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Department of Arctic and Marine Biology

Bivalves as indicators of environmental perturbations related to climate and ocean acidification

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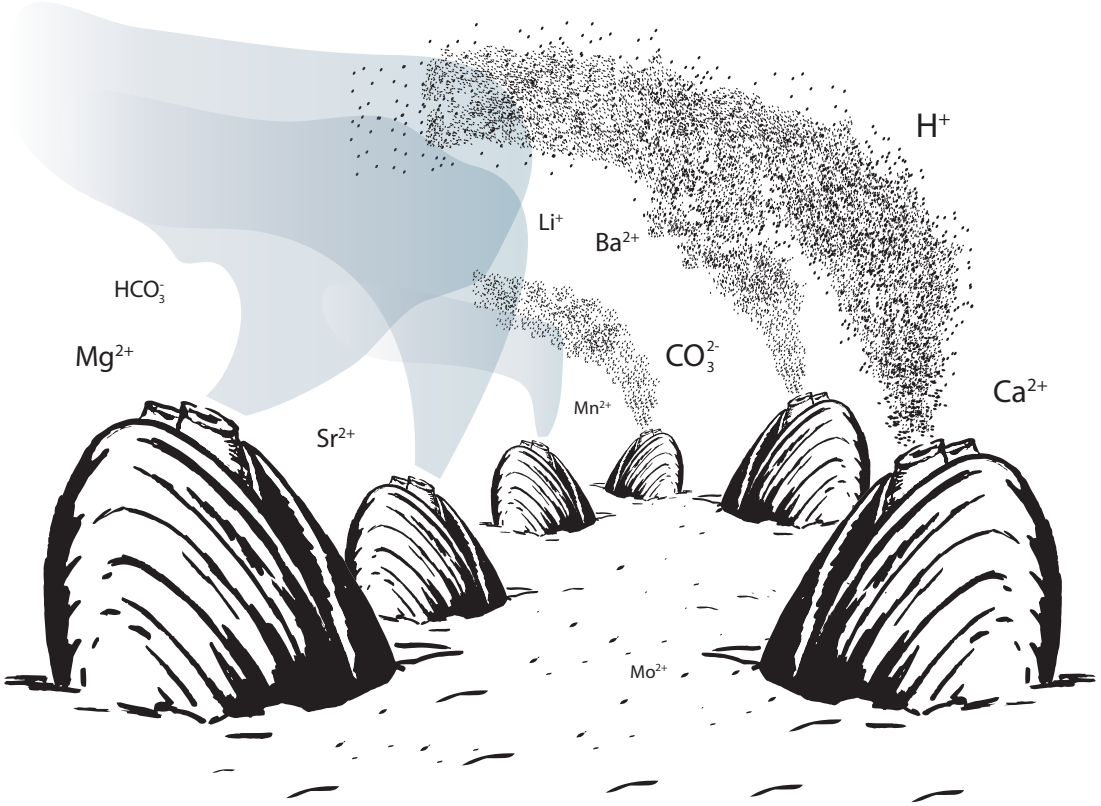
Mikko Vihtakari

A dissertation for the degree of Philosophiae Doctor – December 2014



UNIVERSITY OF TROMSØ

Bivalves in the cover drawing were kindly provided by Malin Daase

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UiT The Arctic University of Norway
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Norwegian Polar Institute
Research Department

Akvaplan-niva AS

ARCTOS Research Network

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Supervisors:

Prof. emer. Bjørn Gulliksen

Faculty of Biosciences, Fisheries and Economics
Department of Arctic and Marine Biology
UiT The Arctic University of Norway
N-9037, Tromsø, Norway

Dr. Haakon Hop

The Norwegian Polar Institute
Fram Centre
N-9296 Tromsø, Norway

Prof. Paul Renaud

Akvaplan-niva AS
Fram Centre
N-9296 Tromsø, Norway

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E-mail: mikko.vihtakari@gmail.com

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Facing what consumes you
is the only way to be free.
Released from those poisonous fears.
Resurrected once and for all.

— Hatebreed (2003)

CONTENTS

SUMMARY

Ocean acidification (OA) together with other anthropogenic perturbations is projected to cause dramatic changes in marine ecosystems over the coming centuries. Calcifying invertebrates, such as marine bivalve mollusks, are threatened by these perturbations. Many filter-feeding bivalves are long-lived organisms, which record geochemical information within their shells. This information can sometimes be used as a proxy to interpret the ambient environment experienced by the bivalves. The aims of this thesis were to increase knowledge on 1) susceptibility of bivalves to environmental perturbations and 2) usage of bivalve shells as sub-annual environmental proxy archives.

These objectives were addressed using two experimental settings: 1) an ocean acidification experiment on bivalve gametes and early larval stages, and 2) year-long bivalve deployments on oceanographic moorings in two fjords in Svalbard followed by geochemical sampling of bivalve shells. Sperm activity was used as an indicator trait in the acidification experiment, examining both the individual- and population-level responses to assess the adaptability and sensitivity of *Mytilus galloprovincialis* and *Macoma calcarea* to OA. Population-level effects of ocean acidification and warming on early larval development of *M. galloprovincialis* were further evaluated using a factorial experiment. The results were compared to the literature using meta-analyses.

Shells of *Serripes groenlandicus* and *Ciliatocardium ciliatum* were examined to assess their value as sub-annual environmental proxy archives. The shells were sampled for $\delta^{18}O$ using secondary ion mass-spectrometry (SIMS). Dynamic time warping (DTW) algorithms were adapted to align SIMS-determined $\delta^{18}O$ values with $\delta^{18}O$ values predicted from continuous mooring instrument recordings of seawater temperature and salinity, to estimate sub-annual growth patterns during the mooring deployment. Further sampling of element-to-calcium ratios was conducted using laser-ablation inductively-coupled-plasma mass-spectrometry (LA-ICP-MS). Models of sub-annual shell growth patterns permitted statistical comparisons of elemental ratios with oceanographic data recorded by the mooring instruments.

The results indicated that *Mytilus* might be sensitive to OA, but also that ocean warming might have a larger impact on the genus than OA scenarios projected for the year 2100: Larval size was negatively affected by moderately conservative OA scenarios for 2100. Sperm activity of *M. galloprovincialis* males was negatively affected by the high-end OA scenarios for 2100, possibly indicating a reduced fertilization success in low-density populations. The larval development experiment indicated that increasing temperature has a larger negative effect on larval performance than reduced pH. Nevertheless, other studies report

negative effects of OA for 2100 climate scenarios. These conflicting results are likely to reflect both experimental conditions and differences in population and species responses. Sperm from *M. galloprovincialis* demonstrated considerable among-individual variability in response to OA indicating that *Mytilus* populations might have the capacity to adapt to moderate reductions in ocean pH. Sperm activity of *Macoma calcarea* males was not significantly affected by the OA treatment on population-level, but the males also demonstrated considerable among-individual variability in sperm activity responses to acidification. These results combined with those from the literature suggest that among-individual variability in responses to environmental perturbations is likely a norm within populations. Consequently, natural selection might mitigate the negative effects of OA, but strong selection pressure can reduce the genetic variability and therefore affect the future adaptability of bivalve populations.

The results from the proxy development indicated that Li/Ca, Mg/Ca, Li/Mg, Mn/Ca, Sr/Ca, Mo/Ca or Ba/Ca cannot be used as straightforward proxies of temperature, salinity, phytoplankton biomass or growth rate in *S. groenlandicus* and *C. ciliatum* shells. Despite this, Li/Ca, Mg/Ca, and Ba/Ca demonstrated relatively consistent patterns across individuals from the same fjord indicating a synchronized environmental or physiological control for these element ratios. Shell growth of *S. groenlandicus* and *C. ciliatum* deployed on oceanographic moorings was estimated to occur from May to December in Kongsfjorden and from mid-June to November in Rijpfjorden. These growth seasons were followed by a slow growth period during which each bivalve deposited a prominent growth line. Shell growth rate of studied bivalves correlated with temperature, and the length of the growth season was likely determined by food availability. Consequently, the prominent winter growth lines in these two bivalve species can likely be used as indicators of time period when food-source was not sufficient to sustain shell growth.

In conclusion, the results of this thesis indicate that bivalves are sensitive to environmental perturbations, specifically to ocean acidification. This sensitivity, however, varies among species, populations, individuals, and life stages with larval stages and fertilization being most affected. Due to the variability and complex ecosystem interactions, it is too early to precisely project possible effects of OA on bivalve populations. A combination of different study approaches and repeated studies are needed to evaluate the potential response mechanisms of populations in the future oceans. Despite the high potential as sub-annual environmental proxies, interpretation of elemental ratios in bivalve shells is complicated by multiple internal and external factors. Understanding the seasonal dynamics of elements, especially Li, Ca, and Ba, in the ocean is required for further development of this potential. The insights into sub-annual growth of Arctic bivalves and the methods to align sample spots along chronologically deposited materials, as provided by this thesis, can be used in further proxy development studies.

LIST OF PAPERS

Paper I Vihtakari, M.; Havenhand, J.N.; Renaud, P.E.; Hendriks I.E. Variable individual- and species-level responses to ocean acidification. Under revision in *PLoS One*.

Paper II Vihtakari, M.; Hendriks I.E.; Holding J.; Renaud, P.E.; Duarte C.M.; Havenhand, J.N. Effects of ocean acidification and warming on sperm activity and early life stages of the Mediterranean mussel (*Mytilus galloprovincialis*). *Water*, **2013**, 5, 1890-1915.

Paper III Vihtakari, M.; Renaud, P.E.; Clarke, L.J.; Whitehouse, M.J.; Hop, H.; Carroll, M.L.; Ambrose Jr, W.G. Decoding the oxygen isotope signal for seasonal growth patterns in Arctic bivalves. Manuscript.

Paper IV Vihtakari, M.; Ambrose Jr, W.G.; Renaud, P.E.; Locke V, W.L; Berge, J.; Cottier, F.; Hop, H.; Carroll, M.L.; Clarke, L.J. Interpreting the seasonal environmental history recorded by Arctic bivalves. Manuscript.

1 INTRODUCTION

1.1 BIVALVES AS INDICATORS OF ENVIRONMENTAL PERTURBATIONS

"Just about everything civilization does has a mixed impact on the natural environment. Man enriches as well as degrades the environment. Very frequently it is a matter of scale."
— Odum *et al.* (1)

In 2014, the atmospheric carbon dioxide (CO_2) concentration reached a new milestone by temporarily exceeding 400 ppm (2) possibly for the first time since the Pliocene 2.6 million years ago (3–5). Further increases to 420 - 940 ppm are anticipated by the end of this century depending on political decisions affecting the rate of anthropogenic emissions and land use (6). Although CO_2 itself is a non-detrimental gas necessary to sustain life on Earth (7), the atmospheric CO_2 is among the main contributors to the Earth's heat regulation system (6) and the partial pressure of CO_2 (pCO_2) in atmosphere affects the bicarbonate balance of oceans (Box 1). The increase of atmospheric pCO_2 is therefore contributing to the processes known as global warming and ocean acidification (OA). Ocean acidification and global warming together with other human-induced perturbations are predicted to cause dramatic changes in the marine ecosystems over the coming centuries (8, 9, Papers I-II).

Based on our understanding from the geological record, the changes in atmospheric pCO_2 themselves are not unique in Earth's history (10, 11). The rate, however, at which CO_2 concentration in the atmosphere is projected to increase during the coming century (12) has not been observed previously in the geological record (3, 11, 13). This might be due to a lack of required temporal and analytical resolution in geological samples, demonstrating the need for development of high temporal-resolution proxies. Nevertheless, it is likely that already the present day atmospheric CO_2 concentrations lead to negative acidification effects in the most sensitive marine organisms and life-stages (14–16). Consequently, human activities subject the marine ecosystems to a grand-scale perturbation experiment (17–19) for which scientific hypotheses have begun to emerge only rather recently (7, 20–23). Understanding and anticipating the complex implications of this experiment requires information on the past climate (3, 24) as well as knowledge of the potential response mechanisms of sensitive marine species in connection with the projected climate scenarios (25). Such knowledge can be acquired through proxy records (3) and indicator species sensitive to the environmental perturbations (26, 27).

Box 1: Ocean acidification and climate projections

"The goal of working with scenarios is not to predict the future, but to better understand uncertainties in order to reach decisions that are robust under a wide range of possible futures."

— Moss *et al.* (28)

Approximately 30-50 % of the total CO_2 released into the atmosphere since the industrial revolution has been dissolved into the oceans (29, 30). Once dissolved in water, CO_2 forms a weak acid increasing the concentration of free hydrogen ions (H^+; Figure 1), consequently, leading to a decrease in ocean pH (pH = $-\log_{10}[H^+]$). The chemical changes in seawater associated with the CO_2 dissolution are called ocean acidification. Ocean acidification alters the saturation state of calcium carbonate ($CaCO_3$) minerals present in shells and exoskeletons of marine taxa (31). This increases the energy required to build and maintain the exoskeletons especially in species consisting of magnesium-calcite or aragonite minerals, which are more soluble than calcite $CaCO_3$ mineral (32, 33). Reduced pH also directly alters the chemical environment of marine organisms with possible detrimental effects on life stages of organisms with low homeostatic control mechanisms (Paper I). Therefore, negative effects of OA are often observed in experiments conducted on fertilization or early life stages of calcifying marine organisms, such as bivalves (27, 34; Papers I-II).

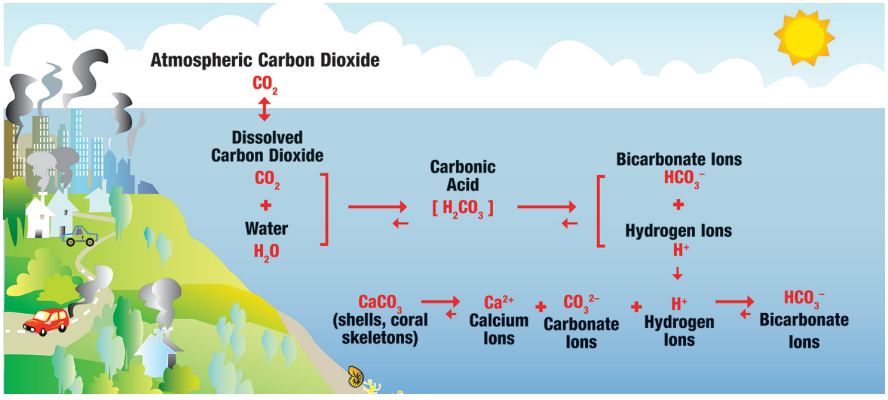

Figure 1: An overview of the most important ocean chemistry changes associated with dissolution of CO_2 into the oceans. Source: National Research Council (35).

Global average ocean surface pH has decreased by 0.1 units (= 30 % increase in H^+ concentration) since the industrial revolution (36). In the Fifth Assessment Report (AR5), the Intergovernmental Panel on Climate Change (IPCC) has used four model scenarios to describe plausible trajectories of climate conditions in the

future. These scenarios attempt to cover the full range of potential socio-economic pathways for this century (Box 1.1 in Cubasch *et al.* 6). The models are separated by radiative forcing (RF) characteristics associated with emission scenarios allowing a flexible intercomparison of models through Coupled Model Intercomparison Project Phase 5 (CMIP5; 28, 37). These scenarios, termed Representative Concentration Pathways (RCPs), are separated by two extreme pathways, RCP2.6 and RCP8.5, and complemented by two equally spaced middle scenarios, RCP4.5 and RCP6.0 (number after an RCP indicates the estimated RF in $W\,m^{-2}$ for 2100; 6). The IPCC models project average atmospheric pCO_2 for the 2100 year as 420, 538, 670, and 936 ppm from RCP2.6 to RCP8.5 (38). This translates as average open ocean pH reductions of 0.07, 0.15, 0.20, and 0.31 units, but the ocean pH will have large geographical variations, the reduction being largest in the Arctic due to sea-ice loss and higher CO_2 dissolution to cold water (Figure 2). During the same time period, global mean surface temperature is modeled to increase by 0.94, 1.68, 2.03, and 3.57 °C (50% quantiles) for RCP2.6, RCP4.5, RCP6.0, and RCP8.5, respectively. Also temperature change will be dependent on the geographical area, the Arctic being most affected (39).

