Vol. 555: 29–47, 2016
doi: 10.3354/meps11820

MARINE ECOLOGY PROGRESS SERIES
Mar Ecol Prog Ser

Published August 18

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Species-specific calcite production reveals *Coccolithus pelagicus* as the key calcifier in the Arctic Ocean

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ABSTRACT: Through the production and export of their calcite coccoliths, coccolithophores form a key component of the global carbon cycle. Despite this key role, very little is known about the biogeochemical role of different coccolithophore species in terms of calcite production, and how these species will respond to future climate change and ocean acidification. Here, we present the first study to estimate species-specific calcite production, from samples collected in the Arctic Ocean and subarctic Iceland Basin in June 2012. We show that although the coccolithophorid *Coccolithus pelagicus* comprised only a small fraction of the total community in terms of abundance (2%), our estimates indicate that it was the major calcite producer in the Arctic Ocean and Iceland Basin (57% of total calcite production). In contrast, *Emiliania huxleyi* formed 27% of the total abundance and was responsible for only 20% of the calcite production. That *C. pelagicus* was able to dominate calcite production was due to its relatively high cellular calcite content compared with the other species present. Our results demonstrate, for the first time, the importance of investigating the complete coccolithophore community when considering pelagic calcite production, as relatively rare but heavily calcified species such as *C. pelagicus* can be the key calcite producers in mixed communities. Therefore, the response of *C. pelagicus* to ocean acidification and climate change has the potential to have a major impact on carbon cycling within the North Atlantic and Arctic Ocean.

KEY WORDS: Coccolithophores · Calcification · Arctic Ocean

INTRODUCTION

Coccolithophores are a major group of phytoplankton, comprising up to 10% of primary production (Poulton et al. 2007), dominating pelagic calcite production and export with their calcite coccoliths (Broecker & Clark 2009), and thus forming a key component of the global carbon cycle (de Vargas et al. 2007, Ziveri et al. 2007). Marine calcifiers, including coccolithophores, face an uncertain future, as they have to contend with the effects of global warming and ocean acidification (The Royal Society 2005, Winter et al. 2014). Culture experiments considering the response of coccolithophores to ocean acidification have produced conflicting results (Iglesias-Rodriguez et al. 2008, Langer et al. 2009, Hoppe et al.

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2011), with long term studies suggesting adaptive evolution could partly compensate for the effects of global warming and ocean acidification (Lohbeck et al. 2012, Schluter et al. 2014). Furthermore, a more mechanistic understanding of coccolithophore responses to variable pH indicates that different species respond differently (Langer et al. 2009) and have different growth-optimum conditions in terms of pH (Bach et al. 2015).

Many of the previous studies on coccolithophores, along with the majority of the current literature, have considered only a single species of coccolithophore: *Emiliania huxleyi*. Although *E. huxleyi* is considered the keystone coccolithophore species due to its global dominance and ability to form large-scale, highly visible blooms (Paasche 2002), there are ~200 extant species of coccolithophore that vary in cell size (2 to 20 µm), and cellular calcite quota (Young et al. 2003). In this context, *E. huxleyi* has a small cell (~5 µm) with a relatively low cellular calcite content (0.2 to 1.1 µmol C cell^{-1}; Paasche 2002, Daniels et al. 2014) and hence relatively low calcification rates; other larger and more heavily calcified species such as *Coccolithus pelagicus*, with ~30 times more calcite per cell than *E. huxleyi* (Daniels et al. 2014), have the potential to be key species in terms of upper ocean calcite production and export (Ziveri et al. 2000, Baumann et al. 2004, Daniels et al. 2014).

The response of coccolithophores to ocean acidification in culture experiments appears to differ between species and strains (Langer et al. 2006, Langer et al. 2009), and culture experiments do not necessarily reflect the response of natural populations to environmental fluctuations (Smith et al. 2012, Poulton et al. 2014, Marañón et al. 2016). Therefore, it is unlikely that *E. huxleyi*'s response to ocean acidification in culture can be applied to multi-species populations of coccolithophores (Bach et al. 2015). In natural communities, the response to variability in pH is often secondary to the effects of light, nutrient availability and growth rate (Zondervan 2007, Charalampopoulou et al. 2011, Poulton et al. 2014). To examine how a diverse coccolithophore community will respond to environmental changes, and to assess the relative biogeochemical importance of different coccolithophore species, field studies considering the whole coccolithophore community are required.

The effect of anthropogenic CO_2 emissions on the Arctic Ocean is expected to be among the largest and most rapid of any region on the globe (ACIA 2004), with the Arctic already experiencing rapid warming (ACIA 2004). Ocean acidification is also expected to be particularly enhanced at high latitudes because of the increased solubility of CO_2 at low temperatures. Within the Nordic Seas (Greenland Sea and Norwegian Sea) of the Arctic Ocean, large natural gradients of environmental variables such as temperature and carbonate chemistry already exist; in the west, the East Greenland Current transports cold (<0°C) Polar Water southwards through the Greenland Sea (see Fig. 1), while in the east, the Norwegian Current carries relatively warm (6 to 10°C) Atlantic water into the Norwegian Sea (Johannessen 1986). Coccolithophores are a key phytoplankton group within these Nordic Seas (Samtleben & Schröder 1992). The highest species diversities are found in the Norwegian Sea (Samtleben & Schröder 1992, Baumann et al. 2000), as the more diverse North Atlantic communities are transported northwards by the Norwegian Current. The Norwegian Sea coccolithophore community is generally numerically dominated by *E. huxleyi* (Samtleben & Schröder 1992, Baumann et al. 2000, Charalampopoulou et al. 2011), with some species such as *Calciopappus caudatus* present throughout, while other species such as *Syracosphaera* spp. are limited to Atlantic surface waters. In contrast, coccolithophore diversity is lower in the Greenland Sea (Samtleben & Schröder 1992); *C. pelagicus* is commonly observed along with other polar species (e.g. *Papposphaera* spp.). The contrast in coccolithophore community structure and diversity, coupled with the strong natural environmental gradients of the Greenland and Norwegian Seas, means that this region is an ideal location to examine the influence of both the environment and the coccolithophore community structure on calcite production.

The aim of this study was to determine whether *E. huxleyi* is the major calcite producer in the Arctic Ocean, and if not, which coccolithophore species are. As only total community calcite production (CP) can be measured from mixed communities (e.g. Charalampopoulou et al. 2011, Poulton et al. 2014), a novel method was developed to determine species-specific calcite production (CP_{sp}) for each individual coccolithophore species. This method incorporates species-specific cellular calcite, growth rates and abundances to partition CP. This is the first study to determine the calcite production rates of individual coccolithophore species within a natural multi-species community. Here, we present results from 19 stations within the Arctic Ocean and the subarctic Iceland Basin (see Fig. 1); CP, coccolithophore cellular abundances, carbonate chemistry parameters and other environmental variables were measured, and CP_{sp} was derived for each station.

MATERIALS AND METHODS

Sampling

Sampling was carried out in the subarctic Iceland Basin, and in the Greenland and Norwegian Seas within the Arctic Ocean (Fig. 1) between 4 and 30 June 2012 during the UK Ocean Acidification Arctic Cruise, aboard the RRS 'James Clark Ross' (JR271). Water samples for rate measurements, coccolithophore community structure and ancillary measurements were collected from a single depth within the middle of the mixed layer at 19 CTD stations. Temperature and salinity were obtained from the CTD. Incidental photosynthetically active radiation (PAR), measured with ship-mounted scalar irradiance sensors (Kipp & Zonen ParLite 0348900, Skye Instruments SK3), was integrated over the incubation periods to calculate daily incidental irradiance (mol photons m^{-2} d^{-1}). The vertical diffuse attenuation coefficient of PAR (k_d) in the water column was calculated from the CTD casts, with the depth of the euphotic zone (z_{eup}) calculated as the depth of 1% incident irradiance.

Calcite production

Daily rates of calcite production were measured using the micro-diffusion technique (Paasche & Brubak 1994, Balch et al. 2000) following Poulton et al. (2014). Unfiltered water samples (70 ml, 3 light, 1 formalin-killed), collected from one depth within the middle of the mixed layer, were inoculated with 25 to 50 µCi ^{14}C-labelled sodium bicarbonate. Samples were incubated for 24 h in an on-deck incubator, chilled with surface seawater and the 55% incidental irradiance light depth was replicated using Misty-blue optical filters (LEE™). When the surface seawater supply was unavailable (at ice stations), samples were incubated in a constant temperature container laboratory (see Richier et al. 2014) with the temperature and photoperiod set to replicate the *in situ* environment. Formalin-killed blanks were prepared by addition of 1 ml of 0.2 µm triple-filtered and sodium-borate buffered formalin solution.

Incubations were terminated by filtration through 25 mm 0.45 µm polycarbonate filters (Nuclepore™). Filters were secured in glass scintillation vials with a gas-tight septum and a bucket containing a CO_2 trap

