



## Freshwater bivalves tell of past climates: But how clearly do shells from polluted rivers speak?

Elena Dunca<sup>a,b</sup>, Bernd R. Schöne<sup>b,\*</sup>, Harry Mutvei<sup>a</sup>

<sup>a</sup>Museum of Natural History, Department of Palaeozoology, Box 50007, 10405 Stockholm, Sweden

<sup>b</sup>Institute for Geology and Paleontology, Bio-INCREMENTS Research Group, J.W. Goethe University, Senckenberganlage 32-34, 60325 Frankfurt/Main, Germany

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### Abstract

Freshwater bivalves, *Margaritifera margaritifera* (Linnaeus) and *Unio crassus* (Philipsson), from rivers in Sweden (79 specimens) and England (one specimen) were used to study the effects of human-induced pollution on shell growth (Table 1). We analyzed variations in annual and daily shell growth rates of 80 specimens from unpolluted and polluted (pH < 5, oxygen depletion and eutrophication) localities. 35% of the variability in annual growth of shells from unpolluted rivers is explained by ambient temperature during June through August. Daily shell growth also co-varies with the temperature during the growth season (approximately April–October). Long-term trends in temperature and growth compare well to each other. A weak correlation was also found for shell growth and the summer North Atlantic Oscillation (NAO) index. However, all of these environmental signals are obscured in specimens from polluted settings. In settings with high human impact, shell growth does not co-vary with summer temperatures or the NAO. Results of our study suggest a judicious sampling strategy when shells are used for climate reconstructions.

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### 1. Introduction

Shells of freshwater bivalves from mid- to high latitudes provide long, uninterrupted, seasonally to inter-annually resolved archives of continental paleo-

climate. Such information can complement climate proxy data derived from tree-rings (e.g., Schweingruber et al., 1991; Grudd et al., 2002; Linderholm et al., 2003) and stalagmites (e.g., McDermott et al., 1999; Frisia et al., 2003; Niggemann et al., 2003). Like bivalve mollusks from marine settings (e.g., Pannella and MacClintock, 1968; Clark, 1975; Jones et al., 1989), freshwater mussels show the following shell growth characteristics: (1) They sensitively record ambient environmental conditions during growth as

\* Corresponding author. Tel.: +49 69 798 22863; fax: +49 69 798 22958.

E-mail address: B.R.Schoene@em.uni-frankfurt.de (B.R. Schöne).

variable increment widths in their shells (Bauer, 1992; Dunca and Mutvei, 2001). In particular, changes in summer (June–August) air temperature can be inferred from variations of shell growth rates (e.g., Mutvei et al., 1996; Schöne et al., 2004). Faster shell growth occurs during warmer temperatures and growth may cease below certain temperatures. For example, the shell growth of freshwater bivalves from Sweden is limited to temperatures above 5 °C. In southern Sweden, shell growth starts in May and ends in October/November (Dunca and Mutvei, 2001). (2) An unrivaled temporal resolution enables precise calendar dating of each shell portion. Freshwater bivalves grow by periodic accretion of calcium carbonate and produce distinct annual (Hendelberg, 1961; Mutvei et al., 1994; Hastie et al., 2000) and daily growth increments (Dunca and Mutvei, 2001); (3) In addition, some freshwater bivalves have an extremely long life span. Some species reach ontogenetic ages of 100 (Bauer, 1992; Bauer and Wächtler, 2001) and up to 280 years (Mutvei and Westermark, 2001). Growth patterns of shells of extant and subfossil bivalves with overlapping life spans can be strung together to form long master chronologies spanning many mollusk generations (e.g., Jones et al., 1989; Marchitto et al., 2000) thereby enabling multi-centennial reconstructions of environmental conditions and past climate variability.

In the context of the development of sclerochronological techniques, the reliability of environmental reconstructions based on variable growth rates of bivalve shells has rarely been addressed, and rules for judicious sampling have not been formulated. In this regard, it is important that the effects of anthropogenic environmental change and shell growth be determined. For example, recent dendrochronological studies demonstrate that wood density became increasingly insensitive to temperature forcing during the late 20th century (Briffa et al., 1998a,b). Annual changes in the density of latewood are considered to provide an excellent proxy for summer air temperature at the site where the trees grew (Schweingruber et al., 1991). During the last decades, however, wood density and summer air temperature have diverged. Although the causes for this divergence remain unknown, it is hypothesized that increased levels of human-induced climate change such as rising levels of atmospheric CO<sub>2</sub> and NO<sub>x</sub>, pollution or UV radiation

(Briffa et al., 1998a,b) might have exerted some influence on the wood formation. Our concern is that changes in water quality may affect shell growth.

Here, we apply sclerochronological (growth rate analytical) methods to determine if the shell growth of freshwater bivalves is affected by human-induced environmental disturbances. We question whether specimens from polluted settings can be used for climate reconstructions. We also question whether freshwater shells can monitor and track anthropogenically induced environmental hazards. Furthermore, sampling strategies are suggested for further environmental studies using freshwater bivalves. Results of the present study can be used to improve environmental and climate reconstructions based on shells of freshwater bivalves.

## 2. Material and methods

Eighty specimens of the freshwater pearl mussel, *Margaritifera margaritifera* (Linnaeus) and one specimen of *Unio crassus* (Philipsson) were collected alive from eight different rivers across Sweden between 1930 (collections stored in the Swedish Museum of Natural History, Stockholm) and 1997 (Fig. 1; Table 1). In addition, we used one specimen collected in 2002 from Borrans Beck, Lake District, England (same latitude, 56°N, as Vramsån). The shells lived in water depths of less than 1 m. Two of the Swedish rivers, the River Slereboån and the River Kvarnbäcken, were affected by human-induced environmental perturbation — in particular during the 1970s — by acid rain that lowered the pH of the water. In the following we refer to the two rivers with strong human impact as ‘polluted’ settings, and to the remaining seven rivers as ‘unpolluted’. As part of mitigation projects since the early 1980s liming was used in both of the polluted rivers to address acidification, and fertilization was used in the in the Kvarnbäcken for the stimulation of the biological activity (Mutvei et al., 1996).

### 2.1. Sample preparation

One valve of each specimen was cut from the umbo to the ventral margin along the axis of minimum growth (i.e. perpendicular to the winter lines;

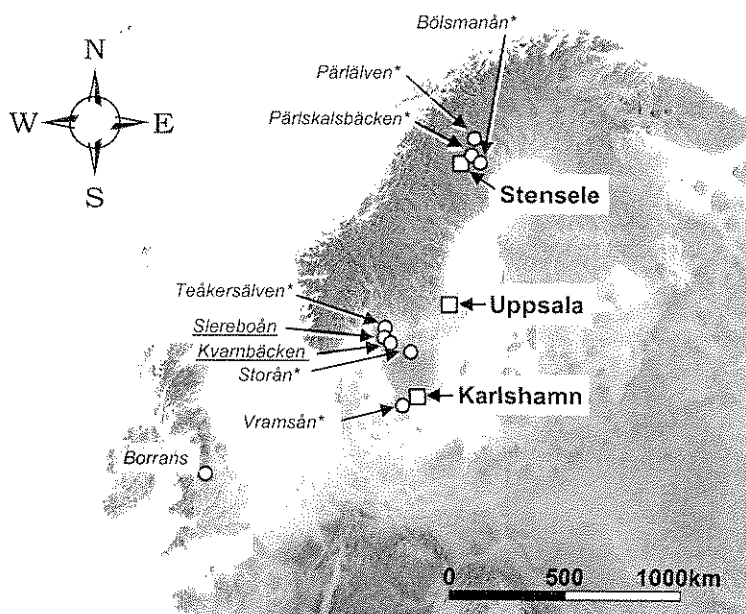


Fig. 1. Map showing sample localities. See Table 1 for details. The asterisk (\*) refers to unpolluted rivers. Specimens of those rivers were used for the construction of the mean chronology. The names of polluted rivers are underlined.