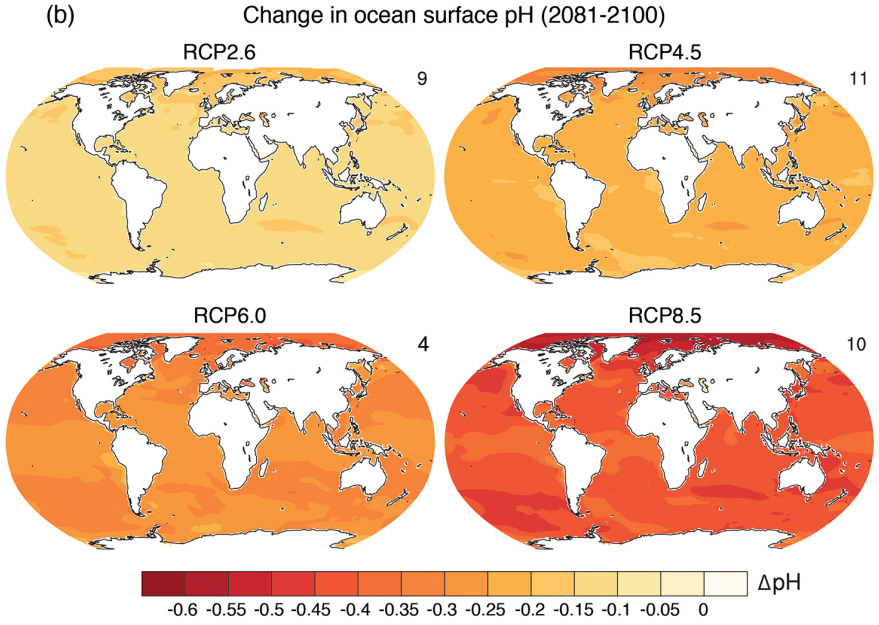

Figure 2: Modeled average ocean surface pH change between 1986-2005 and 2081-2100 following the four IPCC scenarios from the lowest (RCP2.6) to the highest (RCP8.5). The number of CMIP5 models to calculate the multi-model means is indicated in the upper right corner of each panel. Reproduced from Stocker *et al.* (22), Figure TS.20b, page 95, with a permission.

Bivalve mollusks are often long-lived organisms sensitive to environmental perturbations, such as ocean acidification (Papers I-II, 27, 34, 40). A record of bivalve growth can often be reconstructed from sequential bands that form during shell growth (Paper III, 41). Past environmental conditions can sometimes be interpreted from bivalve shells based on the geochemical signature of calcium carbonate ($CaCO_3$) deposited during the life-time of the animal (Paper IV, 42, 43). Furthermore, bivalves are distributed across a wide variety of habitats and latitudes (44–47). Finally, they are well represented in the geological record, the first bivalves already appearing during the Cambrian period (545–488 Ma, million years before present; 44, 48–51). Consequently, filter-feeding bivalves can be excellent indicator species of current and historical environmental perturbations not only because they are often sessile and can be used as monitors of water quality (26), but also because they record the environmental fluctuations within their shells. This thesis examines the susceptibility of bivalves to environmental perturbations using OA as a case study. In addition, the thesis aims to develop techniques to use bivalve shells as environmental proxies on sub-seasonal temporal resolution.

1.2 INDICATOR TRAITS FOR EFFECTS OF OCEAN ACIDIFICATION

The truth is out there. We work with distorted fragments of it.

— Modified from Carter *et al.* (52)

Although bivalves are considered among the most vulnerable taxa to ocean acidification (27), projecting their success in the future oceans is challenging. Many calcifying marine invertebrates, such as bivalves, undergo multiple life-history stages, each of which might respond differently to environmental perturbations (53). In bivalves, larval-stages and fertilization are the most sensitive life-stages to ocean acidification (Papers I-II, 27, 34, 54). The persistence of a population, however, is not determined by the average response of a single life-stage, but by the overall life-history responses of different genotypes within individuals as traits susceptible to OA are subject to natural selection and phenotypic plasticity (55). Although the rate of projected marine climate change might be too rapid to allow long-lived sensitive marine taxa to adapt through new genetic mutation, most populations contain genetic variability, which will be subject to selection and mixing through sexual reproduction (56). This adaptive evolution can strongly influence the responses of populations to future perturbations (55, 57–59). Selection from existing genetic variability is not the only adaptive force affecting future responses of populations. Phenotypic plasticity over single life-stages, over development of individuals, and over generations can all lead to responses within considerably shorter time-frames than selection as plasticity does not involve changes in DNA sequences (56, 60–62).

Environmental variability and habitat changes further complicate feasible projections of species success in the future oceans. Currently, the temporal pH variation in coastal habitats can be within the range of open ocean mean pH scenarios for 2100 (Box 1, 63–66). Therefore, inter-tidal bivalves, such as blue mussels (*Mytilus* genus), could be expected to tolerate lower pH regimes (e.g. see Paper II). Changed ecosystem interactions in future oceans might alter habitats, either increasing or decreasing the performance of each life-stage (53, 67–69). Finally, ocean acidification does not occur separately from other anthropogenic perturbations: increases in temperature (Paper II, 40, 70), alterations in food availability and competition (69, 71), hypoxia (72), pollution (73) and other stressors (74) might subject marine invertebrate populations to further selection pressures. Natural selection will therefore take place in a multidimensional fitness landscape, where multiple perturbations can subject selection to fitness trade-offs among traits affecting the rate and direction of adaptive evolution (55, 56, 60). Therefore, estimating the effects of OA on a species or population can be extremely complicated and might show large local- and among-experiment variability. Consequently, assessing the vulnerability of a species to climate change is not possible by using only one study or method, and a combination of different approaches and repeated studies are needed.

Effects of ocean acidification on marine organisms have traditionally been evaluated using an approach analogous to acute ecotoxicological experiments: individuals are first exposed to different pH regimes acquired through pCO_2 handling of seawater. Average responses are then measured using traits, such as growth, shell structure, survival, or oxygen consumption, that are presumed to indicate the performance of an organism. Finally, treatment effects on the indicator traits are compared to control conditions (e.g. see Paper II). Such experiments are important helping to determine organisms and life-stages that might be most sensitive to OA on population-level (termed "population sensitivity" hereafter, see Glossary) and might give indications of the relative magnitude of selection pressure caused by OA. Nevertheless, population-level responses tell little about the impacts of environmental perturbations over multiple generations (55). Individual variability with respect to traits prone to OA can be used to give clues of adaptability of a population (Paper I). Meta-analyses, on the other hand, can be used to gather information over multiple experiments and might give further information as to the population sensitivity. In this thesis, sperm activity was used as an indicator trait examining both the population- and individual-level responses to assess the sensitivity (Section 3.1) and adaptability (Section 3.2) of *Mytilus galloprovincialis* (Lamarck, 1819) and *Macoma calcarea* (Gmelin, 1791) to ocean acidification (Paper I, Box 2). Further, population-level responses to ocean acidification and warming were evaluated for early-larval development of *M. galloprovincialis* (Paper II) and these responses were compared to the literature using a meta-analysis (Section 2.4.3) to establish a broader view of OA effects on the *Mytilus* genus.

Box 2: Fertilization kinetics in bivalves

Fertilization kinetics in free-spawning marine invertebrates, such as bivalves, is an excellent example of complexity in natural selection. Fertilization of gametes occurs in the water column, and its success depends on gametes being released into an environment favorable for the gametes to remain viable, meet, and fertilize (75). Concentration of gametes is critical for a successful fertilization: in too low concentrations gametes have a small probability for an encounter leading to a low proportion of fertilized eggs, whereas in too high concentrations several sperm cells have time to penetrate the egg envelope before the activation of polyspermy block leading to an inviable zygote (Figure 3, 76–79). Consequently, the fertilization process subjects individuals to a strong selection pressure in a complex environment, where location of an individual in relation to the spawning population and prevailing currents, spawning synchrony, and gamete compatibility cause a high variability in life-time reproductive success among individuals (79–82).

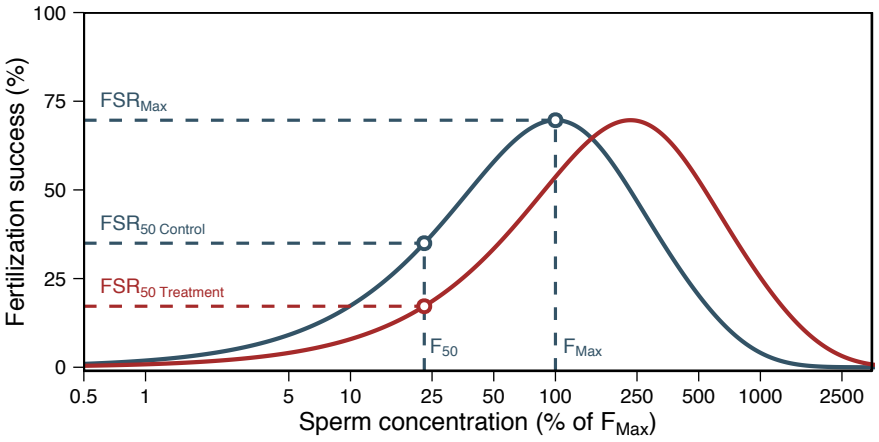

Figure 3: Modeled fertilization success (Fs) for average sperm swimming speed and percent motility of *M. galloprovincialis* exposed to ocean acidification (data from Paper I). Blue line indicates control (pH = 8.1) and red acidification treatment (pH = 7.7). Both control and treatment yield similar maximum Fs, but decreased pH lead to a decreased Fs when sperm concentration is low. FSR_{Max} means the maximum fertilization success (Fs) in control conditions, F_{Max} the sperm concentration that yields FSR_{Max}, $FSR_{50\ Control}$ the Fs that is half of FSR_{Max}, F_{50} the sperm concentration that yields $FSR_{50\ Control}$, and $FSR_{50\ Treatment}$ the Fs in treatment conditions resulting from F_{50}. Polyspermy block activation time of 10 s is used in the figure to illustrate the decreased fertilization success in high sperm concentrations (see 79, 83). The figure is modified from Schlegel *et al.* (84).

Gamete traits are important in determining the reproductive success of an individual (Figure 3, 82). These traits are directly influenced by the environment (Paper I, 84–87) and are presumed to be under genetic control. Fertilization models, such as Model S used in this thesis (Figure 3, 83), can be used to predict fertilization success by controlling gamete traits, such as sperm swimming speed, percent motility and egg size (88, 89). These models often allow more control for estimation of fertilization outcome than laboratory experiments, because fertilization success is dependent on sperm concentration (88). Due to the extremely dynamic environment (90), there is a lack of fertilization success data from natural habitats (78). Fertilization models are therefore often tested only in laboratory conditions. These models should, however, be sufficient to project relative differences in fertilization capability among individuals with differing gamete traits.

1.3 BIVALVE SHELLS AS ENVIRONMENTAL PROXIES

"The World as we know it nears its end, yet the past remains the key to the future..."
— CD Projekt RED (91)

In order to set the ongoing climate change to a context and project its implications, it is vitally important to understand the past climate on Earth (11, 92, 93). Knowledge of historical climate is interpreted from proxy archives formed during the period of interest – often representing long time scales (e.g. see Tables 5.A.2 and 5.A.3 in Masson-Delmotte *et al.* 3). While such information is essential, it has been suggested that long-term climate change events often occur in sudden jumps rather than gradual changes (94, 95). Consequently, most long-term proxy archives do not contain the required temporal resolution to identify the mechanisms causing the abrupt changes in the past climate (24). Proxy archives on seasonal to annual resolution can help to answer these questions, but precise high-resolution archives are rare (41, 96). Development of such archives would be beneficial not only for better understanding of the past climate on Earth, but also for more accurate projections of the ongoing climate change (37).

Bivalve shells offer promising proxy archives on seasonal resolution due to the longevity and relatively fast shell growth rate of many bivalve species (Paper III, 41, 97). Shells from multiple individuals can be combined using visible growth lines providing markers through time (Paper III, 41, 98, 99) to form decadal to multi-centennial chronologies (100). These chronologies can, in turn, be used to hind-cast environmental conditions with the help of geochemical proxies deposited in shell $CaCO_3$ (Box 3). Understanding the timing and environmental factors triggering growth band deposition is a fundamental prerequisite for successful use of geochemical proxies in bivalve shells as the shells do not have a constant growth rate over time (Paper III, 101).

Bivalve shells are complex structures often consisting of several $CaCO_3$ minerals (102), microstructures (51), and organic matrix controlling biomineralization

and stabilizing the shell matrix (51, 103). The bivalve shell matrix contains a relatively consistent concentration of calcium, which is often used as an internal standard by relating other elements to Ca concentration (Paper IV). Consequently, element-to-calcium ratios are commonly used geochemical proxies in biogenic carbonates – including bivalve shells (Box 3, 42, 43, 104–106). The incorporation of elements into bivalve shells occurs in two steps (51, 107): ions are first collected from ambient seawater to hemolymph through gills or the digestive system. These ions are then transported to extrapallial fluid (EPF), which is a thin layer of liquid between the calcifying shell surface and mantle providing optimal conditions for $CaCO_3$ formation, where it is finally precipitated into shell matrix.

Element-to-calcium ratios in bivalve shells are, therefore, complicated proxies subject to several layers of metabolic control before formation of $CaCO_3$. Some ions, such as $2Li^+$, Mg^{2+}, Mn^{2+}, Sr^{2+}, and Ba^{2+}, might substitute Ca^{2+} in $CaCO_3$ minerals (= "lattice-bound elements"; 108–111) in equilibrium with several factors including the growth rate of $CaCO_3$ crystals (= "kinetic processes"; 111–114), temperature (110, 112), salinity (109), and/or ambient element concentration (115). The development of geochemical proxies from bivalve shells is a complicated process with a general aim of finding predictable and consistent patterns in element ratio deposition confounded by multiple factors: element-to-calcium ratio is a proxy of the environment, if the precipitation predictably correlates with the environment regardless of which factors control the element deposition. Nevertheless, controls for element deposition in bivalves are important to identify, but do not necessarily imply whether the element ratio can or cannot be used as a proxy of the environment (e.g. see 116). Therefore, experiments that examine the relationships between shell element-to-calcium ratios and ambient environmental variables are required to evaluate the veracity of these ratios as environmental proxies.

Separating the sampling bias from environmental, metabolic and kinetic factors further complicates the interpretation of geochemical signals in bivalve shells. The organic matrix in bivalve shells can also consists of typical lattice-bound elements, such as Mg, Mn or Sr (117). Conventional *in situ* sampling methods such as LA-ICP-MS and SIMS used in this thesis, cannot differentiate between lattice-bound and organic-bound elements, but measure all elements in the shell matrix complicating the interpretation of element-to-calcium ratios. As a result, the environmental signals stored in bivalve $CaCO_3$ are often not consistent among species, locations or shell layers (113, 117, 118). Spot samples taken along a section of chronologically deposited shell material generate two additional issues that complicate the interpretation: First, sample spot location is difficult to determine using distance from a defined position, such as the shell margin, if the sample spots are not aligned along a sequence consistently perpendicular to growth lines (Paper III). Second, time averaging of sample material is, in principle, always present when shell material is physically sampled, thereby

leading to a systematic underestimation of peaks and troughs in geochemical signals (119, 120). The magnitude of this phenomenon, also referred to as "time-averaging error", depends on the sample size and the growth rate of the sampled material.

This thesis aims to develop new seasonal geochemical proxies using two circumpolar bivalve species (*Serripes groenlandicus* Mohr, 1786 and *Ciliatocardium ciliatum* Fabricius, 1780) that were deployed in oceanographic moorings for one year in two fjords on Svalbard. Both, *S. groenlandicus* and *C. ciliatum*, produce an aragonite shell with prominent annual growth lines following slower growth during winter, but the exact timing and causes of the winter growth line deposition are unknown (99, 121, 122). The sub-annual growth patterns of these bivalves were estimated using oxygen isotopes (Box 3) allowing identification of environmental processes triggering the growth band deposition (Paper III). Models of sub-annual shell growth patterns permitted statistical comparisons of elemental ratios with oceanographic data recorded by the mooring instruments (Paper IV). In order to correct for the spatial sampling bias, a digitized method to align sample spots along chronologically deposited materials with non-linear growth lines was developed (Paper III, 123). This method combined with the growth models also allowed the estimation of time-averaging error of LA-ICP-MS sampling (Paper IV).

Box 3: Geochemical proxies in bivalve shells

The ratio of stable oxygen isotope ^{18}O to the most abundant oxygen isotope ^{16}O (denoted as $\delta^{18}O$, expressed in parts per thousand relative to a reference) is a proxy of temperature in most bivalve shells, when the isotopic composition of water ($\delta^{18}O_W$) is known (124, 125, Paper III). Seawater $\delta^{18}O$ is positively correlated with salinity (126) and varies with time following the glacial cycles (127), and geographically, the signature being higher closer to the equator and lower towards the poles (128, 129). Therefore, acquiring the prior information of $\delta^{18}O_W$ is often guesswork resulting in an uncertainty in water temperature estimates from $\delta^{18}O$ in $CaCO_3$.

Element-to-calcium ratios offer not only potential temperature proxies (130–134), but also a possibility for estimating many ecosystem-relevant parameters from bivalve shell carbonate using relatively small sample sizes (e.g. 104, Paper IV). Due to metabolic control of shell deposition (see Section 1.3), elemental composition of bivalve shells often reflects biological effects (also called "vital effects") or kinetic processes rather than a relationship that consistently follows environmental parameters (113). Element ratios might also vary within a sample depending on the shell layer and structure of $CaCO_3$ minerals (51, 135). Consequently, the results in the literature are often confusing and sometimes conflicting. As a part of this thesis, a literature survey was conducted in order to find reoccurring patterns in element ratios examined in Paper IV (see Section 2.4.3).

