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

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Abstract

Chemical and physical variation in skeletal structures of marine organisms can reflect environmental variability, forming the basis for reconstructing the conditions in which the organism lived. The successful use of these bio-archives for reconstructing seasonal and sub-seasonal environmental conditions is dependent on understanding sub-annual growth patterns and the timing of deposition of visible markers. We studied sub-annual shell growth patterns, as well as the timing and environmental processes initiating winter growth line deposition in two circumpolar bivalve mollusks, Serripes groenlandicus and Ciliatocardium ciliatum. Shell growth deposited during a 1-year deployment on oceanographic moorings in Kongsfjorden and Rijpfjorden, Svalbard, was analyzed in situ for $\delta^{18}$O using high spatial resolution secondary ion mass spectrometry (SIMS). A new digital method was developed to measure the location of SIMS spots along chronologically deposited shell material. 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, in order to derive sub-annual shell growth models. The resulting growth models indicated that the prominent winter growth band was formed during a slow shell growth period lasting from December until May in Kongsfjorden and from November until mid-June in Rijpfjorden. The length of the slow growth period during winter was most likely controlled by food availability. Shell growth rate during the growing season was significantly explained by temperature (marginal $R^2 = 0.29$) indicating that temperature is a major driver of shell growth rate when the food supply is sufficient. The insights of sub-annual growth of Arctic bivalves and the methods developed in our study are important contributions for further development of bivalve shells as proxy archives.
Introduction

Detailed records of oceanic conditions across a range of spatial and temporal scales are needed to understand implications of the rapid climate change on sensitive ecosystems [1]. This is especially true for the Arctic Ocean, where instrumental records are sparse [2] and the climate change is projected to be most pronounced [3–6]. Skeletal structures of calcifying marine organisms provide archives of ocean conditions that can be interpreted based on the geochemical signatures preserved within the calcium carbonate (CaCO₃) deposited during the life-time of the animal [7–11]. Visible growth lines deposited within these skeletal structures can be used as time markers to reconstruct their growth patterns back through time [12–15]. Marine organisms do not have a constant growth rate through time, complicating the dating of geochemical samples from their hard parts [15,16]. Knowledge of species-specific growth patterns and their seasonality is therefore required in order to facilitate interpretation of sub-annual geochemical proxy records [14].

High-latitude bivalve mollusks, such as *Serripes groenlandicus* Mohr, 1786 and *Ciliatocardium ciliatum* Fabricius, 1780, are long lived filter-feeding organisms that record ocean conditions within their aragonitic shells [17–19]. Their shell growth is sensitive to changes in food supply and temperature [15,20], and is regulated by large scale climatic drivers over annual to decadal scales [21–24]. Both species deposit prominent annual growth lines during winter that can be used to construct decadal to multi-centennial chronologies, with the possibility of sampling geochemical information on sub-seasonal resolution [10,14,15,25,26]. The usage of geochemical signatures, such as element-to-calcium ratios [18], as sub-annual proxies of seawater conditions has been difficult in these species, because the processes that trigger the deposition of the annual growth lines or the months during which these growth lines form are unknown [15]. Sejr et al. [20] suggested that shell growth of *C. ciliatum* is controlled by food availability due to significant negative correlations between annual sea-ice cover and shell increment width. The authors were, however, unable to further quantify this relationship due to lack of data on food availability and sub-seasonal growth rates. Furthermore, Ambrose et al. [15] suggested that a shortage of food triggers the deposition of the winter growth lines in *S. groenlandicus* and *C. ciliatum*, but the lack of sub-annual shell growth models prevented testing of that hypothesis and precluded estimating dates when the winter growth line was deposited. Identifying the specific timing of seasonal growth line deposition and the development of sub-annual shell growth models are necessary first steps for further geochemical proxy studies using these species.

The ratio of stable oxygen isotopes (¹⁸O/¹⁶O, denoted as $\delta^{18}O_{VPDB}$ values, expressed in parts per thousand relative to the Vienna PeeDee belemnite international reference) in biogenic carbonates, such as *S. groenlandicus* shells [25], is a well established geochemical proxy that can be used to reconstruct seawater temperatures, when the oxygen-isotope composition of the water ($\delta^{18}O_w$) is known [27–29]. If seawater salinity, temperature, and a salinity–$\delta^{18}O_w$ relationship [30] are known, these variables can be used to calculate the expected $\delta^{18}O$ values in bivalve shell CaCO₃ [31], thereby enabling determination of bivalve shell growth patterns [14,32,33].

Oxygen-isotope ratio analyses on bivalve mollusk shells are conventionally implemented by milling CaCO₃ powder along sectioned shells [18,32,34,35]. Carbon dioxide acquired by phosphoric acid digestion of sampled CaCO₃ powder is then analyzed for $\delta^{18}O$ values using a gas-source isotope ratio mass spectrometer with a typical precision of c. 0.05 ‰ [36]. However, this conventional method, requires relatively large sample sizes (> 10 µg), leading to a spatial
resolution of sampling that is incapable of investigating sub-annual growth banding of Arctic bivalves with a slow growth rate (few mm y\(^{-1}\)). In contrast, surface (= in situ) \(\delta^{18}O\) analysis by secondary ion mass spectrometry (SIMS), with a typical probe diameter of c. 10 \(\mu\)m and limited depth penetration (< 2 \(\mu\)m), consumes a much smaller amount of sample (c. 0.5 ng) compared to the conventional analysis. SIMS analysis is preferred when high spatial resolution sampling is required [e.g. 37], as long as the expected variation in \(\delta^{18}O\) values exceeds the lower precision obtained by the method (ca. 0.2 \%)..

A notable drawback of SIMS analysis of \(\delta^{18}O\), however, is that it sputters oxygen from all phases within the sampled volume, including organic-hosted oxygen [38] and carbonate-hosted sulphate [38, 39]. Further, SIMS-determined \(\delta^{18}O\) values are depleted in the heavy isotopes compared to the true isotopic abundances, because light ions are more easily emitted during the sputtering process relative to heavy ions due to the differences in energy of ions with different masses and bond energy discrepancies among sputtered molecules [37, 40]. Although this effect, called instrumental mass fractionation (IMF), is accounted for by usage of reference materials, the magnitude of IMF is reported to vary among CaCO\(_3\) minerals, such as calcite and aragonite [37, 41, 42]. Consequently, the \(\delta^{18}O\) values acquired by SIMS can differ from values obtained using the conventional phosphoric acid digestion method [31, 43, 44], and thus from predictions generated for the latter.

Spot samples taken along a section of chronologically deposited shell material generate two issues that complicate the interpretation of the measured geochemical signals. 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. Some studies on bivalve mollusk shells overcame this issue by sampling at locations where the sampling sequence can be placed perpendicularly against the growth lines, such as the umbo region [e.g. 45, 46]. This practice, however, limits the sampling resolution since most shell growth occurs at the shell margin. 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 [47, 48]. 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.

In our study, we used two common circumpolar bivalves, the Greenland cockle (S. groenlandicus) and the hairy cockle (C. ciliatum), deployed on oceanographic moorings within two fjords, with different oceanographic conditions, situated in the Svalbard archipelago. The objectives of this study were to: 1) establish sub-annual shell growth models for subsequent element/Ca analyses and interpretations, 2) study the effects of temperature and food availability on sub-annual shell growth patterns, and 3) test the hypothesis that the trigger for the winter growth line is reduced food supply [15], using an oceanographic dataset with sub-annual resolution.

Materials and Methods

Bivalves, mooring sites and oceanographic instrumentation

Oceanographic moorings are maintained in Kongsfjorden (K, 78°57.4′ N 11°49.6′ E, depth 170 m) and Rijpfjorden (R, 80°18′ N 22°20′ E, depth 220 m), situated in Svalbard (Figure 1) [15, 49, 50].
Calcein-marked bivalve mollusks (Serripes groenlandicus and Ciliatocardium ciliatum) were placed in 7 mm plastic-mesh (Vexar L-32) baskets on each mooring in September 2009 and recovered one year later (Table 1). Originally, the bivalve mollusks were collected from Spitsbergenbanken in the Western Barents Sea (Figure 1) in August 2009 using a triangular dredge, after which they were held in flow-through seawater tanks for 4 weeks at the University Centre on Svalbard. Specimens were incubated in seawater with 125 mg L$^{-1}$ of calcein dye for 24 h as described in Ambrose et al. [15] before deployment on the oceanographic moorings within both fjords (Table 1).

Kongsfjorden is located on the west coast of Spitsbergen and is influenced by the West Spitsbergen Current, which brings relatively warm Atlantic water into the fjord (Figure 1) [51]. The mooring site in Kongsfjorden is rarely covered by sea-ice. In contrast, Rijpfjorden is located on Nordaustlandet facing northward to the Arctic Ocean. It has an irregular sill and a broad, shallow shelf (depth 100–200 m) outside the fjord that extends approximately 100 km to the north before the shelf-break of the polar basin [52]. Consequently, Rijpfjorden is strongly influenced by cold Arctic currents and is seasonally ice covered [50,52]. The entire water column in Rijpfjorden has a temperature of $-1.9 \degree C$ for 5-6 months each year during which the fjord is typically ice-covered [52]. Despite the shallow sill, the fjord is episodically affected by Atlantic water inflow events, leading to occasional rapid temperature and salinity fluctuations [53].