Fig. 2; Dunca and Mutvei, 2001). On that axis, two 3-mm-thick sections were cut from the valve with a low-speed saw equipped with a wafer-thin (0.3 mm) diamond blade. The ('mirroring') sections were subsequently mounted on glass slides, ground with metallographic grinding paper (320, 600 and 1200 grit) and polished with 1 and 0.25  $\mu\text{m}$  diamond paste. Finally, the sections were ultrasonically cleaned in 95% ethyl alcohol and air-dried. To help to resolve inter- and intra-annual growth patterns in the outer (prismatic) layer of the shells (Figs. 3–7), one polished section from each specimen was immersed in Mutvei's solution (Schöne et al., this issue) for 5 to 30 min at 37–40 °C. Immediately afterward, the etched sections were rinsed with de-mineralized water and allowed to air-dry.

## 2.2. Sclerochronological analyses

The growth patterns of samples immersed for 30 min in Mutvei's solution were viewed under reflective light stereomicroscopes (Nikon SMZ 1500, Leitz-Wetzlar SM-LUX-POL) using both oblique and axial light, and digitized with an AxioCam HRc Zeiss camera. For scanning electron microscopy

(SEM) studies (Fig. 3), shell portions etched for 5 min were mounted on metal stubs using carbon glue Leit-C, sputter coated with a 30 nm gold layer and studied with a Hitachi S-4300 SEM at acceleration voltages of 15 to 25 keV. Annual growth increment widths were measured to the nearest 1  $\mu\text{m}$  and daily microgrowth increment widths to the nearest 0.2  $\mu\text{m}$  with Scion/NIH version 4.0.2 beta image analysis software (available as freeware from <http://www.scioncorp.com>). Measurements were conducted in the outer shell layer as shown in Fig. 2. Daily growth rates were only determined in one specimen, because previous research (Dunca and Mutvei, 2001) has studied this in detail. The average annual growth increment width is 370  $\mu\text{m}$ . Daily microincrements measure on average 1.3  $\mu\text{m}$ .

## 2.3. Detrending and standardization, mean chronologies

Ontogenetic growth trends must be removed in order to isolate environmental signals from annual growth increment time-series. As the bivalve matures, the growth rate and the year-to-year variance decrease. These ontogenetic trends were removed with statistical

Table 1  
Freshwater shells used in the present study

Locality (River)	Individuals' dates of death (yr AD)	Specimen ID	Number of specimens	Number of specimens
Parlälven	1986	Parlälven86_1 to 5	5	NCS mean chronology, AD 1860–1989 (56) Daily growth patterns (2; Vrams 94_4 and 12)
Pärskalsbäcken	1993	Paerlskal93_1 to 5	5	
Bölsmanån	1997	Balmans97_1 to 5	5	
Teåkersälven	1986	Teakers86_1 to 8	8	
Storån	1988	Stor88_1 to 4	4	
Vramsån	1930, 1989, 1991, 1993, 1994	Vrams93_1 to 10, 12 to 15, 91_1 to 3, 89_1 to 10, 30_1 and 2, Vrams 94_4 and 12	31	
Borrans Beck	2002	Borrans02_1	1	
Kvambäcken	1990	Kvam90_1 to 7	7	
Stereboån	1989, 1995	Stere89_1, 4, 6–10, 12, 14, 16–20	14	

One specimen (Vrams94\_12) from the River Vramsån belongs to *Unio crassus*, all others are *Margaritifera margaritifera*. Numbers in brackets refer to numbers of specimens used. Underlined rivers = polluted settings, all others unpolluted.

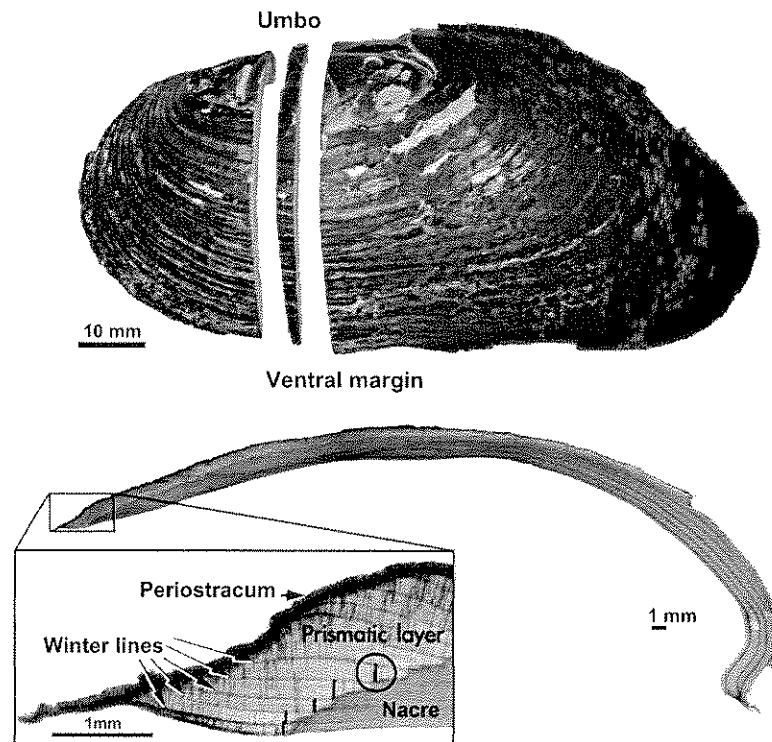


Fig. 2. Sampling and measuring of freshwater bivalves. Two- to three-millimeter-thick slices were cut from the shells along the axis of minimum growth. After immersion in Mutvei's solution, annual growth patterns can easily be studied in the cross-sections. Winter lines (arrows) result from growth interruption below 5 °C and limit annual (May–October) growth increments. Measurements of annual increment widths were completed at the boundary between the prismatic and nacreous layers (circle).

methods for detrending developed by dendrochronologists (Cook and Kairiukstis, 1990). Logistic growth functions are suitable to estimate the growth curves and age trends. Detrending retained most of the decadal and multi-decadal periods of climate variability. We calculated growth indices (GI) by dividing measured growth values by predicted (estimated) growth values at each year. Then, the GI data were standardized by subtracting the mean of the GI time-series and dividing by the standard deviation of the GI time-series. The resulting standardized growth indices (SGI values) detail information on inter-annual changes in shell growth independent of ontogenetic trends.

Where the SGI time-series of individual specimens temporally overlap, results were combined to form a mean chronology. For each year, the arithmetic mean of the SGI values of all shells was calculated. The resulting composite or mean chronology stretches over longer time intervals than individuals' life spans. Low-pass filtering (Savitzky–Goley, window

length 15 years) was used to reveal inherent longer-term (decadal) trends. Such trends were compared to low-pass filtered (window length of 15 years) summer air temperature data (June–August; see below). Prior to spectral analysis, temporal trends in the SGI mean chronology were removed with a cubic model. We also smoothed the mean chronology with Savitzky–Goley low-pass filters (window lengths=4 and 10 years) so that frequencies greater than 0.25 (4 years) or 0.1 (10 years) were cut off, respectively. Prior to spectral frequency analysis with Fast Fourier Transform (Welch method) the low-pass filtered time-series were corrected for autoregression (lag-1, AR-1 process).

