Seasonal and interannual variability of coccolithophore blooms in the North East-Atlantic Ocean from a 18-year time-series of satellite water-leaving radiance

Laurie Perrot^{1,2}, Francis Gohin¹, Diana Ruiz-Pino², Luis Lampert¹

¹IFREMER/DYNECO/PELAGOS Centre Ifremer de Brest, Plouzané, 29280, France

²Laboratoire LOCEAN-CNRS/IPSL, Université de Paris VI, Paris, 75005, France

Correspondence to: L.Perrot (laurie.perrot@ifremer.fr)

Abstract. Evaluate the impact of anthropogenic CO₂ uptake and acidification on the most abundant calcareous phytoplankton, coccolithophores, requires a better knowledge of the temporal and spatial evolution of their blooms. Here we determine, from satellite radiance, the seasonal and interannual variability of coccolithophore blooms for 18 years (1998 to 2015) across the North-East Atlantic region covering the Bay of Biscay and the Celtic Sea. The identification of coccolithophores is carried out using a modified version of the fuzzy method developed by Moore (2009) applied to a time series of SeaWiFS (1998-2003) and MODIS (2003-2015) spectral radiance. After identification of the coccolith pixels, the abundance of coccoliths is assessed from a data base of non-algal Suspended Particulate Matter (SPM), a product initially developed for estimating the content of mineral particles, mainly due to resuspension effects, in coastal waters. The results, in terms of identification and quantification, are consistent with *in situ* observations in the area and with those of algorithms addressing coccolithophore blooms at global scale (CALCITE and PHYSAT). Although a regular pattern in the phenology of the blooms is observed, starting south in April in Biscay and moving northwards until July near Ireland, there is a high seasonal and interannual variability in the extent of the blooms. Year 2014 shows very low concentrations of coccoliths from space (twice less than average) and anomalies point out the maximum level for 2001. The cause of the seasonal and interannual variability of the coccolithophores blooms in this Atlantic region remains an open question.

Keywords: Coccolithophore blooms, Coccoliths, Suspended particulate matter, Ocean Colour, Bay of Biscay, Celtic sea

1 Introduction

Coccolithophores belong to the nanophytoplankton size-class in the *Prymnesiophyceae* algae class. Coccolithophores produce CaCO₃ scales called coccoliths which form the «shell», the coccosphere of the algae cell. Coccoliths are in the size range of a few µm and can also be detached from the cell in the water. On a global scale coccolithophore blooms are studied with regard to CaCO₃ and dimethylsulfide (DMS) production, phytoplankton community structure, and their key role in the ocean carbon flux (Brown and Yoder, 1994). Carbon and Calcite storage as well as CO₂ fluxes produced by coccolithophores

have also climatic feedbacks. This primary producer pumps dissolved CO₂ of the ocean through photosynthesis and produces CO₂ through calcification in coccoliths and creates Calcite that sinks to the ocean bed (Sabine et al., 2004).

Contradictory assumptions have currently been pointed out regarding the evolution of coccolithophore blooms during the anthropogenic era. According to the hypothesis of increasing acidification (pH decrease due to CO₂ increase) of the upper ocean layers (Orr et al., 2005), the calcification, development and extent of the coccolithophore blooms could be positively or negatively impacted (Doney et al., 2009; Beaufort et al., 2011). However, using data from the Continuous Plankton Recorder, Rivero-Calle et al. (2015) showed that coccolithophore occurrence in the North Atlantic increased from about 2% to more than 20% from 1965 through 2010. There is also evidence from plankton samples, sediment traps, and satellite imagery that the coccolithophore *Emiliania huxleyi* is expanding its range poleward in both hemispheres over the last two decades (Winter et al., 2014).