Temperature data loggers (HOBO TidbiT v2, accuracy $\pm 0.2 \degree C$) mounted on the baskets containing bivalves recorded temperature every 10 to 40 min throughout the deployment period. Fluorometers (Seapoint Sensors, Inc.) and CTDs (37-SM MicroCAT, Sea-Bird Electronics) that were located adjacent to the baskets on the moorings (Table 1) measured fluorescence and conductivity (as a proxy for salinity) every second hour. The fluorometers were used with manufacturer’s calibrations to provide estimates of seawater chlorophyll a concentration [54]. Since fouling or instrument drift may have affected the absolute voltage readings, a fluorescence index (FLI) was created by normalizing the voltage acquired from the fluorometers to the highest voltage recorded over the deployment period [15]. Daily averages of temperature and salinity were calculated from the mooring instrument data and used further in predicting $\delta^{18}O$ values for bivalve mollusk shell aragonite and data analyses.

Bivalve shell $\delta^{18}O$ measurements using SIMS

Individuals with the most growth (1.7–4.0 mm in shell length) during the deployment period were selected for $\delta^{18}O$ analyses. Samples from the basket at 15 m depth within Kongsfjorden were not analyzed for $\delta^{18}O$ values due to the lack of salinity measurements adjacent to the basket (Table 1). Shell height of each bivalve was measured from the umbo to the ventral margin, along the maximum growth axis, immediately after recovery (Figure 2). Next, soft tissue was removed, and the shells were dried at room temperature. Shells were stored dry until being embedded in epoxy resin and sectioned along the maximum growth axis using a Buehler Isomet low-speed saw, as described by Ambrose et al. [15] (Figure 2). The sections were then polished with a gradually finer sequence of sandpapers and finished using canvas with 1.5 $\mu$m AlO$_2$ powder. In order to locate the calcein line marking the beginning of the field deployment, the polished shell sections were photographed using fluorescent imaging (460–500 nm) with a Nikon episcopic compound microscope equipped with a Ludl motorized stage. Afterwards, the polished shell...
slabs were glued on petrographic slides using epoxy resin. The shells were cut and the resulting thick sections polished to a thickness of 2.0±0.1 mm. Growth increments during the mooring period were then cut and the pieces mounted on epoxy mounts with a diameter of 25 mm. The mounts were coated with ca. 30 nm of gold and analyzed for in situ δ¹⁸O values using secondary ion mass spectrometer (SIMS). Approximately one SIMS analysis was completed per 100 µm of shell growth until the prominent winter growth band, after which samples were taken at higher spatial resolution until the calcine line indicating the beginning of the mooring deployment period (Figure 3).

In situ oxygen-isotope ratio analyses were conducted using a CAMECA IMS 1280 large geometry SIMS at the NordSIM facility in Stockholm, Sweden. The analytical protocol closely followed the standard δ¹⁸O measurement procedure described for zircon by Whitehouse and Nemchin [55], which is applicable with little modification to other O-bearing minerals. A critically focused Cs⁺ primary beam with 20 keV impact energy and ca. 2 nA beam current was used to sputter the sample, with charge build-up mitigated by use of a normal-incidence, low-energy electron flooding gun. Each analysis consisted of an initial pre-sputter over a rastered 20 µm area to remove the gold coat and attain stable secondary ion emission. A 10 µm primary raster was retained for data acquisition in order to homogenize the primary beam profile on the sample; together with the intrinsic primary beam spot diameter, this approach resulted in average sample spot diameter of 20.2 ± 2.6 µm (SD, n = 450). Following centering of the secondary beam in the field aperture (field of view on the sample of 30 µm with 90x magnification transmission ion optics) the ¹⁸O (c. 2x10⁵ cps) and ¹⁸O ion beams were mass filtered at a mass resolution of ca. 2500 (M/∆M) and analyzed simultaneously by Faraday detectors with amplifiers housed in an evacuated, temperature stabilized chamber. The secondary magnetic field was locked at high stability using an NMR field sensor operating in regulation mode. All pre-sputter, beam centering, and data acquisition steps were automated in the run definition.

Fully automated sequences consisted of 2–3 measurements of the reference carbonate, Brown Yule Marble calcite (BYM, kindly provided by J. Craven, University of Edinburgh, from an original donation by B. Gilletti, Brown University) separating 5–7 measurements of the shell material. The regularly interspersed BYM measurements enabled correction of measured isotope ratios for any drift during the run sequence. Three separate bulk analyses of the BYM calcite yielded an average δ¹⁸OVPDB of -6.55 ± 0.13 ‰ (SD, J. Craven personal communication), in good agreement with other reported values for the same material (e.g. Clemens et al. [56] who report -6.56 ± 0.06 ‰) and this value was used for calculation of instrumental mass fractionation (IMF) and normalization of shell δ¹⁸OVPDB values. The average repeatability (internal precision) of 679 analyses was 0.11 ‰ (SE on 12 4-second cycles; range from 0.05 to 0.31 ‰), while the average reproducibility (external precision) of 229 drift-corrected BYM analyses from four sessions was 0.14 ‰ (SD, range from 0.12 to 0.17 ‰). The uncertainty on each reported δ¹⁸O value results from propagation of the repeatability with the reproducibility for the specific analytical session. SIMS data is available as supplementary information (Data S2). All δ¹⁸O values in this study are given relative to VPDB unless specified otherwise.
Predicted bivalve shell aragonite $\delta^{18}$O values

Predicted $\delta^{18}$O values in biogenic aragonite were calculated from known seawater temperature and salinity records based on a modified version of the equation formed by Grossman and Ku [31]. The equation was corrected for the Vienna normalized scale by subtracting 0.27 [57–59] from the original equation:

$$T = 20.60 - 4.34[\delta^{18}O_{\text{aragonite}} - (\delta^{18}O_{\text{water}} - 0.27)]$$  \hspace{1cm} (1)

where $T$ is temperature in °C and $\delta^{18}$O values for aragonite and seawater expressed relative to Vienna Pee Dee Belemnite (VPDB) and Vienna Standard Mean Ocean Water (VSMOW), respectively. Rearranging this equation and simplifying the constants allows prediction of the $\delta^{18}$O values in bivalve mollusk shell aragonite according to:

$$\delta^{18}O_{\text{aragonite}} \text{ VPDB}(\%o) \approx -0.23 T(°C) + \delta^{18}O_{\text{water}} \text{ VSMOW}(\%o) + 4.48$$  \hspace{1cm} (2)

Temperature measurements were acquired from the temperature loggers deployed in each basket within each fjord. Predicted $\delta^{18}$O values for seawater were calculated using daily salinity measurements from the mooring CTDs (Table 1), and the salinity–$\delta^{18}$O$_{w}$ mixing-line equation for Kongsfjorden from MacLachlan et al. [30]:

$$\delta^{18}O_{\text{water}} \text{ VSMOW}(\%o) = 0.43 \text{ Salinity} - 14.68$$  \hspace{1cm} (3)

Prediction intervals (95% level) for predicted bivalve shell $\delta^{18}$O values were calculated using average propagated external and internal error for each basket as standard deviation and an assumption of normal distribution. Uncertainties in $\delta^{18}$O equations [30,31] were not considered and the actual prediction intervals could be wider.

Measurement of SIMS sample spot distances along bivalve shell sections

The measurement technique developed for this study estimates the location of SIMS $\delta^{18}$O sample spots relative to the historical location of the shell margin along an axis approximately perpendicular to the direction of growth (called measurement axis; Figure 3). In principle, the locations where growth lines reached the shell surface were first projected to the measurement axis ($L_1$ and $L_2$; Figure 4). Next, centroids of each sample spot were related to the closest growth lines on both sides of the sample spot by a distance ratio $d_1/d_2$ (Figure 4). This distance ratio was then scaled to the projected points ($L_1$ and $L_2$) along the measurement axis, such that the relative distance to the adjacent growth lines remained the same ($d_1/d_2 = d_{L_1}/d_{L_2}$). Finally, positions of the scaled sample spots along the measurement axis were measured from the point where the shell margin was projected to the measurement axis (Figure 3). The procedure was repeated for the closest points to $L_1$ and $L_2$ along the perimeter of a sample spot to estimate the extent to which a sample was averaged [48,60].

Growth lines were first identified from high resolution photographs of polished thick sections before SIMS and LA-ICP-MS sampling and compared to high resolution photographs of the sections after sampling. Identified growth lines and sample spots were then marked using ImageJ [61] and imported to R [62] using RImageJROI package [63]. The spatstat package [64]
was used to calculate the distances as described above. The resulting R functions were compiled to the sclero package [65] and the work-flow is described in detail in the associated tutorial [66]. The distances given by the sclero package were afterwards inverted to correspond to the direction of growth (Figure 3). Hence, the distances included in the figures are given along the measurement axis, starting from the calcein line.

Alignment of predicted and measured intra-shell $\delta^{18}$O profiles

Sub-annual growth patterns for *S. groenlandicus* and *C. ciliatum* were determined by aligning the SIMS-measured $\delta^{18}$O values with predicted $\delta^{18}$O values using dynamic time warping (DTW). The DTW method consists of algorithms that align and compare temporal sequences, which might vary in time or speed (i.e. along the x-axis) [67–70]. Dynamic time warping was run using the dtw package [69] for R. In DTW, the alignment of reference (predicted bivalve shell $\delta^{18}$O values in this study) and query (SIMS-measured $\delta^{18}$O profiles) indexes are constrained by algorithms called “step pattern” [69, 71]. The chosen step pattern (*symmetric2*) allowed flexible alignment of a query index along the reference index. A detailed description of DTW alignment procedure used in this study is presented in Text S1.