#### 2.4. Statistical evaluation of correlation between annual shell growth and temperature

The correlation of year-to-year variability between air temperature records and SGI master chronologies

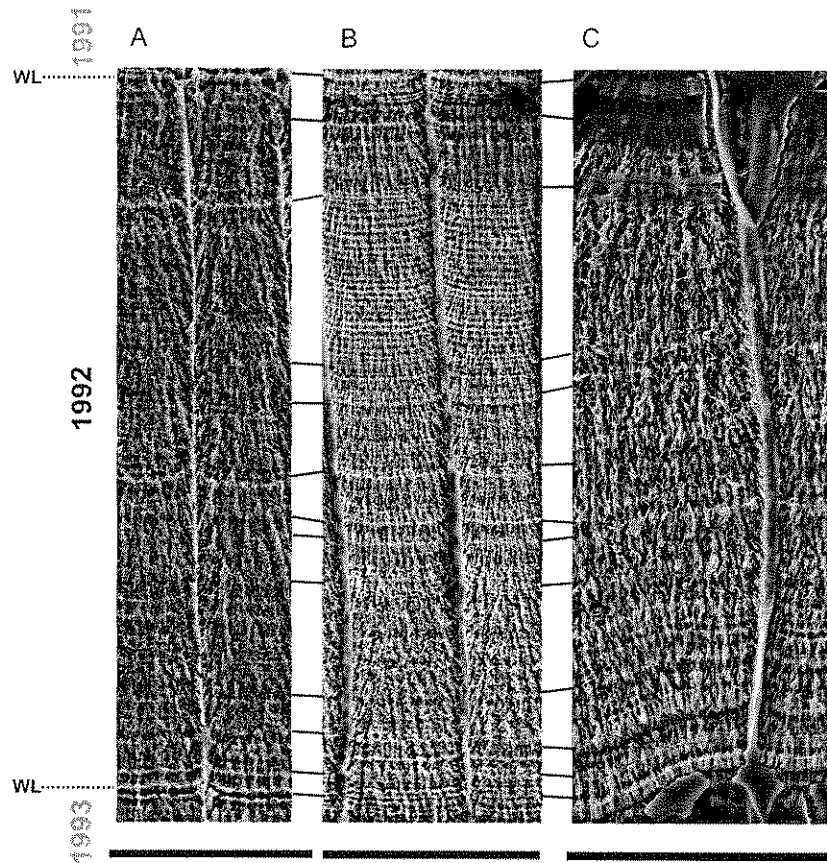


Fig. 3. Daily microincrement patterns for the year AD 1992 from young specimens of *Margaritifera margaritifera* (A, B) and *Unio crassus* (C) that lived in unpolluted rivers and at approximately the same temperature regime. Specimen A came from the Borrans Beck, England and specimens B and C came from the River Vramsån, southern Sweden. Common intra-annual growth patterns among the specimens imply common external forcing on shell growth. WL=winter lines; scale bars=60  $\mu\text{m}$ .

was quantified using three different statistical tests: (1) linear correlation, (2)  $t$ -statistic after Baillie and Pilcher (1973), and (3) running similarity (*Gleichläufigkeit*) test after Huber (1943). Linear correlation analyzes the degree of similarity between two chronologies. SGI values of one chronology are plotted against the SGI values of the second chronology. Baillie and Pilcher's (1973)  $t$ -statistic measures the strength of agreement between two chronologies:

$$t = \frac{r\sqrt{n-2}}{\sqrt{1-R^2}},$$

where  $r$  is the correlation coefficient,  $R^2$  the coefficient of determination and  $n$  the sample size. The running similarity test by Huber (1943) assesses

agreement between the patterns of two chronologies. A value of one is assigned to a year if the growth rate increases (positive sign) or decreases (negative sign) from one to the following year in both chronologies. A value of zero indicates that the growth rate increases in one chronology and decreases in the other. The sum of sign agreements between two chronologies over a given period is expressed as percent agreement. Confidence intervals based on standard deviation units (SD) provide information on the statistical significance of the match between two chronologies where  $1SD \approx 50/\sqrt{n}$  and  $2SD \approx 2 \cdot 50/\sqrt{n}$ :

$$G = 50 + \frac{SD \cdot 50}{\sqrt{n}}.$$

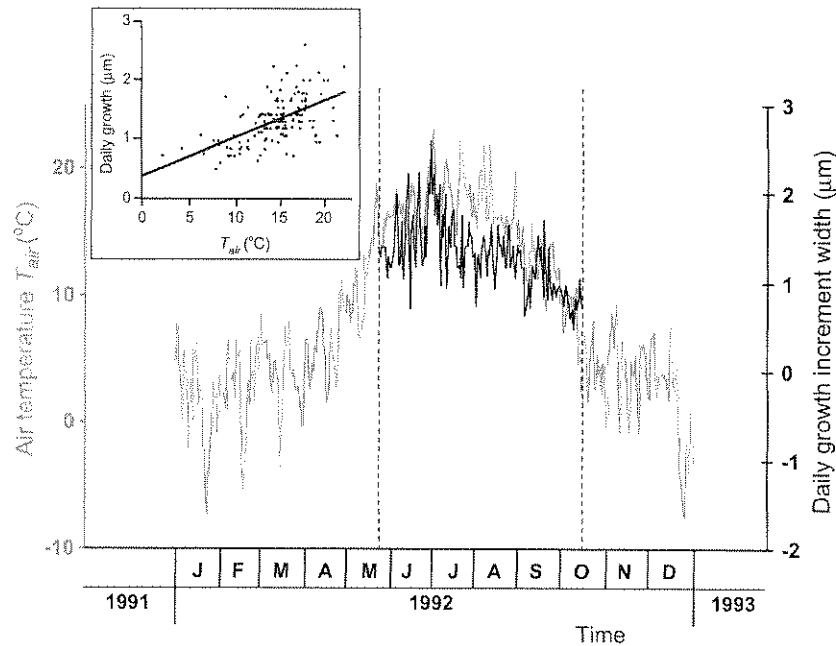


Fig. 4. Comparison between intra-annual shell growth of the *Margaritifera margaritifera* specimen from the River Vramsån depicted in Fig. 3C and daily summer air temperatures. Note the close correlation between growth and temperature ( $R^2=0.31$ ,  $p<0.0001$ ).

Two SGI chronologies are in statistically significant agreement if the percentage of agreement is higher than the running similarity value  $G$ .

### 2.5. Environmental variables and air temperature proxies

Data on river water temperatures are extremely scarce. However a previous study has demonstrated that river water temperatures closely follow air temperatures (Schöne et al., 2004). An increase in air temperature by 1 °C is concurrent with a river water temperature increase of 0.88 °C. Air temperature can thus be used as a proxy for river water temperature. Observational records of air temperature in Sweden reach back until about AD 1860 (note: in what follows all years are 'AD'). Meteorological records for northern (Stensele), central (Uppsala) and southern Sweden (Karlshamn) were obtained from the Swedish Meteorological and Hydrological Institute. We compared annual SGI data of the mean chronologies with observational records of summer air temperatures (June–August averages) to study if and how temperature controlled the growth rates of the bivalves. Previous studies demonstrated that temperature plays an essen-

tial role in shell growth of naiad shells (Hendelberg, 1961; Bauer, 1992; Dunca, 1999; Dunca and Mutvei, 2001). We also compared the low-pass filtered (Savitzky–Goley filter, window lengths: 4 and 10 years) SGI data with low-pass filtered North Atlantic Oscillation (NAO) indices during summer (May–August). Monthly NAO indices were obtained from J. Hurrell's web page at <http://www.cgd.ucar.edu/~jhurrell/nao.stat.ann.html>.