The survey indicates that barium to calcium ratio (Ba/Ca) is by far the most promising environmental proxy followed by Li/Ca and Mo/Ca (Figure 4). Barium profiles are often characterized by abrupt transient peaks, which are often connected to primary production, but the exact environmental cause of Ba peaks is still debated (136, 137). Barium can be incorporated to bivalve shells through, at least, three different pathways: dissolved Ba (115), particular Ba (barite; 138, 139) or diet enriched in Ba (140, 141). Freshwater input and sediment surface redox-processes can increase dissolved Ba concentration in seawater potentially influencing Ba/Ca values in bivalve $CaCO_3$ (137, 142). Fewer Mo and Li studies are available. Mo has been connected with phytoplankton nitrate uptake and also a direct incorporation through diet has been suggested (115, 143, 144), whereas Li shows promising results as a proxy of shell growth rate of bivalves (145**?**). Although Sr/Ca and Mg/Ca have been suggested as alternative temperature proxies to $\delta^{18}O$, biological controls in EPF seem to complicate the incorporation of these elements such that they are useful temperature proxies only in specific cases when temperature correlates with growth rate (133). Mn has also been related to primary production, although the mechanisms of Mn incorporation may be related to several factors, such as sediment surface redox-processes and therefore more complicated than Ba (113, 146).

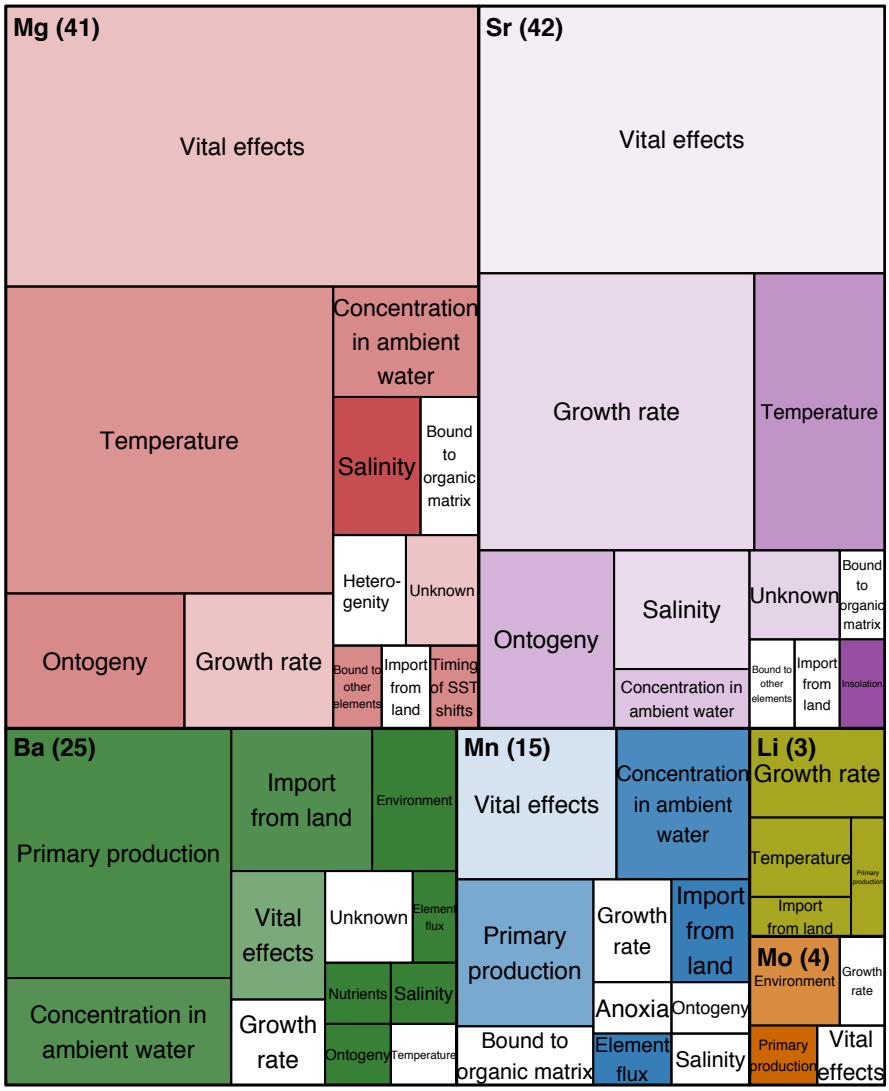

Figure 4: A treemap presenting factors suggested to affect incorporation of element-to-calcium ratios in bivalve shells in 68 articles published 1965 - 2014. Large boxes with different colors illustrate different elements. Area of the boxes is related to the number of studies, total number given in parenthesis after element name. Darker boxes indicate a higher proportion of studies concluding that the corresponding element ratio is a potential proxy affected by the factor inside the box. Growth rate refers to both shell growth and kinetic processes.

1.4 OBJECTIVES

The main objectives of this thesis are to increase knowledge on 1) susceptibility of bivalves to environmental perturbations, and 2) bivalve shells as sub-seasonal environmental proxies. These objectives are divided into research questions, each of which is handled separately in the Summary of results section. The research questions are approached using indicator species and traits to draw overall conclusions (Section 4). These research questions are:

1. Sensitivity on the population level (Papers I-II, *Mytilus* meta-analysis)
    - Are bivalves sensitive to ocean acidification scenarios for 2100?
    - Which life-stages are the most sensitive?
    - At which future pCO_2 levels do bivalves show negative effects of ocean acidification?
    - Can differences in experimental results be addressed to differences among populations and species?
2. Adaptability of bivalve populations (Paper I)
    - Do bivalves show signs of adaptability to climate change?
3. Effects of temperature (Papers I-III)
    - Does increasing temperature affect bivalve reproduction and early-larval stages?
    - How do food availability and temperature affect shell growth of bivalves in the Arctic?
4. Bivalve shells as seasonal environmental proxy archives (Papers III-IV)
    - When are the prominent annual growth bands deposited in *S. groenlandicus* and *C. ciliatum* shells?
    - Which environmental processes trigger the growth band deposition?
    - Are element-to-calcium ratios useful environmental proxies in two aragonitic bivalve species with Arctic distribution?

2 MATERIAL AND METHODS

This PhD thesis consists of four scientific articles. Data for these articles were obtained by two larger experiments: 1) an ocean acidification experiment on bivalve gametes and early larval stages (Section 2.2), and 2) a year-long bivalve deployment on oceanographic moorings (Section 2.3). In addition, the thesis contains a self-standing meta-analysis, which was partly published in connection with Paper II (Section 2.4.2), and a literature survey presented in a scientific conference (Section 2.4.3). Both, the meta-analysis and the literature survey, have been extended and updated since the previous publications.

2.1 STUDY SPECIES

Five filter-feeding bivalve species were used in this thesis (Table 1). *Serripes groenlandicus* (Mohr, 1786), *Ciliatocardium ciliatum* (Fabricius, 1780), *Macoma calcarea* (Gmelin, 1791), and *Chlamys islandica* (Müller, 1776) are cold water species with a circumpolar distribution (147). *Mytilus galloprovincialis* (Lamarck, 1819), on the other hand, is a temperate blue mussel believed to be native to the Mediterranean Sea (148). *Serripes groenlandicus*, *C. ciliatum* and *M. calcarea* are subtidal infaunal species, whereas *Chlamys islandica* and *M. galloprovincialis* are epibenthic species. Further, *M. galloprovincialis* is an intertidal sessile species, and *C. islandica* is a subtidal species capable of swimming short distances.

Table 1: Overview of bivalve species included in this thesis.

Species	Paper	Collection site	Experimental site
Macoma calcarea	I	Balsfjorden, Norway	Mallorca, Spain
Chlamys islandica	I	Balsfjorden, Norway	Mallorca, Spain
Mytilus galloprovincialis	I-II	Menorca, Spain	Mallorca, Spain
Serripes groenlandicus	III-IV	Spitsbergenbanken, Barents Sea	Kongsfjorden and Rijpfjorden, Svalbard
Ciliatocardium ciliatum	III-IV	Spitsbergenbanken, Barents Sea	Kongsfjorden and Rijpfjorden, Svalbard

Sperm activity of *M. calcarea*, *C. islandica*, and *M. galloprovincialis*, as well as early larval development of *M. galloprovincialis* (Table 1) were examined in an ocean acidification experiment conducted at the Mediterranean Institute for Advanced Studies, Mallorca. Artificial seawater (Instant Ocean Sea Salt) was aerated using the annual average atmospheric pCO_2 level of 2005 (380 ppm; 2) and a level close to the RCP8.5 scenario for 2100 (1000 ppm; Box 1). In order to assess the combined effects of ocean acidification and warming, the larval development experiment and parts of the sperm activity experiment were conducted using a factorial design by combining the acidification treatment with a temperature treatment.

Paper I with additional data: Sperm activity (swimming speed and percent motility) of males of three bivalve species (*Chlamys islandica*, *Macoma calcarea* and *Mytilus galloprovincialis*) were used as indicator traits to compare population- and individual-level responses to experimental ocean acidification (OA) using two pH_T levels (control = 8.1, and treatment = 7.7). In addition, a selection of males was exposed to a combination of the acidification treatments and temperature treatments using the ambient collection site temperature (2 °C) as a control and a 6 °C increase as an elevated-temperature treatment. Treatment effects on fertilization success were theoretically quantified by fertilization modeling (Box 2) using Styan's model (Model S; 79, 83) and FSR_{50} values to compare differences between treatment and control (Figure 3). Responses to treatments were assessed using response ratios (see Section 2.4.1). Population-level responses were calculated using mean values for each male as replicates, whereas individual-level response ratios were computed using replicate measurements. *Chlamys islandica* and temperature data were not included to Paper I due to low sample size, but are presented in this thesis (see Section 3.2).

Paper II: Early development of Mediterranean mussel larvae (*M. galloprovincialis*) was examined for population-level effects of ocean acidification (OA) and warming. Developing embryos of *M. galloprovincialis* were exposed to pH_T levels of 8.0 (current pH) and 7.6 (2100 level). Effects of temperature [16–17 °C (ambient) and ≈20 °C (elevated)] and acidification on early larval development were accessed by cultivating embryos until late trochophore/early D-veliger stages in treatment cylinders using a factorial design. Larval size, survival, respiration, and calcification rate were used as response variables.

2.3 BIVALVE DEPLOYMENT ON MOORINGS (PAPERS III-IV)

Sub-seasonal shell growth, $\delta^{18}O$, and element ratio patterns of bivalve shells were studied by deploying *S. groenlandicus* and *C. ciliatum* on oceanographic moorings in two oceanographically different fjords, Kongsfjorden and Rijpfjorden, in Svalbard (see Paper III). The bivalves were placed in 7 mm mesh plastic cages and held at two depths (15 m and 25 m) in both fjords for one year. After mooring deployment, the bivalves were sectioned along maximum growth-axis and further analyzed for growth patterns, $\delta^{18}O$ composition and element ratios. Weekly averaged mooring instrument data of temperature, salinity and fluorescence as a proxy of phytoplankton biomass were used to correlate these predictor variables with shell growth and geochemical composition (response variables). Linear mixed-effects models (LMM) were used to examine overall relationships between response and predictor variables. The marginal R^2 value was used as an indicator of overall variance explained by each predictor variable (149).

Paper III: Subannual shell growth of nine bivalves was estimated by aligning high-resolution secondary ion mass-spectrometer (SIMS) $\delta^{18}O$ measurements with predicted $\delta^{18}O$ values derived from continuous mooring instrument recordings using dynamic time warping (DTW; 150). The estimated weekly growth rates were correlated with temperature and fluorescence index using LMMs to identify environmental factors triggering shell growth.

Paper IV: Potential use of element ratios (Li/Ca, Mg/Ca, Li/Mg Mn/Ca, Sr/Ca, Mo/Ca, and Ba/Ca) as environmental proxies was examined by sampling the mooring shell sections using laser-ablation inductively-coupled-plasma mass-spectrometry (LA-ICP-MS). Subannual growth models from Paper III were used to relate the element ratio patterns in nine shells to weekly growth rate and oceanographic data (temperature, salinity, and fluorescence) using LMMs. Growth rate was logarithmically related to element ratios and log-transformed prior to analyses. An additional dataset containing individuals without growth models (n = 21) was used to examine the variability in peak and trough element-to-calcium values among fjords and depths.

2.4.1 *Response ratios*

Results in Papers I-II, and in the *Mytilus* meta-analysis were expressed as back-calculated log-transformed response ratios as described in Hedges *et al.* (151):

$$R = exp[Ln(\frac{\bar{X}_{Treatment}}{\bar{X}_{Control}})] \times 100\% \quad (1)$$

where $\bar{X}_{Treatment}$ and $\bar{X}_{Control}$ are arithmetic means over the measured response variable in treatment and control, respectively. Response ratios, also called effect sizes or effects in this thesis, display the mean ratio of treatment to control. Therefore R of 100% indicates equal treatment and control means, whereas 200% indicate twice as large mean value for treatment compared to control, and 50%, in turn, twice as large mean for control compared to treatment. The statistical significance between treatment and control was evaluated by 95% confidence intervals (CIs) for response ratios. Confidence intervals were calculated from propagated variances in log-space following Equation 1 in Hedges *et al.* (151). Effect size statistics is a better alternative to more commonly used null hypothesis significance testing, and does not only offer information about the statistical significance (p-value), but also about the magnitude of an effect (R) and precision of the effect estimate (CIs; 152). Response ratios presented in this thesis are statistically significant (at least to 95% confidence level), if CIs do not cross the 100% effect size. This critical level is indicated as a solid black line in effect size figures.

2.4.2 *Mytilus meta-analysis*

Reported OA effects on *Mytilus* genus were compiled from published literature by a Google Scholar search using keywords "Mytilus", and "ocean acidification" (23 relevant studies found on 2014-11-11). Response variables related to size were the only parameters reported by every study and consequently they were further evaluated by extracting pH, mean response and standard deviation for OA treatments and controls from the articles (Table A1). Required information was extracted from tables or the Pangaea database (153) whenever possible. WebPlotDigitizer (154) was used for articles where the information was only available in figures. If the information was not available in required format, authors of corresponding articles were contacted and asked for data. Authors who contributed data to the meta-analysis are mentioned in the acknowledgments. The number of quasi-independent experimental units was used as the replication level for each study. Response ratio and 95% CIs were calculated assuming a normal distribution following Hedges *et al.* (151) directly. The analysis was

separated by life-stages: adults and juveniles were examined together, whereas larvae were analyzed separately. In the larval analysis, time over development was handled as separate data points resulting in several dependent observations per study. Mean daily growth rate (increase in shell length, wet weight, or dry weight of $CaCO_3$; see Table A1) was calculated for adults and juveniles by using the difference in shell size between the beginning and the end of an experiment and dividing by the number of days resulting in independent observations.

Resulting effect sizes (effect on maximum shell length in larval analysis and effect on growth rate in juveniles and adults) were plotted against the experimental pH difference (ΔpH) in each study. Differences in pH were set to context by comparing them to the IPCC ocean acidification projections for 2100 (Box 1) and to the future maximum ΔpH scenarios (155). The average open ocean pH_T value between 1986 and 2005 (8.11) was extracted from Ciais *et al.* (156, Figure 6.28) and used as initial value for ΔpH. The minimum ΔpH value for 2100 (-0.06 pH units) was taken from the minimum confidence level of the average open ocean surface pH change for RCP2.6 scenario (156, p.469). The maximum value (-0.54 pH units), in turn, represents the maximum area-weighted reduction between 1986-2005 and 2081-2100 for the Arctic Ocean (> 70°N) following the RCP8.5 scenario (156, Figure 6.28a). Future maximum ΔpH scenarios followed the IPCC 4th assessment (157) projections for burning all known fossil fuel reserves (5000 Pg C emission scenario, ΔpH = -0.63) and for utilizing all possible future fossil fuel reserves together with methane hydrates (20 000 Pg C emission scenario, ΔpH = -1.28) as reported by Caldeira and Wickett (155). These pH values were extracted from Figure 2 and compared to the 1986-2005 average used in the 5th IPCC report for integrity. Non-relative pH values are expressed against total scale throughout this thesis unless specified otherwise.

2.4.3 *Element ratio review*

Literature survey in element-to-calcium ratios in bivalve shells was conducted by going through all published literature on elements examined in Paper IV (Li, Mg, Mn, Sr, Mo and Ba) by 2014-09-02 using Google Scholar. Results from Paper IV were included in the review resulting to 66 studies (see Table A2). The review was compiled into a figure (Figure 4), with an aim of finding reoccurring patterns in subjectively categorized data: maximum of 3 factors controlling element incorporation into $CaCO_3$ matrix were scored in order of importance listed in each article. Total of 6 points were allocated for each species-study combination. These points were further allocated to controlling factors, giving the most important factor 6, 4, or 3 points depending on the number of controls the authors listed. The next important control received 2 or 1 points and the third factor 1 point. Elements and different bivalve species were used as replicates, leading to a total of 158 element-species-study combinations (rows). The poten-

tial of a proxy was evaluated after information given in articles and contributed to the shading of boxes in Figure 4: if authors clearly stated that an element ratio could have a potential as a proxy, the corresponding row received a score of 1 ("yes" in Table A2). If authors indicated that an element ratio could be used as a proxy, but did not specify of what or were uncertain, the row received a score of 0.5 ("maybe"). If authors stated that an element ratio could not be used as a proxy, the row received a score of 0 ("no"). If an article did not contain sufficient information to evaluate proxy relationship with a specified factor, the row was considered as missing data and did not contribute to the proxy score. The scores were averaged for each element ratio-factor combination. These average values were used to define the shading of boxes giving mean score of zero white color and mean score of one full color of each element ratio. Results were plotted using the *treemap* package for R (158).