Several studies, from both in situ and satellite data evidenced the occurrence of intense and large blooms of coccolithophores on our studied area (Fig. 1) located on the margin of the North East Atlantic shelf (Holligan et al., 1983; Tyrell and Merico, 2004; Morozov et al., 2013; Harlay et al. 2010; Poulton et al., 2014). As soon as 1978, with the launch of the Coastal Zone Color Scanner (CZCS), *E. huxleyi* blooms were observed from ocean colour sensors along the outer margin of the north-west European shelf between 40°N and 60°N (Holligan et al., 1983). The spring and summer phytoplankton blooms, including coccolithophore, in the Bay of Biscay and the Western English Channel were investigated by Garcia-Soto and Pingree (2009) using remote-sensing data and in situ measurement obtained along a ferry-line. Harlay et al. (2010) described from in situ and remote-sensing data different stages of a coccolithophore bloom observed on the Northern Bay of Biscay in June 2004. A similar work was done by Van Oostende et al. (2012) from pigment signatures observed in May and June 2006-2008 on the continental margin of the Celtic Sea. Coccolithophore blooms, represented mainly by *E. Huxleyi*, following a diatom bloom occurring as soon as April in the Bay of Biscay (Lampert et al., 2002) and later, in May and June, on the margin of the Celtic Sea. Coccolithophore blooms have also been observed at the end of June and in July on the continental shelf of the Western English Channel (Smyth et al., 2002; Garcia-Soto and Pingree, 2009). In these cases, patches of the dinoflagellate, *Karenia mikimotoi* are sometimes associated to coccolithophore blooms, but these latter do not occur in this area as regularly as on the shelf break. In order to reduce uncertainties on the long-term evolution of coccolithophore blooms determined from satellite data in the eastern North Atlantic, it is necessary to use an algorithm allowing to : i- separate the spectral signal of coccolithophores from the suspended particulate matter and from other phytoplankton group; ii- do not detect coccolithophore bloom from chlorophyll, as this phytoplankton produced generally a low chlorophyll (<1µg L⁻¹, Tyrrell and Merico, 2004).

To investigate the spatial and temporal (seasonal and interannual) evolution of the blooms, four sub-areas were defined near the 200m-contour of the shelf break (Fig. 1a). These sub-regions are mainly waters optically classified as Case-1; which

means that there is only a marginal effect of resuspended sediments, river outputs, yellow substances of terrestrial origin on the marine radiance. Across this zone, owing to strong backscattering at all wavelengths by detached coccoliths, coccolithophore blooms appear distinctively on the MODIS RGB (Red-Blue-Green) image in spring (25 April 2013, Fig. 1b). This bloom is also visually depicted on the image of the non-algal Suspended Particulate Matter (SPM) derived from the IFREMER procedure (Gohin et al., 2005) applied to MODIS radiance (Fig. 1c), and on the image of calcite (Fig. 1d) obtained from the NASA algorithm (Balch et al., 2005).

The non-algal SPM products, provided by the European project Copernicus Marine Environment Monitoring Service (CMEMS), have been extensively used on the studied area for different purposes, such as sediment model validation (Sykes and Barciela, 2012), to evaluate the impact of climate (waves, solar irradiance) on the availability of light in the water column and its consequences on phytoplankton development (Capuzzo et al., 2013; Gohin et al., 2015) or to investigate the effect of the environment on the development of the Pacific oyster *Crassostrea gigas* along the European coast (Thomas et al., 2016). As the calcite plates of the coccolithophores act optically similarly to non-algal SPM, SPM imagery can provide a useful dataset to investigate the variability of coccolithophore blooms. Coccoliths appear clearly on non-algal SPM images but, due to their mean mass-specific backscattering coefficient higher than the particles generally encountered in coastal waters, their mass is likely to be overestimated by the SPM algorithm. Nevertheless, the signal is very stable in time and the method has proven to be quite robust in providing non-algal SPM or turbidity in coastal waters (Gohin, 2011; Binti Jafar Sidik et al., in revision). In the SPM procedure, the main hypotheses concern the mass-specific backscattering coefficients at 550 and 670 nm. There are three major types of particles whose scattering properties may diverge by nature from the standard ones directly related to pure waters and phytoplankton. Ranked by their increasing scattering cross sections, large aggregated flocs occurring after the phytoplankton blooms in summer, mineral particles in winter, and detached coccoliths contribute to under or over-estimate non-algal SPM when marine radiance is processed through a standard procedure. There is a need for investigating the effect of coccolithophores on the non-algal SPM product.