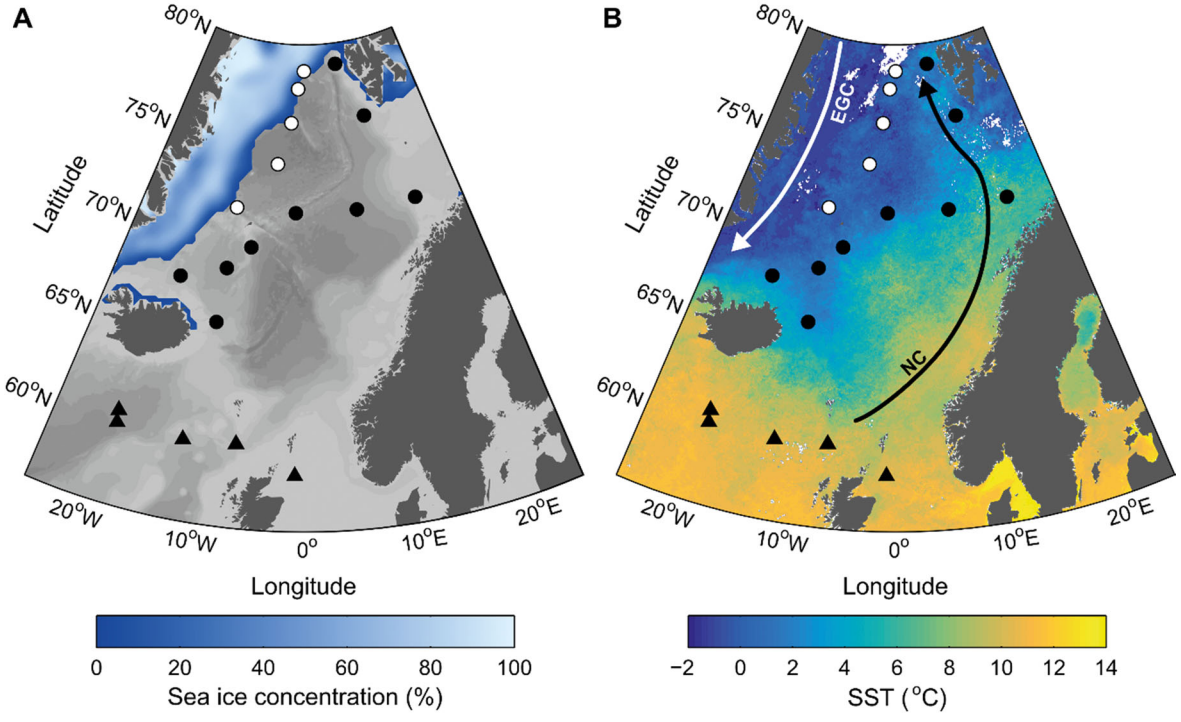

Fig. 1. Sampling locations in the Iceland Basin (triangles), Norwegian Sea (black circles) and Greenland Sea (white circles) showing (A) sea ice concentration in June 2012, taken from www.nsidc.org and (B) MODIS sea surface temperature for June 2012, overlaid with the East Greenland Current (EGC) and Norwegian Current (NC)

(Whatman GFA filter soaked with 200 µl β-phenylethylamine), acidified with a dilute acid (1 ml, 1% phosphoric acid), thus releasing the acid-labile inorganically fixed carbon (i.e. CP) as $^{14}CO_2$ to be absorbed by the CO_2 trap. After 24 h, the GFA filters were removed to separate scintillation vials, and the activity of the filters was determined in Ultima Gold (Perkin-Elmer) and their activity measured using a Tri-Carb 2100 low level liquid scintillation counter. Spike activity was checked following Poulton et al. (2014). The activity in the formalin-killed blanks were subtracted from the triplicate light measurements.

The average coefficient of variation of the triplicate (light) CP measurements was 27% (3 to 113%), and the formalin-killed blank represented on average 26% (7 to 60%) of the CP signal, with generally higher contributions in lower CP signals. These results are comparable to other studies using the same method (e.g. Poulton et al. 2010, 2014).

Coccolithophore community structure

Water samples (100 to 250 ml) for the determination and enumeration of the coccolithophore community were collected following Poulton et al. (2014). Permanent slides were prepared on board using a low viscosity Norland Optical Adhesive (NOA 74) (Poulton et al. 2014). Coccolithophore cell counts and species identification were performed using a Leitz Ortholux polarizing microscope (1000×, oil immersion). A minimum of 54 fields of view were counted per filter for abundant species, with additional fields of view analysed for rarer species. The light microscopy species identification and enumeration were verified and supplemented using scanning electron microscopy (SEM) following Daniels et al. (2012).

Species-specific calcite production

The equation to determine CP_{sp} was adapted from Daniels et al. (2014). CP_{sp} for a given species was calculated as a product of the growth rate (µ), cellular calcite content (C_{sp}) and abundance (N_{sp}) of that species:

$$CP_{sp} = \mu C_{sp} N_{sp} \quad (1)$$

CP_{sp} was estimated from SEM images by combining derived estimates of coccolith calcite (Young & Ziveri 2000) with the number of coccoliths per cell (Table 1). The method of Young & Ziveri (2000) incorporates species-specific coccolith shape factors (k_s). Of the species observed here, only 4 (*E. huxleyi*, *Coccolithus pelagicus*, *Acanthoica quattrospina*, *Syracosphaera* spp.) had a pre-defined k_s. For those species with an undefined k_s, this was estimated from SEM images for the holococcolithophorid (HOL) life stage of *C. pelagicus* and *Calciopappus caudatus* (Table 1), the k_s for *Algirosphaera robusta* was adapted from *E. huxleyi* (Probert et al. 2007), and a 'typical coccolith' k_s was used for *Ophiaster* sp. (Young & Ziveri 2000).

Species-specific growth rates cannot be directly determined from the measurements. However, the growth rate of the bulk community can be calculated by dividing the measured calcite production rate (CP_{bulk}) by the total calcite content of the cells (Poulton et al. 2010, Balch et al. 2014), assuming steady state in terms of cellular quota (Daniels et al. 2014), as shown in Eq. (2):

$$\mu = \frac{CP_{bulk}}{\sum_{i=1}^{n} C_i N_i} \quad (2)$$

This growth rate can then be applied to Eq. (1) to calculate CP_{sp}. This method makes the simplifying assumption that all coccolithophores in the mixed community have the same growth rate. The choice of this method was driven by the lack of data on relative growth rates of coccolithophores in the field or from laboratory experiments (Daniels et al. 2014, 2015). However, this does not account for the fact that growth rates of individual phytoplankton species can vary significantly within the same population (Weiler & Chisholm 1976). To examine whether our results were sensitive to this potential variability in growth rates we performed a sensitivity analysis.

Table 1. Coccolith shape factors, coccolith calcite, number of coccoliths per cell and cellular calcite for the individual coccolithophore species

Species	Coccolith shape factor (k_s)	Coccolith calcite (pmol)	Coccoliths cell^{-1}	Cellular calcite (pmol)
Emiliania huxleyi	0.020	0.024	22	0.52
Coccolithus pelagicus	0.060	1.218	13	15.2
Syracosphaera spp.	0.015	0.012	35	0.40
Acanthoica quattrospina	0.030	0.008	36	0.27
Calciopappus caudatus	0.013	0.002	54	0.09
Ophiaster sp.	0.035	0.001	70	0.09
Algirosphaera robusta	0.045	0.010	43	0.42
Coccolithus pelagicus HOL	0.036	0.008	100	0.78

The growth rates of individual coccolithophore species were manipulated relative to the rest of the community such that the relative growth rates were between 10 and 200% that of the other coccolithophore species. This range encapsulates the variability in maximum growth rate observed by Buitenhuis et al. (2008) and Marañón et al. (2013) for coccolithophores. These growth rates were then applied to the whole dataset using Eq. (3) to model the impact on the CP_{sp} of individual species:

$$CP_{sp} = \frac{\mu_{sp} C_{sp} N_{sp}}{\sum_{i=1}^{n} \mu_i C_i N_i} \times CP_{bulk} \quad (3)$$

This approach is similar to that used in Daniels et al. (2014).

Macronutrients and carbonate chemistry

Macronutrients (nitrate + nitrite, NO_x; phosphate, PO_4; silicic acid, dSi) were determined following Sanders et al. (2007) on a Skalar autoanalyser. The relative concentration of NO_x to PO_4 ($N^\star$; $NO_x - 16 \times PO_4$; Moore et al. 2009) and the relative concentration of dSi to NO_x ($Si^\star$; $dSi - NO_x$; Bibby & Moore 2011, Poulton et al. 2016) were also determined.

Samples for total dissolved inorganic carbon (C_T) and total alkalinity (A_T) were collected into 250 ml borosilicate glass bottles and poisoned with 50 µl of saturated mercuric chloride solution following (Dickson et al. 2007). Using a VINDTA 3C instrument (Marianda), C_T was measured by coulometric titration, and A_T by potentiometric titration and calculated using a modified Gran technique (Bradshaw et al. 1981). The results were calibrated using certified reference material (batch 117) obtained from A. G. Dickson (Scripps Institution of Oceanography). Measurement precision was ±3.8 and ±1.7 µmol kg^{-1} for C_T and A_T respectively. Calcite saturation state (Ω_c), pH on the total scale (pH_T) and seawater partial pressure of CO_2 (pCO_2^{sw}) were calculated using version 1.1 of the CO_2SYS program for MATLAB (Van Heuven et al. 2011) using the carbonic acid dissociation constants of Lueker et al. (2000), the boric acid dissociation constant of Dickson (1990b), the bisulfate ion acidity constant of Dickson (1990a), and the boron:chlorinity of Lee et al. (2010).

Data availability and statistical analysis

All data included in this study are available from the British Oceanographic Data Centre (BODC). Multivariate statistics were used to examine spatial variability in the coccolithophore species composition and CP_{sp} (biotic data), and the environment (abiotic data). Bray-Curtis similarity resemblance matrices were calculated from the standardised biotic data to determine changes in species composition and CP_{sp}. The abiotic data (temperature, salinity, Ω_C, pH_T, $N^\star$, $Si^\star$, daily PAR and z_{eup}) were normalised, and a Euclidean distance resemblance matrix calculated to determine changes in the environmental variables. The species composition of samples via the Bray-Curtis similarity index was then used to cluster samples into groups using non-metric multi-dimensional scaling (NMDS). The species typical of each hydrographic region were identified using a breakdown of similarity percentages (SIMPER routine), calculated in PRIMER-E (Clarke 1993). Spearman's rank correlation (BEST routine) were calculated in PRIMER-E (Clarke 1993) to identify which environmental variables explained most of the variation in the coccolithophore community and CP_{sp}.

Principal component analysis (PCA) of normalised environmental variables was performed using MATLAB, and Pearson product-moment correlations were carried out between the calculated principal components (PC) and coccolithophore community composition and CP_{sp} to further examine the relationship between the biotic and abiotic data.

RESULTS

General oceanography

A wide variety of hydrographic environments were sampled during the cruise throughout the Iceland Basin and the Nordic Seas (Greenland Sea and Norwegian Sea) of the Arctic Ocean (Fig. 1, Table 2), with 2 major fronts dividing the regions: the Norwegian Sea is separated from the Iceland Basin by the Iceland-Faroes Front, while the East Greenland Front separates the Greenland Sea from the Norwegian Sea (Cottier et al. 2014). The Iceland Basin was characterised by the warmest (10 to 10.6°C) and most saline (35.2 to 35.3) waters of the study. The Greenland Sea, with the influence of the East Greenland Current, had the coldest (1 to 3.5°C) and freshest (34.7 to 35.0) waters sampled. The Norwegian Sea lay between the 2 extremes of the Iceland Basin and the Greenland Sea, in terms of both temperature (3.1 to 7.8°C) and salinity (34.8 to 35.2).

