



Coccolithophore surface distributions in the North Atlantic and their modulation of the air-sea flux of CO₂ from 10 years of satellite Earth observation data

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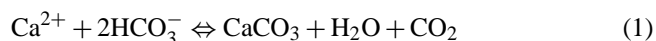
Abstract. Coccolithophores are the primary oceanic phytoplankton responsible for the production of calcium carbonate (CaCO₃). These climatically important plankton play a key role in the oceanic carbon cycle as a major contributor of carbon to the open ocean carbonate pump (~50 %) and their calcification can affect the atmosphere-to-ocean (air-sea) uptake of carbon dioxide (CO₂) through increasing the seawater partial pressure of CO₂ (*p*CO₂). Here we document variations in the areal extent of surface blooms of the globally important coccolithophore, *Emiliania huxleyi*, in the North Atlantic over a 10-year period (1998–2007), using Earth observation data from the Sea-viewing Wide Field-of-view Sensor (SeaWiFS). We calculate the annual mean sea surface areal coverage of *E. huxleyi* in the North Atlantic to be 474 000 ± 104 000 km², which results in a net CaCO₃ carbon (CaCO₃-C) production of 0.14–1.71 Tg CaCO₃-C per year. However, this surface coverage (and, thus, net production) can fluctuate inter-annually by –54/+81 % about the mean value and is strongly correlated with the El Niño/Southern Oscillation (ENSO) climate oscillation index ($r = 0.75$, $p < 0.02$). Our analysis evaluates the spatial extent over which the *E. huxleyi* blooms in the North Atlantic can increase the *p*CO₂ and, thus, decrease the localised air-sea flux of atmospheric CO₂. In regions where the blooms are prevalent, the average reduction in the monthly air-sea CO₂ flux can reach 55 %. The maximum reduction of the monthly air-sea CO₂ flux in the time series is 155 %.

This work suggests that the high variability, frequency and distribution of these calcifying plankton and their impact on *p*CO₂ should be considered if we are to fully understand the variability of the North Atlantic air-to-sea flux of CO₂. We estimate that these blooms can reduce the annual N. Atlantic net sink atmospheric CO₂ by between 3–28 %.

1 Introduction

Understanding the pathways, sources, sinks and impact of CO₂ on the Earth's climate system is essential for monitoring climate and predicting future scenarios. The global ocean is considered the only true net sink of anthropogenic CO₂, annually absorbing ~30 % of emissions, with the North (N.) Atlantic accounting for ~23 % of this global uptake (Sabine et al., 2004). Despite this, it is currently not clear if the global oceanic sink is following the increasing atmospheric levels of CO₂ (Sabine et al., 2004). The N. Atlantic sink in particular has been shown to be highly variable (Watson et al., 2009) and the mechanisms driving this variability are not well understood. Therefore, isolating and reducing the uncertainties in the estimates of the oceanic sink is a crucial goal of climate science (Le Quere et al., 2009).

Coccolithophores are an abundant and widely distributed group of marine phytoplankton that form external calcium carbonate (CaCO_3) scales or platelets, called coccoliths, during their life cycle. Coccolithophores are major calcifiers in the open ocean (Sabine et al., 2004), and play a key role in oceanic carbon cycles (Balch et al., 2005; Iglesias-Rodríguez et al., 2002). Primary production sustains the organic carbon pump that transfers organic carbon from surface waters to the deep ocean and pelagic calcification (such as that from coccolithophores) counter-acts the CO_2 uptake related to primary production (Harlay et al., 2010). CaCO_3 also acts as a ballast mineral for marine organic particles, increasing their sinking rate to deeper waters and is thought to increase the long-term storage of carbon (Honjo et al., 2008). CaCO_3 coccoliths are generated through the calcification reaction:



This leads to a disequilibrium in the ocean carbonate system that can drive an additional flux of CO_2 to the atmosphere; calcification depletes surface CO_3^{2-} , reduces alkalinity and increases the seawater partial pressure of CO_2 ($p\text{CO}_2$). Within this process the deposition of 1 mol of calcium carbonate releases ~ 0.6 mol of CO_2 into seawater (Frankignoulle and Gattuso, 1994). In addition, production and sedimentation of coccoliths is a long-term carbon sink process. As previously mentioned, it is important to note that calcification is not the only biological control on $p\text{CO}_2$. CO_2 uptake by photosynthesis and its release by respiration are major processes by which marine organisms and ecosystems can alter $p\text{CO}_2$ (Gattuso et al., 1995) and the interaction between photosynthesis, respiration and calcification in certain marine systems (e.g., coral reefs) can be tightly coupled (Gattuso et al., 1995). Equally, the biogeochemical history of the body of water is also important.

It has been documented since the early 1980s by the use of satellite imagery that there is a considerable amount of *E. huxleyi* activity in the N. Atlantic (Holligan et al., 1983). Of the numerous coccolithophores, the cosmopolitan species *Emiliania huxleyi*, when present at high concentrations (blooms) in the surface layer, profoundly impacts the optical properties of the upper ocean (Balch et al., 1991; Tyrrell et al., 1999; Holligan et al., 1993). The high reflectance spectral signature of *E. huxleyi* blooms allows the study of their frequency, distribution pattern and areal extent by Earth observation (EO) from satellites (Brown and Yoder, 1994a; Cokacar et al., 2001; Smyth et al., 2004; Merico et al., 2003; Shutler et al., 2010; Iglesias-Rodríguez et al., 2002). Based on EO data the *E. huxleyi* areal coverage in the N. Atlantic has been estimated to account for 40–60% of the global areal coverage of *E. huxleyi* (Brown and Yoder, 1994a), with single blooms covering areas of $>100\,000$ km² (Brown and Yoder, 1994a). Through the use of EO data, these calcifying blooms are known to occur over large areas of the N. Atlantic and the ability of calcification to alter the $p\text{CO}_2$ is well understood. However, the spatial and temporal im-

pact that these blooms have on $p\text{CO}_2$ and on the air-sea flux of CO_2 (F) across the whole of the N. Atlantic has yet to be characterised.