The reference product for calcite derived from space, to which will be compared mineral SPM, is the PIC (Particulate Inorganic Carbon) in mole m^{-3} provided by NASA as part of the standard Level-2 Ocean Color product suite. The PIC concentration (related to the calcite of coccolithophores) is obtained from a two-band method (Balch et al., 2005) in low to moderate concentration switching to a three-band method based on longer wavelengths (Gordon et al., 2001) in highly reflective waters (high concentration). This product covers all pixels, including pixels located on the continental shelf where resuspensions increase the reflectance, similarly to coccoliths, and a coccolithophore flag or a discriminating method is required for classifying the blooms. To that purpose, several methods are available. They all take benefit of the scattering properties of the detached coccoliths. A coccolithophore flag is provided by NASA using a modified version of the procedure defined by Brown and Yoder (1994). Other methods investigate anomaly detection on a time series of images (Shutler et al., 2010) or take into account the inherent properties of coccoliths (Smyth et al., 2002). Moore et al. (2012) developed an identification method extending a classifying procedure of eight major Ocean Water Types encountered globally in remote-sensing. This procedure is based on a fuzzy clustering algorithm applied to the reflectance spectra (Moore et al., 2009). The

fuzzy procedure is selected because, as noted by Moore et al. (2012), this method frees the constraint of specific wavelengths for identifying coccoliths or any other optical water type; hence it has been used with success for distinguishing two dinophyceae blooms producing high biomass on the continental shelf of the Bay of Biscay in summer (Sourisseau et al., 2016).

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By adapting the fuzzy method to a spectra dataset selected on coccolithophore blooms observed in the North-East Atlantic, the variability of their blooms across 4 selected regions (Fig. 1a) will be investigated from 1998 to 2015. Thus, on the one hand, we will have a better understanding of the effect of coccolithophores on the non-algal SPM product, and, on the other hand, we will determine the seasonal and interannual variability of coccolithophore blooms on the margin of the North-East Atlantic continental shelf.

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2 Data and methods

2.1 Satellite data

SeaWiFS (1998-2003) completed by MODIS-Aqua data (2003-2015) have been used. Despite some differences in their wavelengths, SeaWiFS and MODIS provide similar products. Normalised-leaving radiance L2 products (MODIS and SeaWiFS) and Level 3 images (SPM and Calcite) have been used. Both data sets were projected on a common grid at a resolution of about 1.1 km². The semi-analytical model used to retrieve non-algal SPM from satellite reflectance is described in Gohin et al. (2005) and Gohin (2011). Non-algal SPM (NA-SPM hereafter) is estimated from radiance at 550 nm and 670 nm after a preliminary estimation of the Chlorophyll-a concentration. Depending on the NA-SPM level retrieved, the final NA-SPM is chosen at 550 nm if both SPM (at 555nm and 670nm) are less than 4 gm⁻³. This is the general case in relatively clear waters. In other cases it is NA-SPM(670) that is kept. We proceed in two-step for estimating NA-SPM at 555nm and 670nm. In the first step an intermediate reflectance term R' (Eq. (1)) is estimated from the normalised water-leaving radiance nLw.

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$$R' = \alpha_0 + \alpha_1 nLw \quad (1)$$

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Here α_0 and α_1 are two constants defined for each wavelength (555 and 670 nm).

The intermediate reflectance term R' is related to the backscattering and absorption coefficients by

$$R' = \frac{b_b}{a + b_b} \quad (2)$$

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In equation 2, a and b_b are the absorption and backscattering coefficients (wavelength-dependant). These coefficients can be expressed from the concentration of Chl and SPM by equation (3).

$$b_b = b_{bw} + b_{bChl} * Chl + b_{bp} * NA_SPM \quad \text{and} \quad a = a_{aw} + a_{aChl} * Chl + a_{ap} * NA_SPM \quad (3)$$

Where the * quantities represent the mass-specific IOPs (of Chl and SPM) ; w indicates pure water. A specific contribution of coloured dissolved organic material (CDOM) to absorption is ignored in this algorithm.