Macronutrient concentrations of NO_x (0.5 to 10.6 mmol N m^{-3}), PO_4 (0.11 to 0.77 mmol P m^{-3}) and

Table 2. Physicochemical features of the Iceland Basin (ICB), Norwegian Sea (NWS) and Greenland Sea (GS). PAR: photosynthetically active radiation; z_{eup}: euphotic zone depth; pCO_2: partial pressure of CO_2; pH_T: pH on the total scale; Ω_C: calcite saturation state; NO_x: nitrate + nitrite; PO_4: phosphate; dSi: silicic acid; N*: excess NO_x relative to PO_4; Si*: excess dSi relative to NO_x

CTD	Location	Coordinates	Date (2012)	Depth (m)	Temperature (°C)	Salinity	Daily PAR (mol photons m^{-2} d^{-1})	z_{eup} (m)	Carbonate chemistry			Surface macronutrients (mmol m^{-3})				
									pCO_2 (µatm)	pH_T	Ω_C	NO_x	PO_4	dSi	N*	Si*
6	ICB	58.74°N; 0.86°W	04 Jun	9	10.0	35.3	45	40	277	8.2	4.2	0.5	0.11	1.7	−1.3	−1.2
8	ICB	60.13°N; 6.71°W	05 Jun	10	10.4	35.4	33	48	326	8.1	3.8	6.5	0.45	4.3	−0.7	2.3
10	ICB	59.97°N; 11.98°W	06 Jun	20	10.6	35.3	51	28	310	8.1	4.0	2.9	0.21	1.4	−0.4	1.5
12	ICB	60.00°N; 18.67°W	07 Jun	10	10.2	35.2	41	37	340	8.1	3.7	6.1	0.4	1.7	−0.3	4.4
17	ICB	60.59°N; 18.86°W	08 Jun	20	10.4	35.2	10	40	310	8.1	3.9	5.2	0.35	1.3	−0.4	3.9
19	NWS	65.98°N; 10.72°W	10 Jun	24	3.6	34.8	34	23	240	8.2	3.7	0.6	0.22	2.5	−3.0	−1.9
20	NWS	69.90°N; 7.58°W	11 Jun	15	3.1	35.0	53	36	363	8.1	2.7	9.1	0.64	6.1	−1.2	3.0
21	GS	74.12°N; 4.69°W	12 Jun	15	1.0	34.9	40	48	308	8.1	2.8	9.8	0.7	5.7	−1.4	4.0
27	GS	76.18°N; 2.55°W	13 Jun	20	1.5	34.9	42	50	319	8.1	2.7	9.3	0.67	4.7	−1.4	4.6
29	GS	78.72°N; 0.00°	14 Jun	10	3.5	35.0	51	15	209	8.3	4.1	2.6	0.31	5.5	−2.4	−2.9
40	GS	78.25°N; 5.55°W	19 Jun	15	3.1	34.9	20	25	309	8.1	3.0	8.7	0.62	5.6	−1.2	3.1
42	NWS	78.22°N; 6.00°W	20 Jun	15	6.0	35.1	28	22	208	8.3	4.5	4.0	0.38	4.3	−2.1	−0.4
45	NWS	77.82°N; 4.97°W	22 Jun	20	5.7	35.2	19	41	309	8.1	3.3	9.8	0.72	5.8	−1.8	4.0
54	NWS	77.85°N; 1.29°W	25 Jun	13	7.8	35.0	24	41	320	8.1	3.5	6.0	0.49	3.8	−1.8	2.2
56	NWS	78.99°N; 7.98°E	26 Jun	15	6.7	35.2	33	31	305	8.1	3.5	6.8	0.5	5.2	−1.2	1.6
58	NWS	76.26°N; 12.54°E	27 Jun	20	5.4	35.1	35	38	316	8.1	3.2	10.6	0.77	5.7	−1.7	4.9
60	GS	76.16°N; 23.07°E	28 Jun	26	1.4	34.7	49	45	328	8.1	2.6	8.6	0.64	2.2	−1.6	6.5
63	NWS	72.89°N; 26.00°E	29 Jun	20	3.8	34.8	40	32	318	8.1	3.0	8.9	0.65	2.6	−1.5	6.3
65	NWS	71.75°N; 17.90°E	30 Jun	20	5.1	34.9	33	48	246	8.2	3.8	4.0	0.43	4.1	−2.8	0.0

dSi (1.3 to 6.1 mmol Si m^{-3}) were highly variable and no clear spatial patterns were observed (Table 2). The values of N* were negative at all sites indicating that, assuming Redfield stoichiometry (Redfield 1958), NO_x was low relative to PO_4. The values of Si* ranged from −2.9 to 6.5. While generally positive, indicating high residual dSi concentrations, 4 stations exhibited a negative Si*, indicating depleted dSi relative to NO_x. No clear spatial patterns in N* or Si* were identified between sampling sites.

The z_{eup} ranged from 15 to 50 m, and daily incidental PAR varied from 10 to 53 mol photons m^{-2} d^{-1}, with both showing variability within and between regions (Table 2). As the cruise occurred in mid-summer, the stations in the Nordic Seas experienced a 24 h photoperiod, while the Iceland Basin stations experienced a shorter photoperiod (~18 h). The effect of this on daily PAR is not clear (Table 2), suggesting a stronger influence through varying cloud cover. Values of pH_T varied from 8.07 to 8.29 and Ω_C varied from 2.65 to 4.46, with the low Ω_C particularly in the Greenland Sea (Table 2).

Coccolithophore community structure

Total coccolithophore abundance was highly variable, ranging from 5 to 932 cells ml^{-1}. The most commonly observed coccolithophore species were *Emiliania huxleyi* (0 to 425 cells ml^{-1}), *Coccolithus pelagicus* (0 to 33 cells ml^{-1}) and *C. pelagicus* HOL (0 to 223 cells ml^{-1}) (Fig. 2, Table 3). Other species present included *Acanthoica quattrospina*, *Algirosphaera robusta*, *Calciopappus caudatus*, *Ophiaster* sp. and *Syracosphaera* spp. (Fig. 2). While each species was considered individually in determining CP_{sp} and in the environmental analysis, for the purpose of graphical representation, species other than *E. huxleyi*, *C. pelagicus* and *C. pelagicus* HOL were grouped into one category (termed 'others'; see Fig. 5) as they were minor contributors to regional calcite production. SEM identified *Syracosphaera* spp. as including: *S. borealis*, *S. corolla*, *S. dilata*, *S. marginaporata* and *S. molischii*. The cellular calcite contents of the *Syracosphaera* genus, however, are not

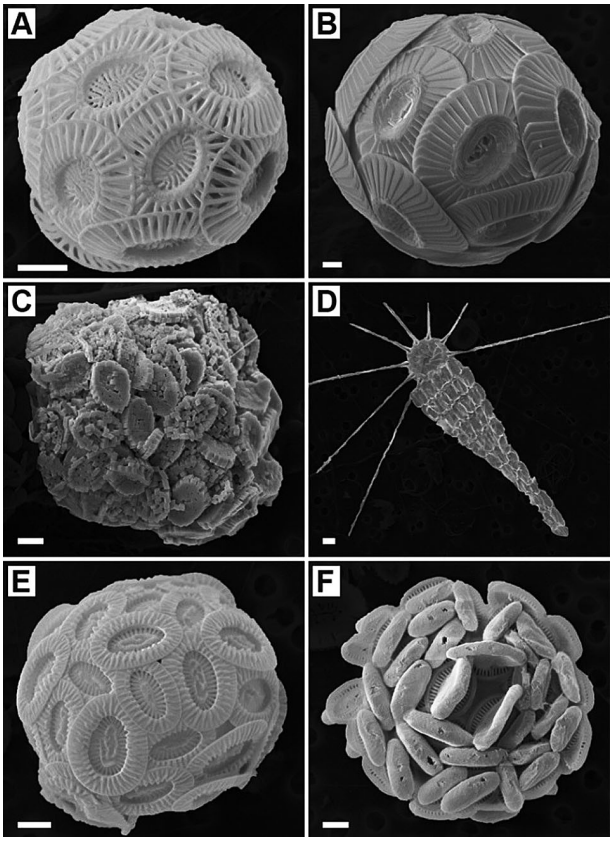

Fig. 2. Scanning electron microscopy (SEM) images of (A) *Emiliania huxleyi*, (B) *Coccolithus pelagicus*, (C) *C. pelagicus* HOL, (D) *Calciopappus caudatus*, (E) *Syracosphaera molischii*, and (F) *Algirosphaera robusta*. Scale bars: 1 µm

well constrained (Young & Ziveri 2000), thus we did not consider these species individually and used a 'small *Syracosphaera*' coccolith calcite (Young & Ziveri 2000) estimate for calculating their cellular calcite. The different coccolithophore species had varying spatial distributions (Table 3, Fig. A1 in the Appendix). *E. huxleyi* was most abundant in the Iceland Basin and Norwegian Sea, *C. pelagicus* HOL was present in the highest latitude stations, while *Syracosphaera* spp. were restricted to the Iceland Basin.

To account for the large variability in coccolithophore abundances between stations, the stations were grouped into the 3 distinct regions (Iceland Basin, Greenland Sea and Norwegian Sea; Fig. 1, Table 2), defined from the characteristic hydrography of each station. Coccolithophore abundances, aggregated over these regions and over the entire study area (Fig. 3A), showed that *E. huxleyi* represented 27% of the total coccolithophore abundance, with a relatively consistent contribution across all regions (19 to 30%; Fig. 3A). In contrast, *C. pelagicus* formed only a small component of the coccolithophore community in terms of abundance (1 to 4%; Fig. 3A) in all regions sampled. The Iceland Basin community was dominated by *C. caudatus* (43%) and *Syracosphaera* spp. (24%); the Norwegian Sea by *C. caudatus* (43%); and the Greenland Sea by *C. pelagicus* HOL (77%, Fig. 3A).