It has already been observed that $p\text{CO}_2$ in regions of the N. Atlantic is highly variable (Watson et al., 2009; Schuster and Watson, 2007) and within some regions of the N. Atlantic sub-polar gyre $p\text{CO}_2$ has unexpectedly increased faster than atmospheric levels (Lefevre et al., 2004; Le Quere et al., 2009). Lefevre et al. (2004) concluded that this unexpected increase was due to a decrease in biological activity as the trend was strongest during spring and summer (Lefevre et al., 2004). Similarly, in general *E. huxleyi* bloom activity in the N. Atlantic increases as spring progresses into summer, exhibiting a peak in bloom activity during late June and early July, with blooms still forming in the north east during August. Due to the slow equilibration time between the surface ocean and the atmosphere (6 months to a year) the increased $p\text{CO}_2$ due to calcification will remain in place long after a bloom has finished, blocking a large portion of the annual CO_2 sink cycle (Robertson et al., 1991). In addition, increased *E. huxleyi* bloom activity at higher latitudes has been reported in the polar Barents sea (Smyth et al., 2004), while an increasing (multi-decadal) trend in multi-taxa coccolithophore abundance has been identified (McQuatters-Gollop et al., 2010) for a sub-set of the north east Atlantic. Therefore, it is possible that calcifying plankton may be playing a role in the unexpected elevated $p\text{CO}_2$ in the waters of the N. Atlantic sub-polar gyre.

Many regional ecosystem models used to produce short to medium term predictions of air-sea CO_2 fluxes in the N. Atlantic do not include the effect that calcifying plankton can have on $p\text{CO}_2$ (e.g., ERSEM; Wakelin et al., 2012). Global biogeochemical/ecosystem models often represent calcification by a highly simplified parameterisation, for instance as a globally constant rain ratio (e.g., OCMIP-2, (Najjar et al., 2007), a latitudinal function (e.g., MEDUSA; Yool et al., 2011) or as a function of the surface saturation state of calcite or aragonite (e.g., BioGEM; Ridgwell et al., 2007). Calcification can alter the $p\text{CO}_2$ and, thus, the air-sea flux and so these simplifications may introduce a large source of uncertainty within the modelled air-sea fluxes.

In this paper, we study the areal extent of *E. huxleyi* blooms in the open ocean (>200 m depth) in temperate to high latitudes of the N. Atlantic (35°N – 68°N , 75°W – 11°E) (Fig. 1).

We use EO derived bloom surface areal coverage data collected over a 10-year period (1998–2007) to (i) study the variability in the coccolithophore surface coverage, (ii) investigate linkages between surface coverage and leading climate oscillators, (iii) determine the CaCO_3 -C standing stock and (iv) estimate the effect that these surface distributions have on modulating $p\text{CO}_2$ and, thus, F in the N. Atlantic.

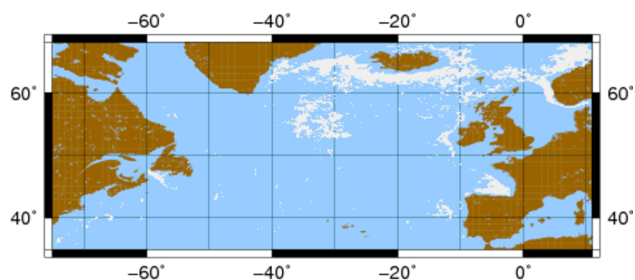


Fig. 1. The study region. The white regions show the spatial distribution of open ocean (>200 m) *Emiliania huxleyi* blooms during April to August as detected by the Earth observation (EO) data used in this study (1998–2007). Land is in brown. A white pixel shows that an instance of an *Emiliania huxleyi* bloom has appeared once or more within the 1998–2007 time series.

2 Methods

E. huxleyi blooms were detected in ocean colour radiometry measurements acquired by the Sea Viewing Wide Field-of-View Sensor (SeaWiFS) from 1998 to 2007 by applying a recently developed technique (Shutler et al., 2010) to estimate their surface areal coverage. We restricted our examination of data to the months from April to August so that the minimum solar elevations were between 30° and 50°. This criterion helps reduce the uncertainties in the optical remote sensing data. It is also ecologically appropriate as low light levels and low temperatures inhibit coccolithophore growth during the Northern Hemisphere winter months (Tyrrell and Merico, 2004). A confusion matrix evaluation between the EO data and a 10 year in situ dataset from the Continuous Plankton Recorder (CPR) (Reid et al., 2003) combined with analytical error propagation were used to determine error estimates in the EO derived surface areal coverage. Net CaCO_3 -C production of *E. huxleyi* blooms was calculated using constant standing stock approaches and a range of parameter quantities. The impact that the *E. huxleyi* distributions have on $p\text{CO}_2$ (and, thus, F) was then determined using the SeaWiFS data, a constant CaCO_3 -C concentration, a carbonate system software toolbox (seacarb) and climatology data (Takahashi et al., 2009).

2.1 Datasets

The SeaWiFS level 2 quality controlled normalised water leaving radiance data covering the region of interest for all complete years (1998–2007) were obtained from the NASA ocean colour website (NASA, 2010). All SeaWiFS data were re-projected using generic tools (Shutler et al., 2005) to an equidistant cylindrical projection with an equatorial pixel size of 4 km. The SeaWiFS data archive also includes data for 2008–2009. However, these data were excluded from analysis due to prolonged gaps in data coverage during these years, as these prolonged gaps fail the requirements for the coccolithophore classification algorithm.