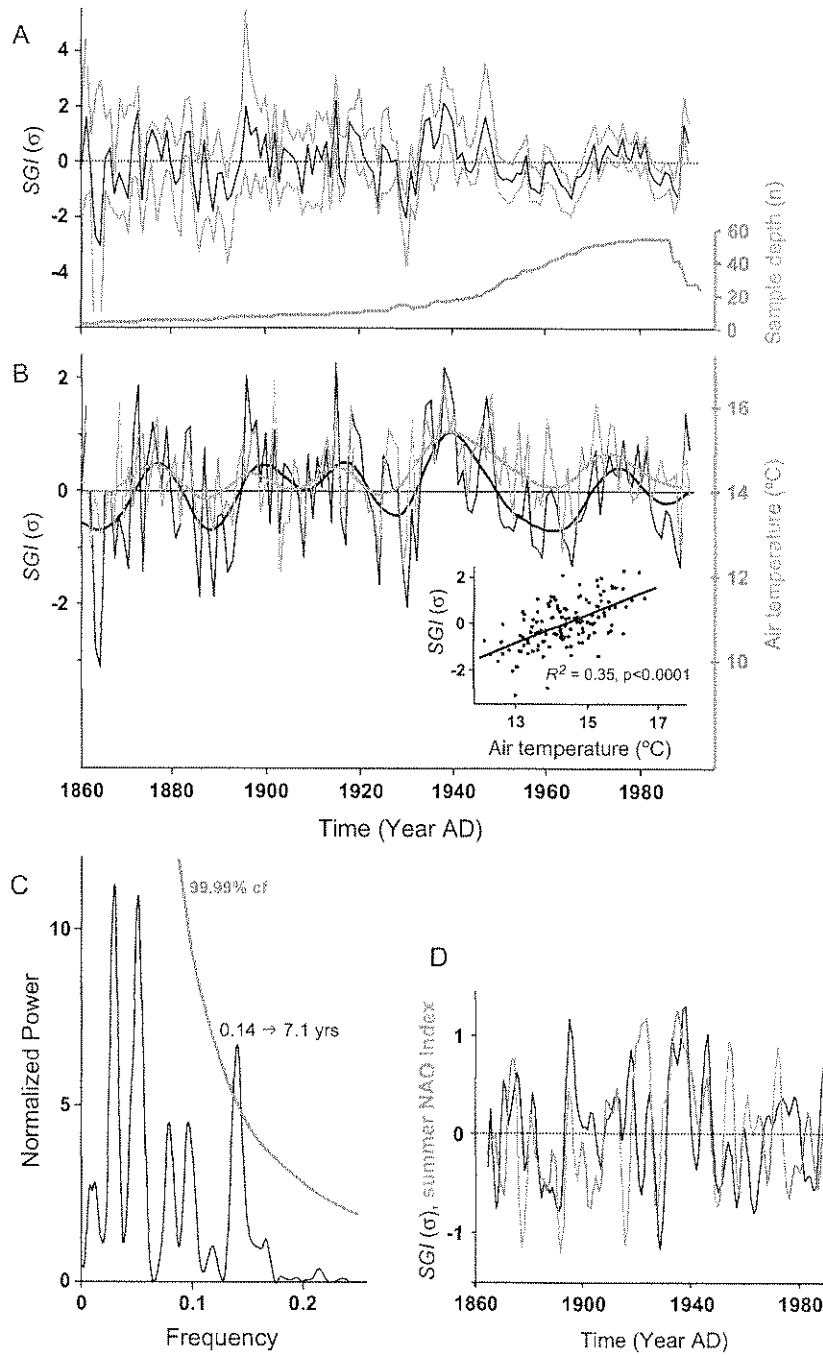
## 3. Results

### 3.1. Daily growth patterns of shells from unpolluted settings

Strikingly similar microgrowth patterns were found in contemporaneous shell portions of two specimens of *M. margaritifera* collected alive from the unpolluted Rivers Vramsån in southern Sweden and the River Borrans Beck, Lake District, England (Fig. 3A,B). Microgrowth increment widths quickly increased after the winter line (ca. 1 to 1.5 µm) and reached maximum values of about 2.6 µm at ca. 40% of the annual increment. Then, growth rates

continuously slowed toward the 1992/1993 winter line (Figs. 3 and 4). Aside from these common overall growth trends, minor fluctuations in micro-growth rates could be visually cross-matched bet-

ween the specimens (Fig. 3). Similar microgrowth trends and fluctuations were observed in year 1992 of another freshwater bivalve species, *Unio crassus*, from the Vramsån (Fig. 3C).





We counted 143 microincrements (intra-annual increments) in the annual increment of the specimens from Vramsån (Figs. 3B and 4). The number of microincrements counted in *M. margaritifera* closely coincided with the number of days expected to occur during the 5- to 6-month growth season in southern Sweden (e.g., Dunca and Mutvei, 2001). A variety of previous studies have demonstrated that these microincrements form on a daily basis (Dunca and Mutvei, 2001). We therefore used the microincrements to assign approximate calendar dates to each portion of the 1992 growth increments and aligned the daily growth record so that shell growth started during May and ended during October 1992. A good agreement was found between daily growth rates and daily air temperatures (Fig. 4; Table 2). About 31% of the variation in daily growth rates is explained by the ambient temperature regime.

### 3.2. Annual growth patterns of shells from unpolluted settings

Shells of *M. margaritifera* from unpolluted rivers in northern (Pärlälven, Pärlskalsbäcken, Bölsmanån), central (Teåkersälven, Storån) and southern Sweden (Vramsån) were combined for the construction of an SGI mean chronology (NCS mean chronology, Fig. 5A). This was because the number of specimens analyzed did not provide continuous records at each locality. The NCS mean chronology consisted of shells from 56 specimens with overlapping life spans and representing the period 1860–1989. 95% confidence intervals of the SGI values of the NCS chronology become smaller in most recent years and with increasing sample depth (defined as the number of specimens arithmetically averaged per year) (Fig. 5A). Narrow confidence bands also reflect a high level of synchronicity among the growth patterns of specimens that lived at the same time interval (Fig. 5). Highest sample depth ( $n = 54$ ) occurred during 1979–

1986. Sample depth and SGI values are not correlated ( $R^2 = 0.001$ ,  $p = 0.68$ ). Ontogenetic ages of the bivalves ranged from 10 to 127 years with a mean of 51 years. Fastest overall growth rates were detected during 1914 (SGI value of +2.27) and 1937 (+2.21), while the bivalves grew at slowest rates during 1864 (−3.12) and 1929 (−2.05). Low-pass filtering (Savitzky–Goley, window length: 15 years; Fig. 5B) leaves distinct periods of faster (peaks at 1877, 1899, 1917, 1939, 1975) and slower growth (peaks at 1864, 1888, 1906, 1927, 1962, 1984) which coincide well with the longer-term temperature trends (Fig. 5B). Visual comparison reveals a high degree of synchronicity between the growth and temperature curves, an observation that is supported by statistics (Table 2).