10 After making these substitutions, the NA_SPM concentration is then obtained by inverting equation 2:

$$NA_SPM = \frac{R'[\alpha_w + b_w + (a_{Chl} * + b_{bChl} *)Chl] - [b_{bw} + b_{bChl} * Chl]}{b_{bp} * -(\alpha_p * + b_{bp1} *)R'} \quad (4)$$

The constants α_0 and have been obtained by minimization of the variance of the errors derived from Eq. (4) applied to a data set of coastal SPM and satellite reflectance spectra, see Gohin et al. (2005) for details. $b_{bs}^*[550]$ and $b_{bs}^*[670]$ have been arbitrarily set from the literature to 0.0074 and 0.0058 m^2g^{-1} respectively. b_{bChl} and a_{Chl} come also from the literature. They are applied in Eq. (3) to Chl previously obtained from the OC5 algorithm (Gohin, 2002). Corresponding to standard coastal waters, the mass-specific coefficients b_{bs}^* are likely to be lower than those of detached coccoliths. In consequence, SPM could be overestimated in case of coccoliths.

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2.2 The fuzzy method

The first step to apply the fuzzy method is to define the typical spectra of nLw derived from monospecific « coccolithophore » blooms. Figure 1.c shows the SPM concentration for the entire area on April 25th 2013. A coccolithophore bloom is apparent on this image on the continental shelf of the Bay of Biscay, and is confirmed by the RGB MODIS picture (Fig. 1b). The pixels in the bloom area along the shelf-break can be selected for defining a reference data set of spectral radiance on coccoliths.

By selecting radiance from coccolith pixels as described above, we built up a reference SeaWiFS dataset of 361 spectra, from 412 to 670 nm over the period 1998-2002, and a MODIS dataset of 2343 spectra for the period 2003-2015. The means of these spectral radiance datasets are considered as the characteristic coccolith spectra.

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The next step concerns the application of the fuzzy method to every MODIS and SeaWiFS pixel of the area in order to distinguish coccoliths from other components in the water. This is done by comparing each local nLw spectrum with the reference spectrum of coccoliths. The fuzzy index, defined in Moore et al. (2012), is based on the Mahalanobis distance Z (Rencher et al, 1995) as described in Eq. (5) taking into account the covariance matrix V and the vector of mean values (μ) from the spectra database (nLw).

$$Z^2 = (\overrightarrow{nLw} - \vec{\mu})^t V^{-1} (\overrightarrow{nLw} - \vec{\mu}) \quad (5)$$

Vector (μ) is the reference coccolith spectrum. In Moore et al (2012), the likelihood that a radiance spectrum belongs to the coccolithophore class is defined in Eq. (6):

$$1 - F_n(Z^2) \quad (6)$$

$F_n(Z^2)$ is a cumulative χ^2 with 6 degrees of freedom corresponding to the 6 wavelengths available (412, 443, 490, 510, 550 and 670 nm) for SeaWiFS and for MODIS (412, 443, 488, 532, 555 and 667 nm). Equation (6) gives the fuzzy index whose values are included within [0-1] range.

The fuzzy index describes the proximity between the spectrum of each pixel in regard to the reference spectrum of a coccolithophore bloom: the higher the fuzzy index is close to 1, the higher the probability of pixel to be a coccolith bloom is. Fuzzy indices are calculated for all the pixels of daily images between April to August from 1998 to 2015 on the full area and on the four sub-areas indicated in red squares (Fig. 1a). A threshold is applied to the fuzzy index for selecting coccolith pixels in the SPM signal giving an evaluation of the coccolith mass (SPM_{fc} hereafter). The threshold (0.4 here) is chosen to select pixels with a sufficient probability.

3 Results

3.1 Applying the fuzzy method to SeaWiFS and MODIS radiance

3.1.1 The reference spectra for applying the fuzzy method

The selected radiance (nLw) spectra for SeaWiFS are normalised by the normalised water-leaving radiance at 490 nm. The criterion $nLw(490) > nLw(550)$ is applied to the spectra for distinguishing the coccolith pixels. This criterion is based on the general shape of the coccolith spectra.