The mean Multivariate El Niño/Southern Oscillation (ENSO) Index data for each year were calculated using monthly data downloaded from NOAA (2011). The N. Atlantic Oscillation (NAO) data used here are the result of Principal Component Analysis of sea-level pressure over the N. Atlantic sector for months from December to March (Hurrell et al., 2001) and were obtained from Hurrell (2011). Correlations between the climate indices (ENSO and NAO) and the total season surface areal coverage of *E. huxleyi* blooms were determined using the Pearson correlation coefficient in python using the SciPy (v0.7.1) toolbox. Climatological data of the partial pressures of CO_2 in seawater ($p\text{CO}_2$) and air, salinity, solubility and the gas transfer velocity (Takahashi et al., 2009) at $5^\circ \times 4^\circ$ global grid resolution were used as the basis to calculate F . Two grid cells south of Iceland were missing from the climatology data and this is a region of annual *E. huxleyi* activity e.g., (Raitso et al., 2006). Therefore, to allow this area to be studied the climatology values within these cells were generated by linearly interpolating the data from the adjacent cells. In situ measurements of coccolithophore cell numbers for the same geographical region and temporal period as the SeaWiFS data were obtained from the CPR survey (Reid et al., 2003).

2.2 *Emiliania huxleyi* surface areal coverage

For each satellite pass in the time series a map of *E. huxleyi* surface areal coverage was generated (Shutler et al., 2010). The first stage of this algorithm exploits temporal correlation to remove the background signal (referred to as background subtraction and relies upon the data from preceding months to characterise the region of interest). This approach reduces the number of false positives caused by suspended sediment and allows the spectral classification stage to focus on just the areas of ocean that have recently changed (e.g., due to the formation of a bloom). The second stage of the algorithm uses a spectral algorithm (Brown and Yoder, 1994a) to classify the remaining data into areas that contain *E. huxleyi* and those that do not. The algorithm default parameters and data quality control thresholds followed those of Shutler et al. (2010). All maps of *E. huxleyi* surface coverage were masked based on bathymetry and only regions with depth >200 m were included in the analysis as these are considered open ocean waters. Additionally, the enclosed region of the Laurentian Channel (in the Gulf of St Lawrence in the region of 48.0° N 61.5° W) was not included as this is only 290 m deep and was considered susceptible to influences from river run off. Monthly maps of counts of *E. huxleyi* elements (CC_{xy}) and cloud-free elements (CF_{xy}) at pixel position (x, y) were generated from the EO data for all data for years 1998–2007. A cloud free element is defined as any pixel with a water leaving radiance >0 at any of the wavelengths of interest (443, 510 and 555 nm). For each month, m , the estimated area of *E. huxleyi* surface coverage (AC_{my}) at each latitudinal image

line (y) is determined using:

$$AC_{my} = \left(\frac{\sum_{x=1}^X CC_{xy}}{\sum_{x=1}^X CF_{xy}} \right) A_y NV_y \quad (2)$$

where x is the longitudinal grid point, NV_y is the number of valid ocean pixels in the longitudinal row and A_y is the pixel area at that latitude. The pixel area was determined by representing the Earth as an ellipsoid. The use of NV_y stops regions of land from consistently biasing the results and the normalisation using the cloud free element count (CF_{xy}) is done to make a first order correction for cloud cover by assuming that the proportion of *E. huxleyi* under the clouds is the same as that observed in cloud free areas. Summing this over all latitudes gives the monthly surface coverage; summing across all months (April–August) gives the yearly total surface coverage (in km^2).

2.3 Standing stock and air-sea flux calculations

Previous $\text{CaCO}_3\text{-C}$ standing stock calculations (e.g., Brown and Yoder, 1994a) have relied on an estimate of the cell density, the $\text{CaCO}_3\text{-C}$ content of each coccolithophore cell and a mixed layer depth (and have assumed a fixed number of coccoliths per cell). In this study, we are interested in the impact that calcification can have on the $p\text{CO}_2$ and to do this we require a $\text{CaCO}_3\text{-C}$ concentration. Therefore, our $\text{CaCO}_3\text{-C}$ standing stock calculations use a coccolith concentration and a quantity of $\text{CaCO}_3\text{-C}$ per coccolith, which allows a $\text{CaCO}_3\text{-C}$ concentration to be determined without the need to assume a fixed number of coccoliths per cell. Bloom coccolith concentrations in the literature range 75 000–300 000 coccoliths ml^{-1} (Balch et al., 1991, 1992, 1996). Similarly, $\text{CaCO}_3\text{-C}$ per coccolith quantities range between 0.2–0.6 $\text{pg CaCO}_3\text{-C}$ (Balch et al., 1992, 1996; Brown and Yoder, 1994b; Holligan et al., 1983). For our indicative net $\text{CaCO}_3\text{-C}$ content estimates and our flux analyses we use the middle values in each of these ranges, so 187,500 coccoliths ml^{-1} and 0.4 $\text{pg CaCO}_3\text{-C}$ per coccolith; this results in a $\text{CaCO}_3\text{-C}$ concentration of 0.075 mg l^{-1} . For consistency with previous studies, we adopt the mixed layer depth of 20 m as used in previous work (Brown and Yoder, 1994a). To investigate the sensitivity of these parameter choices, we also calculate net production rates across the complete range (quoted above) of coccolith concentrations, $\text{CaCO}_3\text{-C}$ coccolith weights and a range of mixed layer depths. The volume of water below a 1 km^2 area of ocean assuming a (mixed layer) depth of 20 m is 20×10^{12} ml. Multiplying this by an areal coverage and the per coccolith $\text{CaCO}_3\text{-C}$ weight gives the standing stock estimate of the net $\text{CaCO}_3\text{-C}$ content for a given areal coverage. Next a bloom residence time for each bloom is required. Examples of residence values from the literature range between 5–40 days, e.g., (Berelson et al., 2007; Brown and Yoder,

1994b; Harlay et al., 2010). Previous work for the region of study used EO from the Coastal Zone Color Scanner (CZCS) data to estimate mean and median bloom durations of 36 ± 25 days and 31 days, respectively (Brown and Yoder, 1994b). From studying our SeaWiFS data, example blooms in a range of locations were on average visible in the same approximate location for ~ 3 weeks (21 days). The EO data is only able to detect bloom conditions under cloud free conditions and is also unable to detect the development of the bloom. Therefore, the period of initial growth will not be within the 21 days. To include the period of growth prior to the blooms being visible in the EO data, and to cover the potential biasing due to cloud, the estimated residence time was extended to 30 days. Therefore, it was assumed that *E. huxleyi* blooms would be present in the water for an average of 1 month or ~ 30 days meaning that a monthly surface coverage map represents the mean monthly conditions.