Linear regression revealed a positive correlation between annual shell growth and observed summer air temperature over the period 1860–1989 (Fig. 5B; Table 2). June through August air temperatures were calculated from the arithmetic mean of three different stations in northern, central and southern Sweden (NCS) closest to localities where the shells grew, Stensele (northern), Uppsala (central) and Karlshamn (southern Sweden), respectively. 35% of the variation in mean annual shell growth rates over the period is explained by summer air temperature (Fig. 5B; Table 2). For comparison with data presented further below, we also conducted linear regression analyses for the period of 1946 to 1989 and found that 32% of the variability in growth is explained by temperature (Table 2). These results were also supported by running similarity tests (Table 2).

Spectral analysis (FFT) of the detrended and low-pass filtered (cutoff periods: <4 years) NCS mean chronology (1860–1989) revealed frequencies of 0.14 corresponding to periods of about 7.1 years (significant at  $p < 0.0001$ ; Fig. 5C). A weak but significant correlation exists between low-pass filtered NCS mean chronology and low-pass filtered Hurrell's NAO index

Fig. 5. Annual shell growth of *Margaritifera margaritifera* from unpolluted rivers in northern, central and southern (NCS) Sweden (Fig. 1; Table 1,2) and NAO and air temperature data. (A) NCS mean chronology composed of 56 specimens. The narrow 95% confidence intervals (grey) around average SGI values at each year (black) reflect the high degree of running similarity between individual SGI curves. Confidence intervals become narrower as sample depth increases. (B) NCS mean chronology (black) and summer (June–August) air temperatures (grey) agree well with each other. Low-pass filtering (Savitzky–Goley SG filter, window length  $w$ : 15 years) reveals longer-term trends in growth and temperature data. (C) Fast Fourier Transform of the low-pass filtered (SG,  $w$ : 4) and AR-1 corrected NCS mean chronology reveals significant spectral power at frequencies of 0.14 corresponding to periods of 7.1 years. This period is typical for the North Atlantic Oscillation (NAO). (D) Low-pass filtered summer (May–August) NAO and NCS mean chronology (AR-1 corrected) show weak correlation of  $R^2 = 0.13$  (SG,  $w$ : 4) to 0.17 (SG,  $w$ : 10).

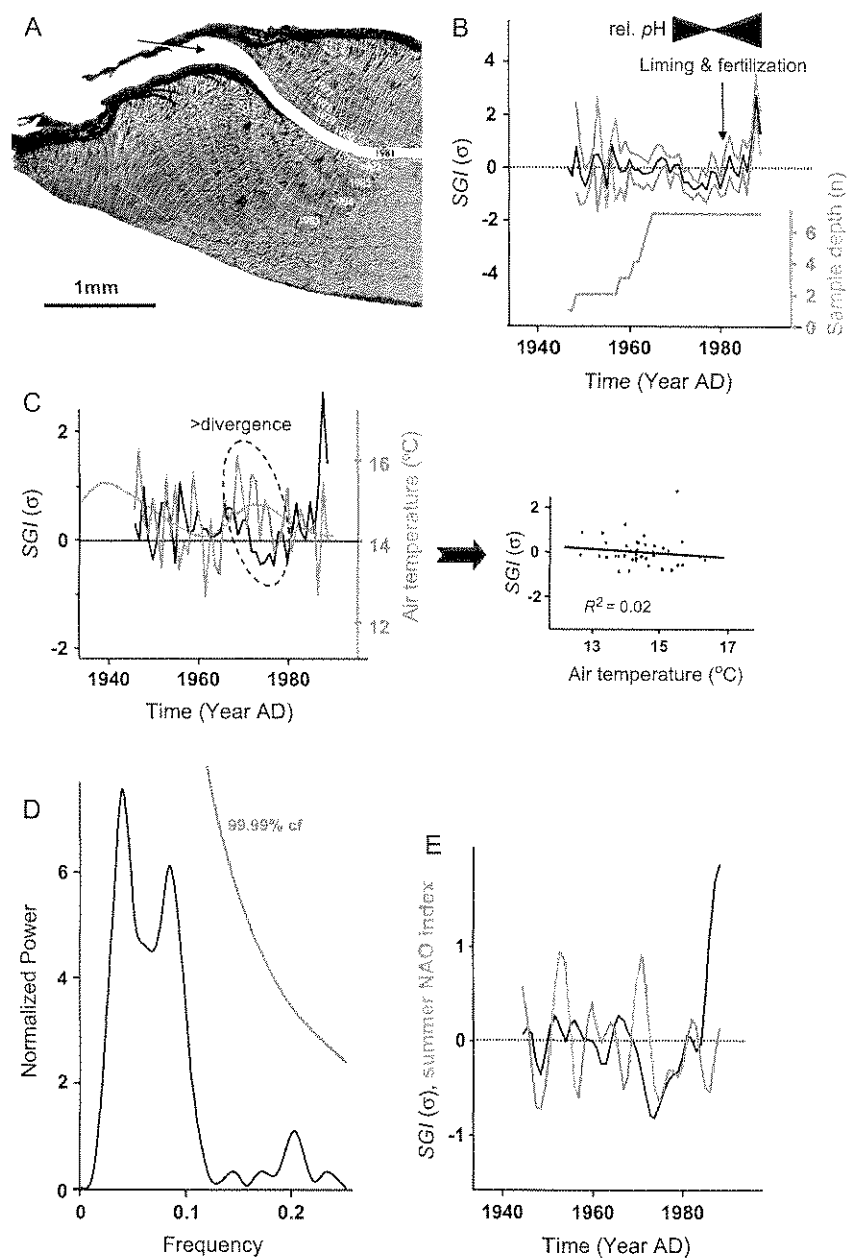


Fig. 6. Annual shell growth of *Margaritifera margaritifera* from the polluted River Kvarnabäcken. Specimen ID: Kvarn90\_2. (A) Declining annual shell growth during the 1970s was caused by decreasing alkalinity. Major growth disruption during 1981 results from liming and addition of fertilizers to the water. Note strong increase in shell growth in following years. (B) Mean chronology consisting of seven specimens. Black: average SGI values at each year, grey: 95% confidence levels. Strongly reduced growth occurred during the 1970s. Shell growth was stimulated by increased pH after liming and eutrophication (nitrogen and phosphorous) during 1981. (C) No correlation exists between shell growth (black) and temperature (grey). Note strong divergence between shell growth and temperature during the 1970s, i.e. during increased environmental stress due to low alkalinity. For details on mathematical pre-treatment see caption of Fig. 5B. (D) Power spectrum (FFT) shows no significant frequencies. For details on mathematical pre-treatment see caption of Fig. 5C. (E) Visual comparison reveals no obvious running similarity between shell growth and summer NAO. For details on mathematical pre-treatment see caption of Fig. 5D.