2.4.4 *R packages*

Two packages for R statistical programming environment (159) were developed to align sample spots along bivalve cross-sections in Papers III and IV. The method is described in Paper III and in further detail by Vihtakari (160). Briefly, subseasonal growth lines in bivalve sections were marked using ImageJ (161). The *RImageJROI* package (162) was necessary to export the obtained coordinates from ImageJ to R. The location of each sample spot was then related to growth lines and projected along the historical location of shell margin using the *sclero* package (123).

3 SUMMARY OF RESULTS

3.1 SENSITIVITY ON THE POPULATION LEVEL (PAPERS I-II, AND META-ANALYSIS)

Are bivalves sensitive to ocean acidification levels projected for 2100?

Experimental acidification using RCP8.5 levels for 2100 significantly reduced average sperm swimming speed of *M. galloprovincialis* by 26%, percent motility by 42%, and modeled fertilization success by 46% (Paper I). Such reductions can lead to lower fertilization rates in sperm-limited low-density populations (76, 78, 88, Box 2, Paper I). Further, the meta-analysis indicated that growth of *Mytilus* larvae is negatively affected in experiments using acidification levels following the projected emission scenarios for the year 2100, and that the larvae are already likely to experience pH values reducing larval growth (Figure 5, Paper II, 65, 163). The former observation could contribute to site-specific and annual variation in recruitment of *Mytilus* (Paper II). Future reductions in pH might expose *Mytilus* larvae to larger pH fluctuations possibly causing larger variations in recruitment (164). Therefore, *Mytilus* genus is likely sensitive to OA levels projected for 2100, but the ecological consequences of this sensitivity are uncertain: In Paper II we did not observe significant negative effects of ocean acidification on *M. galloprovincialis* larvae using RCP8.5 levels, but found significant reduction in larval performance with 3°C increase in temperature (see Section 3.3). The non-significant OA effects could demonstrate differences in responses among populations, species, or experiments (34, 40, 165). The meta-analysis indicated that shell growth rate of adult and juvenile stages will likely not be sensitive to projected acidification levels for 2100 (Figure 6), although the analysis did not consider shell structure, which might be negatively affected by 2100 OA-levels in adults (166–168). Further, we did not detect significant population level effects in sperm activity or modeled fertilization success of *M. calcarea* in response to RCP8.5 acidification levels (Paper I). Consequently, bivalves might be sensitive to acidification levels projected for 2100, but the sensitivity varies between species, populations and life-stages.

Which life-stages are the most sensitive?

We found signs of a negative effect of acidification on sperm activity for one bivalve species indicating that projected 2100 pCO_2 levels might decrease fer-

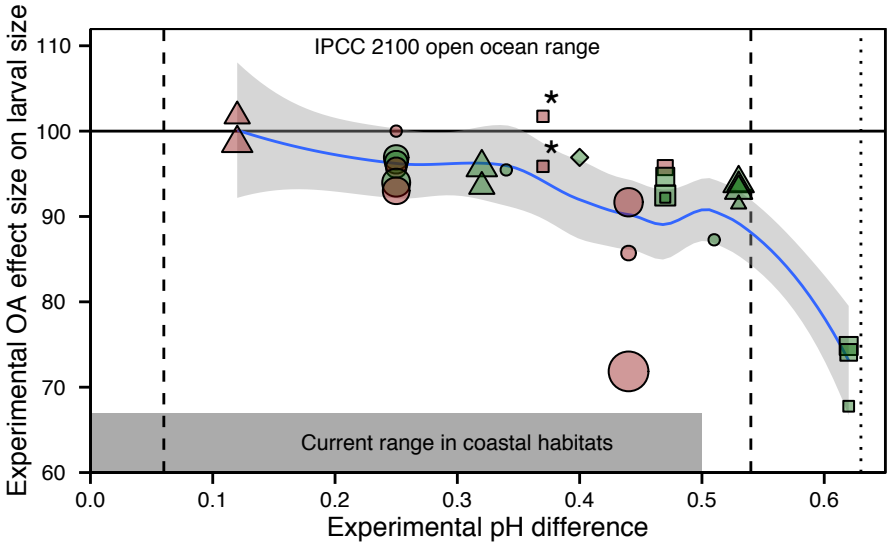

Figure 5: Mean effect sizes of ocean acidification on size of *Mytilus* larvae 2-64 d after fertilization extracted from seven experimental studies (○ = *M. edulis*, □ = *M. galloprovincialis*, ◇ = *M. trossulus*, and △ = *M. californianus*). Green and red symbols indicate significant and non-significant effect sizes, respectively. Symbols with the same x-axis value are from a same study representing dependent measurements over time. Symbol size is related to the development time larger sizes indicating a longer time from fertilization (see Table A1). Blue line with shading illustrates a local regression smoothing (± 95% CIs) for average values. Grey horizontal bar indicates the range of current stochastic pH fluctuations experienced by mussels in coastal habitats (see 65). Dashed lines indicate the range of open ocean pH reduction predicted to occur by the year 2100 depending on an IPCC scenario and geographical location (see Section 2.4.2 and Box 1). Dotted line illustrates an estimated average open ocean pH reduction assuming that all known fossil fuel reserves were burned. Points marked with ⋆ are results from Paper II.

tilization success of some bivalve species in low density populations (Paper I). Other studies, however, indicate that fertilization might not be affected in all species (27). The meta-analysis on short term OA experiment results indicates that shell growth of *Mytilus* larval stages is more affected by OA than that for adults (Figures 5 and 6). This observation is well supported by the literature also for other calcifying taxa (27, 34). *Mytilus* are already present in habitats with a highly fluctuating pH regime (163, Figure 6) and have developed adaptations to cope with low pH environments. For example, the shell of adult blue mussels is covered by a thick organic periostracum (169), which protects the shell

from dissolution of calcite in under-saturated environments (170). Larval stages of blue mussels lack such adaptations partly explaining the sensitivity to ocean acidification. Recruitment of *Mytilus* might be negatively affected by low pH levels (Figure 5), but as *Mytilus* evidently inhabit such habitats already, low pH might select for individuals that better tolerate OA. Future pH reductions could, therefore, expose *Mytilus* populations to a higher selection pressure.

At which experimental pCO_2 levels do bivalves show negative effects of ocean acidification?

We observed substantial negative effects of OA on sperm activity of *M. galloprovincialis* using RCP8.5 scenario pCO_2 levels (~1000 ppm, Paper I). The meta-analysis on population-level effects of ocean acidification (Figure 5) indicates that size of *Mytilus* larvae might be negatively affected already by projected medium emission scenarios for 2100 (RCP6.0, equivalent to 0.2 unit drop in pH or atmospheric pCO_2 of 670 ppm; see Box 1). In contrast, the meta-analysis does not generally indicate that shell growth of adult or juvenile stages would be negatively affected by 2100 open ocean levels: Significant negative effects are reported for populations of adult and juvenile *Mytilus chilensis* (171, 172). Apart from these two studies, significant negative effects of OA on shell growth of adult or juvenile *Mytilus* have been connected with temperatures outside the thermal tolerance range of a population or species (173, 174). Keppel *et al.* (175), on the other hand, report a positive effect of acidification on *Mytilus edulis* shell growth with ΔpH levels corresponding to the RCP6.0 scenario, even though these estimates are associated with a large variability. Shell growth of adults might be negatively affected by ΔpH values >0.6 equivalent to atmospheric pCO_2 values >2000 ppm (Figure 6), although there is a high variability in responses.

As sensitivity of bivalves varies among species and life-stages, establishing a general threshold for the entire class is not sensible. There is evidence that parts of the life-cycle in some genera may have reduced performance even under IPCC medium emission scenarios for 2100 (Figures 5 and 6; 27). It should be noted, however, that shell shell growth is only one response variable among many others which might be better indicators of environmental perturbations. Fitzer *et al.* (166), for instance, reported a significant decrease in shell aragonite content of *M. edulis* adults exposed to ocean acidification, but their study did not yield a significant decrease in shell growth in the meta-analysis (Table A1). The ecological meaning of this observation is unclear as pH values for that particular study were very low (7.7 for control, and 7.7–7.2 for treatments; 176), but indicates that estimates of population sensitivity are not straightforward. Responses of populations in the future oceans might differ from these estimates due to natural selection and changed ecosystem interactions further complicating the estimation of critical levels of ocean acidification.

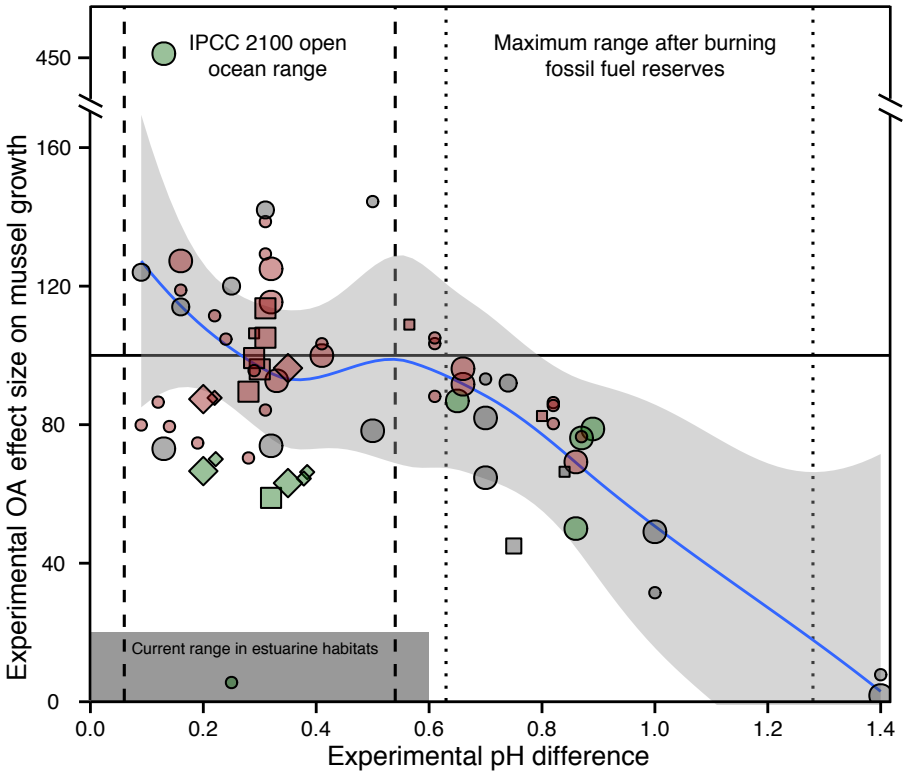

Figure 6: Mean effect sizes of short-term (44-202 d) ocean acidification on growth rate of *Mytilus* adults and juveniles extracted from 16 studies (○ = *M. edulis*, □ = *M. galloprovincialis*, and ◇ = *M. chilensis*). Green symbols indicate significant effect sizes, red symbols non-significant effect sizes, and gray symbols studies where replication was not sufficient to calculate confidence intervals. Small symbols refer to juveniles, large symbols to adults, and medium sized symbols to studies where both groups were mixed. Blue line with shading illustrates a local regression smoothing (± 95% CIs) for average values. Grey horizontal bar indicates the range of current stochastic pH fluctuations experienced by mussels in estuarine habitats (163). Dashed lines indicate the range of open ocean pH reduction predicted to occur by the year 2100 depending on an IPCC scenario and geographical location (see Section 2.4.2 and Box 1). Dotted lines from the left illustrate an estimated average open ocean pH reduction assuming that all known fossil fuel reserves were burned, and that possible future fossil fuel reserves and methane hydrates were utilized.

Can differences in experimental results be addressed to differences among populations and species?

Mytilus growth responses to ocean acidification vary widely with similar ΔpH values in the meta-analysis (Figures 5 and 6). The variability could partly be explained by differences in experimental conditions. For instance, in Papers I and II we used artificial sea-water leading to higher total alkalinity values than in the natural habitat (Paper II, 177, 178). This, in turn, led to aragonite saturation state (Ω_{Ar}) being well over the undersaturation limit seemingly explaining the small pCO_2 effect on larval growth (Figure 7). Nevertheless, such explanations are not straightforward: Ω_{Ar} values used in Paper II were actually within the range projected for 2100 (179). Furthermore, the only larval development study published with comparable ΔpH values and development time for *M. galloprovincialis* (180) yielded effect sizes that were relatively similar to our study (Table A1). Duarte *et al.* (172) report different responses to OA between two *M. chilensis* populations suggesting that the non-significant effect of acidification in Paper II could also be explained by differences in population responses. Also the short development time in Paper II is a likely contributor to the observed non-significant effect (Table A1, 180).

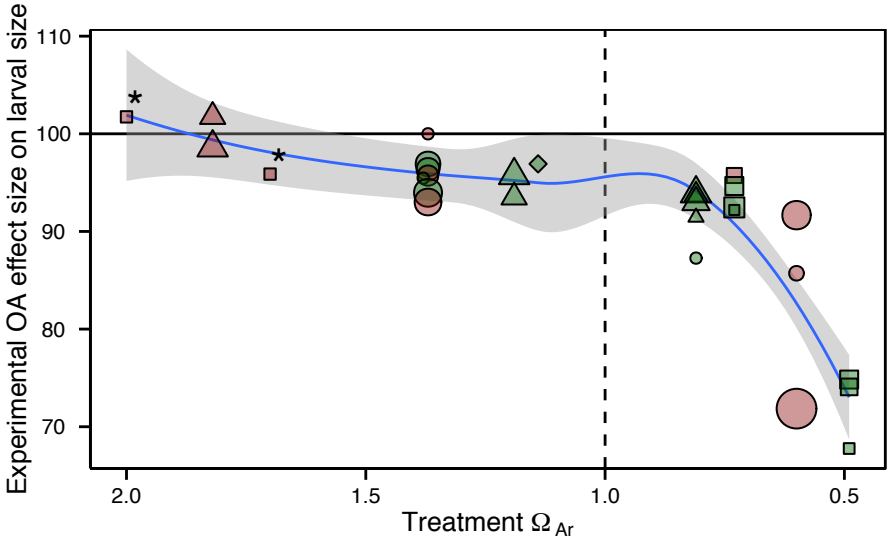

Figure 7: Mean effect sizes of ocean acidification on size of *Mytilus* larvae against treatment aragonite saturation state. Points marked with ⋆ are results from Paper II. Dashed vertical line indicates the undersaturation limit for aragonite. See Figure 5 for details.

Separating experimental differences from population and species differences among *Mytilus* genus through meta-analyses is difficult, because experimental conditions and exposure time vary widely in the literature (Table A1). Experiments with unrealistic water chemistry values might lead under- or overestimating the population sensitivity, yet accurate control of all water chemistry values during experiments is difficult. Furthermore, data on seasonal variations with respect to water chemistry is lacking for many habitats. Therefore, it is challenging to establish a set of "right" values prior to experiments. Meta-analyses can reveal important patterns in population responses, and more published studies will improve the results of these analyses. Therefore, rejecting studies because of water chemistry values is a loss of information and should be avoided. Nevertheless, reviewers and researches alike should pay special attention to accurately measuring and reporting these values.

3.2 ADAPTABILITY OF BIVALVE POPULATIONS (PAPER I)

Do bivalves show signs of adaptability to climate change?

Results from Paper I indicate that males possess different tolerances for climate change within a population: sperm of *M. galloprovincialis*, *M. calcarea* and *C. islandica* demonstrated substantial among-individual variability in responses to simulated ocean acidification or warming (Figure 8). The variability in response to acidification was greatest in *M. galloprovincialis* ranging from statistically non-significant to a 10-fold reduction in percent sperm motility, 2-fold reduction in sperm swimming speed, and 6.5-fold reduction in modeled fertilization success (Figure 8C, F, and I). Acidification had a significant negative effect on modeled fertilization success in 10 of 13 *M. galloprovincialis*, and in three of 10 *M. calcarea* males (Figure 8G-I). The effects of acidification among *M. calcarea* males varied from significantly positive in one male to significantly negative in two males in percent sperm motility, and from non-significant to a 1.5-fold reduction in sperm swimming speed (Figure 8B and E). The two studied *Chlamys islandica* males demonstrated opposite responses to increased temperature, but were not significantly affected by simulated ocean acidification (Figure 8A, D, and G).