The net air-sea fluxes were calculated for each year using the Takahashi climatology dataset as perturbed by the *E. huxleyi* distributions (1998–2007) and following the methods in the original climatology publication (Takahashi et al., 2009) to produce F in teragrams of carbon per month (Tg C month^{-1}). The air-sea flux of CO_2 is estimated as the difference in $p\text{CO}_2$ between the surface water and the atmosphere modulated by an exchange coefficient (Takahashi et al., 2009). As we follow the methods of Takahashi et al. (2009) for deriving the air-sea fluxes, the solubility coefficient and gas transfer velocity parameterisations follow those of the original publication. The water and atmospheric $p\text{CO}_2$ are estimated from Takahashi et al. (2009) by increasing the climatological values linearly with year by $1.5 \mu\text{atm yr}^{-1}$ to account for the known underlying global trend (Takahashi et al., 2009). We assume that the ratio of particulate inorganic carbon to particulate organic carbon (rain rate) remains the same between years. The corrected seawater $p\text{CO}_2$ within each grid cell was then modified based on *E. huxleyi* bloom activity using (i) a fixed concentration of $\text{CaCO}_3\text{-C}$ within each bloom of 0.075 mg l^{-1} ; (ii) the ratio (ψ) of CO_2 released ($\Delta[\text{CO}_2]$) to precipitated CO_3 as calculated using the methods in the seacarb software package (Lavigne and Gattuso, 2011; Dickson et al., 2007; DOE, 1994; Frankignoulle and Gattuso, 1994; Zeebe and Wolf-Gladrow, 2001) and (iii) the change in $p\text{CO}_2$ dissolved into the water due to calcification was calculated using $\Delta[\text{CO}_2]/k_0$, where k_0 is the temperature dependent CO_2 seawater constant. The modified $p\text{CO}_2$ was then used to determine F assuming also that the *E. huxleyi* blooms raise the temperature of the surface water within the bloom by 1°C (Holligan et al., 1993). This process was then repeated for all months and years, and the average and maximum percentage differences in seawater $p\text{CO}_2$, the air-sea partial pressure difference ($\Delta p\text{CO}_2$) and F in each month (with respect to the original climatological values) were determined. The analysis was also repeated excluding the years with strong mean multivariate ENSO index values (1998 and 1999). This additional analysis was conducted to determine

if the modulation of the air-sea fluxes is primarily driven by strong ENSO events.

3 Results

3.1 Uncertainties in *E. huxleyi* surface coverage estimates

The accuracy of the coccolithophore (*E. huxleyi*) maps was analysed using the method of (Shutler et al., 2010) and all available CPR (26 738 in situ data points) and SeaWiFS data for 1998–2007. This analysis provides a measure of the methods ability to correctly detect *E. huxleyi* blooms. This analysis using an in situ database resulted in $N = 2102$ coincident data points (265 bloom instances, 1837 no-bloom instances) and a correct classification rate (CCR) of 78 % with a false alarm rate (FAR) of 14 %. (An ideal case would be $CCR = 100\%$ and $FAR = 0\%$). The $FAR = 14\%$ can be partially explained by the mesh size characteristics of the CPR instrument. The mesh size of the CPR is 270 μm , whereas an *E. huxleyi* coccosphere diameter is of the order of $\sim 5\ \mu\text{m}$ and coccolith diameters are of the order of 2.5 μm . Therefore, the CPR is only able to detect the presence of coccolithophores when the coccospheres or coccoliths become caught on the strands of the mesh. As the blooms subside the spectral signal from the coccoliths will still be significant and due to the smaller coccolith size they are less likely to be caught on the strands of the CPR mesh. Following this analysis, we estimate the total uncertainty in the *E. huxleyi* surface coverage data as having an upper limit of 22 % (100–CCR). To investigate how much of this uncertainty was due to the spectral algorithm, we performed a theoretical error propagation analysis of the spectral algorithm. This analysis was performed by propagating the known uncertainties of the individual spectral channels and their ratios (Hooker et al., 2001) through the classification algorithm using standard error propagation approaches (Taylor, 1997). These known uncertainties are all random perturbations as the sensor spectral channels have been shown not to exhibit significant bias (Hooker et al., 2001). The resultant uncertainty in the spectral classification algorithm due to the known uncertainties in the input spectral data was calculated to be 11 %, which equates to half of the total uncertainty.