Table 3. Coccolithophore abundances (cells ml^{-1}) in the Iceland Basin (ICB), Norwegian Sea (NWS) and Greenland Sea (GS). (–) species absence

CTD	Location	*Emiliania huxleyi*	*Coccolithus pelagicus*	*C. pelagicus* HOL	*Syracosphaera* spp.	*Acanthoica quattrospina*	*Calciopappus caudatus*	*Ophiaster* sp.	*Algirosphaera robusta*
6	ICB	31.7	–	–	–	1.5	–	–	–
8	ICB	21.2	2.6	–	24.2	–	3.0	1.5	3.0
10	ICB	64.1	2.3	3.0	7.9	2.4	0.6	2.4	–
12	ICB	76.2	7.7	–	179.6	10.9	348.3	27.2	–
17	ICB	91.2	4.2	5.4	84.4	12.2	179.6	50.3	–
19	NWS	1.9	2.8	–	–	–	–	–	–
20	NWS	–	0.6	59.9	–	–	359.2	–	5.4
21	GS	–	0.4	3.8	–	–	–	–	–
27	GS	–	–	6.0	–	–	–	–	–
29	GS	17.0	0.4	0.9	–	–	–	–	0.9
40	GS	1.9	–	11.3	–	–	–	–	–
42	NWS	25.2	–	–	–	–	–	–	–
45	NWS	69.5	0.1	1.5	–	–	1.5	–	4.5
54	NWS	19.7	–	–	–	–	–	–	4.5
56	NWS	424.5	7.1	223.1	–	–	157.8	–	119.7
58	NWS	33.1	15.4	2.2	–	–	72.8	–	47.4
60	GS	–	2.8	54.8	–	–	–	–	–
63	NWS	20.8	32.7	–	–	–	274.0	–	–
65	NWS	2.8	2.9	–	–	–	–	–	–

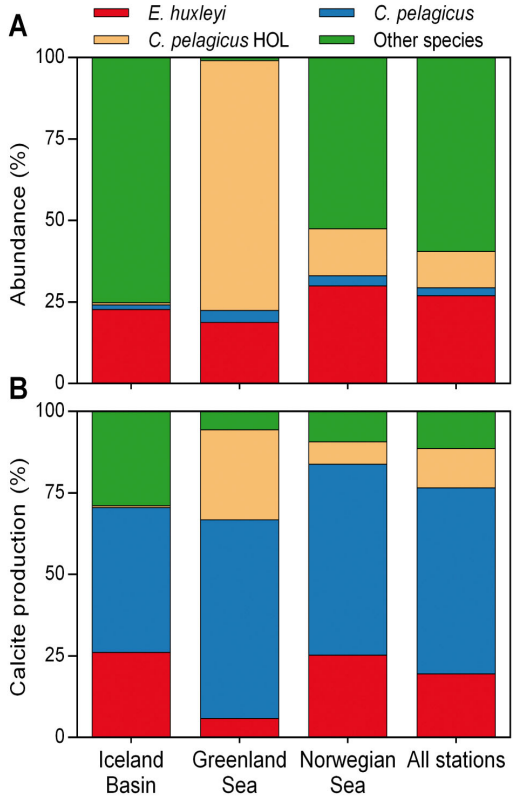

Fig. 3. Percentage contribution of coccolithophore species to (A) abundance and (B) calcite production, aggregated over each hydrographic region and the entire study area

Species-specific calcite production

Total community calcite production was highly variable throughout the study (from 2 to 202 µmol C m^{-3} d^{-1}), with no clear spatial patterns in the distribution of calcite production. The largest calcite production (202 µmol C m^{-3} d^{-1}) was measured in the central Norwegian Sea (Fig. 4), with the lowest rates in the Greenland Sea (<10 µmol C m^{-3} d^{-1}). Bulk coccolithophore community growth rates had a geometric mean of 0.33 d^{-1} (0.1 – 3.0 d^{-1}; Table 4). Two stations showed growth rates >1 d^{-1} (1.4 and 3.0 d^{-1}), which are unrealistic (Marañón et al. 2013) and are most likely due to underestimates in the calcite content of the coccolithophore species present.

At each individual station, the major calcite producers were *E. huxleyi* (0 to 100%), *C. pelagicus* (0 to 98%) and *C. pelagicus* HOL (0 to 100%). However, there was significant variability between the stations (Table 4, Fig. 5), and when considering each station individually, *E. huxleyi* was the largest contributor at 6 stations, *C. pelagicus* at 10 stations and *C. pelagicus* HOL at 3 stations. Of the other species present, *Syracosphaera* spp. were also a significant source in the Iceland Basin (0 to 27%), and *C. caudatus* was generally a small source (0 to 12%) except at Stn 20 in the Norwegian Sea where it contributed 37% of the total calcite production. When present, *A. robusta* was a minor contributor to calcite production in the Norwegian Sea (3 to 16%).

Considering the percentage calcite production of each species on a per station basis, however, does not account for the high variability in the measured total calcite production. Incorporating total calcite production and aggregating over the 3 regions and the entire cruise reveals that *C. pelagicus* was the major calcifier, responsible for 57% of total calcite production (Fig. 3B), with a higher contribution in the Nordic Seas (59 to 61%) than in the Iceland Basin (44%). In contrast, *E. huxleyi* represented only 20% of total calcite production (Fig. 3B), with a much smaller contribution in the Greenland Sea (6%) than in the Norwegian Sea (26%) and Iceland Basin (25%). *C. pelagicus* HOL was a significant calcite producer in the Greenland Sea (28%), but less so in the other regions, resulting in a total contribution of only 12% (Fig. 3B). The contribution of the other species to calcite production was greatest in the Iceland Basin (29%), of which *Syracosphaera* spp. (19%) and *C. caudatus* (7%) were the major calcifiers. In the Arctic, *C. caudatus* (2 to 5%) and *A. robusta* (0 to 7%) were the largest calcite producers of the other coccolithophore species present.

Sensitivity analysis

To examine the impact of growth rates on CP_{sp}, a sensitivity analysis was applied to the 3 main calcifiers (*E. huxleyi*, *C. pelagicus* and *C. pelagicus* HOL), with CP_{sp} aggregated over the entire region as used above (Fig. 6). Varying the growth rate of *E. huxleyi* (Fig. 6A) had little impact on the overall result, with *C. pelagicus* responsible for 53 to 63% of total calcite production. When the relative growth rate of *E. huxleyi* was <39% of the rest of the community, *C. pelagicus* HOL becomes the second largest calcite producer. Varying the growth rate of *C. pelagicus* (Fig. 6B) to <15% of the rest of the community produced the only scenario in which *C. pelagicus* was not the major calcifier (25 to 30%), with *E. huxleyi* then becoming the greater calcite producer (30 to 32%) by a small margin. The relative growth rate of *C. pelagicus* HOL did not affect the overall pattern, with *C. pelagicus* dominating calcite production throughout (56 to 59%).

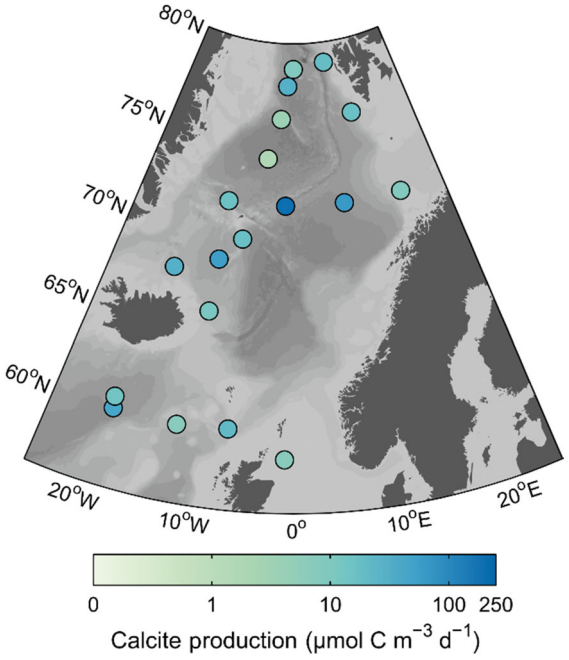

Fig. 4. Distribution of total calcite production throughout the study area in the Nordic Seas

As *E. huxleyi* and *C. pelagicus* were the 2 main contributors to calcite production in our dataset, a further sensitivity analysis was performed where the relative growth rates of these 2 species were varied concurrently (Fig. 7). In this model scenario, *C. pelagicus* is the dominant calcifier (>50% calcite production) in 74% of the model (Fig. 7B), while it remains the greatest single species contributor to calcite production in 96% of the model. If the growth rate of *E. huxleyi* is increased to 200% that of the rest of community, then *C. pelagicus* did not dominate calcite production, with a growth rate <30% of the total community growth rate.

Coccolithophore species composition, CP_{sp} and environmental variables

In order to explore the relationship between the environmental variables and the species composition of the coccolithophore community and their contribution to CP_{sp}, a PCA was carried out using normalised environmental variables (temperature, salinity, Ω_C, pH, N^*, Si^*, daily PAR and z_{eup}). The first principal component (PC-1) explained 40.1% of the variance between stations in terms of the environmental conditions, while the second principal component (PC-2) explained a further 33.3% of the variance. Therefore, the combination of PC-1 and PC-2 explained 73.4% of the total environmental variability.

Eigenvalues from the PCA (Table 5) indicate the relative weight of the environmental variables in influencing each of the PCs. Pearson moment correlations showed that PC-1 was strongly related to Ω_C, pH, Si^* and z_{eup} while PC-2 was related to temperature, salinity and N^* (Table 5). Correlated with latitude ($r = 0.68$, $p < 0.005$, $n = 19$), PC-2 essentially describes the north–south environmental gradient, with warmer, more saline and high N^* waters in the south. Correlations between PCs and both coccolithophore composition and CP_{sp} indicated significant correlations ($p < 0.005$) between PC-1 and the contribution of *E. huxleyi* and *C. pelagicus* HOL to species composition, and between PC-1 and the percentage contribution to CP_{sp} by *E. huxleyi* ($p < 0.005$) and *C. pelagicus* HOL ($p < 0.05$). PC-2 was significantly correlated ($p < 0.005$) with the composition and percentage contribution to CP_{sp} of *Syracosphaera* spp., *A. quattrospina* and *Ophiaster* sp. These species were found only in the Iceland Basin samples, further demonstrating the link between PC-2 and the north–south environmental gradient.