In brief, SIMS-measured $\delta^{18}$O values were first standardized to the maximum and minimum value of a predicted $\delta^{18}$O profile due to the offset between SIMS-measured and predicted $\delta^{18}$O values (possible reasons for the offset are discussed later). Such standardization required two assumptions: 1) maximum and minimum $\delta^{18}$O value over the predicted $\delta^{18}$O profile (mooring deployment period) was sampled along each shell, and 2) SIMS-measured $\delta^{18}$O values along each shell section were consistently related to predicted $\delta^{18}$O values within some random error (signal noise). Oxygen-isotope values predicted for the first and last day of mooring deployment were added as standardized $\delta^{18}$O values for the initial calcein mark ($d = 0 \, \mu m$) and the shell margin in the measured profile, respectively. Next, measured $\delta^{18}$O profiles were linearly interpolated such that the length a query index matched with that of the reference index, which consisted of daily predictions of $\delta^{18}$O in shell aragonite. Finally, the uncertainty of aligned query index values was estimated by random sampling of distances within the minimum and maximum extent of each SIMS sample spot (Figure 4). The resulting permutation allowed estimation of uncertainty introduced by several possible DTW alignments in resulting growth models: the procedure not only changed the distance of each measured value, but could also change the order, if maximum and minimum limits of adjacent samples overlapped. The permutation was repeated 500 times to estimate the maximum and minimum extent of each SIMS sample spot over time and to calculate 95% quantiles for growth models.

Predicted $\delta^{18}$O values for 26 m depth within Rijpfjorden decreased sharply during the ice-formation, increased again a week later only to decrease to the levels lasting until mid-July (Figure 5). This $\delta^{18}$O fluctuation interfered with $\delta^{18}$O profile alignment resulting to inadequate DTW alignments. It is likely that this fluctuation was not representatively sampled in analyzed shells, as the fluctuation took place during the winter with assumed slow shell growth (likely over couple of $\mu m$ [15]). Consequently, this fluctuation was removed from the reference index used for DTW by linearly interpolating daily values using a $\delta^{18}$O value before (2009-12-23) and after (2010-02-07) the fluctuation. The fit of the SIMS-measured $\delta^{18}$O values with predicted $\delta^{18}$O values after DTW alignment was quantified using linear regressions (Predicted $\delta^{18}$O ∼
Measured $\delta^{18}$O). Additionally, the range of days for which each measured $\delta^{18}$O value was aligned during 500 permutations (referred to as “accuracy”) was used to illustrate the uncertainty in the resulting DTW alignments.

**Relationships between shell growth rate and mooring recordings**

Weekly growth rates for each bivalve shell were calculated from the sub-annual growth models using the DTW aligned centroid values. Two individuals (J and K) showed anomalously low $\delta^{18}$O values towards the ventral margin leading to implausible growth models; these two specimens were removed from further growth rate analyses. The effects of seawater temperature, fluorescence index and salinity values (fixed effects) to shell growth rate (response variable) were assessed using linear mixed effects regression models (LMM). Relationships between shell growth rate and the fixed effects was logarithmic, and consequently shell growth rate was logarithm transformed prior analyses. Individual bivalve shells (termed “Samples” in consequent tables) and weeks from deployment were used as crossed random effects assuming a random intercept and a constant slope in LMMs [72]. Marginal coefficient of variation ($R^2_m$; [73]) was used to examine the variance explained by each response variable separately (Model 1; see Text S2). The significance and relative effects of each fixed effect on the growth rate were calculated using standardized values of temperature, fluorescence index and salinity (each variable was centered to their means and scaled to their standard deviations). The significance of these model parameters was estimated using Satterthwaite approximation for denominator degrees of freedom [74] (Model 2; see Text S2). Linear mixed models were calculated using the lme4 [75] and lmerTest packages [76], and $R^2_m$ values using the MuMIn package [77]. All statistics were run using R statistical programming environment [62].

**Results**

**Seawater mooring data**

Seawater temperature in Kongsfjorden ranged between −1.8 and 5.1 °C, with highest temperatures at the beginning (September to December 2009) and the end (mid-July to September 2010) of the mooring deployment period (Figure 5A). Kongsfjorden experienced three Atlantic water inflow events during October 2009 and January and March 2010 (Figure 5A), as indicated by abrupt increases in seawater temperature and salinity (Figure 5B). Except for these events, salinity was relatively stable throughout the deployment period, ranging between 33.3 and 35.0. The mooring site in Kongsfjorden was not ice-covered during the deployment period. Rijpfjorden experienced seawater temperatures between −1.9 and 4.8 °C, and the fjord was ice-covered from mid-February until mid-July (inferred from mooring ADCP data as described in Wallace et al. [50], and confirmed from ice charts [78]). Seawater temperatures were similar at both measured depths within Rijpfjorden until late August, when the surface layer cooled by approximately 3 °C relative to the deeper (25 m) layer. Rijpfjorden experienced melt-water induced salinity fluctuations ranging between 30.5 and 33.5 from September 2009 until November and again from July to September 2010 (Figure 5B).

Predicted $\delta^{18}$O in bivalve shell aragonite varied between 2.9 and 4.9 % in Kongsfjorden,
between 2.1 and 5.1 ‰ at 10 m depth in Rijpfjorden, and between 3.1 and 5.0 ‰ at 26 m depth in Rijpfjorden (Figure 5C). The δ18O profiles gradually increased from September 2009 until mid-April 2010 in Kongsfjorden, and until July 2010 in Rijpfjorden. After the maximum values were attained, predicted δ18O values started to gradually decline in Kongsfjorden, whereas Rijpfjorden showed a more dramatic drop: predicted δ18O values decreased from maximum values to minimum values within two months (Figure 5C). Prediction intervals (95% level) for predicted δ18O values were ±0.40 ‰ in Kongsfjorden, 0.35 ‰ for the 15 m basket in Rijpfjorden, and 0.41 ‰ for the 25 m basket in Rijpfjorden. These values indicate likely non-detectable δ18O differences for SIMS-measured values in Kongsfjorden from mid-January until May and for Rijpfjorden from December until mid-June. The fluorescence index was close to zero prior to a dramatic increase during the spring to early summer (Figure 5D); the first fluorescence peak occurred earlier in Kongsfjorden (April) than in Rijpfjorden (mid-June).

**SIMS-measured shell δ18O profiles and alignment**

SIMS-measured bivalve shell δ18O values exhibited an increase from the calcein mark until or right after the winter growth band, after which δ18O values decreased gradually until the margin (Figure 6). This decrease was of higher magnitude in Rijpfjorden than in Kongsfjorden. Maximum δ18O values among shell sections within a basket were relatively consistent, the maximum difference of 1.14 ‰ between sample A and C from Kongsfjorden (Table 2). In contrast, minimum values varied more: two C. ciliatum shells (J and K; Table 2) from the 25 m basket in Rijpfjorden had an anomalously low minimum δ18O values (down to -8.7 ‰) towards the shell margin.

Trends in the SIMS-measured shell δ18O profiles matched with the changes in the predicted δ18O values, assuming relatively slow growth until the winter growth band and faster growth during the time of decrease in SIMS-measured δ18O values (Figures 7 and 8). The dynamic time warping (DTW) procedure provided significant regressions (p < 0.001) between predicted δ18O values for shell aragonite and the centroids of SIMS-measured δ18O values, with coefficients of determination (R²) ranging from 0.53 to 0.99 (Table 2). SIMS-measured δ18O values were on average 5.45 ± 0.22 (SE, n = 12) ‰ lower than the predicted δ18O values (Table 2). The average accuracy of aligned SIMS spots varied between 4 and 35 days among samples (Table 2).

**Sub-annual bivalve shell growth models**

Shell growth models resulting from DTW alignment exhibited two growth seasons during the mooring deployment: autumn (September to November–December) and summer (May–July to September; Figure 9). Two shells from Kongsfjorden [a C. ciliatum (C) and a S. groenlandicus (B)] grew considerably during the autumn growth season (43 and 52 % of their total annual growth increments), whereas the other shells grew the most during July–August, irrespective of fjord or species (Figures 9 and 10). There were no apparent differences in modeled growth patterns between species (Figure 9). Estimation of the timing of the start and end of the prominent winter growth band was associated with a high uncertainty for both fjords due to slow shell growth rate and resulting low number of SIMS δ18O sample spots taken adjacent to the winter growth bands (Figure 6). Relatively constant predicted δ18O values between February
and mid-June in Rijpfjorden further increased the uncertainty in assigning dates to the start and end of the winter growth band (Figure 5). Nevertheless, the growth models indicated that winter growth bands could have ended approximately simultaneously with the onset of summer growth in both fjords (Figure 9). Samples J and K (C. ciliatum from the basket at 25 m depth within Rijpfjorden) with anomalously low measured δ18O values have different growth models than other samples from Rijpfjorden, with most of their estimated shell growth occurring in the middle of the winter (Figures 8 and 9).