This selection provides a database of spectra (Fig. 2a) for SeaWiFS. The red line shows the characteristic spectral shape of coccoliths for SeaWiFS, which is used as the spectral reference for the fuzzy method. The green lines indicate the standard deviations for the SeaWiFS (Fig. 2a) spectra database.

Radiance spectra selected from the MODIS database are normalised by radiance at 488 nm with the criterion $nLw(488) > nLw(550)$. This criterion is applied similarly to what has been done for SeaWiFS spectra, considering that 488 nm is close to 490 nm. This selection provides a MODIS database of spectra with the characteristic spectrum of coccoliths in red in Fig. 2b and considered as the reference for calculating the fuzzy index.

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3.1.2 Sensitivity test

Figure 3 illustrates the application of the fuzzy index in the case of 25 April 2013 (Fig. 3a), allowing the selection of pixels of coccoliths on SPM image (Fig. 3b). Two sensitivity test were performed. The first sensitivity test was performed on the threshold criterion of the fuzzy index used to discriminate pixels of coccoliths. The test was done on the number of coccolith pixels on 25 April 2013 (Fig. 4a, blue line) resulting from thresholds varying between 0.1 and 0.9. As expected, the number of pixels reached maximum ($15 \cdot 10^5$ pixels of coccoliths) for 0.1, and the highest value of the threshold (0.9) was the most discriminative. The threshold of 0.4, which corresponded well with the mean number of coccolith pixels detected (Fig. 4a, black line), was chosen as a good compromise. Moreover, the fuzzy index showed a low variability between 0.3 and 0.5. The second sensitivity test was carried out on the number of radiance spectra used to build up the radiance database. Figure 4b shows the resulting number of coccolith pixels after the application of the fuzzy method for 25 April 2013, depending on the number of spectra in the radiance database. The sum of coccolith pixels is close to 0 for a number of spectra below 1290 in input (Fig. 4b). The maximum value of the number of coccolith pixels ($\sim 5 \cdot 10^5$ pixels) was reached for 1935 spectra in input and remained stable up to 2343 spectra. In consequence, it is the set of 2343 spectra that will be used as input in the method.

3.1.3 SeaWiFS and MODIS continuity in the products

To analyse the variability over the 18-year period, a good agreement was required between the results of the fuzzy method applied to SeaWiFS (1998-2003) and MODIS (2003-2015) radiance. To check the consistency of the two sensors, the SPM_{fc} budget ($mg\ L^{-1}$) in 2003, a common year between MODIS and SeaWiFS was considered. The correlation between the daily sums of SPM_{fc} derived from SeaWiFS and MODIS for the season considered in this study (April-August 2003) indicates a $r^2=0.89$. This good agreement suggests that it is possible to use time series as continuous over the period 1998-2015.

3.2 Analysis of the 18-year time-series

Coccolith pixels were estimated from daily radiance on the full area and for 4 sub-areas (A,B,C,D zones in Fig. 1a.), from 1998 to 2015, focusing on the major season of the coccolith blooms, from April to August. These blooms were quantified by the number of coccolith pixels and by their SPM_{fc} concentration, which allowed us to investigate the spatial extent and the concentration budget of the blooms at seasonal and interannual scales.

3.2.1 Seasonal variability

After the application of the fuzzy method, the monthly means of coccoliths provided an overview of the blooms between 1998 and 2014 (Fig. 5) from April to June. Climatologies (Fig. 5a,b,c) show the seasonal variability between 1998 and 2014, which are expressed with a factor 10 for concentrations to make the monthly variability clearer. The monthly average maps of coccoliths show that blooms generally start in April in the southern part of the Bay of Biscay (Fig. 5a) between 45°N and 47°N. In May they extend along the shelf-break, with stronger concentrations in the Northern part of Bay of Biscay between 45°N and 47.5°N and in the Celtic zone between 47.5°N and 48.5°N (Fig. 5b). Blooms move northward in June in the Celtic sea area and in the Irish zone between 49.5°N and 53.5°N (Fig. 5c). Coccolith bloom starts in 2013 between 44°N and 48°N in the Bay of Biscay (Fig. 5d) and moves northward along the margin of the shelf in May between 45°N and 52°N (Fig. 5e), to the Celtic sea and southwest of Ireland between 48°N and 53.5°N in June (Fig. 5f). The variability of the monthly averages in 2013 is close to the climatological variability (Fig. 5a,b,c) whereas the year 2014 is particular as it shows a very weak signal during the 3 months (Fig. 5g,h,i), with a low concentration of SPM_{fc} that does not exceed 2 mg L^{-1} in June, between 47.5°N and 48.5°N (Fig. 5i).