3.2 Surface coverage, CaCO_3 production and correlation with climate indices

The mean annual surface areal coverage of the *E. huxleyi* blooms in the study area during the 10 years examined was $474\,000 \pm 104\,000\ \text{km}^2$ with a highly variable inter-annual surface coverage, varying from -54% to $+81\%$ of the mean value in 2000 and 1998, respectively. Based on our mid range standing stock parameter choices (see Sect. 2.3) this areal extent results in a standing stock production of 0.71 ± 0.16 teragrams $\text{CaCO}_3\text{-C}$ per year ($\text{Tg CaCO}_3\text{-C yr}^{-1}$). Using the

full range of parameter values and the mean areal estimate ($474\,000\ \text{km}^2\ \text{yr}^{-1}$) produces a $\text{CaCO}_3\text{-C}$ standing stock of $0.14\text{--}1.71\ \text{Tg CaCO}_3\text{-C yr}^{-1}$. The monthly surface areal coverage and our mid range net $\text{CaCO}_3\text{-C}$ production for the months April to August for the study period are shown in Fig. 2a and b shows the net surface coverage for each year (April to August). The yearly (April–August) total *E. huxleyi* surface areal coverage (1998–2007) was found to be strongly positively correlated with the mean multivariate ENSO index ($r = 0.75$, $p < 0.02$) (Fig. 2d). A negative correlation with the winter NAO was also observed ($r = -0.47$) (Fig. 2c), but the correlation was not statistically significant ($p = 0.17$).

3.3 Modulation of the air-sea flux of CO_2

We calculated values of $p\text{CO}_2$, $\Delta p\text{CO}_2$ and F in each year from 1998 to 2007 and each month from April to August as perturbed by the *E. huxleyi* activity. Figure 3 shows the average percentage change in these variables observed in each cell over all years and months. Figure 4 shows the maximum of these monthly values over the five months. In regions where *E. huxleyi* blooms are prevalent, monthly $p\text{CO}_2$ is increased on average by 14 % (Fig. 3a), reducing the monthly $\Delta p\text{CO}_2$ by 77 % (Fig. 3b), and consequently reducing F by 55 % (Fig. 3c). However, the monthly $p\text{CO}_2$ can be increased by a maximum of 35 % (Fig. 4a), reducing $\Delta p\text{CO}_2$ by 231 % (Fig. 4b) and consequently reducing F by 155 % (Fig. 4c). As can be seen from Figs. 3 and 4, the greatest impacts occur in the north east sub-polar regions where bloom activity is greatest. The results from repeating the analysis excluding the years with strong ENSO (1998 and 1999 – maps not included) resulted in comparable spatial distributions and percentages to those shown in Figs. 3 and 4. There were small differences in the upper limits of the percentages. When 1998 and 1999 are excluded the maximum impact on the monthly $\Delta p\text{CO}_2$ is slightly reduced to 215 % and the corresponding maximum reduction in F is 147 %. While the average and maximum impacts are very similar when 1998 and 1999 data are excluded, the spatial regions that exhibit the maximum impacts vary. Figure 5 shows the sensitivity of the maximum (localised) reduction in air-sea CO_2 flux to the selected $\text{CaCO}_3\text{-C}$ concentration. It shows that even a bloom with the lowest $\text{CaCO}_3\text{-C}$ concentration from the literature ($0.015\ \text{mg l}^{-1}$ from $0.2\ \text{pg CaCO}_3\text{-C}$ per coccolith and $75\,000$ coccoliths per ml^{-1}) has the potential to produce a 25 % reduction in the (localised) air-sea CO_2 flux (dependent of course upon the temperature and salinity of the region in question). Referring to Fig. 4 these maximum reductions in the air-sea CO_2 flux occur in the waters around southern Iceland and northern Norway (i.e., those regions that appear red).