If sperm activity is a heritable trait, genotypes of individuals that demonstrated robust responses to climate change could become more abundant in the gene pool. It is unknown whether sperm robust to environmental change carry genes that are similarly beneficial during the remainder of the life-cycle. It is possible that these traits are not related and that strong selection early on during the life-cycle leads to carry-over effects during the later life-cycle as a form of reduced phenotypic plasticity (56, 181). Our results together with other recent studies on marine invertebrates (84, 87, 182) indicate that among-individual variability in response to environmental perturbations is likely to be a norm, rather

than an exception. Therefore, bivalves do show signs of adaptability to climate change and the population responses in future oceans are unpredictable.

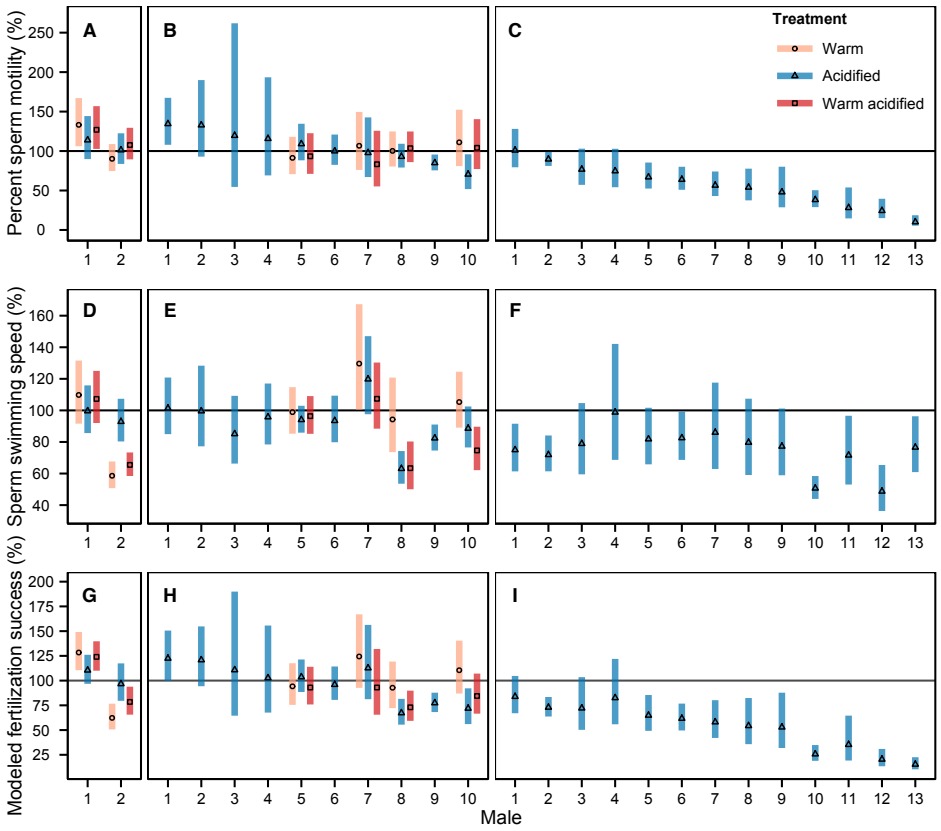

Figure 8: Treatment effect sizes (i.e. response ratios) on percent sperm motility (**A-C**), swimming speed (**D-E**), and modeled fertilization success (**G-I**). **A, D & G** = *C. islandica*, **B, E & H** = *M. calcarea*, and **C, F & I** = *M. galloprovincialis*. Error bars represent 95% confidence intervals. Males are ordered based on average percent sperm motility in response to *Acidified* treatment. Figure is modified from Paper I with additional data.

3.3 EFFECTS OF TEMPERATURE AND FOOD AVAILABILITY (PAPERS I-III)

Does increasing temperature affect bivalve reproduction and early-larval stages?

In additional data for Paper I six males (2 *C. islandica* and 4 *M. calcarea*) were studied for temperature effects on sperm activity. A six degree increase in temperature alone yielded no significant population-level effects, but the combination of temperature and pCO_2 treatments reduced sperm swimming speed in *M. calcarea* leading to a marginally non-significant interaction effect (pCO_2 × Temp; $\bar{R}$ = 90%, CIs 79-101%) and to a significant 14% reduction in modeled fertilization success ($\bar{R}$ = 86%, CIs 76-97%). Results from Paper II indicated that the temperature treatment had a larger effect on early larval stages of *M. galloprovincialis* than the OA treatment: We observed no significant effects of acidification, whereas temperature significantly reduced larval performance ($\bar{R}_{Size}$ = 90% and $\bar{R}_{Survival}$ = 70%) and increased energy demand ($\bar{R}_{Respiration}$ = 429%). Temperature tolerance of *Mytilus* larvae varies widely among species, populations, and studies even within relatively small geographical area: *M. galloprovincialis* larvae from Mallorca (Paper II) demonstrated different optimal temperature than a study conducted in the Mediterranean Sea approximately 900 km from our study site (183). Large variation in thermal tolerance might indicate that populations could have an adaptation capacity to small temperature changes caused by global warming. However, extreme temperatures in summer and winter are likely to define the future population limits (184). Therefore, increasing temperature does affect bivalve reproduction, but the responses are likely to vary such that general conclusions as to the directions of this effect are difficult to establish based on data presented in this thesis.

How do food availability and temperature affect shell growth of bivalves in the Arctic?

The results from Paper III indicate that food availability sets the limits for the timing of shell growth periods, whereas temperature controls the growth rate, when a food source is sufficient. These conclusions, however, are based solely on shell growth, and seasonal allocation to somatic or gonadal tissue is not considered here. Studied shells demonstrated variable growth rates during the mooring deployment growth season lasting from May until December in Kongsfjorden and from mid-June until November in Rijpfjorden (Figure 9). Shell growth commenced two weeks to a month after the returning food source as indicated by mooring fluorescence readings (Figure 9) possibly due to replenishment of energy reserves and energy allocation to growth of soft tissue. Shell growth models indicated a period with slower growth in Kongsfjorden in June that could be related to spawning (see Section 4.1 in Paper III). Shell growth rate of all studied bivalves with an adequate DTW fit demonstrated significant correlations with

temperature, marginal R^2 value ranging from 0.30 to 0.40 among baskets (see Table 3 in Paper III).

3.4 BIVALVE SHELLS AS SEASONAL ENVIRONMENTAL PROXY ARCHIVES (PAPERS III-IV)

When are the prominent annual growth bands deposited in S. groenlandicus and C. ciliatum shells?

Prominent winter growth bands were likely deposited during the slow shell growth period lasting from November until May in Kongsfjorden and from October until late-June in Rijpfjorden (Figure 9).

Which environmental processes trigger the growth band deposition?

The growth cessation and growth band deposition was likely controlled by food availability (see Section 3.3). Fluorescence data, however, indicated a low biomass of photosynthesizing phytoplankton during autumn in Kongsfjorden, but the water column could have contained degraded phytoplankton and heterotrophic plankton (185, 186) that could have functioned as a food source for the bivalves. Consequently, results from Paper III suggest that winter growth bands can be used as a proxy of the timing of food availability in *S. groenlandicus* and *C. ciliatum*.

Are element-to-calcium ratios useful environmental proxies in two aragonitic bivalve species with Arctic distribution?

Despite the high potential of element-to-calcium ratios as seasonal environmental proxies (see Box 3), the results from Paper IV mostly reflected the metabolic controls of element deposition in bivalves (Figure 10). Consequently, Li, Mg, Mn, Mo, Sr or Ba to calcium ratios are not straightforward proxies of temperature, shell growth rate, food availability or salinity in *S. groenlandicus* or *C. ciliatum* shells. Nevertheless, deposition of Li/Ca and Ba/Ca is likely synchronized within shells from a same location and these ratios can be used as subannual chronological markers. Li/Ca might be a useful proxy of shell and/or crystal growth rate, Mg/Ca appears to loosely follow temperature, and Ba/Ca might reflect elemental concentrations in seawater. Despite this, the relationships are not strong enough to precisely reconstruct these parameters due to among and within shell variability. These results do not exclude the use of element-to-calcium ratios as environmental proxies in studied bivalves, but merely indicate that the deposition of element ratios is controlled by internal and external several factors.

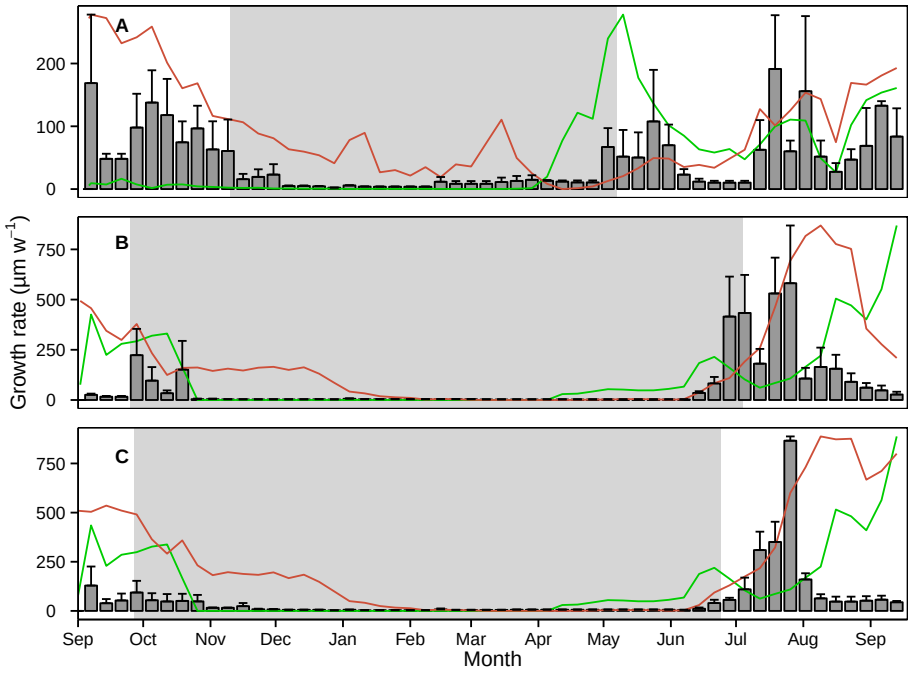

Figure 9: Weekly averaged growth rate for *S. groenlandicus* and *C. ciliatum* deployed in oceanographic moorings (gray bars) + 1 standard error (error bars) related to weekly averages of fluorescence index (green line) and temperature (red line). **A)** Kongsfjorden 25 m basket, **B)** Rijpfjorden 15 m basket, and **C)** Rijpfjorden 25 m basket. Fluorescence and temperature values are scaled to the maximum growth rate, including error bars, in each subplot. Fluorescence measurements are from 36 m in Kongsfjorden and from 10 m in Rijpfjorden. Grey shading indicates the estimated maximum extent for winter growth bands. The figure and the caption are from Paper III.

Barium-to-calcium profiles were characterized by low background values broken by abrupt unimodal peaks in 27 of 30 examined shells (Paper IV). These peaks occurred approximately 2.5 months after the peak in phytoplankton bloom in Kongsfjorden and two weeks to a month after the spring bloom in Rijpfjorden. Despite the relatively synchronous timing within fjords, peak values within baskets varied as demonstrated by the high coefficient of variation (24-140%). This and the variable time-lag from bloom between fjords indicated that Ba/Ca peaks cannot be used as a proxy of primary production in studied species, as suggested by early literature (Box 3). None of the environmental factors in our dataset explained variability in Ba/Ca values satisfactorily, marginal R^2 being

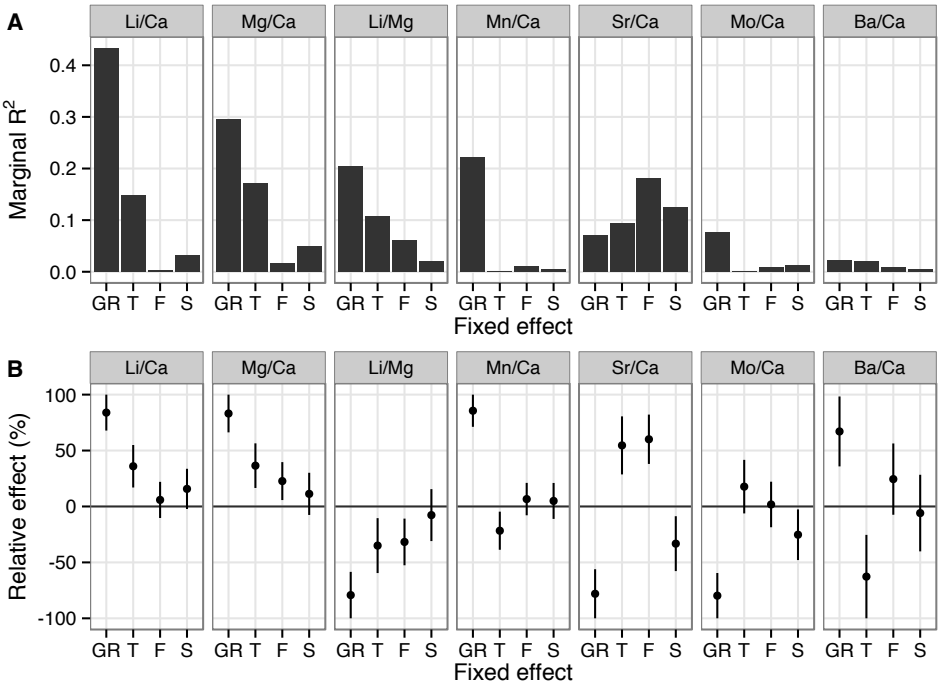

Figure 10: Overall relationships between element ratios and predictor variables (Fixed effect: GR = logarithm of growth rate, T = temperature, F = fluorescence index, and S = salinity) estimated using linear mixed-effect models. **A)** Marginal coefficient of variation indicating the variation in an element ratio explained by a predictor variable across nine growth modeled shells samples from Kongsfjorden and Rijpfjorden. **B)** Relative effect (i.e. the slope using intercepts from the random effect) of predictor variables indicating the relative magnitude and direction of correlations. Error bars represent 95% confidence intervals (CIs) for relative effects. Relative effects are scaled to absolute value of maximum CI. If a CI does not cross the horizontal line at 0, the effect is significantly different from 0 at 95% confidence-level. The figure and caption are from Paper IV.

0.02 for logarithm of shell growth rate and temperature, and 0.01 for salinity and fluorescence (Figure 10). Furthermore, bivalve age, shell height, or length of the growth increment during mooring deployment did not yield significant slopes in a regression model with Ba/Ca peak values, but Ba/Ca peak values were significantly lower in the 25 m basket in Rijpfjorden compared to other baskets. Therefore we were unable to conclude what causes the prominent Ba/Ca peaks in *S. groenlandicus* and *C. ciliatum*. Even though we lack the evidence, it is possible that increased Ba concentration in seawater could contribute to the observed peaks as suggested by Tabouret *et al.* (115). The incorporation could

also be affected by particular Ba or diet particles enriched in Ba (138–141), seasonal Ba dynamics in the water column partly explaining the variable time-lag from bloom between fjords. Nevertheless, the consistent incorporation could be disturbed by bivalve metabolism as Ba/Ca peaks tended to occur simultaneously with fast shell growth periods, and the incorporation further affected by yet unknown processes as suggested by recent studies (137, 142, 187). Despite the unidentified controlling factors, it seems plausible to use Ba/Ca peaks as sub-annual temporal anchors in studied shells, because the deposition likely occurred simultaneously within baskets.

Li/Ca and Mg/Ca values followed patterns in logarithm of growth rate as illustrated by marginal R^2 of 0.43 and 0.30, respectively (Figure 10). These ratios were also linearly related to temperature with marginal R^2 of 0.15 and 0.17 for Li/Ca and Mg/Ca respectively. Li/Ca was consistent among samples allowing the use as a sub-annual temporal anchor, but the relationships with shell growth rate or temperature did not yield consistent enough results for use as a proxy. Variability among individuals in Mg/Ca indicated that this ratio was biologically controlled or affected by shell micro-structure to the extent that it could not be used as a proxy of temperature, shell growth rate, salinity or primary production using LA-ICP-MS.

Samples from a same basket demonstrated variability with respect to Mn/Ca and Sr/Ca maximum and minimum values (see Figure 2 and Table 3 in Paper IV). This indicates that the incorporation of these elemental ratios was likely controlled by metabolic processes to the extent that separating potential environmental signals in these element ratios was difficult. Nevertheless, Mn/Ca profiles demonstrated peaks in 24 of 30 analyzed shells, but these peaks were not as consistent as the Ba/Ca peaks and therefore did not allow the usage as sub-annual temporal anchors. Mo/Ca patterns were similar within baskets, but did not demonstrate meaningful relationships with mooring instrument data (Figure 10).

It should be noted that the bivalves in Paper IV were deployed to oceanographic moorings in open water column and therefore might not have recorded elemental ratios similarly to their natural habitat. The mooring deployment likely excluded the effect of sediment-surface redox-processes, which have been suggested as important contributors for the seasonal dynamics of, at least, Mn (105, 143, 146, 188). Even though LA-ICP-MS sampling was able to capture the Ba/Ca peaks, it is possible that time-averaging contributed to profiles of some elements such that no meaningful environmental correlations were found (118). Also the imprecision in the growth models (Paper III) could have contributed to the relatively low marginal R^2 values as a shift in peak values of elemental ratios by a month could have considerably changed the relationships between elemental ratios and predictor variables.