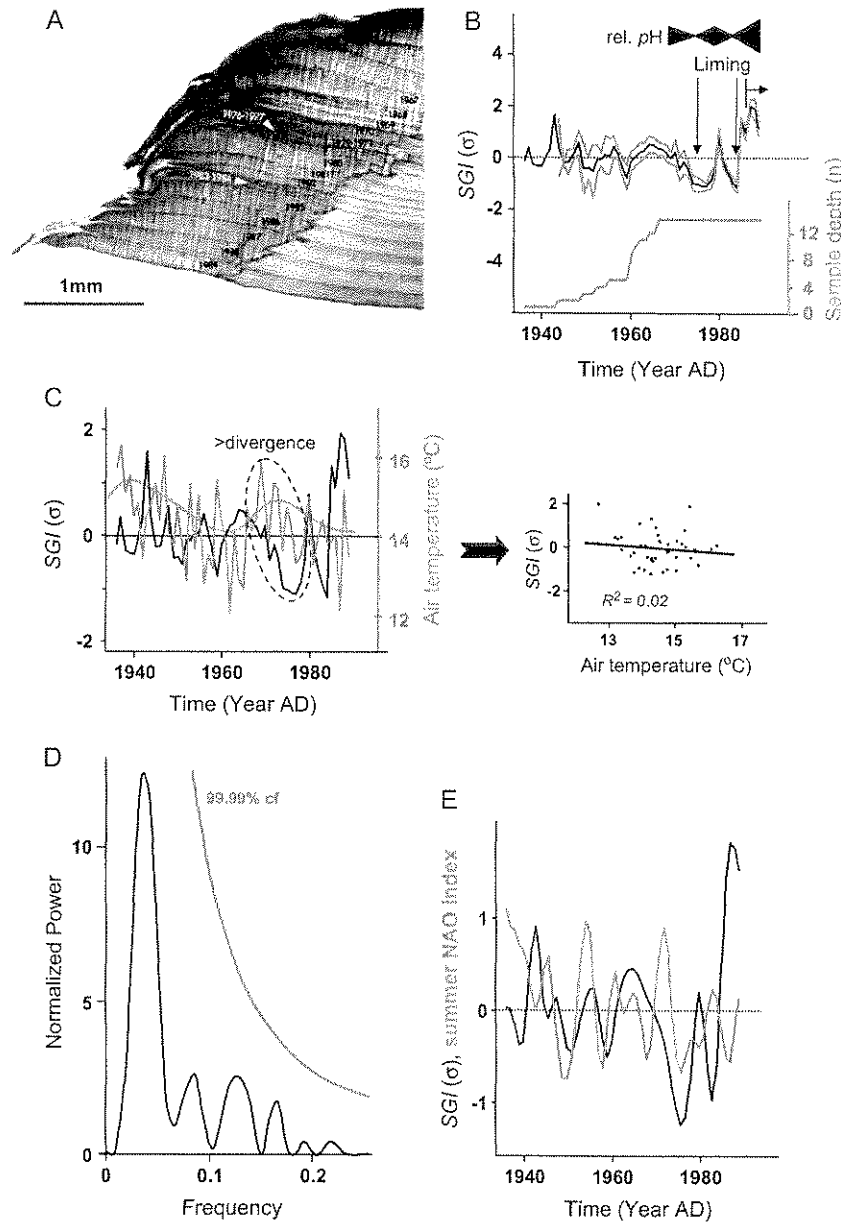


Fig. 7. Annual shell growth of *Margaritifera margaritifera* from the polluted River Slereboån. Specimen ID: Slere89\_6. (A) Strongly reduced growth rates during the 1970s and early 1980s as a result of low pH values. Note strong increase in shell growth after liming. (B) Mean chronology consisting of 14 specimens. Black: average SGI values at each year, grey: 95% confidence levels. Strongly reduced growth occurred during the 1970s. Shell growth was stimulated by reduced acidity after liming in 1974, 1984 and since 1986. (C) No correlation exists between shell growth (black) and temperature (grey). Note strong divergence between shell growth and temperature during the 1970s, i.e. during increased environmental stress due to low alkalinity. For details on mathematical pre-treatment see caption of Fig. 5B. (D) Power spectrum (FFT) shows no significant frequencies. For details on mathematical pre-treatment see caption of Fig. 5C. (E) Visual comparison reveals no obvious running similarity between shell growth and summer NAO. For details on mathematical pre-treatment see caption of Fig. 5D.

Table 2

Statistics are based on variations in growth rates of the bivalve mollusk *M. margaritifera* (daily growth, annual growth: SGI) and summer (June–August) air temperature ( $T_{\text{air}}$ )

Compared data	Unpolluted settings			Polluted settings	
	Vramsån, $T_{\text{air}}$ —daily growth	NCS mc		Kvambäcken mc $T_{\text{air}}$ —SGI	Slereboån mc
Period (years AD)	1992	1860–1989	1946–1989	1946–1875	1946–1989
Length of period	143 days	130 years	44 years	44 years	44 years
Number of shells per year	1	2 to 56	20 to 37	7	14
Correlation coefficient $r$	0.56*	0.59*	0.56*	−0.13 <sup>ns</sup>	−0.13 <sup>ns</sup>
Variance explained $R^2$	0.31*	0.35*	0.32*	0.02 <sup>ns</sup>	0.02 <sup>ns</sup>
Baillie and Pilcher's $t$ -test	8.04*	8.28*	4.43*	−0.84 <sup>ns</sup>	−0.87 <sup>ns</sup>
Huber's running similarity (or sign) test ( $G$ value, hit/miss) <sup>1</sup>	56.34 80/62 <sup>&gt;SD1</sup>	65.12 84/45 <sup>&gt;SD2</sup>	65.12 28/15 <sup>&gt;SD2</sup>	41.86 18/25 <sup>ns</sup>	51.16 22/21 <sup>ns</sup>

$T_{\text{air}}$  data represent averages of three meteorological stations in northern, central and southern Sweden (see Fig. 1). \* $p < 0.0001$ , <sup>1</sup> $p < 0.01$ , ns=not significant. NCS=northern, central and southern Sweden (see Fig. 1 and Table 1); mc=mean chronology.

during summer (average over May through August; Fig. 5D). Applying a Savitzky–Goley filter with a window length of 4 years to both series results in an  $R^2$  value of 0.13 ( $r = 0.36$ ,  $p < 0.0001$ ), while comparison of longer-term trends (filter length of 10 years) gives slightly higher correlation values:  $r = 0.41$ ,  $R^2 = 0.17$ ,  $p < 0.0001$ .

### 3.3. Annual growth patterns of shells from polluted settings

All seven specimens of *M. margaritifera* from the River Kvambäcken (Fig. 6) exhibited a continuous decrease in annual shell growth during the 1970s and a major growth decline in 1981 (Fig. 6A,B). Thereafter, shell growth increased by several hundred percent (Fig. 6A,B). The 1981 growth disturbance coincides with a liming project in the Rammsjön Lake, about 4 km upstream. Liming was applied to address the acidity of the lake and river waters. To stimulate bio-production and reduce mercury levels at the bottom of the Rammsjön Lake nitrogen and phosphorous were also added (Mutvei et al., 1996).

Like the Kvambäcken, the River Slereboån had pH values below 5 during the early 1970s. Liming was applied in 1974 and 1984 and has been continuously applied since 1986 (installation of a dozer). In response to the liming, pH values reached levels close to 7 in a few years (Mutvei et al., 1996). The mean chronology of the 14 shells of *M. margaritifera* from the Slereboån (Fig. 7) exhibited relatively stable

growth rates during the 1950s and 1960s with alternating narrower and broader increments (Fig. 7A,B). During the early 1970s, however, growth rates strongly decreased and remained below the long-term average, in particular during 1973–1974 and during 1976–1977. A strong increase in shell growth during 1980 was followed by extremely low rates in 1983–1984. Since then the shells grew very fast. SGI values remained positive until the shells were captured in 1989 (Fig. 7A,B).