To visualise the multivariate patterns in similarity between the individual stations in terms of community composition, NMDS analysis was applied to both species composition data (Fig. 8A) and CP_{sp} (Fig. 8B). The stress values of the 2-dimensional NMDS plots were low (<0.08), indicating that they are a good representation of the high-dimensional patterns (Clarke 1993). The NMDS plots revealed different patterns of similarity between the stations whether species composition or CP_{sp} were considered. To examine the underlying factors driving the similarity between stations, individual species contributions to community composition (Fig. 8B–D) and CP_{sp} (Fig. 8F–H) were overlaid on to the NMDS plots. In terms of species composition, the spatial pattern was generally explained by the contributions of *E. huxleyi* (Fig. 8B) and *C. pelagicus* HOL (Fig. 8D) to community composition. The majority of Greenland Sea samples clustered distinctly away from other stations (Fig. 8A), with their coccolithophore communities comprised of a large contribution from *C. pelagicus* HOL and a small contribution from *E. huxleyi*.

The dissimilarities in species contribution to community composition between stations in the different hydrographic regions were tested statistically using a SIMPER analysis. The high dissimilarity between stations in the Greenland Sea and those in both the Iceland Basin (average dissimilarity = 85.6%) and the Norwegian Sea (average dissimilarity = 82.3%) was driven by *C. pelagicus* HOL (43 to 44% of dissimilarity) and *E. huxleyi* (26 to 27% of dissimilarity), as

Table 4. Total calcite production (µmol C m^{-3} d^{-1}) and species-specific calcite production (%) in the Iceland Basin (ICB), Norwegian Sea (NWS) and Greenland Sea (GS). (–) species absence

CTD	Location	Total calcite production (µmol C m^{-3} d^{-1})	Community growth rate µ (d^{-1})	% Calcite production						
				Emiliania huxleyi	*Coccolithus pelagicus*	*C. pelagicus* HOL	*Syracosphaera* spp.	*Acanthoica quattrospina*	*Calciopappus caudatus*	*Ophiaster* sp.	*Algirosphaera robusta*
6	ICB	7.25	0.4	97.6	–	–	–	2.4	–	–	–
8	ICB	21.65	0.3	17.7	64.1	–	15.5	–	0.5	0.2	2.0
10	ICB	7.06	0.1	44.4	47.1	3.1	4.2	0.9	0.1	0.3	–
12	ICB	42.51	0.2	14.9	43.9	–	27.0	1.1	12.3	0.9	–
17	ICB	13.56	0.1	27.2	37.0	2.4	19.3	1.9	9.7	2.5	–
19	NWS	11.31	0.3	2.3	97.7	–	–	–	–	–	–
20	NWS	17.45	0.2	–	9.8	50.8	–	–	36.8	–	2.5
21	GS	1.65	0.2	–	70.0	30.0	–	–	–	–	–
27	GS	3.54	0.8	–	–	100.0	–	–	–	–	–
29	GS	9.04	0.6	54.8	38.2	4.5	–	–	–	–	2.5
40	GS	29.64	3.0	10.0	–	90.0	–	–	–	–	–
42	NWS	18.96	1.4	100.0	–	–	–	–	–	–	–
45	NWS	16.69	0.4	88.6	3.5	2.9	–	–	0.3	–	4.7
54	NWS	8.61	0.7	84.2	–	–	–	–	–	–	15.8
56	NWS	63.93	0.1	38.9	19.0	30.5	–	–	2.6	–	8.9
58	NWS	201.55	0.7	6.2	83.6	0.6	–	–	2.4	–	7.1
60	GS	16.21	0.2	–	50.3	49.7	–	–	–	–	–
63	NWS	55.87	0.1	2.0	93.2	–	–	–	4.8	–	–
65	NWS	29.58	0.6	3.2	96.8	–	–	–	–	–	–

observed in the NMDS plots. The spatial patterns in the CP_{sp} NMDS plots contrasted that of species composition (Fig. 8E), being influenced by *E. huxleyi* (Fig. 8F), *C. pelagicus* (Fig. 8G) and *C. pelagicus* HOL (Fig. 8H). The Greenland Sea stations did not cluster separately in this case, as they did for analysis of their coccolithophore community composition; SIMPER analysis found that that the hydrographic regions were more similar in terms of CP_{sp} (average dissimilarity < 71%) than in terms of species composition.

To determine which environmental variables best explain the patterns in species composition and CP_{sp}, Spearman's rank correlations (r_S) were calculated between resemblance matrices of abiotic and biotic data (Clarke 1993; see Charalampopoulou et al. 2011). The variability in species composition between stations was best explained by temperature, Ω_C, and N^* (r_S = 0.55, $p < 0.01$; Table 6), while the single variable that explained most of the variability was Ω_C (r_S = 0.55, $p < 0.01$). The variability in CP_{sp} was best correlated with Ω_C (Table 6), though the relationship was slightly weaker (r_S = 0.37, $p < 0.01$) than for species composition.

DISCUSSION

A robust measure of species-specific calcite production?

As CP_{sp} cannot be directly determined, its calculation requires assumptions with associated potential errors. The 2 main sources of error are the estimates of both cellular calcite and growth rates. With the natural variability in coccolith size and shape, the error in determining cellular calcite is estimated to be ~30 to 50% (Young & Ziveri 2000, Daniels et al. 2012). We have minimised this error by using species-specific shape factors together with measurements of coccolith length in SEM images, and our estimates of cellular calcite for *Coccolithus pelagicus* (15.2 pmol C $cell^{-1}$) and *Emiliania huxleyi* (0.52 pmol C $cell^{-1}$) are comparable to literature values (16.6 pmol C $cell^{-1}$ and 0.22 to 1.1 pmol C $cell^{-1}$ respectively; see Paasche 2002, Daniels et al. 2014). That the majority of bulk community growth rates (89%) as estimated by Eq. (2) were <1 d^{-1} (similar to that observed by Balch et al. 2014) suggests that

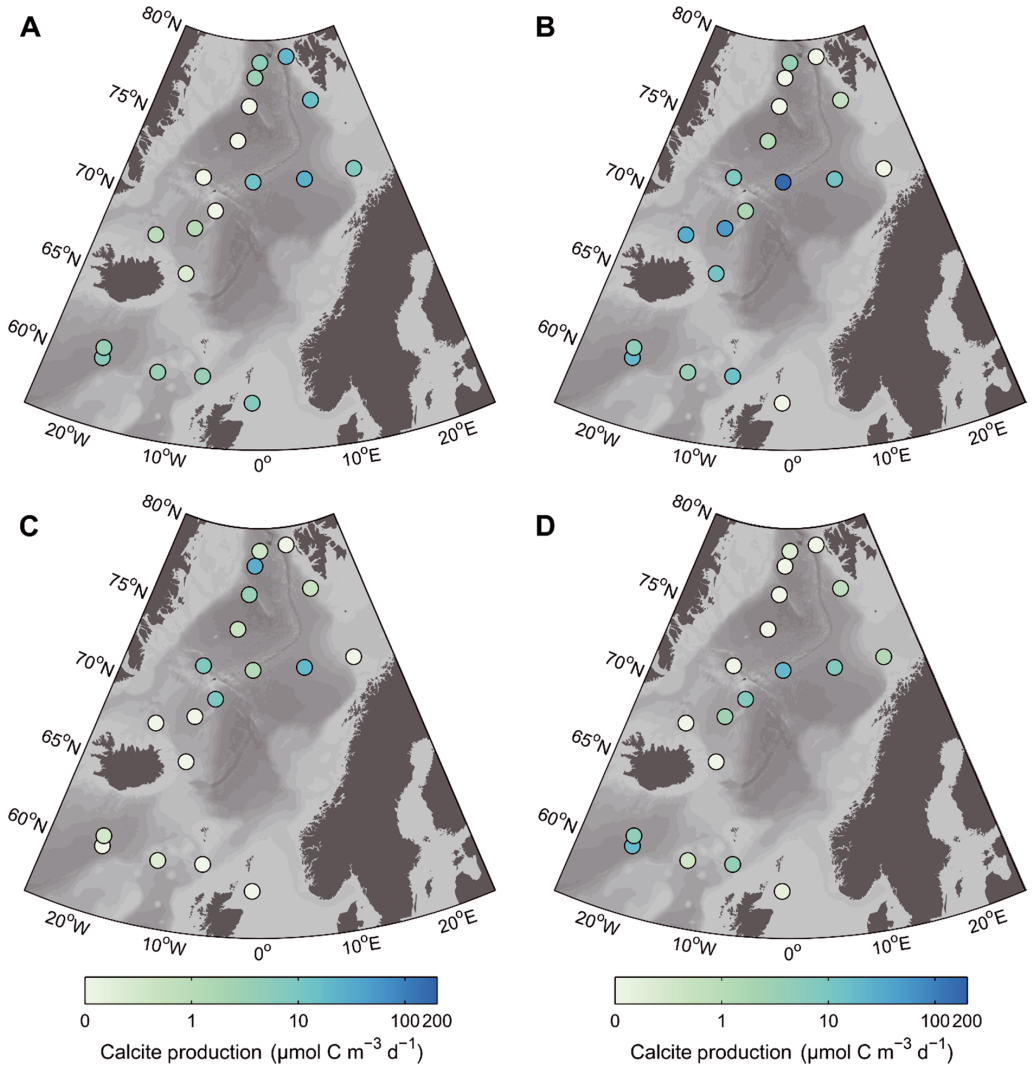

Fig. 5. Distribution of species-specific calcite production by (A) *Emiliania huxleyi*, (B) *Coccolithus pelagicus*, (C) *C. pelagicus* HOL, and (D) other coccolithophore species