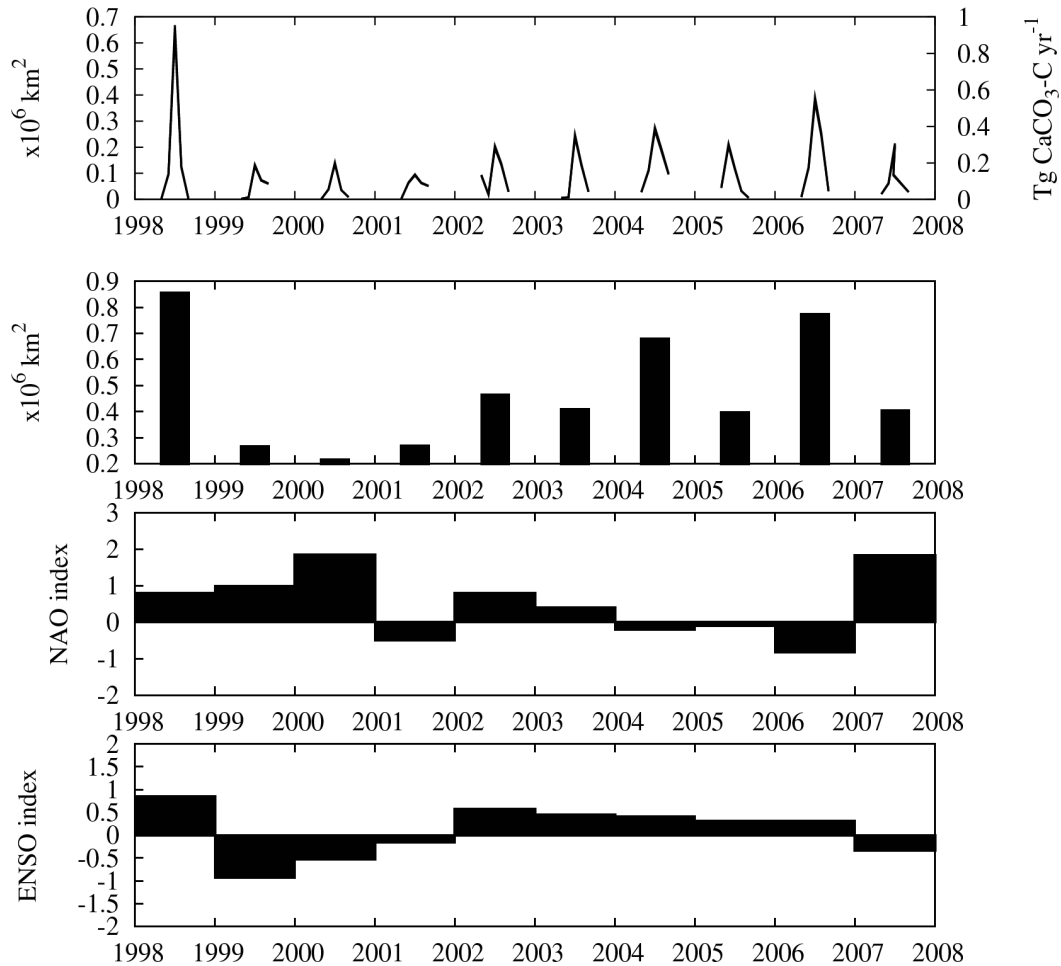


Fig. 2. Time series data (1998–2007). (a) Monthly *Emilia huxleyi* surface coverage and $\text{CaCO}_3\text{-C}$ production for April to August of each year using the mid-range parameter set; (b) yearly total (sum of April to August) of surface coverage for each year; (c) mean NAO index for each year and (d) mean multivariate ENSO index for each year.

4 Discussion

The strong positive correlation found here between total *E. huxleyi* surface coverage and ENSO illustrates climate-driven variability of *E. huxleyi* blooms, although the exact impact and processes of the ENSO acting on the *E. huxleyi* in the N. Atlantic are unclear. Furthermore, this correlation also suggests a linkage between the ENSO and variations in $p\text{CO}_2$ and air-sea CO_2 flux in the N. Atlantic (via the *E. huxleyi*). However, the similar upper value of percentage change in $p\text{CO}_2$ (and, thus, flux) reported for the results excluding the strong ENSO years of 1998 and 1999 show that the large modulation of $p\text{CO}_2$ and flux is not simply a function of strong ENSO variation. Previous work has shown that the ENSO can drive the NAO (Mokhov and Smirnov, 2006) and, thus, impact on the physical conditions in the Atlantic (i.e., temperature, precipitation and storm activity). It has also been shown that $>89\%$ of variation in *E. huxleyi* surface coverage in the N. Atlantic (between 1997–2004) can be

explained by the variations in the physical conditions of solar radiation, mixed layer depth and water temperature (Raitso et al., 2006). Therefore, we suggest that the ENSO is influencing the physics (e.g., heat budget and sea state) in the N. Atlantic and, thus, influencing *E. huxleyi* surface coverage.

Previous studies have found a negative correlation between phytoplankton abundance and the NAO (Boyce et al., 2010; Fromentin and Planque, 1996). Therefore, we suggest that continued monitoring (i.e., to create a longer time series) will increase the significance of the negative correlation found here between the *E. huxleyi* surface coverage and the NAO. The SeaWiFS sensor is no longer in orbit, but alternative and equivalent optical sensors, such as the Moderate Resolution Imaging Spectroradiometer (MODIS), the Visible Infrared Imaging Radiometer Suite (VIIRS) and the soon-to-be launched Ocean Land Colour Instrument (OLCI) could be used to extend the time series analysis into the next decade.

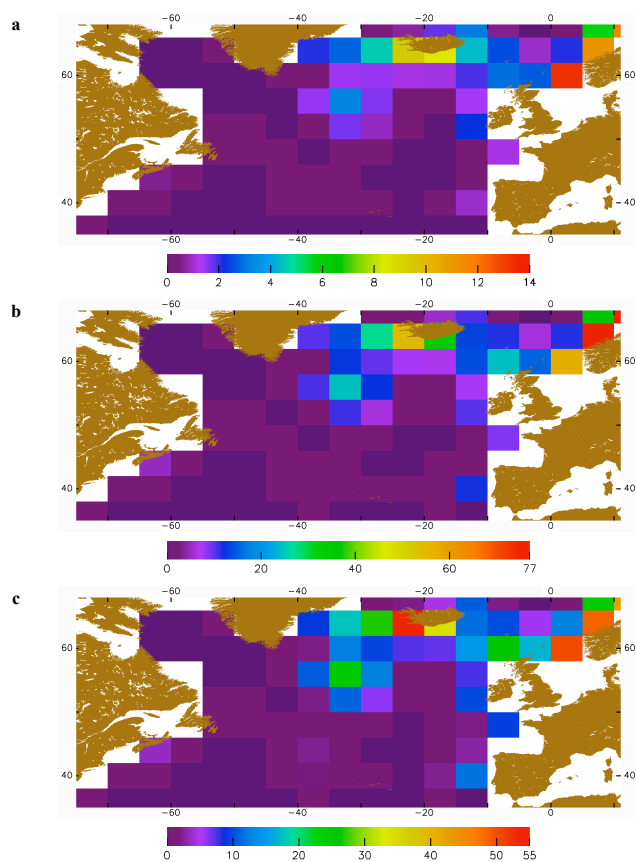


Fig. 3. Spatial distribution of the average impact of *Emiliania huxleyi* on the monthly air-sea CO_2 flux for years 1998–2007. (a) Percentage increase in seawater partial pressure $p\text{CO}_2$ (0–14 %); (b) Percentage decrease in air-water partial pressure difference $\Delta p\text{CO}_2$ (0–77 %); (c) Percentage decrease in air-sea CO_2 flux (0–55 %).

The linear nature of the net $\text{CaCO}_3\text{-C}$ standing stock calculations means that they are highly sensitive to the input values. Doubling the coccolith concentration will result in doubling of the net standing stock estimate. Similarly doubling the coccolith $\text{CaCO}_3\text{-C}$ weight will double the net standing stock estimate. That said, our net $\text{CaCO}_3\text{-C}$ estimates of $0.14\text{--}1.71 \text{ Tg CaCO}_3\text{-C yr}^{-1}$ are comparable to those of (Balch et al., 2005) where they report a euphotic zone aerally integrated value of $1.65 \text{ Tg CaCO}_3\text{-C yr}^{-1}$ for 2002 in the N. Atlantic (sum of Atlantic Subarctic, Gulf Stream and N. Atlantic Drift Longhurst regions from Table 6 of (Balch et al., 2005)). Although our values (and those of Balch et al., 2005) are both lower than the model-derived $150 \text{ Tg CaCO}_3\text{-C yr}^{-1}$ estimated for the year 1990 by an earlier study (Lee, 2001).