4 SYNTHESIS

Population- and individual-level responses to environmental perturbations can be used to study the vulnerability of a population to future perturbation scenarios from different angles. Individual-level responses give indications about the possible adaptability of a population to the future environment (Paper I), whereas population-level responses tell about the general sensitivity of a population to the projected perturbation (Paper II). We observed considerable among-individual variability in sperm activity of three bivalve species in response to climate change (Paper I, Section 3.2). These findings together with other recent studies on marine invertebrates (84, 87, 182) indicate that among-individual variability in response to environmental perturbations is likely a norm, rather than an exception, within populations of marine taxa. If this variability is caused by heritable traits, the responses of a species in the future might differ from the average response of a population today, but this might come at the cost of reduced genetic variability.

Today, spatial variability in surface ocean pH varies by 0.6 units (36), and seasonal and diurnal fluctuations can be in the range of 0.5 – over 1.0 pH units, respectively (65, 66, 72). Thus, natural variability in many systems is larger than the open ocean pH decreases projected for 2100 (Box 1), and inter-tidal mussels may be expected to tolerate such fluctuating pH regimes. The results presented in this thesis indicated that although adult and juvenile *Mytilus* are generally robust to the range of average open ocean pH reductions projected to occur by 2100 (Figure 6), sperm activity and fertilization of *M. galloprovincialis* (Papers I-II), and larval stages of the *Mytilus* genus in general (Figure 5, Paper II), are sensitive to such pH reductions. Anthropogenic OA might expose *Mytilus* populations to larger pH fluctuations than they experience today (164), therefore subjecting the populations to a stronger selection pressure. Marine invertebrates undergo several different life-stages on which selection pressure can act differently (54, 67). If genes determining performance at one life-stage are different to those at the next stage, *strong* selection early on during the life-cycle may lead to carry-over effects during the later life-cycle as a form of reduced phenotypic plasticity, hence potentially reducing the persistence of a population to further environmental changes (56, 181, 189).

Due to complex species-interactions (68, 69), natural selection (55), differing responses depending on life-stage (34, 53), and potential effects of other perturbations (70) projecting ocean acidification effects on *Mytilus* genus, and bivalves in general, is not feasible based on the current knowledge: Overarching ecological meaning of the ~ 30% and ~ 50% reductions in sperm swimming speed and

percent motility of *M. galloprovincialis*, respectively (Paper I), as well as the ~ 10% reductions in *Mytilus* larval size (Figure 5) in response to the OA scenarios projected for the year 2100 is difficult to estimate. Repeated studies, population modeling, and a combination of different study approaches, including studies that combine the measured traits with genetics and heritability, are needed to evaluate the potential response mechanisms of populations in the future oceans (e.g. see 190). Nevertheless, results from Paper II suggest that the OA may cause smaller negative effects on *Mytilus* larvae than increased temperature projected for the year 2100.

It should be noted, however, that OA is only one process caused by increasing CO_2 levels in the atmosphere. Larval and adult stages of *Mytilus* might be sensitive to global warming, but the temperature tolerance varies among *Mytilus* species and populations, leading to possible shifts in distribution range of species (Paper II, Section 3.3). Therefore, projecting the consequences of global warming on marine bivalves is more complicated than the effects of ocean acidification. Nevertheless, if temperature will not change outside species' thermal tolerance window and if the food source remains unaffected or positively affected by the climate change, increased temperatures are expected to increase shell growth, and potentially also productivity of bivalve populations (Paper III).

Although not within the scope of data presented in this thesis, the geological record indicates that increases in atmospheric pCO_2 have been involved in mass extinctions previously in Earth's history (191–194). The current trajectory of CO_2 emissions might lead to rates of climate change that are unprecedented in the geologic record, for which sufficiently preserved records are available (11). Together with other anthropogenic perturbations, unabated CO_2 emissions may lead to conditions that cause, if not a mass extinction, at least dramatic changes in marine ecosystems during the coming centuries (8, 9, 191, 193). Although these scenarios are pure speculation due to lack of resolution in geological records comparable with the rate of the projected climate change for the coming centuries, using information from the past to project possible future scenarios deserves more attention. More research efforts should be directed in finding methods that allow interpretation of high-resolution records of climate change in the past, using these records to compare the ecological response mechanisms in the past to the current ecosystems, and finally attempts in projecting the consequences of IPCC climate scenarios to marine ecosystems.

The meta-analysis approach used in this thesis revealed that finding comparable indicator traits among OA studies on *Mytilus* genus is challenging. Traits related to size were the only response variables reported by every study, allowing to generate a meaningful biological response in the meta-analysis (Figures 5-6). Yet, growth is likely not a trait most indicative of reduced fitness in bivalves, nor a trait best reflecting responses in short-term experiments. Furthermore, the studies included in the meta-analysis did not consider the effects of phenotypic

plasticity or adaptive evolution (55, 56). Varying seawater chemistry parameters and exposure times caused difficulties in separating the high variability in responses to species or population-level differences. Despite these shortcomings, short-term experiments are relatively easy to conduct and useful for pinpointing critical thresholds when a current population could become negatively affected by an increased perturbation (1). Such thresholds fall short in their level of realism if the goal is to estimate species success in the future oceans. Nevertheless, these thresholds are useful indicators of sensitivity of species and life-stages to perturbations and might provide clues of relative magnitude of natural selection due to a perturbation. Consequently, it is beneficial to report easily measurable traits, such as growth, along with traits that are better indicators of OA effects in future studies.

Whereas individual variability in bivalve populations potentially increases persistence of a population to environmental perturbations, it complicates the interpretation of environmental information recorded in bivalve shells. Results from Paper III demonstrated large variability in growth rate of *S. groenlandicus* and *C. ciliatum* within season, between fjords, and among individuals. Understanding such growth patterns is a prerequisite for using bivalve shells as sub-annual proxy archives. Our results indicated that the prominent growth bands are deposited during the slower growth period in winter. Furthermore, the time of the winter growth line deposition is likely to reflect low food availability. The insights on winter growth line deposition in two circumpolar bivalve species commonly used as environmental proxies (e.g. see 99, 195, 196) are a good starting point for further environmental proxy studies.

Separating individual variability, sampling bias, and effects of multiple environmental and chemical factors from elemental ratio signal was challenging (Paper IV). Consequently, we were unable to conclude whether our negative results were related to sampling bias and whether the studied element ratios could work as proxies, if for instance organic matter was removed prior ICP-MS analyses. Nevertheless, further efforts studying Li/Ca, Mg/Ca and Ba/Ca as environmental proxies in Arctic bivalve shells are recommended. Furthermore, high-latitude bivalve species that deposit a calcitic shell, such as *C. islandica*, should be evaluated for elemental ratios, as Mg/Ca appears to exhibit stronger correlations with seawater temperature in calcitic bivalve shells (Paper IV). The ongoing mooring project on Svalbard (Papers III-IV) combined with a high-frequency non-invasive valvometry (197) to record *in situ* shell growth would offer an optimal setup for such studies. The tool to align sample spots along chronologically deposited material along the growth axis, as developed for this thesis, is a stating point for an improved identification of time-averaging error and bias caused by the inconsistent location of sampling spots in relation to growth lines (123). Nevertheless, this tool cannot identify the sampling bias caused by inconsistencies in the shell matrix (see Section 1.3). Experiments controlling ambient element concentration together with environmental factors (115) and diet (198) are needed to further

evaluate the veracity of element ratios as environmental proxies in *S. groenlandicus* and *C. ciliatum* shells. This thesis demonstrates the need for understanding seasonal dynamics of elements in the water column because many elemental ratios might be deposited in a relationship with seawater concentration. Furthermore, many elemental ratios in bivalve shells reflect the metabolism of the organism. Some studies have demonstrated that consistent metabolic processes interpreted from elemental ratios in bivalve shells can be used as proxies of the environment (e.g. 116). Therefore, finding relationships between environment and metabolic processes of bivalves that are reflected in elemental ratios could be beneficial for the field.

4.1 CONCLUDING REMARKS: BEYOND NEGATIVITY

Publishing negative results (i.e. studies that confirm the null-hypothesis) is important for advancement of science (199): such results might initiate improved solutions, increase understanding of nature, and avoid duplication of efforts that do not advance scientific fields of study. In this thesis, I presented two papers with partially negative results. In Paper II we used a factor with an expected strong effect as a scale to demonstrate that increased pCO_2 had a smaller effect on larval performance than temperature. The meta-analysis was necessary to examine whether our non-significant acidification results could be explained by differences in the experimental design – and they could, but the answer was not conclusive (see section 3.1). Consequently, a dataset that at a first glance seemed to lack any exciting effects led to an improved understanding of variability among experiments and populations. In Paper IV, we could not identify useful all-encompassing elemental ratio proxies of the ambient environment. Nevertheless, the study demonstrated that some element ratios, such as Li/Ca and Ba/Ca, are likely deposited simultaneously across individuals from a same location, and that more controlled experiments are needed to identify the potential multiple factors affecting the elemental ratios.

Importantly, it should be remembered that perturbations with no effects on a population are ecologically as important to identify as perturbations that have an effect. Consequently, despite the generic name, negative results are not always negative, but they merely confirm the complexity, uncertainty, and variability of nature – aspects that should excite rather than repulse scientists. The most valuable lesson I learned during my PhD period is to see beyond negativity, because negativity exists only in our minds. Complex, unexpected or inconclusive results must be seen as an opportunity to understand more about the world around us leading to improved interpretations, methods, and study designs in the long run.

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ABBREVIATIONS

AR5 ..... IPCC Fifth Assessment Report (see Box 1)

CI(s) ..... confidence interval(s)

CMIP5 ..... Coupled Model Intercomparison Project Phase 5 (see Box 1)

$CaCO_3$ ..... calcium carbonate

$\delta^{18}O$ ..... a measure of the ratio of oxygen isotopes 16 and 18 (see Box 3)

ΔpH ..... pH difference between future and the present day values (average of 1986-2005) for climate scenarios or between treatment and control for experiments

DTW ..... dynamic time warping [see SI1 for Paper III and Giorgino (150)]

EPF ..... extrapallial fluid

FSR_{50} ..... fertilization success that yields 50% of theoretical maximum fertilization success in control conditions (see Box 2)

IPCC ..... Intergovernmental Panel on Climate Change

LA-ICP-MS . laser-ablation inductively-coupled-plasma mass-spectrometry

LMM(s) ..... linear mixed-effects model(s)

Ma ..... unit of time, million years before present

Ω ..... saturation state of calcium carbonate

OA ..... ocean acidification (see Box 1)

pCO_2 ..... partial pressure of carbon dioxide

RCPs ..... Representative Concentration Pathways. Model scenarios based on future RF used in IPCC AR5 (see Box 1)

RF ..... radiative forcing ($W\,m^{-2}$, see Box 1)

SIMS ..... secondary ion mass spectrometry

GLOSSARY

Definitions in science tend to be flexible and depend on context. In this thesis I have selected definitions from the literature whenever possible with source in brackets after a definition. Several definitions are specific for population- or individual-level. In such cases the level is underlined.

ACCLIMATION reversible process whereby an individual organism adjusts to short-term changes in the environment, for instance through physiology, morphology, or behavior. Refers often to experimental conditions, but can as well occur in the natural habitat.

ACCLIMATIZATION a long-term process by which an individual organism adjusts to environmental conditions. Refers often to conditions experienced in the natural habitat, but can also occur under experimental conditions.

ADAPTABILITY also persistence. The capability of a population or species to persist and adapt to environmental changes through plasticity and evolution.

ADAPTATION genetic-based changes in a population increasing fitness in the new environment.

BIOGENIC CARBONATE calcium carbonate mineralized by marine organisms.

BIOMINERALIZATION formation of minerals, such as $CaCO_3$, by organisms (102).

CLIMATE CHANGE referred solely as a combination of ocean acidification and warming throughout the thesis (Box 1).

EPIGENETIC changes in gene expression and regulation through chemical reactions as a response to development, physiology or environment.

EVOLUTION any change in DNA sequences within a population that is inherited from one generation to the next (55).

- ADAPTIVE * evolution that is driven by natural selection and mixing of already existing genetic material. Excludes genetic mutation (56).

FITNESS also Darwinian fitness. The capacity of an individual organism to survive and reproduce in a particular environment.

GENOTYPE the genetic composition of an individual.

INDICATOR SPECIES an organism that can be used to infer conditions or population responses in a particular habitat.

KINETIC PROCESS element partitioning dependent on crystal growth rate of e.g. $CaCO_3$ minerals.

LATTICE-BOUND ELEMENT an ion that substitutes Ca^{2+} in $CaCO_3$ matrix.

LIFE CYCLE also life history. The series of changes in form during the life of an organism. Includes reproduction.

LIFE-STAGE also life history stage. A single form or phase of an organism during the life cycle.

NATURAL SELECTION also selection. The process by which organisms better adapted to their environment tend to survive and produce more offspring.

PHENOTYPE a set of traits of an individual resulting from the interaction of its genotype with the environment.

PLASTICITY the capacity of an individual to persist in a new environment through acclimation and acclimatization. Does not involve changes in DNA sequences.

- PHENOTYPIC * "the capacity of individual genotypes to produce different phenotypes when exposed to different environmental conditions" — Munday *et al.* (55).
- DEVELOPMENTAL * irreversible phenotypic plasticity resulting from environmental cues experienced during the development of an organism (55, 200).
- TRANSGENERATIONAL * "phenotypic plasticity resulting from environmental cues experienced during the parental, or previous, generations. It can occur via epigenetic inheritance or through the transmission of nutrition, proteins, hormones or other bioactive materials from parents to their offspring." — Munday *et al.* (55).

POPULATION SENSITIVITY relative consequences of changing environmental factors for a population or species. Typically identified through acute short term experiments. Does not consider population's capacity for adaptation (adaptability).

PRECIPITATION (in biomineralization context). Mineralization of elements to $CaCO_3$ skeletons of marine organisms usually taking place in a chemically and metabolically controlled space.

PROXY a measurable parameter that bears a known relationship to an environmental factor or process of interest.

* RECORD also proxy archive. A temporally resolved material, such as a calcium carbonate skeleton, containing proxy information. Can also consist of several cross-dated pieces of material.

* SIGNAL measured pattern in proxy data excluding signal noise.

SIGNAL NOISE variability in proxy signal that cannot be attributed to defined predictor variables.

TRAIT also phenotypic trait. A measurable or observable characteristic in an individual organism that is a product of the interaction between genotype and the environment.

INDICATOR * a trait that has traditionally been used in experiments to quantify responses to environmental perturbations.

VITAL EFFECTS undefined factors contributing to signal noise that cannot be attributed to predefined predictor variables.

VULNERABILITY also susceptibility. A general estimate indicating how susceptible a population or species could be to changes in the environment. In contrast to population sensitivity, vulnerability considers adaptability of a population.

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5

PAPERS

Paper I

Variable individual- and species-level responses to ocean acidification

Vihtakari, M.; Havenhand, J.N.; Renaud, P.E.; Hendriks I.E.

Under revision in *PLoS One*.

Paper II

Effects of ocean acidification and warming on sperm activity and early life stages of the Mediterranean mussel (*Mytilus galloprovincialis*)

Vihtakari, M.; Hendriks I.E.; Holding J.; Renaud, P.E.; Duarte C.M.; Havenhand, J.N.

Water, **2013**, 5, 1890-1915.

Paper III

Decoding the oxygen isotope signal for seasonal growth patterns in Arctic bivalves

Vihtakari, M.; Renaud, P.E.; Clarke, L.J.; Whitehouse, M.J.; Hop, H.; Carroll, M.L.; Ambrose Jr, W.G.

Manuscript.

Paper IV

Interpreting the seasonal environmental history recorded by Arctic bivalves

Vihtakari, M.; Ambrose Jr, W.G.; Renaud, P.E.; Locke V, W.L.; Berge, J.; Cottier, F.; Hop, H.; Carroll, M.L.; Clarke, L.J.

Manuscript.

APPENDIX

Table A1: Data extracted for *Mytilus* meta-analysis. Species: MG = *M. galloprovincialis*, ME = *M. edulis*, MCa = *M. californianus*, MT = *M. trossulus*, MCh = *M. chilensis*; Stage = life stage (Larv = larvae, Juv = juvenile, Ad = adult); Time = duration of experiment in decimal days; Equivalent to time from fertilization in larvae; pH_C = pH in control ($\star$ = total scale, † = seawater scale, otherwise on NBS scale); pH_T = pH in treatment (on the same scale than control); ΔpH = difference in pH between control and treatment; Temp = temperature in °C; Resp = response variable (L = shell length, A = shell area, Li = increase in shell length, LRi = Relative increase in shell length, Wi = increase in weight, DWi = increase in dry weight of shell weight); Unit = unit of measurement for the response variable (iL = initial shell length); N = number of relevant experimental replicates; LnRR = back-calculated logarithm transformed response ratio in %; CI_{min} and CI_{max} = minimum and maximum 95% confidence intervals (CI) for the response ratio, respectively; Sign: indicating whether the response ratio was significant (+ = significant, – = non-significant, empty = insufficient replication to calculate CIs.)