Shell growth rate was more seasonal in Rijpfjorden, with higher growth rates during July–August (up to 1141 μm w⁻¹), than in Kongsfjorden where the growth season lasted longer than in Rijpfjorden (7 months, Figure 10). Weekly averaged temperature provided significant regressions with logarithm of weekly shell growth marginal $R^2$ ranging from 0.30 to 0.40 among baskets (Table 3). By comparison, weekly averages of the fluorescence index demonstrated significant relationships with logarithm of shell growth rate only in Kongsfjorden marginal $R^2$ being 0.11. Salinity did not demonstrate significant relationships with shell growth rate. The relative effect estimates demonstrated that the effect of temperature on logarithm of growth rate was twice the magnitude compared to the effect of fluorescence (Table 3).

Discussion

The dynamic time warping (DTW) alignment of SIMS-measured δ18O values to predicted δ18O values lead to adequate sub-annual growth models in 9 of 11 studied shells (Figure 7, Table 2). These growth models suggested that shell growth occurs between May and December within Kongsfjorden and between mid-June and November in Rijpfjorden (Figure 9). Furthermore, the winter growth band most likely marked the slow growth periods in both fjords (Figure 9). The results further indicated that food availability sets the temporal limits for growth season, whereas temperature partly controls shell growth rate when a food source is sufficient (Figure 10, Table 3). Consequently, our results confirm the hypothesis by Ambrose et al. [15] that the winter growth lines can be used as proxies of the time period when a food supply is not sufficient to sustain shell growth.

Sub-annual shell growth patterns

Alignment of measured δ18O profiles with predicted δ18O values suggested sub-annual growth patterns that are characterized by shell growth commencing during the phytoplankton bloom, highest shell growth rate one to two months after the peak of the phytoplankton bloom, and a growth cessation starting from November–December in both fjords and both bivalve species (Figures 9 and 10). Growth of both species commenced approximately six weeks earlier within the warmer and more Atlantic water influenced Kongsfjorden, compared to Rijpfjorden (Figure 10) which is typically dominated by Arctic water masses [51–53]. Most of the shell growth in Rijpfjorden specimens occurred over a short time period, from late June until mid-August, whereas the growth season within Kongsfjorden was longer, i.e. beginning in May and ending in November (Figure 10). Consequently, shell growth models indicated a shorter duration shell growth hiatus for Kongsfjorden (∼5 months) compared to Rijpfjorden (∼7–8 months). These shell growth patterns, modeled from measured versus predicted δ18O profiles, are plausible. S.
groenlandicus and C. ciliatum are both filter-feeders and, because these bivalves were deployed on moorings in the water column, most of their diet likely consisted of phytoplankton and/or ice-algae. Primary production in the Arctic region is highly seasonal; very low or no food for filter-feeding bivalves is available during winter, before light-levels sufficient for photosynthesis return in the spring and lead to a phytoplankton or ice-algae bloom [53, 79]. Timing of the phytoplankton bloom at high latitudes is regulated by the angle of the Sun at a given latitude, by water-mass stratification, and by the occurrence of sea-ice, which can delay the bloom by several months [80]. The fluorescence index in our study can be used as a proxy for the timing and relative intensity of phytoplankton bloom events within the two fjords that were investigated [15, 81].

Since Kongsfjorden was ice-free during the deployment period, the timing of its phytoplankton bloom was relatively predictable, beginning in April, reaching its maximum in early May, and turning to post-bloom by June, as indicated by mooring fluorescence readings at 36 m depth (Figure 10). Such bloom dynamics correspond to what is known for Kongsfjorden from previous studies [79, 82, 83]. Our reconstruction of shell growth starting a month after the return of a food source is reasonable considering that somatic growth and replenishment of energy reserves is likely to precede shell growth [84, 85]. Growth models for shells deployed within Kongsfjorden show a slower growth during June, compared to earlier in the season (Figure 10). Although this could be explained by an imprecision in the DTW alignment, maturation of gonads could also have affected the shell growth rate: S. groenlandicus specimens deployed in Kongsfjorden were 31 mm and 39 mm in shell length during deployment and therefore likely sexually mature [86]. The timing of spawning in S. groenlandicus on Svalbard is not well documented, but specimens collected in May and held in aquaria at 2 °C spontaneously spawned in mid-June (Vihtakari, personal observation). The timing within laboratory aquaria matches with the time of slower shell growth observed in this field study. The C. ciliatum specimen from Kongsfjorden was likely not sexually mature (shell height only 21 mm) and differed from S. groenlandicus specimens in its shell growth pattern, with most shell growth taking place during the autumn (Figure 9). This interpretation of a large proportion of annual shell growth during autumn in Kongsfjorden is interesting (Figure 10). Even though our fluorescence data indicated a low abundance of chlorophyll a and thus photosynthesizing algae during the autumn (Figure 5D), the water column was likely to contain degrading phytoplankton and heterotrophic plankton that could have functioned as a food source for the bivalves [82, 87]. Bivalve shell growth during October to November has previously been reported for Chlamys islandica from South-East Greenland [88] and for Arctica islandica from the North Sea [14].

Rijpfjorden was ice covered until mid-July and consequently the timing of the phytoplankton bloom there was more difficult to establish than in Kongsfjorden. Seawater temperature records indicated sea-ice melt beginning in mid-June, coinciding with a peak in fluorescence index (Figure 5A and D). It is possible that this first peak in fluorescence was caused by ice-algae released to the water column due to sea ice melt. Alternatively, the phytoplankton bloom could have taken place during the ice melt in crevasses formed during the melting process [53]. Nevertheless, the fluorescence peak in mid-June clearly indicates a major food source for bivalves deployed on the Rijpfjorden mooring (Figure 5D). Shell growth in Rijpfjorden commenced almost simultaneously with this food occurrence, suggesting that the bivalves could have already replenished their energy reserves (Figure 10). Low fluorescence readings prior to June could indicate that some ice-algae was available within the water column, starting from April–May (Figure 5D), as also
reported by Leu et al. [53]. High fluorescence index values within Rijpfjorden from mid-August until the end of the bivalve deployment period (mid-September) are difficult to explain, but indicate a supply of an algal food source throughout the summer period. Highest shell growth rates during late-July for both baskets in Rijpfjorden are likely over-estimates due to poor fitting of measured $\delta^{18}\text{O}$ values in the DTW alignments and therefore the low shell growth rates during August to September should be treated with caution. One S. groenlandicus specimen from the basket at 25 m depth within Rijpfjorden was likely mature (shell length 40 mm), whereas other specimens were not. Nevertheless, the sub-annual growth pattern did not remarkably differ from the other specimens from the same basket (solid line with the highest growth rate in October in Figure 9C).

Two samples from Rijpfjorden had anomalously low $\delta^{18}\text{O}$ values towards the ventral margin. The growth models for these two shells indicated 42 to 50 % of the total annual shell growth occurred during the period between November and February, which is an unlikely scenario given the likely absence of food source. Even though we are unable to specify the definitive reasons for these low in situ SIMS-measured $\delta^{18}\text{O}$ values based on our dataset, growth models for these two shells are unrealistic and the samples thus have been excluded from further shell growth rate analyses (Figure 10, Table 3).

Effects of temperature and food availability on sub-annual shell growth rates

The overall weekly shell growth rates demonstrated significant relationships with measured weekly seawater temperature for all baskets with overall marginal $R^2$ of 0.29 (Table 3). Annual standardized shell growth index (SGI) of S. groenlandicus and C. ciliatum has previously been used as an environmental proxy, which has been linked to various climatic oscillation indexes [20–24]. Many of these studies have raised a fundamental question, i.e. whether it is food availability or seawater temperature that is the driving factor of shell growth rate and thereby correlation with the climatic indexes. Our data indicate that seawater temperature is an important contributing factor to shell growth in Arctic bivalves by controlling the metabolic rate of ectotherms, whereas food availability sets the limits for growth season, but does not necessarily correlate well with growth rate (Figure 10). Therefore, both food availability and seawater temperature are important factors regulating shell growth of Arctic bivalve mollusks, but if a food source is sufficient then, shell growth is likely to reflect variations in seawater temperature. Consequently, SGI is likely influenced by both, temperature and food availability, but also other factors that were not identified in this study.