It is apparent from Fig. 2a that *E. huxleyi* bloom surface distributions vary in extent between years and so their impact cannot be assumed to be identical each year. We note that it is common practice (as done here) for air-sea exchange studies to exploit climatology datasets for values of $p\text{CO}_2$, under

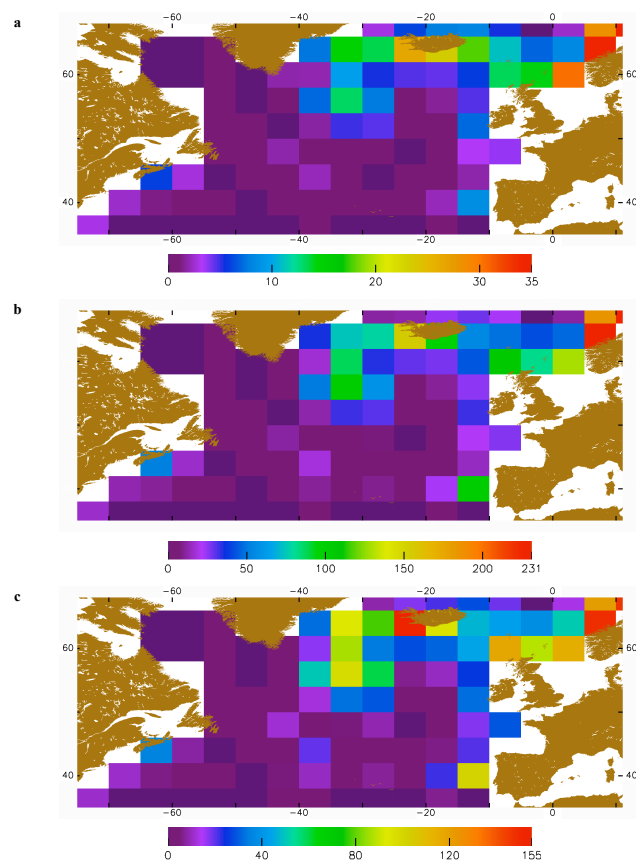


Fig. 4. Spatial distribution of the maximum impact of *Emiliania huxleyi* on the monthly air-sea CO_2 flux for years 1998–2007. (a) Percentage increase in seawater partial pressure $p\text{CO}_2$ (0–35 %); (b) Percentage decrease in air-water partial pressure difference $\Delta p\text{CO}_2$ (0–231 %); (c) Percentage decrease in air-sea CO_2 flux (0–155 %).

the assumption that $p\text{CO}_2$ is changing linearly with time. The high variability of *E. huxleyi* blooms between years in the N. Atlantic, their correlation with the ENSO, their modulation of $p\text{CO}_2$ and the observed increase in *E. huxleyi* activity in some sub-polar regions, suggests that such an assumption can introduce large errors in regional estimates of air-sea CO_2 flux. These blooms are not unique to the N. Atlantic and the influence that these blooms can have upon $p\text{CO}_2$ and, thus, F in other oceanic regions will be dependent on the specific environmental conditions, spatial extent and temporal variability of the blooms. We, therefore, suggest that air-sea CO_2 flux studies that exploit climatological fields of $p\text{CO}_2$ identify any regions that are known to exhibit *E. huxleyi* activity. For the same reasons applying the simplifications detailed in the introduction when modelling the specific regions of the N. Atlantic for short- to medium term predictions has the potential to introduce a large source of error within any estimated air-sea CO_2 fluxes. Reasons for the simplifications within models include a lack of knowledge

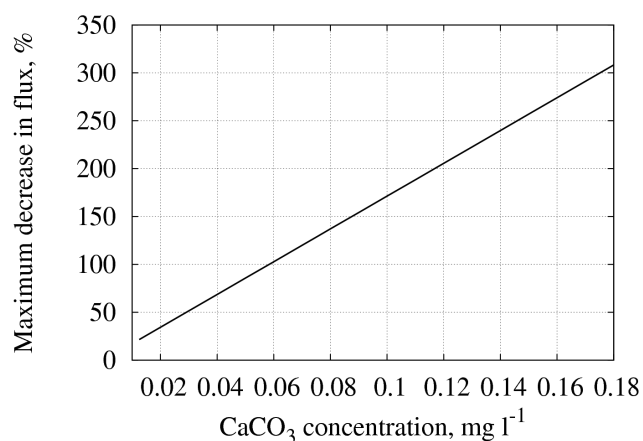


Fig. 5. Sensitivity of the maximum percentage reduction in air-sea flux of CO₂ (across all years, months and regions) to concentration of CaCO₃-C (in mg l⁻¹). Figures 3 and 4 are calculated for a CaCO₃-C concentration of 0.075 mg l⁻¹.

about the biogeochemical and ecological factors that fundamentally govern calcification rates, disparate responses of different calcifying organisms to these factors, and also the lack of appropriate data to evaluate model output at the appropriate scale. Towards this last issue, the advantages of using EO to evaluate ecosystem model output has recently been highlighted (Shutler et al., 2011) and the maps of *E. huxleyi* surface coverage generated within this study would provide a suitable dataset for model evaluation.

