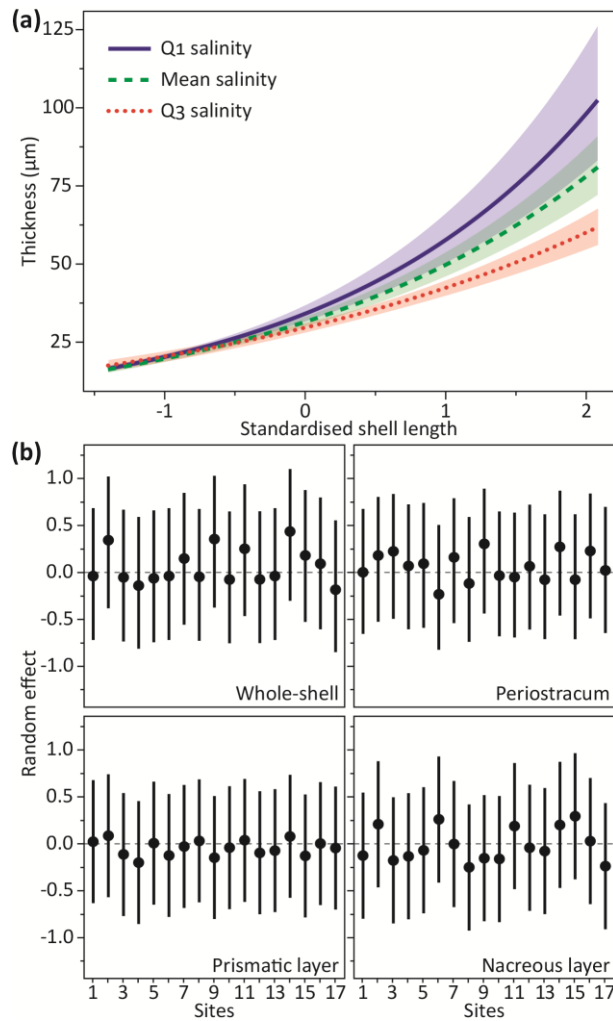
852  $-7.10, p < 0.0001, \text{pseudo}R^2 = 0.40$ ). Predicted values (solid line) and confidence intervals  
 853 (shaded area) were estimated for mussels of mean shell length (52 mm).



854

855 **Figure 4.** Environmental influence on shell production and composition. (a) Predicted multiple  
 856 relationships between the thickness of prismatic (solid margin plane) and nacreous (dashed  
 857 margin plane) layers, and standardised salinity [mean (SD) = 25.52 psu (10.29)], shell length  
 858 [mean (SD) = 47.42 mm (16.20)] and their interactions. (b) Shell thickness is modelled as a  
 859 function of salinity for the 1st quartile ( $Q_1 = 31.50$  mm), mean value (47.42 mm) and 3rd quartile  
 860 ( $Q_3 = 63.90$  mm) of the shell lengths sampled. For medium-sized mussels, we detected a

861 decreasing proportion of the prismatic layer (calcite) with increasing salinity and the deposition  
862 of relatively thicker nacreous layers (aragonite) at salinities  $> 27.67$  psu. (c) Thickness is  
863 modelled as a function of length for the 1st quartile ( $Q_1 = 18.92$  psu), mean value (25.52 psu)  
864 and 3rd quartile ( $Q_3 = 33.13$  psu) of salinity. At mean salinity, we detected an inversion of the  
865 relative layers' thickness for shell length  $> 55.30$  mm. Across the entire range of shell lengths,  
866 the model predicts formation of prismatic layer-dominated shells under low salinities and  
867 nacreous layer-dominated shells under higher salinities. Mean values (lines) and confidence  
868 intervals (shaded areas) are predicted while controlling for temperature ( $13.03$  °C) and Chl-*a*  
869 ( $2.48$  mg m<sup>-3</sup>).  
870



871

872 **Figure 5. (a)** Interacting effects of salinity and shell length on periostracum. Periostracum

873 thickness is modelled as a function of standardised shell length [mean (SD) = 47.42 mm (16.20)]

874 for the 1st quartile ( $Q_1 = 18.92$  psu, solid line), mean (25.52 psu, dashed line) and 3rd quartile

875 ( $Q_3 = 33.13$  psu, dotted line) of water salinity. Predicted values (lines) and confidence intervals

876 (shaded areas) indicate higher rates of exponential periostracal thickening with decreasing

877 salinity. Smaller individuals (shell length < 48.38 mm) were characterised by non-significant

878 thickness differences under different salinity regimes. **(b)** Among sites shell variation. GLMMs'

879 conditional modes (filled circles) and variances (solid lines) of the random effect estimated for

880 individual shell layers. Modes represent the difference between the average predicted response

881 (layer thickness) for a given set of fixed-effects values (mean environmental covariates and shell  
882 length) and the response predicted at a particular site. These suggest no detectable residual effect  
883 of species (*Mytilus edulis* or *M. trossulus*) and level of hybridisation on shell thickness among  
884 sites.

885