Visual comparison of the data from both rivers indicated that shell growth and temperature strongly diverged during the 1970s (Figs. 6B and 7B). Although growth rates reached abnormally high values subsequent to this period of divergence, a greater synchronicity occurred between SGI values and temperature since the mid-1980s (Figs. 6C and 7C).

Shell growth of the Slereboån and Kvambäcken populations was not correlated to summer temperature (Figs. 6C and 7C). Over the period 1946–1989 we found  $R^2$  values of 0.02 for populations from the Slereboån and Kvambäcken (Figs. 6C and 7C; Table 2). Similarly, the running similarity tests of Baillie and Pilcher (1973) and Huber (1943) did not return significant relationship between growth and temperature (Table 2). Spectral analyses of detrended and low-pass filtered SGI mean chronologies from Slereboån and Kvambäcken did not reveal any significant cycles (Figs. 6D and 7D). Also, the shell growth of the bivalves from both rivers did not coincide with summer NAO dynamics (Figs. 6E and 7E).

## 4. Discussion

### 4.1. Shell growth in unpolluted settings

In settings with little to no human impact on the environment, temperature exerts a clear control on daily and annual shell growth of freshwater bivalves (Figs. 4 and 5; Table 2). The amount of variance explained by annual shell growth is approximately as high as that found for April–August air temperatures and tree-ring widths in Fennoscandian Scots pines (Briffa et al., 1990). Our findings on the close relation between shell growth and temperature is true for daily and annual time scales and supports many previous studies on bivalve mollusk species from various different freshwater (e.g., Bauer, 1992; Dunca and Mutvei, 2001) and marine ecosystems (e.g., Davenport, 1938; Kennish and Olsson, 1975). The correlation between shell growth and temperature probably reflects a causal link between the two parameters. Many biological processes are temperature-dependent. Within certain limits, higher temperatures stimulate metabolism and also shell production (Sprung, 1984; Lewis and Cerrato, 1997).

Annual shell growth at unpolluted settings is weakly positively correlated with summer (May–August) NAO index of Hurrell (1995) (Fig. 5; Table 2). In addition, the NCS mean chronology contains the typical NAO-type spectral peaks at frequencies corresponding to periods of about 7.1 years ( $p < 0.0001$ ). The NAO is to a major climate phenomenon that occurs over Europe and North America and is most pronounced during winter. Its measure of state, the NAO index is typically defined as the pressure difference between the Azores and Iceland (Hurrell, 1995). In general, positive NAO indices correspond to greater pressure level differences and coincide with mild temperatures, strong winds and wet conditions over northern Europe. We suggest that shell growth of freshwater bivalves in Sweden is elevated during positive NAO years due to higher average temperatures. Covariance between decadal climate cycles such as the NAO and shell growth was also observed in marine shells of *Arctica islandica* from the Northeast Atlantic (Schöne et al., 2003).

### 4.2. Shell growth in polluted settings

Ecosystems severely affected by human activities that resulted in low pH levels or oxygen depletion have detrimental effects on shell growth (Figs. 6 and 7; Table 2). This is well documented for contemporary and fossil aquatic ecosystems. Animals with calcareous hard parts such as brachiopods and bivalve mollusks living under reduced oxygen and lowered pH values remain thin-shelled and small and grow very slowly (Schmidt, 1931; Rhoads and Morse, 1971). Physical stunting in such environmental settings is well known (Tasch, 1953; Hallam, 1965; Bretsky and Bermingham, 1970) and largely results from reduced metabolic rates (Rhoads and Morse, 1971). Mutvei et al. (1996) demonstrated that oxygen depletion and elevated levels of phosphorous and organic carbon in Lake Krasnoe, Russia, cause significant reductions in the shell growth of *Unio tumidus* (Philipsson, 1788) and *Unio pictorum* (Linnaeus, 1758). In the present study, sclerochronological analyses also clearly demonstrated that increasing acidification of the Sierboån and Kvarnbäcken resulted in diminished growth rates. Mitigation of the negative effects of low pH on the ecosystem stability was accomplished by liming during the mid- and end 1970s and since the early 1980s, after which pH returned to normal levels and shells grew much faster than during times of environmental stress (Figs. 6 and 7). Unusually high shell growth rates of bivalve mollusks living in the Kvarnbäcken after 1981 can also be explained by fertilization concurrent to liming in the early 1980s. Similar observations were made by Mutvei et al. (1996) on specimens of *U. crassus* from the eutrophicated River Soodla, Estonia.

Although our findings of shell growth under human-induced environmental stress are not entirely new or unexpected, it has not been statistically evaluated if shells living under unnatural conditions still function as good temperature recorders. We found that growth rates of bivalves that were exposed to low pH or artificially fertilized ecosystems did not show any correlation with ambient temperatures or the summer NAO index (Figs. 6 and 7; Table 2). In the shells we studied, anthropogenic forcing completely obscured the environmental signals.

## 5. Implications and conclusions

In unpolluted settings, ambient water temperature exerts a large control on shell growth of freshwater bivalves. We found a significant correlation between growth rates and temperature on intra- and inter-annual time scales. The shells sensitively react to human-induced changes of the ecosystem. In settings with lowered pH values, depleted oxygen and increased food supply, shell growth and water temperature do not co-vary with each other. These findings have significant implications for climate reconstructions based on variable growth rates of bivalve shells. Only shells from undisturbed environmental settings provide reliable records of past environmental variables. Only those shells should be used for climate reconstructions.

Further studies should focus on reconstructing the pollution history of lakes and rivers using the shell carbonate (e.g., reconstruction of pH of the water with boron isotopes; Vengrosh et al., 1991; Hemming and Hanson, 1992). The development of new proxies for human-induced pollution might also be useful. In combination with the calendric shell record of the bivalves, such proxies would produce a powerful bio-monitoring tool for ecosystems without environmental records. Additional studies should also quantify the effect of specific human-induced environmental changes, e.g., how and why is shell growth altered by changing pH conditions? Such methods could be useful for archeological studies, for example, for tracking mining activity in history.