Despite their uncertainties, our shell growth models indicate that the prominent winter growth bands were likely formed simultaneously with the shell growth cessation lasting from December until May within Kongsfjorden and from November until mid-June within Rijpfjorden (Figure 9). Food availability was the likely determinant controlling when shell growth commenced in the spring, as well as the slowing of shell growth during the autumn (Figure 10), and therefore our data support the hypothesis by Ambrose et al. [15] that winter growth bands can be used as a proxy of the timing of food abundance in S. groenlandicus and C. ciliatum grown on moorings in Svalbard. However, this finding does not necessarily apply directly to bivalve molluscs sampled from their natural habitats, since wave action and storms might resuspend sediments with potential food particles during winter and therefore provide a food source that can support shell
The offset between SIMS-measured $\delta^{18}O$ and predicted $\delta^{18}O$ values

Measured $\delta^{18}O$ values in bivalve shell aragonite were on average 5.5‰ lower compared to predicted $\delta^{18}O$ values (Table 2), the latter derived by combining continuous seawater temperature and salinity records with an established paleothermometry equation (Equation 2) [31, 59] and a salinity-$\delta^{18}O$ relationship for Kongsfjorden (Equation 3) [30]. All paleothermometry equations for biogenic carbonates have been generated using phosphoric acid digestion, which liberates oxygen only from carbonate phases [31,36,43,44]. In contrast, secondary ion mass spectrometry (SIMS) is a surface (in situ) technique that sputters oxygen ions from all phases contained within a shell sample, and SIMS-determined $\delta^{18}O$ values are often lower compared to the true isotopic abundances due to instrumental mass fractionation (IMF) [37, 40]. Because reliable aragonite SIMS-standards were not available, a calcite standard (Brown Yule Marble) was used in this study, even though the bivalve shells consisted of aragonite. Previous SIMS studies of oxygen isotope ratios in carbonates using similar instrumentation and analytical protocols to those in this study have yielded conflicting results regarding the relative instrumental mass fractionation between calcite and aragonite. Rollion-Bard et al. [37] report an average IMF$_{\text{arg}}$ − IMF$_{\text{calc}}$ of −2.8 ‰, further noting that this differed from session to session (range: −2.0 to −4.2 ‰) as a result of unspecified instrument conditions. In contrast, Gabitov [42] report IMF$_{\text{arg}}$ − IMF$_{\text{calc}}$ of 0.8 and 2.1 ‰ from two different sessions. It cannot be discounted that the marked differences in IMF might be attributed to undetected heterogeneity in the aragonite reference materials used or a dependence on trace element geochemistry as noted by Allison et al. [41] who report a range in IMF$_{\text{arg}}$ (calibrated using calcite) from 0.2 to −2.8 ‰ that might be dependent on Sr/Ca ratios. The range of reported $\delta^{18}O$ values for S. groenlandicus shells using the phosphoric acid digestion technique further indicate that the lower than predicted $\delta^{18}O$ values measured in this study were likely due to the in situ SIMS technique: Measured $\delta^{18}O$ values in this study ranged from −8.68 to 0.82 ‰ (Table 2), whereas predicted $\delta^{18}O$ values varied between 2.1 and 5.1 ‰. Khim [25] reported annual mean S. groenlandicus shell $\delta^{18}O$ values to range from 0.1 to 3.4 ‰, whereas predicted $\delta^{18}O$ values within a year varied between 0.3 and 2.7 ‰. Further, Carroll et al. [18] reported $\delta^{18}O$ values ranging from 0 to 4 ‰ in S. groenlandicus shell aragonite. It is possible that also other factors than differences in IMF contributed to the offset. These could include organic-hosted oxygen [38] and carbonate-hosted sulphate [39]. SIMS sampling of these two phases would supply oxygen to the measured signals and resultant $\delta^{18}O$ values, whereas the predicted $\delta^{18}O$ values are only for the carbonate oxygen within aragonite.

Conventional oxygen-isotope ratio analyses would have required $> 10 \mu g$ of aragonite powder [32,89], which could not have been milled at the required sampling resolution due to the small growth increments and thin shells (200–500 µm) of the Arctic bivalves investigated in this study (Figure 2). The aim of the present paper is not to use intra-shell SIMS-derived $\delta^{18}O$ values as an absolute seawater temperature proxy, but instead to use the observed intra-shell variations to estimate the sub-annual shell growth patterns. Therefore, these causes of inaccuracy when calibrating aragonite to calcite are not critical to our overall conclusions but, nonetheless, highlight some of the pitfalls of the in situ methodology that remain to be properly resolved by the SIMS analytical community. Oxygen-isotope ratio analysis of biogenic carbonates is a
standard method to reconstruct relationships between temperature and isotopic composition of seawater in paleoceanography [29,31], and it has successfully been used for a wide range of bivalves [32,47,90] including *S. groenlandicus* [18,25]. Therefore, shells from the same basket should in principle have similar $\delta^{18}$O values at a given time during the mooring deployment.

Despite the limitations of the *in situ* SIMS measurements used in this study, the similarities in intra-shell $\delta^{18}$O patterns (Figures 6) shell $\delta^{18}$O profiles were characterized by similarities with respect to patterns adjacent to the winter growth band and margin (Figures 6 and 8). The resulting feasible sub-annual shell growth models (Figure 9), allow us to confidently assume that possible oxygen that was not in paleothermometric equilibrium was randomly distributed along the studied shells. This assumption is further supported by us failing to detect any obvious deviations in measured $\delta^{18}$O values in association with sub-annual growth lines (Figures 6 and 8), which were considerably darker in color, and therefore likely to have contained more organic matrix [91]. Consequently, the methodology used was adequate to estimate sub-annual shell growth patterns of *S. groenlandicus* and *C. ciliatum*.

**Novel methodology**

Matching SIMS-measured $\delta^{18}$O values to values predicted for aragonite could have been done in many different ways due to the offset in the magnitude of measured and predicted $\delta^{18}$O values (Table 2), low variability in predicted $\delta^{18}$O values during winter (Figure 5C), and noise in the measured $\delta^{18}$O signal (Figures 6 and 8). Predicted and measured $\delta^{18}$O profiles are often aligned manually in similar studies [32,92], but manual alignment of $\delta^{18}$O values is rarely an objective or reproducible method. The dynamic time warping (DTW) method was chosen for alignment because of its reproducibility and objective alignment of $\delta^{18}$O values among bivalve shells (see Text S1). This study introduces a new digital method to align sample spots along chronologically deposited materials [65,66]. The method not only aligns sample spots in relation to growth lines, which can vary in angle, curvature and distance, but also helps to estimate the area covered by a sample spot (Figure 4). Dynamic time warping combined with permutations using the spatial range each SIMS sample covered along the measurement axis allowed an incorporation of time-averaging error to the growth model outcomes [47,48]. SIMS $\delta^{18}$O measurements were associated with a considerable signal noise component, which led to an imprecision in dynamic time warping (DTW) alignments (Figures 7 and 8) and the resulting shell growth models (Figure 9). Consequently, the DTW alignment method performed with a variable degree of success, depending on the variability of the measured $\delta^{18}$O profile, but in general resulted to plausible sub-annual shell growth models. We believe that the combination of the two methods used in this study will be useful for a wider research community, especially for time-series records with considerable intra-annual variability, and that these methods can be developed further.

**Conclusions**

Information on the sub-annual timing of visible growth band deposition is especially valuable for paleoclimate proxy studies, since these marks can be used as anchors to create chronologies, if their deposition is connected to processes that take place predictably from year to year. Our results demonstrate that the prominent winter growth band in *S. groenlandicus* and *C. ciliatum*
is likely deposited during the time of low food supply, and this information can be used further in proxy studies to constrain seasonal shell growth patterns. Our results also indicate that when food supply is sufficient, the shell growth rate of studied bivalves is likely to correlate with seawater temperature indicating that temperature is an important driver of Arctic bivalve mollusk shell growth. Dynamic time warping is a promising approach to better containing sub-seasonal and annual growth patterns in bivalves. Together with the sample spot alignment method developed for this study, DTW algorithms allowed an estimation of time-averaging error associated with geochemical sampling of bivalve shells. This study demonstrates a need for calibration of in situ SIMS $\delta^{18}O$ analyses in biomineral carbonates, such as aragonitic bivalve mollusk shells, including comparisons with the conventional phosphoric acid digestion and gas source isotope ratio mass spectrometry methods, the evaluation of the contribution of organic- and sulfate-hosted oxygen [38,39] to the SIMS $\delta^{18}O$ signals, and most importantly, addressing the potential inaccuracy in IMF of aragonite reference materials for SIMS.