Reference	Species	Stage	Time	pH_C	pH_T	ΔpH	Temp	Resp	Unit	N	LnRR	CI_{min}	CI_{max}	Sign
Frieder *et al.* (180)	MCa	Larv	2	8.04*	7.51	0.53	16.5	L	µm	3	91.4	89.4	93.5	+
Frieder *et al.* (180)	MCa	Larv	4	8.04*	7.51	0.53	16.5	L	µm	3	93.7	92.5	95.0	+
Frieder *et al.* (180)	MCa	Larv	6	8.04*	7.51	0.53	16.5	L	µm	3	93.0	90.6	95.5	+
Frieder *et al.* (180)	MCa	Larv	8	8.04*	7.51	0.53	16.5	L	µm	3	93.9	91.9	95.9	+
Gaylord *et al.* (201)	MCa	Larv	5	8.07	7.95	0.12	15.4	A	mm^2	6	101.8	99.7	103.9	–
Gaylord *et al.* (201)	MCa	Larv	5	8.07	7.75	0.32	15.4	A	mm^2	6	93.5	90.6	96.6	+
Gaylord *et al.* (201)	MCa	Larv	8	8.07	7.95	0.12	15.4	A	mm^2	6	98.6	95.9	101.3	–
Gaylord *et al.* (201)	MCa	Larv	8	8.07	7.75	0.32	15.4	A	mm^2	6	95.7	93.0	98.6	+
Bechmann *et al.* (202)	ME	Larv	3	8.06	7.62	0.44	10	A	mm^2	7	85.7	72.9	100.8	–
Bechmann *et al.* (202)	ME	Larv	16	8.06	7.62	0.44	10	A	mm^2	7	91.7	75.5	111.2	–
Bechmann *et al.* (202)	ME	Larv	64	8.06	7.62	0.44	10	A	mm^2	7	71.9	47.6	108.4	–
Gazeau *et al.* (203)	ME	Larv	2	8.03	7.78	0.25	19.4	L	µm	3	100.0	86.7	115.3	–
Gazeau *et al.* (203)	ME	Larv	2	8.15	7.81	0.34	16.5	L	µm	3	95.5	94.1	96.8	+
Gazeau *et al.* (203)	ME	Larv	2	8.09	7.58	0.51	16.5	L	µm	3	87.3	86.3	88.2	+
Gazeau *et al.* (203)	ME	Larv	6	8.03	7.78	0.25	19.4	L	µm	3	95.7	91.0	100.7	–
Gazeau *et al.* (203)	ME	Larv	8	8.03	7.78	0.25	19.4	L	µm	3	96.4	92.9	100.0	+

Table A1: (continued)

Reference	Species	Stage	Time	pH_C	pH_T	ΔpH	Temp	Resp	Unit	N	LnRR	CI_{min}	CI_{max}	Sign
Gazeau *et al.* (203)	ME	Larv	10	8.03	7.78	0.25	19.4	L	µm	3	96.9	94.2	99.8	+
Gazeau *et al.* (203)	ME	Larv	13	8.03	7.78	0.25	19.4	L	µm	3	93.0	86.1	100.5	–
Gazeau *et al.* (203)	ME	Larv	15	8.03	7.78	0.25	19.4	L	µm	3	94.0	89.0	99.2	+
Frieder *et al.* (180)	MG	Larv	2	7.95*	7.48	0.47	15.7	L	µm	3	92.2	87.5	97.2	+
Frieder *et al.* (180)	MG	Larv	4	7.95*	7.48	0.47	15.7	L	µm	3	95.7	89.8	102.0	–
Frieder *et al.* (180)	MG	Larv	6	7.95*	7.48	0.47	15.7	L	µm	3	94.7	89.8	99.7	+
Frieder *et al.* (180)	MG	Larv	8	7.95*	7.48	0.47	15.7	L	µm	3	92.5	88.1	97.0	+
Kurihara *et al.* (204)	MG	Larv	2.2	8.05	7.43	0.62	13	L	µm	5	67.8	64.5	71.2	+
Kurihara *et al.* (204)	MG	Larv	5	8.05	7.43	0.62	13	L	µm	5	74.1	69.8	78.6	+
Kurihara *et al.* (204)	MG	Larv	6	8.05	7.43	0.62	13	L	µm	5	74.8	70.2	79.8	+
Paper II	MG	Larv	2.4	7.97*	7.60	0.37	16.8	L	µm	3	95.9	89.7	102.5	–
Paper II	MG	Larv	2.4	8.02*	7.65	0.37	19.6	L	µm	3	101.7	94.0	110.1	–
Sunday *et al.* (182)	MT	Larv	2.7	8.30	7.90	0.40	12	L	µm	10	96.9	96.3	97.5	+
Duarte *et al.* (171)	MCh	Juv	60	8.06*	7.84	0.22	12	Wi	% d^{-1}	5	70.1	53.5	91.8	+
Duarte *et al.* (171)	MCh	Juv	60	8.00*	7.78	0.22	16	Wi	% d^{-1}	5	87.7	59.3	129.8	–
Duarte *et al.* (171)	MCh	Juv	60	8.06*	7.67	0.38	12	Wi	% d^{-1}	5	66.3	49.1	89.6	+
Duarte *et al.* (171)	MCh	Juv	60	8.00*	7.62	0.38	16	Wi	% d^{-1}	5	64.5	47.1	88.2	+
Berge *et al.* (205)	ME	Juv	44	8.10	7.60	0.50	19.1	Li	µm d^{-1}	1	144.4			
Berge *et al.* (205)	ME	Juv	44	8.10	7.40	0.70	19.1	Li	µm d^{-1}	1	93.2			
Berge *et al.* (205)	ME	Juv	44	8.10	7.10	1.00	19.1	Li	µm d^{-1}	1	31.5			
Berge *et al.* (205)	ME	Juv	44	8.10	6.70	1.40	19.1	Li	µm d^{-1}	1	7.8			
Hiebenthal *et al.* (173)	ME	Juv	91	7.85	7.76	0.09	7.5	Li	µm d^{-1}	4	79.9	63.3	100.9	–
Hiebenthal *et al.* (173)	ME	Juv	91	7.98	7.86	0.12	25	Li	µm d^{-1}	4	86.5	53.0	141.3	–
Hiebenthal *et al.* (173)	ME	Juv	91	7.96	7.82	0.14	16	Li	µm d^{-1}	4	79.5	48.2	131.0	–

Table A1: (continued)

Reference	Species	Stage	Time	pH_C	pH_T	ΔpH	Temp	Resp	Unit	N	LnRR	CI_{min}	CI_{max}	Sign
Hiebenthal *et al.* (173)	ME	Juv	91	7.96	7.80	0.16	10	Li	$\mu m\,d^{-1}$	4	118.8	81.2	173.9	–
Hiebenthal *et al.* (173)	ME	Juv	91	8.01	7.82	0.19	20	Li	$\mu m\,d^{-1}$	4	74.7	51.3	108.7	–
Hiebenthal *et al.* (173)	ME	Juv	91	7.85	7.63	0.22	7.5	Li	$\mu m\,d^{-1}$	4	111.4	87.5	142.0	–
Hiebenthal *et al.* (173)	ME	Juv	91	7.96	7.72	0.24	16	Li	$\mu m\,d^{-1}$	4	104.7	68.6	159.7	–
Hiebenthal *et al.* (173)	ME	Juv	91	7.98	7.73	0.25	25	Li	$\mu m\,d^{-1}$	4	5.6	1.0	29.6	+
Hiebenthal *et al.* (173)	ME	Juv	91	8.01	7.73	0.28	20	Li	$\mu m\,d^{-1}$	4	70.4	41.7	118.8	–
Hiebenthal *et al.* (173)	ME	Juv	91	7.96	7.67	0.29	10	Li	$\mu m\,d^{-1}$	4	95.6	71.3	128.3	–
Thomsen *et al.* (163)	ME	Juv	60	8.13	7.72	0.41	13.8	Li	$\mu m\,d^{-1}$	4	103.4	84.3	126.8	–
Thomsen *et al.* (163)	ME	Juv	60	8.13	7.26	0.87	13.8	Li	$\mu m\,d^{-1}$	4	76.5	56.7	103.2	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.70	0.31	17.2	Li	$\mu m\,d^{-1}$	7	138.6	99.1	193.9	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.70	0.31	17.2	Li	$\mu m\,d^{-1}$	7	129.3	89.5	186.8	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.70	0.31	17.2	Li	$\mu m\,d^{-1}$	7	84.2	68.6	103.4	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.40	0.61	17.2	Li	$\mu m\,d^{-1}$	7	103.4	74.1	144.2	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.40	0.61	17.2	Li	$\mu m\,d^{-1}$	7	105.1	71.0	155.4	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.40	0.61	17.2	Li	$\mu m\,d^{-1}$	7	88.2	71.3	109.1	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.19	0.82	17.2	Li	$\mu m\,d^{-1}$	7	86.4	65.2	114.3	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.19	0.82	17.2	Li	$\mu m\,d^{-1}$	7	80.3	53.5	120.6	–
Thomsen *et al.* (71)	ME	Juv	49	8.01*	7.19	0.82	17.2	Li	$\mu m\,d^{-1}$	7	85.5	68.3	107.1	–
Bressan *et al.* (168)	MG	Juv	202	8.21	7.41	0.80	11.9	Li	$\mu m\,d^{-1}$	3	82.5	51.1	133.3	–
Bressan *et al.* (168)	MG	Juv	94	8.23†	7.39	0.84	10.8	Li	$\mu m\,d^{-1}$	1	66.4			
Range *et al.* (178)	MG	Juv	84	7.88*	7.59	0.29	17.1	Li	$\mu m\,d^{-1}$	3	106.4	91.5	123.6	–
Range *et al.* (178)	MG	Juv	84	7.88*	7.31	0.57	17.1	Li	$\mu m\,d^{-1}$	3	108.9	88.6	133.8	–
Heinemann (206)	ME	Juv/Ad	110	8.02	7.93	0.09	12.1	Li	$\mu m\,d^{-1}$	1	124.0			
Heinemann (206)	ME	Juv/Ad	110	8.02	7.86	0.16	12.1	Li	$\mu m\,d^{-1}$	1	114.0			

Table A1: (continued)

Reference	Species	Stage	Time	pH_C	pH_T	ΔpH	Temp	Resp	Unit	N	LnRR	CI_{min}	CI_{max}	Sign
Heinemann (206)	ME	Juv/Ad	110	8.02	7.77	0.25	12.1	Li	$\mu m\,d^{-1}$	1	120.0			
Heinemann (206)	ME	Juv/Ad	110	8.02	7.71	0.31	12.1	Li	$\mu m\,d^{-1}$	1	142.0			
Heinemann (206)	ME	Juv/Ad	110	8.02	7.28	0.74	12.1	Li	$\mu m\,d^{-1}$	1	92.0			
Michaelidis *et al.* (207)	MG	Juv/Ad	90	8.05	7.30	0.75	18	Li	$\mu m\,d^{-1}$	1	45.0			
Duarte *et al.* (172)	MCh	Ad	20	8.05*	7.85	0.20	16	DWi	$\mu g\,d^{-1}$	5	87.4	71.3	107.0	–
Duarte *et al.* (172)	MCh	Ad	20	8.05*	7.85	0.20	16	DWi	$\mu g\,d^{-1}$	5	66.6	47.5	93.3	+
Duarte *et al.* (172)	MCh	Ad	20	8.05*	7.70	0.35	16	DWi	$\mu g\,d^{-1}$	5	96.3	74.5	124.5	–
Duarte *et al.* (172)	MCh	Ad	20	8.05*	7.70	0.35	16	DWi	$\mu g\,d^{-1}$	5	63.1	50.4	79.0	+
Berge *et al.* (205)	ME	Ad	44	8.10	7.60	0.50	19.1	Li	$\mu m\,d^{-1}$	1	78.2			
Berge *et al.* (205)	ME	Ad	44	8.10	7.40	0.70	19.1	Li	$\mu m\,d^{-1}$	1	81.9			
Berge *et al.* (205)	ME	Ad	44	8.10	7.10	1.00	19.1	Li	$\mu m\,d^{-1}$	1	49.1			
Berge *et al.* (205)	ME	Ad	44	8.10	6.70	1.40	19.1	Li	$\mu m\,d^{-1}$	1	1.0			
Keppel *et al.* (175)	ME	Ad	70	8.10*	7.94	0.16	20	Li	$\mu m\,d^{-1}$	4	450.0	142.3	1423.0	+
Keppel *et al.* (175)	ME	Ad	70	8.10*	7.94	0.16	24	Li	$\mu m\,d^{-1}$	4	127.3	39.5	410.2	–
Melzner *et al.* (208)	ME	Ad	45	8.01	7.69	0.32	5.4	Li	$\mu m\,d^{-1}$	4	125.0	87.4	178.8	–
Melzner *et al.* (208)	ME	Ad	45	8.01	7.69	0.32	4.8	Li	$\mu m\,d^{-1}$	4	115.4	68.4	194.5	–
Melzner *et al.* (208)	ME	Ad	45	8.01	7.35	0.66	5.4	Li	$\mu m\,d^{-1}$	4	91.7	63.8	131.7	–
Melzner *et al.* (208)	ME	Ad	45	8.01	7.35	0.66	4.8	Li	$\mu m\,d^{-1}$	4	96.2	59.6	155.2	–
Melzner *et al.* (208)	ME	Ad	45	8.01	7.15	0.86	5.4	Li	$\mu m\,d^{-1}$	4	50.0	33.2	75.2	+
Melzner *et al.* (208)	ME	Ad	45	8.01	7.15	0.86	4.8	Li	$\mu m\,d^{-1}$	4	69.2	40.8	117.4	–
Ries *et al.* (209)	ME	Ad	60	8.15	8.02	0.13	25	DWi	$\mu g\,d^{-1}$	1	73.1			
Ries *et al.* (209)	ME	Ad	60	8.15	7.83	0.32	25	DWi	$\mu g\,d^{-1}$	1	73.9			
Ries *et al.* (209)	ME	Ad	60	8.15	7.45	0.70	25	DWi	$\mu g\,d^{-1}$	1	64.7			

Table A1: (continued)

Reference	Species	Stage	Time	pH_C	pH_T	ΔpH	Temp	Resp	Unit	N	LnRR	CI_{min}	CI_{max}	Sign
Thomsen and Melzner (210)	ME	Ad	56	8.03	7.70	0.33	8.8	Li	µm d^{-1}	4	92.7	81.9	104.9	–
Thomsen and Melzner (210)	ME	Ad	56	8.03	7.38	0.65	8.8	Li	µm d^{-1}	4	86.9	82.7	91.2	+
Thomsen and Melzner (210)	ME	Ad	56	8.03	7.14	0.89	8.8	Li	µm d^{-1}	4	78.8	74.3	83.5	+
Thomsen *et al.* (163)	ME	Ad	60	8.13	7.72	0.41	13.8	Li	µm d^{-1}	4	99.9	91.2	109.6	–
Thomsen *et al.* (163)	ME	Ad	60	8.13	7.26	0.87	13.8	Li	µm d^{-1}	4	76.2	67.4	86.0	+
Kroeker *et al.* (174)	MG	Ad	28	7.95*	7.67	0.28	16	LRi	µm d^{-1} / iL	3	89.5	57.8	138.6	–
Kroeker *et al.* (174)	MG	Ad	28	7.99*	7.70	0.29	24	LRi	µm d^{-1} / iL	3	99.2	75.6	130.2	–
Kroeker *et al.* (174)	MG	Ad	28	7.96*	7.66	0.30	20	LRi	µm d^{-1} / iL	3	96.0	73.4	125.5	–
Kroeker *et al.* (174)	MG	Ad	28	7.97*	7.66	0.31	12	LRi	µm d^{-1} / iL	3	113.6	81.1	159.1	–
Kroeker *et al.* (174)	MG	Ad	28	7.96*	7.65	0.31	18	LRi	µm d^{-1} / iL	3	105.1	83.4	132.5	–
Kroeker *et al.* (174)	MG	Ad	28	7.99*	7.67	0.32	14	LRi	µm d^{-1} / iL	3	58.9	38.2	90.6	+

Table A2: Articles reviewed for the element ratio figure. Columns from the left: El = Element-to-calcium ratio; $CaCO_3$ = type of $CaCO_3$ mineral (Ar = aragonite, Ca = calcite); 1. control = the most important factor controlling element-to-calcium deposition. Growth rate includes both shell growth and kinetic processes; 2. control = the second most important factor; 3. control = third most important control; Proxy = Number indicating whether the article concludes if corresponding element-to-calcium ratio could be used as a proxy (regardless of the article identifying the proxy relationship). 1 = "yes", 0.5 = "maybe", 0 ="no", missing value = insufficient data.