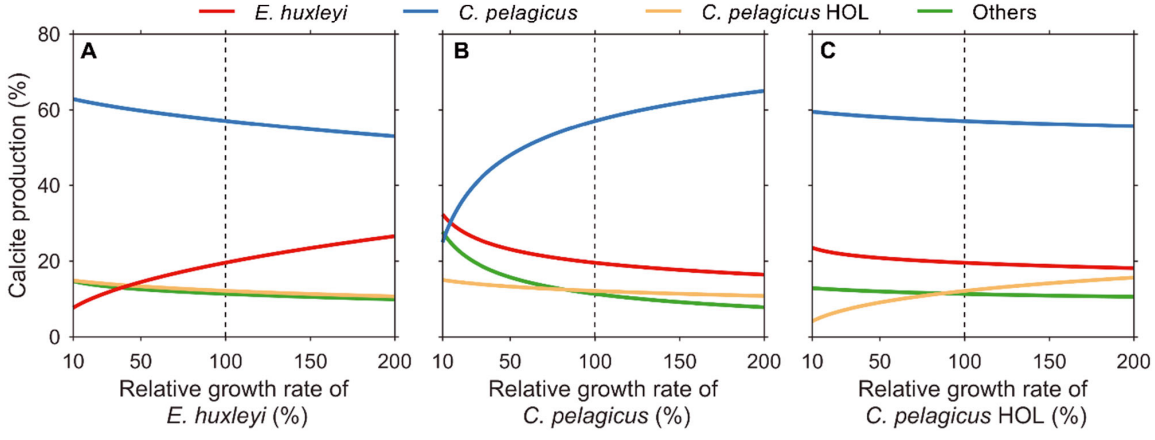

Fig. 6. Effect of varying the relative growth rate of one species on the species' contribution to calcite production. The growth rates of (A) *Emiliania huxleyi*, (B) *Coccolithus pelagicus*, and (C) *C. pelagicus* HOL were singly varied whilst all other species had a relative growth rate of 100 %

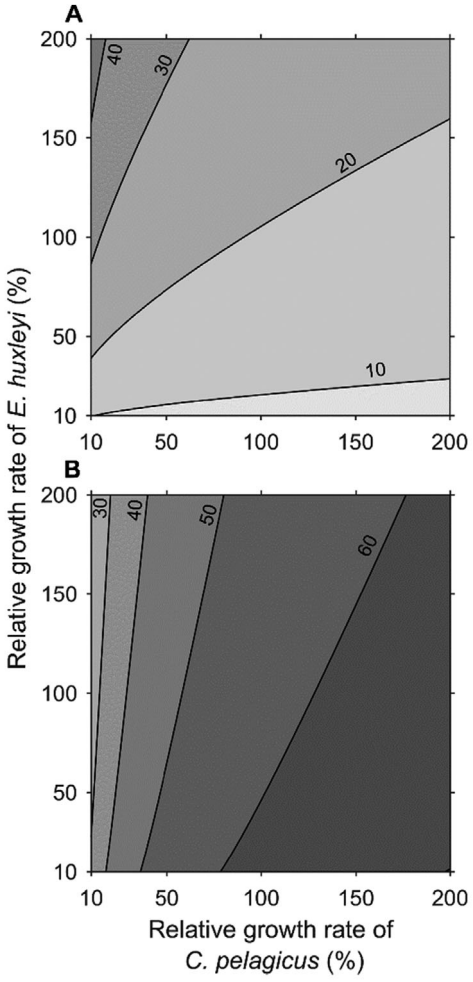

Fig. 7. Effect of varying the relative growth rates of both *Emiliania huxleyi* and *Coccolithus pelagicus* on the contribution to calcite production by (A) *E. huxleyi* and (B) *C. pelagicus*

both the estimates of cellular calcite and the method of calculating bulk community growth rates are valid. The station with the highest, and unrealistic, growth rate (3.0 d^{-1}; Table 4) was dominated by *C. pelagicus* HOL, which has relatively poorly constrained cellular calcite content, and may be greater and/or more variable than that estimated here.

The assumption that all coccolithophores in a mixed community have the same growth rate is unlikely to always hold true as individual phytoplankton species have been shown to vary significantly within natural populations (Weiler & Chisholm 1976), with the growth rates of individual species of phytoplankton primarily set by cell size (Finkel et al. 2010, Marañón et al. 2013, Marañón 2015). Although *E. huxleyi* is perceived to be fast growing relative to other coccolithophore species (Paasche 2002, Tyrrell & Merico 2004), little data exists concerning relative *in situ* growth rates of coccolithophores in mixed communities to test this perception. However, as the range in cell size of the coccolithophores present in the samples was small (~4 to 12 µm) compared to other phytoplankton groups, and the maximum growth rates in culture of similar species of coccolithophores have been shown to be almost identical (Marañón et al. 2013), it suggests that the growth rates of individual coccolithophore species within a mixed community may be similar. Furthermore, with such a narrow range in cell size, it is difficult to see how these coccolithophore species would be selectively grazed by different-sized grazers (i.e. micro- vs. meso-zooplankton).

Table 5. Results of the principal component analysis (PCA), including eigenvectors and Pearson's correlation coefficients for the relationships between PC scores, environmental variables and individual species contributions to both species composition and species-specific calcite production (CP_{sp}). Ω_C: calcite saturation state; pH_T: pH on the total scale; z_{eup}: euphotic zone depth; N^*: excess NO_x relative to PO_4; Si^*: excess dSi relative to NO_x; PAR: photosynthetically active radiation. **Bold** — significant: $^{***}p < 0.005$; $^{**}p < 0.01$; $^{*}p < 0.05$

Variables	Variables vs. principal components PC-1 (40.1%)	PC-2 (33.3%)
Environmental		
Temperature	0.23 (0.41)	0.53 (**0.87*****)
Salinity	0.19 (0.34)	0.53 (**0.87*****)
Ω_C	0.51 (**0.92*****)	0.21 (0.34)
pH_T	0.48 (**0.87*****)	−0.26 (−0.42)
N^*	−0.19 (−0.35)	0.51 (**0.83*****)
Si^*	−0.50 (**−0.90*****)	0.12 (0.19)
PAR	−0.06 (−0.12)	−0.14 (−0.22)
z_{eup}	−0.35 (**−0.62*****)	0.17 (0.27)
Latitude	−0.08	**−0.68*****
Longitude	0.16	−0.12
Species composition		
Emiliania huxleyi	**0.85*****	0.20
Coccolithus pelagicus	0.12	−0.43
C. pelagicus HOL	**−0.60****	−0.32
Syracosphaera spp.	0.04	**0.78*****
Acanthoica quattrospina	0.24	**0.66*****
Calciopappus caudatus	−0.35	0.32
Ophiaster sp.	0.06	**0.75*****
Algirosphaera robusta	0.02	0.13
% CP_{sp}		
E. huxleyi	**0.67*****	0.37
C. pelagicus	−0.08	−0.12
C. pelagicus HOL	**−0.57***	−0.27
Syracosphaera spp.	0.02	**0.75*****
A. quattrospina	0.22	**0.66*****
C. caudatus	−0.32	0.31
Ophiaster sp.	0.04	**0.69*****
A. robusta	−0.00	0.12

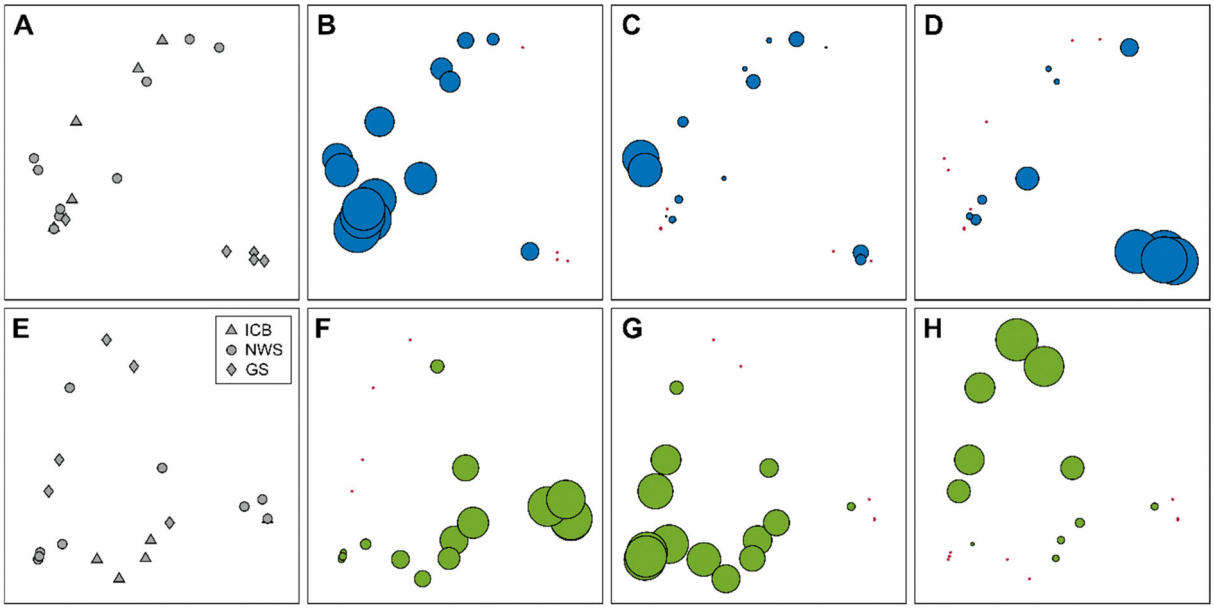

Fig. 8. Non-metric multidimensional scaling (NMDS) ordination of (A–D) coccolithophore species composition and (E–H) species-specific calcite production based on Bray-Curtis similarity. (A) and (E) are labelled according to the hydrographic province of the stations; (B–D) are overlaid with bubble plots of the composition of (B) *Emiliania huxleyi*, (C) *Coccolithus pelagicus*, and (D) *Coccolithus pelagicus* HOL; (F–H) are overlaid with bubble plots of the species-specific calcite production of (F) *E. huxleyi*, (G) *C. pelagicus*, and (H) *C. pelagicus* HOL. Red dots: absence of the species at those stations

Recent culture experiments of *E. huxleyi* and *C. pelagicus* that replicated the light and temperature conditions observed in the Arctic found that the relative differences in growth rates of the Arctic strains of these 2 species were small, with the growth rate of *C. pelagicus* averaging 85% that of *E. huxleyi* (Daniels et al. 2014). Furthermore, in pre-spring bloom conditions in the North Atlantic, the net growth rate of *C. pelagicus* was slightly higher than that of *E. huxleyi* (Daniels et al. 2015). Moreover, the observation that *C. pelagicus* is able to bloom with high cell densities (>1000 cells ml^{-1}; Milliman 1980, Tarran et al. 2001) requires *C. pelagicus* to be competitive, and implies that *C. pelagicus* does not have a vastly slower growth rate relative to other coccolithophore species. Therefore, the initial assumption that all coccolithophore species in a mixed community were growing at the same rate is considered a valid initial assumption. However, the robustness of the method for deriving CP_{sp} must be further evaluated by measuring how sensitive the results are to the relative growth rates.