Acknowledgments

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References


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Figure 1. Map over Svalbard and Barents Sea. Bivalves were deployed on moorings in two fjords on Svalbard (marked with red and blue dot). Red arrows indicate Atlantic water brought by the West Spitsbergen current and blue arrows the cold currents dominated by Arctic water masses. Svalbard is located to the upper left. The map was made by Norwegian Polar Institute.
Figure 2. Schematics of growth line deposition in bivalves. A) *Serripes groenlandicus* shell with visible external growth lines. Black line demonstrates the maximum growth axis along which cross-sections were cut. B) Shell cross-section along the maximum growth axis showing annual growth lines starting from the inner edge and curving upwards towards the visible shell surface. C) Growth during the mooring deployment showing a disturbance growth line formed during deployment (blue arrow), a prominent opaque winter growth line (red arrow) and several weaker subannual growth lines. Growth lines are deposited in an angle to direction of growth (DoG). Each growth line marks an approximate historical location of the shell margin and can be traced along the shell section.
Figure 3. Example of SIMS sample spot alignment. A) Shell cross-section after SIMS (small black dots) sampling. Large holes are laser-ablation craters (unpublished data). Colored lines on the shell section indicate growth lines marked with ImageJ. The blue line represents the calcein mark, and red lines the beginning and the end of the winter growth band. Yellow lines are sub-annual growth lines used to align sample spots. Purple horizontal line represents the measurement axis along which sample spots were aligned. DoG = direction of growth. B) Resulting digital representation of the shell section. Black dots indicate the SIMS samples and blue numbers the sample number. Sample spots are aligned along the measurement axis (purple line) relative to growth lines as explained in Figure 4. Open large circles indicate the laser-ablation craters. Scales are similar for both sub-figures and axes scales in B indicate distance in µm. X-axis scale is converted and begins from the deployment (= calcein mark; blue line). Sample spot sizes are presented in actual size in B. SIMS spots are larger than in reality in A to make them visible.
Figure 4. Alignment of SIMS sample spots along the measurement axis. Grey lines represent marked growth lines and open circle a sample spot. Centroid of the sample spot and the closest points to the growth lines along the perimeter (blue dots) were aligned such that $d_1/d_2 = d_{L_1}/d_{L_2}$ resulting to a segment along the measurement axis (blue dot with error bars). The dot represents the estimated location of the centroid and error bars the extent over which the SIMS sample was averaged.
Figure 5. Seawater mooring data and predicted $\delta^{18}$O values. A) Seawater temperature, B) salinity, C) predicted $\delta^{18}$O, and D) fluorescence index over the deployment period as measured by mooring instruments. Red solid line indicates Kongsfjorden 36 m depth, dark blue solid line Rijpfjorden 26 m depth and light blue dashed line Rijpfjorden 10 m depth. Shading for $\delta^{18}$O values illustrates 95% prediction intervals for observed $\delta^{18}$O values assuming a normal distribution and using propagated external and internal errors of SIMS samples averaged over each basket. The gray bar indicates the time of sea-ice cover in Rijpfjorden.
Figure 6. Measured $\delta^{18}$O values scaled along the measurement axis. (A-C): The basket at 25 m depth in Kongsfjorden. (D-F): the basket at 15 m depth in Rijpfjorden. (G-I) The basket at 25 m depth in Rijpfjorden. Specimens C, F, and I are C. ciliatum, and the rest are S. groenlandicus. Vertical error bars indicate 2 standard deviations of the propagated internal and external instrument precision for SIMS $\delta^{18}$O measurements. Horizontal error bars represent the estimated width of a sample spot along the measurement axis. First black vertical line from the left illustrates the calcein mark (=deployment in September 2009) and the second the shell margin (=recovery in September 2010). Dashed black vertical lines represent the beginning and the end of the prominent winter growth band. Grey vertical lines illustrate visible growth lines used in sample spot alignment (see Figure 3). Blue line represents interpolation trajectory that was used to align measured $\delta^{18}$O values with predicted $\delta^{18}$O values (see Text S1).
Figure 7. Dynamic time warping aligned measured SIMS $\delta^{18}$O values. (A-C) specimens from the basket at 25 m depth in Kongsfjorden, (D-F) the basket at 15 m depth in Rijpfjorden, and the (G-I) basket at 25 m depth in Rijpfjorden. Specimens C, F, and I are *C. ciliatum*, and the rest are *S. groenlandicus*. Predicted $\delta^{18}$O values are given as black line and follow the left Y-axis scale. Grey shading illustrates the 95% prediction interval for predicted $\delta^{18}$O values. Red dots represent the DTW aligned dates for centroids of measured $\delta^{18}$O values and follow the right Y-axis scale. Horizontal error bars indicate the maximum and minimum date assigned for each measured $\delta^{18}$O value after 500 DTW permutations. Vertical dashed lines from the left indicate the DTW aligned beginning and end of the dark winter growth band, respectively. Grey shading under the dashed lines indicate the minimum and maximum dates assigned for the winter growth band during 500 DTW permutations.
Figure 8. Samples with anomalously low $\delta^{18}$O values. Measured SIMS $\delta^{18}$O values (left panels) and dynamic time warping aligned measured SIMS $\delta^{18}$O values (right panels) for two *C. ciliatum* specimens (*J* and *K*) with anomalously low $\delta^{18}$O values towards the end of the shell growth from the basket at 25 m depth in Rijpfjorden. See Figures 6 and 7 for further explanation.
Figure 9. Growth models after matching $\delta^{18}$O values. A) Kongsfjorden 25 m basket, B) Rijpfjorden 15 m basket, C) samples from Rijpfjorden 25 m basket with a decent DTW alignment fit, and D) samples from Rijpfjorden 25 m basket with anomalously low $\delta^{18}$O values towards the end of the shell growth. Solid lines represent estimated growth models using centroids of SIMS samples for *S. groenlandicus* and dashed lines for *C. ciliatum*. Grey shading illustrates 2.5 and 97.5 % quantiles of 500 permutation runs. Red line illustrates the estimated maximum extent of winter growth band using quantiles.
**Figure 10. Shell growth rate related to mooring instrument data.** Weekly averaged growth rate (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 growth rate. Fluorescence measurements are from 36 m in Kongsfjorden and from 10 m in Rijpfjorden. The gray shaded area represents the estimated maximum extent for winter growth bands.
Tables

**Table 1. Overview of analyzed bivalve shells and mooring instruments.** Columns from the left: the fjord; abbreviation used in figures (Abbr.); depth of bivalve baskets (Depth); depth of mooring instruments (CTD) for salinity (S) and fluorescence (F) measurements; number of $\delta^{18}$O analyzed bivalves (n) per species (Ser = *S. groenlandicus*, Cil = *C. ciliatum*); average shell height of bivalve species (±1SD); and deployment (In) and recovery dates (Out). Temperature was measured using loggers attached to each basket.

<table>
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<th>Height (mm)</th>
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**Table 2. Goodness-of-fit indicators for aligned $\delta^{18}$O profiles:** Regression parameters between predicted and measured $\delta^{18}$O values ($\beta_0$ = intercept, $\beta_1$ = slope, $R^2$ = coefficient of determination. P was < 0.001 for all regressions); Minimum, median and maximum measured SIMS $\delta^{18}$O value for each sample; Number of SIMS sample spots (N); Average offset ± SE (% VPDB) between measured and predicted $\delta^{18}$O values (Offset); and average accuracy estimate for aligned measured SIMS $\delta^{18}$O ± SE in days (Accuracy).

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<td>RB</td>
<td>Cil</td>
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<td>-1.38</td>
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<td>Cil</td>
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<td>0.18</td>
<td>0.56</td>
<td>-8.68</td>
<td>-2.28</td>
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Table 3. Linear mixed effect regression parameters between logarithm of growth rate and temperature (T), fluorescence index (F), and salinity (S). Individual shells and weeks from deployment were used as random effects. Treatment represents different baskets (see Table 1), “All” expressing the overall model for all data. Parameter refers to the model parameter (T, S, F = fixed effect, and $\beta_0 = \text{intercept}$). $R^2_m$ gives the marginal coefficient of determination (variation explained by the fixed effect), whereas $R^2_c$ indicates the conditional coefficient of determination (variation explained by the fixed effect as well as by the random effects). Estimate indicates estimated standardized intercept for $\beta_0$ and slope for T, F, and S. Remaining columns represent t-statistics estimated based on Satterthwaite’s approximation for denominator degrees of freedom: standard error (SE), the approximate denominator degrees of freedom (DF), t-statistics (t-value) and the associated p-value from a t-distribution (p-value). $R^2$ values were calculated using Model 1, whereas the ANOVA statistics were estimated using standardized predictor variables and Model 2 (see Text S2 for details).

<table>
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<th>Treatment</th>
<th>Parameter</th>
<th>$R^2_m$</th>
<th>$R^2_c$</th>
<th>Estimate</th>
<th>SE</th>
<th>DF</th>
<th>t-value</th>
<th>p-value</th>
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<tr>
<td></td>
<td>S</td>
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</tbody>
</table>
Supporting Information Legends

Text S1. Alignment of predicted and measured intra-shell $\delta^{18}$O profiles. Detailed information about dynamic time warping (DTW) procedure used to align SIMS-measured $\delta^{18}$O profiles with predicted $\delta^{18}$O profiles.

Text S2. Linear mixed model formulations used to examine the relationships between shell growth rate and mooring instrument recordings.

Data S1. Example dataset for DTW alignment presented in Text S1. The dataset is in R format (.rda) and can be opened in R using the `load()` function.

Data S2. SIMS $\delta^{18}$O data. The file is in Excel format (.xlsx). Analyzed SIMS samples from bivalve shells are separated to “Shell_samples” tab, whereas Brown Yule Marble standards are presented in “BYM_standards” tab. See “Information” tab for column explanations.

Code S1. R scripts presented in Text S1. The file is in R script format (.R) and can be opened using R.

Code S2. R functions used in Code S1 and Text S1. The file is in R script format (.R) and can be opened using R.
Text S1: Alignment of predicted and measured intra-shell $\delta^{18}O$ profiles

SI1.1 Alignment example

Subannual growth patterns of *Serripes groenlandicus* and *Ciliatocardium ciliatum* were determined by aligning *in situ* SIMS-measured $\delta^{18}O$ profiles with predicted $\delta^{18}O$ profiles, the latter derived by combining continuous mooring seawater temperature and salinity records with a salinity–$\delta^{18}O$ relationship (see Material and Methods). One key aim of the alignment procedure was to establish a reproducible method, which would facilitate objective, rather than subjective, alignment of $\delta^{18}O$ profiles. Establishing such a method was, however, challenging due to signal noise in the measured SIMS $\delta^{18}O$ profiles and an offset between measured and predicted values. The signal noise and offset were likely caused by the surface (*in situ*) $\delta^{18}O$ measurement technique used in this study (see Discussion). The explanation here describes how the alignment method was chosen and applied using a subset of the data as a worked example. This example consists of data for the sample named “E” (*S. groenlandicus* specimen from the basket at 15 m depth deployed in Rijpfjorden; Table 2 in the main article). Data analyses were implemented using R statistical programming environment [1]. R scripts and data to reproduce all plots presented here are included as Supplementary Material. Example data is included as Data S1, the R script as Code S1, and the functions needed to run the script as Code S2. These functions are used to make the example code shorter and easier to read, but since the source code is available the functions can be freely modified. R packages *dtw* [2], *ggplot2* [3], *gridExtra* [4], *plyr* [5], and *scales* [6] have to be installed for the example scripts to work.