Reference	El	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Paper IV	Li	*Serripes groenlandicus*	Ar	Growth rate	Temperature		1
Paper IV	Li	*Ciliatocardium ciliatum*	Ar	Growth rate	Temperature		1
Thébault *et al.* (145)	Li	*Arctica islandica*	Ar	Growth rate	Import from land		1
Thébault and Chauvaud (211)	Li	*Pecten maximus*	Ca	Growth rate	Temperature	Primary production	1
Carre *et al.* (113)	Mg	*Mesodesma donacium*	Ar	Unknown			0
Carre *et al.* (113)	Mg	*Chione subrugosa*	Ar	Unknown			0
Carroll *et al.* (212)	Mg	*Serripes groenlandicus*	Ar	Vital effects			0
Dodd (213)	Mg	*Mytilus californianus*	Ca	Vital effects			0
Dodd (213)	Mg	*Mytilus edulis*	Ca	Vital effects			0
Dodd (213)	Mg	*Mytilus edulis*	Ca	Salinity	Vital effects		1
Dodd (213)	Mg	*Mytilus galloprovincialis*	Ca	Vital effects	Temperature		1
Dodd and Crisp (214)	Mg	Several species	Ca	Vital effects	Unknown	Salinity	0.5
Dodd and Crisp (214)	Mg	Several species	Ar	Vital effects	Unknown	Salinity	0.5
Paper IV	Mg	*Serripes groenlandicus*	Ar	Growth rate	Vital effects	Temperature	0
Paper IV	Mg	*Ciliatocardium ciliatum*	Ar	Growth rate	Vital effects	Temperature	0

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Elliot *et al.* (215)	Mg	*Tridacna gigas*	Ar	Ontogeny	Temperature	Vital effects	0
Foster *et al.* (216)	Mg	*Arctica islandica*	Ar	Vital effects			0
Freitas *et al.* (217)	Mg	*Pinna nobilis*	Ca	Temperature	Ontogeny	Vital effects	1
Freitas *et al.* (218)	Mg	*Pecten maximus*	Ca	Vital effects	Temperature		0
Freitas *et al.* (219)	Mg	*Pecten maximus*	Ca	Heterogenity	Vital effects	Temperature	0
Freitas *et al.* (219)	Mg	*Mytilus edulis*	Ca	Vital effects	Temperature		0
Freitas *et al.* (220)	Mg	*Mytilus edulis*	Ca	Vital effects			0
Freitas *et al.* (220)	Mg	*Pecten maximus*	Ca	Temperature			1
Freitas *et al.* (134)	Mg	*Pecten maximus*	Ca	Temperature	Growth rate	Bound to other elements	0.5
Heinemann *et al.* (221)	Mg	*Mytilus edulis*	Ca	Growth rate	Vital effects		0
Immenhauser *et al.* (222)	Mg	*Rudist; Vaccinites*	Ca	Vital effects	Temperature		0
Izumida *et al.* (223)	Mg	*Hyriopsis* sp.	Ar	Vital effects			0
Jimenez-Berrocoso *et al.* (224)	Mg	*Atrina rigida*	Ca	Growth rate	Temperature		1
Klein *et al.* (130)	Mg	*Mytilus trossulus*	Ca	Temperature	Vital effects		1
Lazareth *et al.* (131)	Mg	*Isognomon ephippium*	Ca	Temperature	Vital effects		1
Lazareth *et al.* (135)	Mg	*Protothaca thaca*	Ar	Heterogenity			0
Lorens and Bender (225)	Mg	*Mytilus edulis*	Ca	Vital effects			0
Lorens and Bender (225)	Mg	*Mytilus edulis*	Ar	Concentration in ambient water			0.5
Lorrain *et al.* (226)	Mg	*Pecten maximus*	Ca	Vital effects			0
Mouchi *et al.* (227)	Mg	*Crassostrea gigas*	Ca	Vital effects	Ontogeny	Temperature	0.5

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Mouchi *et al.* (227)	Mg	*Ostrea edulis*	Ca	Vital effects	Ontogeny		0
Pearce and Mann (228)	Mg	*Ensis siliqua*	Ar	Temperature	Ontogeny	Vital effects	1
Pérez-Huerta *et al.* (229)	Mg	*Trachycardium procerum*	Ar	Vital effects	Timing of SST shifts		0.5
Richardson (132)	Mg	*Pinna nobilis*	Ca	Temperature			1
Sano *et al.* (116)	Mg	*Tridacna derasa*	Ar	Vital effects	Temperature		0
Schöne *et al.* (117)	Mg	*Arctica islandica*	Ar	Bound to organic matrix	Vital effects	Temperature	0
Schöne *et al.* (133)	Mg	*Arctica islandica*	Ar	Vital effects	Ontogeny	Temperature	1
Schöne *et al.* (114)	Mg	*Arctica islandica*	Ar	Vital effects			0
Shirai *et al.* (230)	Mg	*Bathymodiolus platifrons*	Ar	Vital effects	Import from land		
Silina (231)	Mg	*Atrina vexillum*	Ca	Growth rate	Ontogeny	Temperature	0
Steuber and Rauch (232)	Mg	*Rudists; several species*	Ca	Temperature	Vital effects	Concentration in ambient water	0
Strasser *et al.* (233)	Mg	*Mya arenaria*	Ar	Vital effects			0
Surge and Lohmann (234)	Mg	*Crassostrea virginica*	Ca	Vital effects	Temperature	Concentration in ambient water	0.5
Takesue and van Geen (235)	Mg	*Protothaca staminea*	Ar	Vital effects	Temperature		0
Takesue *et al.* (236)	Mg	*Corbula amurensis*	Ar	Bound to organic matrix			0
Toland *et al.* (237)	Mg	*Arctica islandica*	Ar	Vital effects			0
Ullmann *et al.* (238)	Mg	*Crassostrea gigas*	Ca	Temperature	Concentration in ambient water		1

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Vander Putten *et al.* (239)	Mg	*Mytilus edulis*	Ca	Vital effects	Temperature		0
Wanamaker *et al.* (240)	Mg	*Mytilus edulis*	Ca	Vital effects	Temperature	Salinity	1
Wisshak *et al.* (241)	Mg	*Neopycnodonte zibrowii*	Ca	Vital effects			0
Barats *et al.* (146)	Mn	*Pecten maximus*	Ca	Concentration in ambient water			1
Carre *et al.* (113)	Mn	*Mesodesma donacium*	Ar	Growth rate			0
Carre *et al.* (113)	Mn	*Chione subrugosa*	Ar	Growth rate			0
Carroll *et al.* (212)	Mn	*Serripes groenlandicus*	Ar	Primary production			1
Dunca *et al.* (242)	Mn	*Arctica islandica*	Ar	Bound to organic matrix	Anoxia		
Paper IV	Mn	*Serripes groenlandicus*	Ar	Growth rate	Vital effects	Primary production	0
Paper IV	Mn	*Ciliatocardium ciliatum*	Ar	Growth rate	Vital effects	Primary production	0
Freitas *et al.* (218)	Mn	*Pecten maximus*	Ca	Concentration in ambient water	Vital effects		1
Freitas *et al.* (220)	Mn	*Mytilus edulis*	Ca	Vital effects			0
Freitas *et al.* (220)	Mn	*Pecten maximus*	Ca	Vital effects			0
Krause-Nehring *et al.* (142)	Mn	*Arctica islandica*	Ar	Primary production	Element flux		1
Langlet *et al.* (243)	Mn	*Pleiodon spekii*	Ar	Concentration in ambient water			1
Lazareth *et al.* (131)	Mn	*Isognomon ephippium*	Ca	Import from land	Primary production		1
Shirai *et al.* (230)	Mn	*Bathymodiolus platifrons*	Ar	Vital effects	Import from land		
Strasser *et al.* (233)	Mn	*Mya arenaria*	Ar	Ontogeny	Vital effects	Salinity	0

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Takesue *et al.* (236)	Mn	*Corbula amurensis*	Ar	Bound to organic matrix			0
Ullmann *et al.* (238)	Mn	*Crassostrea gigas*	Ca	Vital effects	Concentration in ambient water		0.5
Vander Putten *et al.* (239)	Mn	*Mytilus edulis*	Ca	Primary production	Concentration in ambient water		1
Ambrose Jr *et al.* (244)	Sr	*Serripes groenlandicus*	Ar	Growth rate			0.5
Bailey and Lear (245)	Sr	*Margaritifera margaritifera*	Ar	Vital effects			0
Cardoso *et al.* (246)	Sr	*Macoma balthica*	Ar	Vital effects			0
Carre *et al.* (113)	Sr	*Mesodesma donacium*	Ar	Growth rate			0
Carre *et al.* (113)	Sr	*Chione subrugosa*	Ar	Growth rate			0
Carroll *et al.* (212)	Sr	*Serripes groenlandicus*	Ar	Vital effects			0
Dodd (213)	Sr	*Mytilus californianus* & *M. galloprovincialis*	Ca	Growth rate	Temperature	Ontogeny	1
Dodd (213)	Sr	*Mytilus californianus* & *M. galloprovincialis*	Ar	Vital effects	Temperature	Ontogeny	1
Dodd and Crisp (214)	Sr	Several species	Ca	Vital effects	Unknown	Salinity	0.5
Dodd and Crisp (214)	Sr	Several species	Ar	Vital effects	Unknown	Salinity	0.5
Dunca *et al.* (242)	Sr	*Arctica islandica*	Ar	Growth rate			
Paper IV	Sr	*Serripes groenlandicus*	Ar	Unknown			0
Paper IV	Sr	*Ciliatocardium ciliatum*	Ar	Unknown			0
Elliot *et al.* (215)	Sr	*Tridacna gigas*	Ar	Vital effects			0

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Foster *et al.* (247)	Sr	*Arctica islandica*	Ar	Vital effects	Growth rate		0
Freitas *et al.* (217)	Sr	*Pinna nobilis*	Ca	Vital effects			0
Freitas *et al.* (218)	Sr	*Pecten maximus*	Ca	Growth rate			0
Freitas *et al.* (220)	Sr	*Mytilus edulis*	Ca	Vital effects			0
Freitas *et al.* (220)	Sr	*Pecten maximus*	Ca	Vital effects			0
Freitas *et al.* (134)	Sr	*Pecten maximus*	Ca	Growth rate	Temperature		0
Gillikin *et al.* (248)	Sr	*Saxidomus giganteus*	Ar	Growth rate	Vital effects	Ontogeny	0
Gillikin *et al.* (248)	Sr	*Mercenaria mercenaria*	Ar	Vital effects			0
Heinemann *et al.* (221)	Sr	*Mytilus edulis*	Ca	Growth rate	Vital effects		0
Izumida *et al.* (223)	Sr	*Hyriopsis* sp.	Ar	Growth rate			0
Jimenez-Berrocoso *et al.* (224)	Sr	*Atrina rigida*	Ca	Growth rate	Temperature		1
Klein *et al.* (249)	Sr	*Mytilus trossulus*	Ar	Vital effects	Salinity		0
Lazareth *et al.* (131)	Sr	*Isognomon ephippium*	Ca	Vital effects	Temperature		0
Lorens and Bender (225)	Sr	*Mytilus edulis*	Ca	Concentration in ambient water			0.5
Lorens and Bender (225)	Sr	*Mytilus edulis*	Ar	Concentration in ambient water			0.5
Lorrain *et al.* (226)	Sr	*Pecten maximus*	Ca	Growth rate			0.5
Pearce and Mann (228)	Sr	*Ensis siliqua*	Ar	Growth rate	Vital effects	Salinity	0
Pérez-Huerta *et al.* (229)	Sr	*Trachycardium procerum*	Ar	Vital effects			0
Purton-Hildebrand *et al.* (250)	Sr	*Venericardia planicosta*	Ar	Ontogeny	Growth rate		0

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Purton *et al.* (251)	Sr	*Venericardia planicosta*	Ar	Vital effects	Ontogeny	Growth rate	0
Richardson (132)	Sr	*Pinna nobilis*	Ca	Temperature			1
Sano *et al.* (116)	Sr	*Tridacna derasa*	Ar	Insolation	Temperature	Growth rate	1
Schöne *et al.* (117)	Sr	*Arctica islandica*	Ar	Vital effects	Bound to organic matrix		0
Schöne *et al.* (133)	Sr	*Arctica islandica*	Ar	Vital effects	Temperature	Ontogeny	1
Schöne *et al.* (114)	Sr	*Arctica islandica*	Ar	Vital effects			0
Shirai *et al.* (230)	Sr	*Bathymodiolus platifrons*	Ar	Vital effects	Import from land		
Stecher *et al.* (252)	Sr	*Mercenaria mercenaria*	Ar	Vital effects			0
Stecher *et al.* (252)	Sr	*Spisula solidissima*	Ar	Vital effects			0
Strasser *et al.* (233)	Sr	*Mya arenaria*	Ar	Vital effects			0
Takesue and van Geen (235)	Sr	*Protothaca staminea*	Ar	Growth rate	Vital effects		0
Takesue *et al.* (236)	Sr	*Corbula amurensis*	Ar	Growth rate			
Toland *et al.* (237)	Sr	*Arctica islandica*	Ar	Vital effects			0
Ullmann *et al.* (238)	Sr	*Crassostrea gigas*	Ca	Vital effects	Ontogeny	Temperature	0
Vander Putten *et al.* (239)	Sr	*Mytilus edulis*	Ca	Vital effects			0
Wanamaker *et al.* (240)	Sr	*Mytilus edulis*	Ca	Vital effects	Salinity	Temperature	0
Wisshak *et al.* (241)	Sr	*Neopycnodonte zibrowii*	Ca	Growth rate	Concentration in ambient water	Bound to other elements	0
Barats *et al.* (144)	Mo	*Pecten maximus*	Ca	Primary production			1
Paper IV	Mo	*Serripes groenlandicus*	Ar	Vital effects	Growth rate		

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Paper IV	Mo	*Ciliatocardium ciliatum*	Ar	Vital effects	Growth rate		
Tabouret *et al.* (115)	Mo	*Pecten maximus*	Ca	Environment			0.5
Thébault *et al.* (143)	Mo	*Comptopallium radula*	Ca	Environment			1
Barats *et al.* (136)	Ba	*Pecten maximus*	Ca	Concentration in ambient water			1
Carre *et al.* (113)	Ba	*Mesodesma donacium*	Ar	Unknown	Growth rate		0
Carre *et al.* (113)	Ba	*Chione subrugosa*	Ar	Unknown			0
Carroll *et al.* (212)	Ba	*Serripes groenlandicus*	Ar	Import from land			1
Paper IV	Ba	*Serripes groenlandicus*	Ar	Vital effects	Primary production	Concentration in ambient water	0.5
Paper IV	Ba	*Ciliatocardium ciliatum*	Ar	Vital effects	Primary production	Concentration in ambient water	0.5
Elliot *et al.* (215)	Ba	*Tridacna gigas*	Ar	Primary production	Vital effects		1
Gillikin *et al.* (141)	Ba	*Mytilus edulis*	Ca	Primary production	Salinity		1
Gillikin *et al.* (187)	Ba	*Saxidomus giganteus*	Ar	Environment			1
Gillikin *et al.* (187)	Ba	*Pecten maximus*	Ca	Environment			1
Goodwin *et al.* (137)	Ba	*Crassostrea gigas*	Ca	Environment			1
Hatch *et al.* (253)	Ba	*Donax gouldii*	Ar	Concentration in ambient water	Nutrients	Primary production	1
Izumida *et al.* (223)	Ba	*Hyriopsis* sp.	Ar	Growth rate			0
Jimenez-Berrocoso *et al.* (224)	Ba	*Atrina rigida*	Ca	Primary production	Import from land		0.5
Krause-Nehring *et al.* (142)	Ba	*Arctica islandica*	Ar	Concentration in ambient water	Element flux	Primary production	1
Lavaud *et al.* (254)	Ba	*Senilia senilis*	Ar	Primary production	Import from land	Ontogeny	1
Lazareth *et al.* (131)	Ba	*Isognomon ephippium*	Ca	Import from land	Primary production		1

Table A2: (continued)

Reference	Element	Species	$CaCO_3$	1. control	2. control	3. control	Proxy
Pearce and Mann (228)	Ba	*Ensis siliqua*	Ar	Primary production			1
Pérez-Huerta *et al.* (229)	Ba	*Trachycardium procerum*	Ar	Primary production	Concentration in ambient water		1
Sano *et al.* (116)	Ba	*Tridacna derasa*	Ar	Unknown			0
Schöne *et al.* (114)	Ba	*Arctica islandica*	Ar	Environment			1
Shirai *et al.* (230)	Ba	*Bathymodiolus platifrons*	Ar	Vital effects	Import from land		
Stecher *et al.* (252)	Ba	*Mercenaria mercenaria*	Ar	Primary production			1
Stecher *et al.* (252)	Ba	*Spisula solidissima*	Ar	Primary production			1
Strasser *et al.* (233)	Ba	*Mya arenaria*	Ar	Temperature			0
Tabouret *et al.* (115)	Ba	*Pecten maximus*	Ca	Concentration in ambient water			1
Thébault *et al.* (143)	Ba	*Comptopallium radula*	Ca	Primary production			1
Toland *et al.* (237)	Ba	*Arctica islandica*	Ar	Primary production			1
Vander Putten *et al.* (239)	Ba	*Mytilus edulis*	Ca	Primary production			1