Sensitivity analysis revealed that the dominance of calcite production by *C. pelagicus* was unaffected by the relative growth rate of both *E. huxleyi* (Fig. 6A) and *C. pelagicus* HOL (Fig. 6C) when the growth rate of only 1 species was manipulated. Only when the growth rate of *C. pelagicus* was <15% of the community did *E. huxleyi* become the greater calcite producer (Fig. 6B). Taking the geometric average community growth rate estimated from bulk CP and community calcite (0.33 d^{-1}), such a relative growth rate would equate to ~0.05 d^{-1}. Even when the growth rate of *E. huxleyi* was increased to 200% (e.g. 0.66 d^{-1}), *C. pelagicus* remained the major calcifier at growth rates down to 30% of the total community growth rate (~0.1 d^{-1}). In order for *E. huxleyi* to be the major calcifier when *C. pelagicus* had the same relative growth rate as the rest of the coccolithophore community, the relative growth rate of *E. huxleyi* had to be increased to 633% (e.g. 2.1 d^{-1}),

Table 6. Spearman's rank correlation (r_S) of environmental variables with coccolithophore species composition and species-specific calcite production (CP_{sp}). Ω_C: calcite saturation state; N^*: excess NO_x relative to PO_4; PAR: photosynthetically active radiation

Coccolithophore species composition		Species-specific calcite production (% CP_{sp})	
Environmental variables	r_S ($p < 0.01$)	Environmental variables	r_S ($p < 0.03$)
Temperature, Ω_C, N^*	0.553	Ω_C	0.368
Temperature, Ω_C	0.553	Temperature, Ω_C	0.308
Ω_C	0.546	Ω_C, PAR	0.256

which is both well beyond the modelled scenario, and unrealistic. These extreme scenarios far exceed allometric theory (Finkel et al. 2010, Marañón 2015), as well as the relative growth rates observed both in culture (Daniels et al. 2014) and in the field (Daniels et al. 2015). Furthermore, even in these unrealistic scenarios, *C. pelagicus* re-mained a significant single-species calcifier (>20%).

The sensitivity analysis demonstrated that variable growth rates will affect CP_{sp}, and further research is required to constrain both cellular calcite quotas and coccolithophore growth rates. However, in all but the most extreme (and unlikely) scenarios, *C. pelagicus* remained the dominant calcifier in the Arctic Ocean.

C. pelagicus as a key calcifier

Total community calcite production rates were similar to those measured previously in the North Sea and Arctic Ocean (<1 to 300 µmol C m^{-3} d^{-1}; Charalampopoulou et al. 2011), and in the subtropics (0.4 to 102 µmol C m^{-3} d^{-1}; Poulton et al. 2006), but generally lower than those previously measured on the northwest European shelf (2 to 825 µmol C m^{-3} d^{-1}; Poulton et al. 2014). Estimating CP_{sp} reveals that *C. pelagicus* is likely to be the major calcifier in this Arctic study, responsible for 57% of the calcite production in the Arctic Ocean and sub-polar Iceland Basin, despite forming only 2% of the total coccolithophore community abundance (Fig. 3). The influence of *C. pelagicus* on calcite production was further confirmed by a significant correlation between *C. pelagicus* abundance and total calcite production ($r = 0.55$, $p < 0.02$, $n = 19$); no other species correlated significantly with total calcite production. That *C. pelagicus* is able to dominate calcite production at such low relative abundances is due to its significantly higher cellular calcite quota compared to the rest of the coccolithophore species present in the community (Table 1). This potential to dominate community calcite production has been previously identified in a simplified 2-species model of *C. pelagicus* and *E. huxleyi* (Daniels et al. 2014). Although the natural communities in our samples were more complex and species-rich, *C. pelagicus* still had at least a 20-fold greater cellular calcite quota than the rest of the community (Table 1). Thus, when *C. pelagicus* is present in coccolithophore communities, it has the potential to dominate coccolithophore calcite production if its relative growth rate is high enough.

The dominance of *C. pelagicus* on calcite production in our study was not dependent on any single station. Removing the station (CTD 58) that had the highest rate of calcite production (202 µmol C m^{-3} d^{-1}), and therefore the largest influence over CP_{sp}, did not change the overall result. Although removing this station from the analysis resulted in a reduction of *C. pelagicus*-derived calcite production from 57 to 43%, *C. pelagicus* remained the single species with the largest source of calcite in the mixed communities of the Arctic Ocean and Iceland Basin. The effect of removing any other station from the analysis was minimal, with *C. pelagicus* remaining the dominant calcifier.

Although *E. huxleyi* is often perceived to be the most abundant and the keystone coccolithophore species (Paasche 2002), we found that it was neither the most abundant (27% total abundance; Fig. 3A), nor the major calcifier (20% of total calcite production; Fig. 3B), suggesting that it may not be the keystone species of coccolithophore in the North Atlantic and Arctic. However, previous studies have identified *E. huxleyi* as the most abundant coccolithophore in the Norwegian Sea (0 to 3000 cells ml^{-1}), although *C. pelagicus* was still an important component (0 to 30 cells ml^{-1}) of the communities studied (Baumann et al. 2000, Charalampopoulou et al. 2011). This change in dominance between studies is possibly due to seasonal (Baumann et al. 2000) or interannual variability occurring within the coccolithophore community. However, an increase in the abundance of *E. huxleyi*, coupled with a reduction in the abundance of other species such as *C. caudatus* and *A. robusta*, would be unlikely to change the overall result observed here, as *C. pelagicus* is the key calcifier (57%) despite forming only a small fraction (2%) of the coccolithophore community.

Despite dominating calcite production in this study, *C. pelagicus* is unlikely to be a globally dominant calcite producer, as its global distribution is constrained to the Arctic Ocean and sub-polar regions of the North Atlantic and North Pacific (McIntyre & Bé 1967, Ziveri et al. 2007). While other heavily calcified species (e.g. *Calcidiscus leptoporus*, *Helicosphaera carteri*) are more widely distributed (Ziveri et al. 2007) and thus have the potential to dominate calcite production (Daniels et al. 2014), here we show the biogeochemical importance of holococcolith-bearing coccolithophores (i.e. *C. pelagicus* HOL) and relatively weakly calcified but highly abundant coccolithophore species (i.e. *C. caudatus*). Further research into these lesser-studied species is required in order to improve our understanding of the role of different species in calcite production.

How *C. pelagicus* dominates Arctic community CP

It is well established that *C. pelagicus* is commonly found in the Arctic Ocean, but forms only a small component of the overall coccolithophore community abundance (Samtleben & Schröder 1992, Baumann et al. 2000, Charalampopoulou et al. 2011), as observed here. Yet, the importance of *C. pelagicus* as a calcite producer has not previously been recognised. That *C. pelagicus* is a disproportionately larger contributor to calcite production than abundance is due to the significantly higher cellular calcite content of *C. pelagicus* than other coccolithophore species. But how is it able to dominate calcite production — is it due to the absence of *E. huxleyi* or is it due to *C. pelagicus* being present in relatively high enough cellular abundances? Furthermore, what environmental characteristics determine these 2 factors?

To examine these competing factors, we can compare and contrast the compositional analysis based on species composition in terms of cell abundances and species-specific calcite production. The NMDS plots of species composition showed that the relative abundance of *E. huxleyi* in the community was a major driver of the variability in species composition between stations (Fig. 8B), whereas *C. pelagicus* had little influence (Fig. 8C). This is due to *C. pelagicus* being present in almost all samples but forming only a small fraction of the community. In contrast, *E. huxleyi* numerically dominated at some stations, but was totally absent from others (Table 3). This would suggest that as *C. pelagicus* dominates calcite production at stations where *E. huxleyi* is both present and absent, it is the relative abundance of *C. pelagicus* that allows it to dominate calcite production.

The pattern in the NMDS plots of CP_{sp}, however, with *E. huxleyi* (Fig. 8F) and *C. pelagicus* (Fig. 8G) both strongly influencing variability in CP_{sp}, suggest that *C. pelagicus* is responsible for a greater proportion of calcite production when the contribution of *E. huxleyi* is low. The difference between species composition and species contributions to calcite production between stations suggests that the dominance of *C. pelagicus* in terms of calcification is a combination of both the relative abundance of *C. pelagicus* compared to all other species of coccolithophore, and the relative absence of *E. huxleyi*, particularly from stations within the Greenland Sea (Fig. 8). Therefore, species composition has a significant impact on calcite production and which species dominate calcification in the Arctic Ocean.

In terms of understanding variability in calcite production in the Arctic Ocean, it is then important to determine what drives the variability in species composition throughout the Arctic. Variability in the physicochemical environment is clearly recognised as influencing the biogeography of coccolithophores (e.g. Charalampopoulou et al. 2011, Poulton et al. 2011). However, the relationship between species composition and environmental variables is complex and difficult to directly elucidate. Other studies have linked variability in coccolithophore community composition and calcite production to carbonate chemistry (Charalampopoulou et al. 2011, Smith et al. 2012), irradiance (Poulton et al. 2010, Charalampopoulou et al. 2011, Poulton et al. 2014) and nutrient availability (Poulton et al. 2011, 2014).

Using the same multivariate statistical approach as used by Charalampopoulou et al. (2011) on the data collected in this study, Spearman's rank correlations identified temperature, Ω_C and N^* as the environmental variables that could best explain species composition (Table 6). This contrasts with the results from Charalampopoulou et al. (2011) who found that pH and irradiance were the main drivers of coccolithophore species abundance along a transect from the North Sea to the Arctic Ocean. The influence of temperature and N^* on species composition is likely to be due to the contrasting community composition in the warmer (>10°C) and less nitrate-depleted (N^* of −0.4 to −1.3) Iceland Basin compared to the colder (<8°C) and more nitrate-depleted (N^* of −1.2 to −3.0) Norwegian and Greenland Seas. The fact that PC-2 — which was related to temperature ($r = 0.87$, $p < 0.005$, $n = 19$) and N^* ($r = 0.83$, $p < 0.005$, $n = 19$) and correlated with latitude ($r = 0.68$, $p < 0.005$, $n = 19$) — correlated with those species found only in the Iceland Basin (*Syracosphaera* spp., *Acanthoica quattrospina* and *Ophiaster* sp.) further confirms the role of temperature in influencing species composition. However, temperature did not significantly affect CP_{sp}, with Ω_C alone best explaining the contribution of species to CP_{sp}. Those species limited only to the Iceland Basin, thus strongly influenced by temperature, were relatively minor contributors to calcite production (0 to 27%) and had little impact on the variability in CP_{sp}.