Dynamic time warping (DTW) was used to align the predicted and measured SIMS $\delta^{18}O$ profiles [2,7]. The DTW method consists of algorithms that were developed to align and compare temporal sequences, which might vary in time or speed (i.e. along the x-axis) [2,8–10], and was therefore a suitable method to achieve the alignment required by this study. In DTW the alignment of reference (predicted $\delta^{18}O$ values in this study) and query (measured SIMS $\delta^{18}O$ profiles) indexes are constrained by algorithms referred as “step patterns” [2,7]. A step pattern was chosen to allow the query index to be aligned along the reference index as flexibly as possible. The chosen step pattern, symmetric2, constrains the alignment by giving one diagonal step the same cost as two equivalent steps along the sides (see Figure 3 in Giorgino [2]). It allows assignment of one query index value to several reference index values, but also aligns several query index values to one reference index value.
SI1.2 Patterns in measured and predicted $\delta^{18}$O profiles

Depending on the position along graphs’ x-axes, predicted $\delta^{18}$O values were 1 to 6 $\%_\circ$ (all $\delta^{18}$O values in this document are reported relative to VPDB) higher compared to measured SIMS values (Figure S1). In order to align the two $\delta^{18}$O profiles it was necessary to assume that the measured SIMS $\delta^{18}$O values along the shell section were consistently related to predicted $\delta^{18}$O values within some random error (signal noise). In other words, the relationship between a predicted and measured value was assumed to be:

$$\delta^{18}\text{O}_{\text{predicted}} = \beta_1 \delta^{18}\text{O}_{\text{measured}} + \beta_0 + \epsilon$$

(S1)

where $\beta_1$ and $\beta_0$ are the slope and intercept of the linear regression between all predicted and measured SIMS $\delta^{18}$O values within a shell sample (see Table 2), and $\epsilon$ is the random error or noise in the measured $\delta^{18}$O signal for each SIMS measurement separately (analogous to residuals). It should be noted that the noise component does not necessarily indicate biased $\delta^{18}$O measurements, but is simply assigned to measured values which do not fit the resulting DTW alignment model.

The predicted $\delta^{18}$O profile for the worked example was characterized by lower values at the beginning of the deployment (September 2009 until January 2010) and again late during the deployment (July 2010 until recovery, Figure S1). Measured SIMS $\delta^{18}$O values were low until approximately 600 $\mu$m from calcein mark indicating that this part of the shell was deposited relatively soon after deployment. After this $\delta^{18}$O values demonstrated a large increase over a short distance (600 - 640 $\mu$m), possibly indicating cessation, or slowing down, of shell growth. After reaching maximum at 640 $\mu$m, measured SIMS $\delta^{18}$O values fluctuated between -1.44 and 0.21 $\%_\circ$ until a gradual decline that started after 1500 $\mu$m. The cause for the fluctuation is unknown. Nevertheless, judging from two other samples from the same basket deployed in the basket at 15 m depth within Rijpfjorden (Figure 6 in the main article), where similar fluctuation after the growth line was not present, this fluctuation could be assigned to noise component ($\epsilon$) in the measured $\delta^{18}$O signal. After reaching a minimum at 2870 $\mu$m measured SIMS $\delta^{18}$O values increased again – a pattern that mirrored well the predicted $\delta^{18}$O values from August until recovery in mid-September.

Identifying the position along the shell section where measured SIMS $\delta^{18}$O values started to decrease ties that shell distance to mid-June 2010 coinciding with a sharp decline in predicted shell $\delta^{18}$O values. Due to the SIMS-measured $\delta^{18}$O fluctuation between 640 and 1500 $\mu$m there were two feasible scenarios: 1) Measured SIMS $\delta^{18}$O values would start decreasing at 1500 $\mu$m. Values between 1000 and 1200 would contain large amount of negative signal noise ($-\epsilon$). 2) Measured values would start decreasing after 640 $\mu$m. Values between 1200 and 1500 would contain some positive signal noise, and values between 1000 and 1200 would be associated with some negative signal noise. The first scenario is supported by specimen D, where measured SIMS $\delta^{18}$O values remain high until the middle of the shell section (~2000 $\mu$m from calcein line, Figure 6). On the other hand, the second scenario is supported by specimen F (C. ciliatum from basket at 15 m depth in Rijpfjorden), where measured SIMS $\delta^{18}$O values begin to decline right after the measured $\delta^{18}$O maximum at distance of 664 $\mu$m, only one fifth of the distance from the shell margin (Figure 6). Replicate samples, therefore, do not shed more light into the problem at hand and it is necessary to acknowledge that both scenarios are equally valid by purely looking...
Figure S1. A) Predicted aragonite bivalve shell $\delta^{18}O$ values at 10 m depth in Rijpfjorden. X-axis represents time through the deployment period ticks specifying the first day of each month. B) In situ measured SIMS $\delta^{18}O$ values for specimen E (*Serripes* specimen from basket at 15 m depth in Rijpfjorden; Table 2). Dashed vertical lines indicate the beginning and the end of the dark winter growth band. Dotted vertical lines represent the calcein line that denotes the start of the field deployment and the margin of the shell section, on the left and right respectively. X-axis represents distance in $\mu$m measured from deployment, i.e. the calcein line.

SI1.3 Standardization of measured SIMS $\delta^{18}O$ profiles and the initial DTW

Due to the offset between measured SIMS and predicted $\delta^{18}O$ values, the measured $\delta^{18}O$ profile was standardized to the minimum and maximum of predicted $\delta^{18}O$ profiles before running the DTW alignments. The chosen standardization procedure resulted in maximum measured SIMS $\delta^{18}O$ values being aligned with maximum predicted $\delta^{18}O$ values and enhanced the assignment of non-fitting values as noise component ($\epsilon$) in measured signal following the first assumption (Equation S1). Further, the procedure also lead to an assumption that maximum and minimum $\delta^{18}O$ values were measured along each shell section. The bias introduced by this assumption, however, was evaluated small as the resulting DTW alignments were primarily driven by the period of decrease in measured SIMS $\delta^{18}O$ values aligned with the rapid decline in predicted $\delta^{18}O$ values from July to August (Figure S1).
An initial DTW alignment was run using the standardized measured SIMS $\delta^{18}$O values as query index and the predicted $\delta^{18}$O values as the reference index (Figure S2A). The model resulted in an alignment following scenario 1, as explained in the previous section. Fluctuation in measured SIMS $\delta^{18}$O values between 640 and 1200 µm was interpreted as negative signal noise ($-\epsilon$) as demonstrated by eight query index values being assigned to one day in mid-February (Figure S2A). Consequently, the resulting growth model indicated approximately 600 µm of shell growth for the same day in mid-February (Figure S2B). Sudden shell growth in mid winter is not likely in a high Arctic fjord with an extreme seasonality and food source returning at the earliest in April-May in the form of ice algae [11–13]. This initial DTW alignment performed poorly with several non-fitting values, including at the both ends of the reference index. Therefore, the DTW alignment had to be improved.

**Figure S2.** A) Initial DTW output that aligned standardized measured SIMS $\delta^{18}$O values (red dots, query index) overlaid with the predicted $\delta^{18}$O values (black line, reference index). Horizontal error bars indicate query index values that were assigned to several reference index values, thereby giving the minimum and maximum date for each query index value. Red dots on top of each other represent instances where several query index values were assigned to single reference index value, suggesting that these measured values became modeled as signal noise ($\epsilon$). B) Resulting sub-annual bivalve shell growth model (black line). Horizontal dotted line indicates the shell margin. Red dots and horizontal error bars are as described in A.
SI1.4 Addition of margins

The placing of early and late query index values can be improved by encouraging the DTW algorithm to do an open-ended alignment [2]. Since the predicted δ\textsuperscript{18}O values for the deployment calcein mark (dist = 0 µm) and shell margin (dist = 3607 µm) had been estimated, the predicted δ\textsuperscript{18}O values of the first and last day of mooring deployment were added as standardized δ\textsuperscript{18}O values for the beginning and the end of the measured SIMS profile (query index). In the resulting second DTW alignment, the non-fitting values at the beginning and end of the measured SIMS δ\textsuperscript{18}O profile were moved to mid-September and early-August respectively (Figure S3). Lower measured SIMS δ\textsuperscript{18}O values between 640 and 1200 µm were still assigned to one day in mid-February suggesting a higher growth rate that time. As stated in the previous section, such growth is not believable and the DTW alignment required further improvements.