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## References

- Baillie, M.G.L., Pilcher, J.R., 1973. A simple cross-dating program for tree-ring research. *Tree-ring Bull.* 38, 35–43.
- Bauer, G., 1992. Variation in the life span and size of the freshwater pearl mussel. *J. Anim. Ecol.* 61, 425–436.
- Bauer, G., Wächtler, K., 2001. Ecology and evolution of the freshwater mussels Unionoidea. *Ecological Studies*, vol. 145. Springer, Berlin, Heidelberg. XXII+394 pp.
- Bretsky, P.W., Bermingham, J.J., 1970. Ecology of the Paleozoic scaphopod genus *Plagioglypta* with special reference to the Ordovician of eastern Iowa. *J. Paleontol.* 44, 908–924.
- Briffa, K.R., Bartholin, T.S., Eckstein, D., Jones, P.D., Karlén, W., Schweingruber, F.H., Zetterberg, P., 1990. A 1400-year tree-ring record of summer temperatures in Fennoscandia. *Nature* 346, 434–439.
- Briffa, K.R., Schweingruber, F.H., Jones, P.D., Osborn, T.J., Shiyatov, S.G., Vaganov, E.A., 1998a. Reduced sensitivity of recent tree-growth to temperature at high northern latitudes. *Nature* 391, 678–682.
- Briffa, K.R., Schweingruber, F.H., Jones, P.D., Osborn, T.J., Harris, I.C., Shiyatov, S.G., Vaganov, E.A., Grudd, H., 1998b. Trees tell of past climates: but are they speaking less clearly today? *Philos. Trans. R. Soc. Lond., B* 353, 65–73.
- Clark II, G.R., 1975. Periodic growth and biological rhythms in experimentally grown bivalves. In: Rosenberg, G.D., Runcorn, S.K. (Eds.), *Growth Rhythms and the History of the Earth's Rotation*. J. Wiley and Sons, New York, pp. 103–117.
- Cook, E.R., Kairiukstis, L.A., 1990. *Methods of Dendrochronology. Applications in the Environmental Sciences*. Kluwer, Dordrecht, Netherlands. 394 pp.
- Davenport, C.B., 1938. Growth lines in fossil peccans as indicators of past climates. *J. Paleontol.* 12, 514–515.
- Dunca, E., 1999. Bivalve shells as archives for changes in water environment. *Vatten* 55, 279–290.
- Dunca, E., Mutvei, H., 2001. Comparison of microgrowth pattern in *Margaritifera margaritifera* shells from south and north Sweden. *Am. Malacol. Bull.* 16, 239–250.
- Frisia, S., Borsato, A., Preto, N., McDermott, F., 2003. Late Holocene annual growth in three Alpine stalagmite record the influence of solar activity and the North Atlantic Oscillation on winter climate. *Earth Planet. Sci. Lett.* 216, 411–424.
- Grudd, H., Briffa, K.R., Karlén, W., Bartholin, T.S., Jones, P.D., Kromer, B., 2002. A 7400-year tree-ring chronology in northern Swedish Lapland: natural climatic variability expressed on annual to millennial timescales. *Holocene* 12, 657–665.
- Hallam, A., 1965. Environmental causes of stunting in living and fossil marine benthonic invertebrates. *Palaeontology* 8, 132–155.

- Hastie, L.C., Young, M.R., Boon, P.J., 2000. Growth characteristics of freshwater pearl mussels, *Margaritifera margaritifera* (L.). *Freshw. Biol.* 43, 243–256.
- Hemming, N.G., Hanson, G.N., 1992. Boron isotopic composition and concentration in modern marine carbonates. *Geochim. Cosmochim. Acta* 56, 537–543.
- Hendelberg, J., 1961. The freshwater pearl mussel. Report of the Institute of Freshwater Research Drottningholm, vol. 41, pp. 149–171.
- Huber, B., 1943. Über die Sicherheit jahrringchronologischer Datierung. *Holz* 10/12, 263–268.
- Hurrell, J.W., 1995. Decadal trends in the North Atlantic oscillation: regional temperatures and precipitation. *Science* 269, 676–679.
- Jones, D.S., Arthur, M.A., Allard, D.J., 1989. Sclerochronological records of temperature and growth from shells of *Mercenaria mercenaria* from Narragansett Bay, Rhode Island. *Mar. Biol.* 102, 225–234.
- Kennish, M.J., Olsson, R.K., 1975. Effects of thermal discharges on the microstructural growth of *Mercenaria mercenaria*. *Environ. Geol. (Springer)* 1, 41–64.
- Lewis, D.E., Cerrato, R.M., 1997. Growth uncoupling and the relationship between shell growth and metabolism in the soft shell clam *Mya arenaria*. *Mar. Ecol., Prog. Ser.* 158, 177–189.
- Linderholm, H.W., Solberg, B.Ø., Lindholm, M., 2003. Tree-ring records from central Fennoscandia: the relationship between tree growth and climate along a west–east transect. *Holocene* 13, 887–895.
- Marchitto, T.A., Jones, G.A., Goodfriend, G.A., Weidman, C.R., 2000. Precise temporal correlation of Holocene mollusk shells using sclerochronology. *Quat. Res.* 53, 236–246.
- McDermott, F., Frisia, S., Huang, Y., Longinelli, A., Spiro, S., Heaton, T.H.E., Hawkesworth, C., Borsato, A., Keppens, E., Fairchild, I., van Borgh, C., Verheyden, S., Selmo, E., 1999. Holocene climate variability in Europe: evidence from  $\delta^{18}\text{O}$ , textural and extension-rate variations in speleothems. *Quat. Sci. Rev.* 18, 1021–1038.
- Mutvei, H., Westermark, T., 2001. How environmental information can be obtained from Naiad shells. In: Bauer, G., Wächtler, K. (Eds.), *Ecology and Evolution of the Freshwater Mussels Unionoidea*, Ecological Studies, vol. 145. Springer, Berlin, Heidelberg, pp. 367–379.
- Mutvei, H., Westermark, T., Dunca, E., Carell, B., Forberg, S., Bignert, A., 1994. Methods for the study of environmental changes using the structural and chemical information in molluscan shells. *Bull. Mus. Océanograph. Monaco, Num. Spéc.* 13, 163–186.
- Mutvei, H., Dunca, E., Timm, H., Slepukhina, T., 1996. Structure and growth rates of bivalve shells as indicators of environmental changes and pollution. *Bull. Mus. Océanograph. Monaco, Num. Spéc.* 14, 65–72.
- Niggemann, S., Mangini, A., Richter, D.K., Wurth, G., 2003. A paleoclimate record of the last 17,600 years in stalagmites from the B7 cave, Sauerland, Germany. *Quat. Sci. Rev.* 22, 555–567.
- Pannella, G., MacClintock, C., 1968. Biological and environmental rhythms reflected in molluscan shell growth. *Paleontol. Soc. Mem.* 42, 64–81.
- Rhoads, D.C., Morse, J.W., 1971. Evolutionary and ecologic significance of oxygen-deficient marine basins. *Lethaia* 4, 413–428.
- Schmidt, H., 1931. Die ursprünglichen Zusammenhänge zwischen Harz und Rheinischem Schiefergebirge. *Naturwissenschaften* 18, 911–916.
- Schöne, B.R., Oschmann, W., Rössler, J., Freyre Castro, A.D., Houk, S.D., Krönecke, I., Dreyer, W., Janssen, R., Rumohr, H., Dunca, E., 2003. North Atlantic Oscillation dynamics recorded in shells of a long-lived bivalve mollusk. *Geology* 31, 1237–1240.
- Schöne, B.R., Dunca, E., Mutvei, H., Norlund, U., 2004. A 217-year record of summer air temperature reconstructed from freshwater pearl mussels (*M. margaritifera*, Sweden). *Quat. Sci. Rev.* 23, 1803–1816, 2057.
- Schöne, B.R., Dunca, E., Fiebig, J., Pfeiffer, M., 2005. Mutvei's solution: an ideal agent for resolving microgrowth structures of biogenic carbonates. *Palaeogeogr. Palaeoclimatol. Palaeoecol.* 228, 149–166, doi:10.1016/j.palaeo.2005.03.054.
- Schweingruber, F.H., Briffa, K.R., Jones, P.D., 1991. Yearly maps of summer temperatures in Western Europe from A.D. 1750 to 1975 and Western North America from 1600 to 1972. *Vegetatio* 92, 5–71.
- Sprung, M., 1984. Physiological energetics of mussel larvae (*Mytilus edulis*): I. Shell growth and biomass. *Mar. Ecol., Prog. Ser.* 17, 283–293.
- Tasch, P., 1953. Causes and paleoecological significance of dwarfed fossil marine invertebrates. *J. Paleontol.* 27, 356–444.
- Vengrosh, A., Kolodny, Y., Starinsky, A., Chivas, A.R., McCulloch, M.T., 1991. Coprecipitation and isotopic fractionation of boron in modern biogenic carbonates. *Geochim. Cosmochim. Acta* 55, 2901–2910.