That both species composition and CP_{sp} were affected by Ω_C can be further examined using the results from the PCA: PC-1, which is positively correlated with Ω_C ($r = 0.92$, $p < 0.005$, $n = 19$), is also positively correlated with the contribution of *E. huxleyi* to both species composition ($r = 0.85$, $p < 0.005$, $n = 19$) and CP_{sp} ($r = 0.67$, $p < 0.005$, $n = 19$), but is negatively correlated with the contribution of *C. pelagicus* HOL to both species composition ($r = -0.60$, $p < 0.01$,

n = 19) and CP_{sp} (r = −0.57, p < 0.05, n = 19). This suggests that *E. huxleyi* represents a smaller fraction of the coccolithophore community in regions of lower saturation state, whereas *C. pelagicus* HOL represents a higher fraction in these conditions. This could be interpreted to suggest that the expected decline in saturation state in the future would reduce the abundance of *E. huxleyi*. However, our analysis does not allow us to conclude that Ω_C is directly affecting species composition, but rather that within the present day Arctic Ocean, *E. huxleyi* forms a smaller component of the coccolithophore community in regions of lower Ω_C. It should be noted that Ω_C was above the saturation point at all stations and that the gradient in saturation state was much lower (2.6 to 4.2) than in other environmental variables, such as the gradient in temperature (1.0 to 10.6°C) and NO_x (0.5 to 10.6 mmol N m^{-3}). Furthermore, Ω_C is mainly influenced by temperature, as well as salinity, and changes in C_T and A_T are due to biological productivity (Tynan et al. 2016). Temperature is recognised to have a significant control on coccolithophore distributions; for example, there is a well recognised 2°C limit to the range of *E. huxleyi* (Holligan et al. 2010), while *C. pelagicus* is able to persist in sub-zero temperatures (Braarud 1979). Therefore, it may be that temperature is a key driving factor with both direct and indirect influences on the species composition and CP_{sp}.

The relationship between the environment, the coccolithophore community and calcite production is likely to be more complex than presented here; we found no significant environmental influence on total calcite production (p = 0.09), or the contribution of *C. pelagicus* to species-specific calcite production (p = 0.1), implying that other ecophysiological and environmental interactions exist and may influence species biogeography. Furthermore, correlations of individual environmental variables with abundance and CP_{sp} did not produce any significant results, further demonstrating the complexity of the interaction between coccolithophore abundance, calcite production, and environmental variables (Poulton et al. 2014). While the influence of some environmental variables (e.g. temperature) on coccolithophore physiology are well established, we are only beginning to get a mechanistic understanding of the influence of carbonate chemistry; for example, calcite production appears dependent on bicarbonate as its primary substrate, and is inhibited by protons (Bach et al. 2015), with Ω_C not directly affecting calcite formation (Bach 2015). However, we still have very little basic understanding of coccolithophore physiology; for example, until we understand why coccolithophores calcify, and the energetic costs associated with it, we cannot fully understand how cellular calcification will respond to a changing ocean, and the impact this will have on the coccolithophore community in terms of species composition or competitive fitness.

Wider implications

Research into the effect of ocean acidification and climate change on coccolithophores has been dominated by studies of *E. huxleyi* as it is globally abundant and forms large-scale blooms of significant biogeochemical importance (Holligan et al. 1993, Poulton et al. 2013). However, *E. huxleyi* can be considered an atypical coccolithophore species in terms of its genetic lineage, physiology and ecology (de Vargas et al. 2007), and therefore the response of *E. huxleyi* to climate change and ocean acidification may not apply to other coccolithophore species. Few studies have examined the impact of ocean acidification on other species of coccolithophore (but see e.g. Langer et al. 2006, Fiorini et al. 2011, Krug et al. 2011), and very little is known about the Arctic species *C. pelagicus*. This study shows that unless *C. pelagicus* grows extremely slowly (<15%) compared to the rest of the coccolithophore community, it is the key calcifier in a region considered particularly vulnerable to ocean acidification and warming, and the response of *C. pelagicus* to climate change and ocean acidification could have a major effect on calcite production in the Arctic and sub-polar Iceland Basin. Examination of the fossil record of *C. pelagicus* during the Palaeocene–Eocene Thermal Maximum, arguably the best geological equivalent of modern-day climate change, found that it was not able to maintain optimum growth during this period (Gibbs et al. 2013), and had reduced calcification rates (O'Dea et al. 2014). If *C. pelagicus* exhibits a similar response in the modern ocean to current perturbations, it could cause a significant reduction in calcite production within the Arctic Ocean and Iceland Basin, with a major impact on carbon cycling in the North Atlantic.

Acknowledgements. We acknowledge the UK Natural Environmental Research Council (NERC; Grant references NE/H017097/1 and NE/H017348/1), Department of Environment, Food and Rural Affairs (Defra), and Department of Energy and Climate Change (DECC) for funding the research cruise via the UK Ocean Acidification research programme, and to the Danish, Icelandic and Norwegian diplomatic authorities for granting permission to travel and work in Greenland, Iceland and Svalbard coastal and offshore waters. We thank the officers and crew of the RRS 'James Clark Ross'; Mark Stinchcombe for assistance with

nutrient measurements, Fred Le Moigne for assistance with sea ice data. MODIS SST data were obtained from the NASA Ocean Color distributed Archive (http://oceancolor.gsfc.nasa.gov/). Sea ice concentration data from the Nimbus-7 SMMR and DMSP SSM/I-SSMIS passive microwave sensors were obtained from the National Snow and Ice Data Center (www.nsidc.org). Author contributions: C.J.D and A.J.P designed the research; C.J.D., A.J.P., J.R.Y., M.E., M.P.H., M.R-R., and E.T. performed research; C.J.D. and A.J.P. analysed data; and C.J.D. wrote the paper, with input from all co-authors.

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Appendix

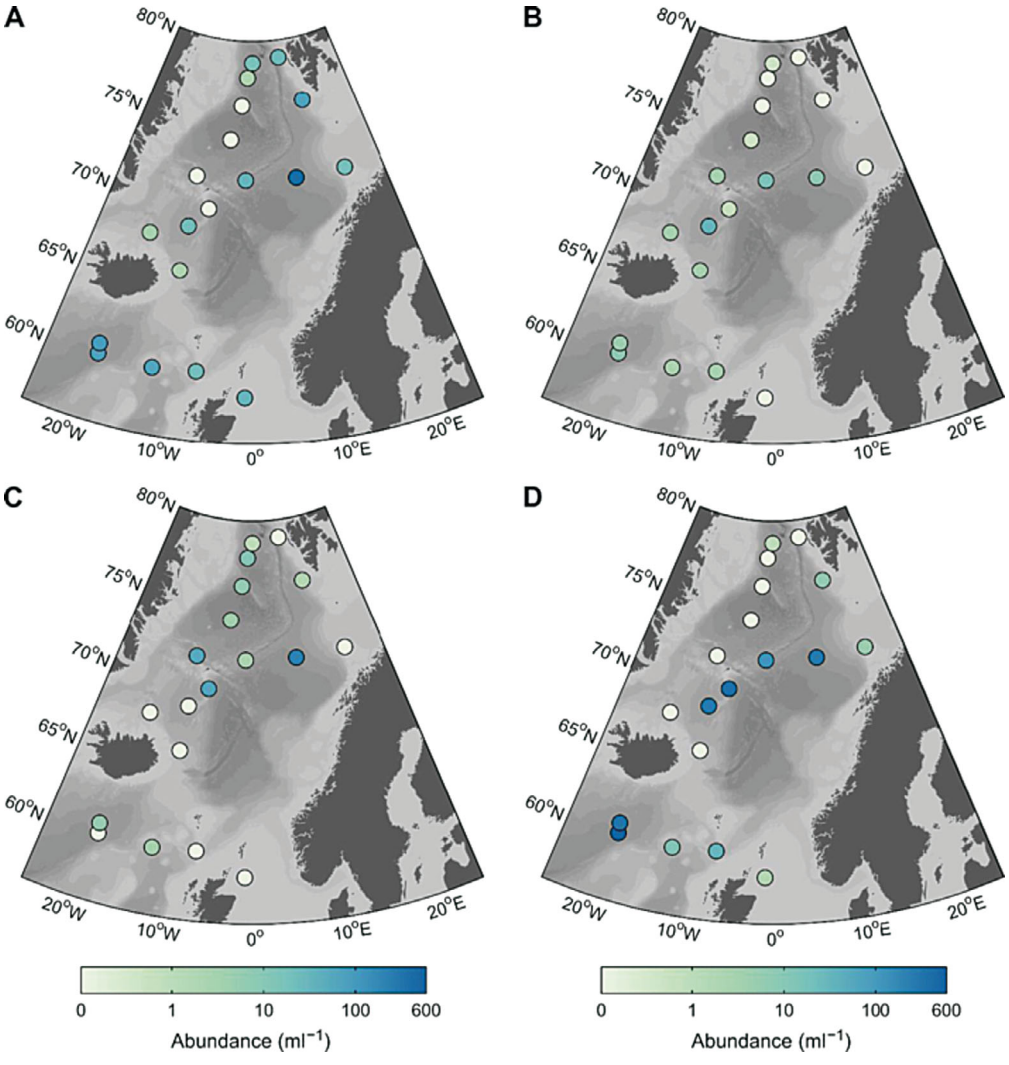

Fig. A1. Distribution of coccolithophore abundances: (A) *Emiliania huxleyi*, (B) *Coccolithus pelagicus*, (C) *C. pelagicus* HOL, and (D) other coccolithophore species

Editorial responsibility: Katherine Richardson, Copenhagen, Denmark

Submitted: November 12, 2015; Accepted: June 24, 2016
Proofs received from author(s): August 3, 2016