**Figure S3.** A) Second DTW alignment of standardized measured SIMS δ\textsuperscript{18}O values (red dots) overlaid with predicted δ\textsuperscript{18}O values (black line), after inclusion of the first and last predicted value (blue dots). See Figure S2 for detailed explanation. B) Resulting regression relating predicted δ\textsuperscript{18}O values as a function of measured SIMS δ\textsuperscript{18}O values (Equation S1). Blue line indicates the regression model and grey shading the 95 % confidence intervals for the model. C) Resulting subannual growth model (black line). Horizontal dotted line indicates the shell margin. Red dots and horizontal error bars are as described in A, except that the first and last predicted δ\textsuperscript{18}O values are removed from the plot.
SI1.5 Matching the query index length with the reference index length

In the previous DTW runs, the alignment procedure did not consider the distance between measured SIMS $\delta^{18}O$ samples (X-axis in Figure S1B), because the query index was represented as a sequence of 60 values (58 SIMS $\delta^{18}O$ measurements and margins), which were then aligned with 380 daily values of predicted $\delta^{18}O$. Further, DTW algorithms might not always perform optimally in cases when the length of query and reference indexes differ drastically [14]. Consequently, it was possible to improve the DTW alignment by making the indexes the same length and simultaneously adding some of the distance information to the query index (measured SIMS $\delta^{18}O$ profile). This was done by upsampling the query index (measured SIMS $\delta^{18}O$ values) using linear interpolation and a constant distance along the distance axis for measured SIMS $\delta^{18}O$ values (Figure S1B). Since this study was also attempting to estimate the timing for the winter growth band (Figure 7 in the main article; see also [12]), intra-shell growth lines were first added as distances for linear interpolation and then complimented with additional points using a constant interval such that the length of query index (measured SIMS $\delta^{18}O$ values) matched with the length of the reference index (predicted daily $\delta^{18}O$ values).

The resulting third DTW alignment generally demonstrated a much better fit compared to the two previous models (Figures S4A-B, S3A-B and S2A). Two measurements at 398 and 589 $\mu$m were assigned to late October and associated with large amount of negative noise ($-\epsilon$, Figure S4A). This alignment is believable since both SIMS measurements had lower $\delta^{18}O$ values compared to proximal values (Figure S1A). Furthermore, 10 measured SIMS $\delta^{18}O$ values were all assigned to late October, which would mean approximately 500 $\mu$m of shell growth within a couple of days (Figure S4C). Although the magnitude of the growth rate is likely an overestimate, the general timing for this growth is feasible: Leu \textit{et al.} [11] reported 28\% of maximum chlorophyll-a concentration in pelagic particular organic matter measured in Rijpfjorden in 2007-2008 to take place in October indicating that bivalves would still have get enough nutrition to sustain shell growth during the autumn (September to November). The resultant subannual growth model estimated a growth cessation to last from November until mid-June (Figure S4C). Due to the 10 values assigned within couple of days in late October, the timing for the beginning of the growth hiatus is associated with a low confidence, but the general subannual shell growth pattern seems feasible since the mooring fluorescence data indicated the return of an algal food source in mid-June (Figure 5). The winter growth band coincides with the growth cessation which is estimated to have commenced in January and to have ended in July. Due to almost invariant predicted $\delta^{18}O$ values, the beginning of the winter growth band cannot be estimated accurately, as it could take place any time between January and June. The end of the winter growth band, on the other hand, is coincident with decreasing predicted $\delta^{18}O$ values and consequently is a relatively robust estimation within the model framework. The non-fitting measured SIMS $\delta^{18}O$ values in earlier models were now assigned to early July (Figure S4A). Although many of these values were still associated with one reference index value, they varied on both sides of the predicted values (both negative and positive residuals) increasing the fit of the regression model ($R^2 = 0.87$ as opposed to 0.76 of the second DTW alignment). Hence, it is possible to conclude that the alignment, which follows the scenario 2 introduced in Section SI1.2, does perform relatively well with these data, although the resulting growth rates caused by assignment of multiple measured SIMS $\delta^{18}O$ values to one day are likely overestimates. Another drawback
Figure S4. A) Third DTW alignment after linear interpolation of measured SIMS δ¹⁸O values. See Figure S2A and text for detailed explanation. B) Resulting regression relating predicted δ¹⁸O values as a function of measured SIMS δ¹⁸O values. See Figure S3B for detailed explanation. C) Resulting sub-annual growth model. The plot is the same as Figure S3C, but the beginning and end of the winter band are now added as dashed vertical lines. Grey shading represents the estimated minimum and maximum date for each line.

Figure S5. Linear interpolation for centroid, maximum and minimum distance point of each SIMS sample spot.
with this DTW model is that it does not assign enough uncertainty to assigned values as by eye. One can see that some measured $\delta^{18}$O values could be easily moved by several months without changing the fit dramatically (the one assigned to late February in Figure S4A, for instance).

**Figure S6.** A) Permuted DTW alignment using linear interpolation and symmetric 2 step pattern. Red dots indicate the assigned date for centroids of SIMS sample spots. Horizontal error bars represent the minimum and maximum date encountered during 500 DTW runs using randomly sampled distance values within the minimum and maximum extent of SIMS sample spots. Blue dots with error bars indicate the alignment of added deployment and recovery predicted $\delta^{18}$O values. B) Resulting regression fit for the centroids. Identical to non-permuted DTW (Figure S4B). C) Subannual growth model for 500 permutation runs. Each black line is transparent and indicates a separate DTW run. Dashed vertical lines are also transparent and indicate the estimated average date for the beginning and the end of the winter growth band for each DTW run.

**SI1.6 Estimating the uncertainty for aligned measured SIMS $\delta^{18}$O values**

The third DTW modeling approach has been shown to result in feasible subannual growth model. Consequently, it is possible to proceed to estimation of the uncertainty of aligned query index values. Even though the SIMS sampling spot diameter was only 20.0 $\mu$m on average, sampled shell material was likely significantly time-averaged wherever shell growth rate was low (Figure S5, [15,16]). The maximum and minimum extent of each SIMS sample spot was used to randomly
sample distance values between these limits for each measured SIMS δ\textsuperscript{18}O value. The resulting
permutation not only changes the distance of each measured SIMS δ\textsuperscript{18}O value, but might also
change the order, if maximum and minimum limits of adjacent samples overlapped (Figure S5).
A new DTW alignment model was calculated by repeating the permutation a sufficient number
of times (n = 500). The DTW model demonstrated an increased uncertainty for measured δ\textsuperscript{18}O
value alignment (Figure S6). The vertical error bars covered almost the entire extent where a
measured value could be placed by eye.

The final DTW alignment estimated a mean offset between predicted and measured δ\textsuperscript{18}O
values as −5.56 ‰, and provided a significant regression between the predicted and measured
δ\textsuperscript{18}O values with an $R^2$ of 0.87 (Figure S6B). The offset of −0.05 ‰ in standardized values
demonstrated a relatively good average fit. Estimated regression parameters ($\beta_0$ and $\beta_1$) were
used to calculate the signal error term (= residuals, $\epsilon$) for each measured value. The error term
ranged between −0.50 and 0.62 ‰ with two values < −0.4 ‰, four values between 0.4 and 0.6 ‰
and two values > 0.6 ‰. Ten measured standardized δ\textsuperscript{18}O values were 10–20 % lower, five values
10–20 % higher and two values 20–30 % higher compared to predicted values. Deposition of the
winter growth band was estimated to begin sometime between January and March and end in
July. The general subannual growth pattern was almost identical to the third DTW alignment
(Figure S4) with most of the annual growth occurring from July until recovery in mid-September.

References


2. Giorgino T (2009) Computing and visualizing dynamic time warping alignments in R :


1–29.


Text S2: Linear mixed model formulations used to examine the relationships between shell growth rate and mooring instrument recordings

Linear mixed models were calculated using lme4 package [1] for R [2].

Model 1

The first model was used to examine the variance explained by each response variable separately. The model was formulated using R notation:

$$lmer(log(GR) \sim X + (1|Smp) + (1|W), \ data = D)$$  \hspace{1cm} (S1)

Where D is the data frame. GR represents a column of D containing information of weekly growth rate, X weekly averaged temperature, fluorescence index or salinity, Smp sample names of individual bivalve shell sample names as factor, W a vector of weeks from the deployment as numeric. S and W represent random effects. The notation $(1|S)$ specifies that random intercepts and constant slopes should be used for a random effect [1, 3]. The marginal and conditional $R^2$ were calculated from the lmer() output (object called model1):

$$r.squaredGLMM(model1)$$  \hspace{1cm} (S2)

Using MuMIn package [4] and the method described in Johnson [5].

Model 2

The second model was used to examine the relative effect of each fixed effect (temperature, fluorescence index, and salinity) on logarithm of growth rate. Fixed effects were first scaled to centered to their means and scaled to their standard deviations:

$$scale(D[, c(“T”, “F”, “S”))]$$  \hspace{1cm} (S3)

Where D is the data frame, and T, F and S the columns containing temperature, fluorescence index and salinity information respectively. The model was calculated using the scaled dataset (Ds):

$$lmer(log(GR) \ T + F + S + (1|Smp) + (1|W), \ data = Ds)$$  \hspace{1cm} (S4)

The significance of T, F and S was estimated by Satterthwaite approximation for denominator degrees of freedom [6] using lmerTest package [7]:

$$summary(model2)$$  \hspace{1cm} (S5)

Where model2 is the output from S4.
References


   r-project.org/package=MuMIn.

5. Johnson PC (2014) Extension of Nakagawa & Schielzeth’s $R^2_{GLMM}$ to random slopes


   for random and fixed effects for linear mixed effect models (lmer objects of lme4 package).