The N. Atlantic is an important and variable sink of atmospheric CO₂ (Watson et al., 2009). Figure 1 shows that the *E. huxleyi* are prevalent in the north east Atlantic sub-polar regions. Additionally, Figs. 3 and 4 shows that these blooms are having an impact on the air-sea CO₂ flux in these sub-polar regions. These results combined with the knowledge of the slow equilibration time between the surface ocean and the atmosphere (6 months to a year) suggests that calcifying plankton may be one reason for the unexpected *p*CO₂ concentrations that have been previously reported in the sub-polar N. Atlantic (Lefevre et al., 2004). As a simple illustration, we provide an estimate of the average impact that the coccolithophores can have on the N. Atlantic CO₂ sink. This used the complete 1998–2007 time series of flux estimates, assumed a simplified one month period of existence for the increased seawater *p*CO₂ due to the coccolithophore activity (i.e., no equilibrium over many months) and the methods and data from Takahashi et al. (2009) for calculating the net CO₂ sink. We found that on average using CaCO₃-C concentrations of 0.01–0.18 mg l⁻¹ reduced the annual net sink by ≤ 0.013 Gt C yr⁻¹. For the region studied this is a reduction of the annual net sink of CO₂ of $\leq 4\%$ based on an annual CO₂ sink of 0.352 Gt C yr⁻¹ (as determined from the Takahashi climatology data). However, due to the previously mentioned slow equilibration times between the ocean and the

atmosphere, this simple one month estimate is likely to be very low. This equilibrium time will vary dependent on a number of factors including the location of the bloom, and the sea state and weather conditions in the months following the bloom, all of which can be highly variable. This suggests that the true impact of the coccolithophores on the net sink of CO₂ in the N. Atlantic is likely to be much greater than 4%. To illustrate this point, we repeated the calculation using a six month equilibrium time, using the maximum change in *p*CO₂ within a region, ignoring the impact of multiple blooms within the same region and assuming that the elevated *p*CO₂ remains constant for six months and then returns to its climatological value on month seven. For the region studied the average reduction of the annual net sink of CO₂ was 0.01–0.05 Gt C yr⁻¹. This equates to a 3–14% reduction of the CO₂ sink based on an annual sink of 0.352 Gt C yr⁻¹. The range in values is due to the range in CaCO₃-C concentrations. The maximum reduction (which occurred in 1998, a strong El Niño year) is 0.01–0.1 Gt C yr⁻¹, which equates to a 3–28% reduction in the net sink of CO₂.

In the natural marine environment the *E. huxleyi* cell concentrations will generally gradually decrease at the edge of each bloom. However, as already discussed, the EO *E. huxleyi* detection algorithm (Shutler et al., 2010) classifies areas of ocean into bloom and non-bloom regions. The output is a binary classification defining clear boundaries between regions of bloom and non-bloom. Whilst the spatial component of the classification algorithm is able to detect regions of gradually varying concentrations (through a ramp detector, see Shutler et al., 2010), the spectral component of the algorithm consists of a series of spectral thresholds which are defined to minimise confusion between *E. huxleyi* and other spectrally similar particulates. These conditions set by the spectral model mean that the algorithm is more likely to miss regions of reduced concentrations of *E. huxleyi*, such as those that can exist around the edge of a bloom. Therefore, the estimates of *E. huxleyi* surface coverage and the resultant net CaCO₃-C presented in this study are likely to be underestimates.

This work assumed that the *p*CO₂ climatology data (Takahashi et al., 2009) does not include the impact of coccolithophores. The original publication makes no mention of the effects of calcification and since we have no a priori way of evaluating the effect of coccolithophores on the climatology, we adopt the pragmatic approach of using it as a baseline for our sensitivity study. That said, the climatology has been created from in situ observations and if a bloom existed during or prior to the collection of any in situ data used within the climatology, then the impact of such a bloom could exist within the climatological dataset. It should also be noted that this work is primarily concerned with characterising the range in sensitivity of the air-sea CO₂ fluxes to populations of the calcifying plankton *E. huxleyi*. The use of the SeaWiFS (binary) coccolithophore areal maps allowed the regional uncertainties to be assessed across the complete EO time series

using the largest in situ phytoplankton database currently in existence (the CPR data). For these reasons this study relied upon $\text{CaCO}_3\text{-C}$ standing stock calculations. An equally valid approach could use the particulate inorganic carbon (PIC) data product as developed for the MODIS sensor (Balch et al., 2005) which would allow variations in $\text{CaCO}_3\text{-C}$ to be accounted for. However, robustly assessing the regional uncertainties (across the complete time series) of such a dataset is likely to be more problematic as the CPR data appear unsuitable.

Whilst this study has focussed on the open ocean (>200 m depth), coccolithophore blooms are also known to occur in shallower water and their impact on shelf sea, shelf edge and coastal carbonate systems has been documented in a number of in situ studies (Harlay et al., 2010; Suykens et al., 2010; Buitenhuis et al., 1996; Purdie and Finch, 1994) and their spatial impact on the air-sea CO_2 fluxes in these regions has yet to be analysed.

5 Conclusions

The results presented in this study show a 10-year time series study of *E. huxleyi* bloom surface distributions in the N. Atlantic, estimated to a precision of $\sim 22\%$. There is a large inter-annual variation in surface area of satellite-detected *E. huxleyi* blooms in the N. Atlantic and this variability is positively correlated with the ENSO. Using climatology data, the time series of EO data, and the seacarb package, we have evaluated the impact that these *E. huxleyi* blooms can have on $p\text{CO}_2$ in the surface water. They have the ability to increase $p\text{CO}_2$, which in turn reduces the air-sea flux; this reduction in the flux can exist for six or more months after the bloom has dispersed and we estimate that it has the potential to reduce the N. Atlantic net sink of CO_2 by between 3–28%. The 10-year times series has illustrated the widespread impact that these calcifying plankton can have on the air-sea flux of CO_2 in the N. Atlantic. The work has also shown that these effects are likely to be more widespread during years that exhibit a strong positive ENSO signal. Our analysis highlights the need for the continued study and monitoring of these phytoplankton if we are to fully understand the inter-annual variability of the N. Atlantic sink of CO_2 .

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