Biweekly climatologies of the coccolith pixels were built up for each sub-area (Fig. 6a and Fig. 6b) and for the full area (Fig. 6c). Climatologies of the 4 sub-areas confirm a northward progression of maximum coccolith blooms from the Bay of Biscay in April to Ireland in June. Considering the number of coccolith pixels, the climatologies of Fig. 6a show that the surface maximum occurs in both the South (6000 pixels) and North areas (10000 pixels) in the Bay of Biscay (red and blue line) in the second fortnight of May. In the Northern sub-areas, the sums of coccolith pixels reach the maximum in the first fortnight of June (Fig. 6a) for both central Celtic Sea (13000 pixels) and Ireland (15000 pixels). The same seasonal scheme is observed on the climatologies of the budget of coccoliths (SPM_{fc}) (mg L^{-1} by pixels) between the 4 sub-areas (Fig. 6b). The maximum in the second fortnight of May for the Celtic zone shows a two fold higher budget ($4.1 \cdot 10^4 \text{ mg L}^{-1}$ by pixels) than in the Bay of Biscay. For both climatologies (number of coccolith pixels and SPM_{fc} budget), the peaks occur in the second fortnight of May for the Southern areas (Bay of Biscay) and in the first fortnight of June for the Ireland sub-area. Only the Celtic Sea sub-area shows a time-lag between the climatologies of the number of coccolith pixels and their budget. Indeed, the bloom intensity reaches its maximum value two weeks before the maximum bloom extent, whereas in the North and South Bay of Biscay and Ireland sub-areas, the maximum bloom development (SPM_{fc} on Fig. 6b) and the maximum extent are concomitant (Fig. 6a).

In the full area, climatologies show that the number of coccolith pixels exceeds about +17% the mean value ($1.1 \cdot 10^5$ pixels) from the second April fortnight (Fig. 6c), and maximum occurs during the second fortnight of May ($3 \cdot 10^5$ pixels). The number of coccolith pixels declines below the mean value in the second fortnight of June (Fig. 6c). This temporal evolution suggests a northward evolution of the blooms with the season, in agreement with the general evolution of the environmental conditions during summer in the full zone. Warmer temperatures, higher solar irradiance and stratification in summer should enhance the development of coccolithophore blooms (Thierstein et al., 2004).

3.2.2 Interannual variability

The mean bi-weekly number of coccolith pixels over the period 1998-2015 is shown in Fig. 7a. The time-series anomalies were calculated from the differences between biweekly levels and climatologies, and then normalised by the percentage of cloud-free pixels of each sub-area (Fig. 7b). Slopes in the timing of the blooms were also calculated between the beginning and the peak of blooms for each sub-area. Fortnight anomalies point out the maximum levels for 2001 and the lowest levels for 2014. 2001 shows the largest blooms (Fig. 7a) and three synchronised peaks in the southern Bay of Biscay ($6.2 \cdot 10^4$ pixels), in the Celtic sub-area ($5.7 \cdot 10^4$ pixels) and in the Ireland sub-area ($1.8 \cdot 10^4$ pixels). The 2001 event is the only bloom with a positive anomaly in the time-series for the 3 sub-areas (Fig. 7b), in the Southern Bay of Biscay (anomaly of $+7 \cdot 10^4$ pixels), in the Celtic area ($+6 \cdot 10^4$ pixels) and in the Ireland sub-area ($+1.3 \cdot 10^4$ pixels). At the contrary, 2014 is the only year when the lowest bloom signal occurs in all sub-areas, with a maximum of $6.9 \cdot 10^2$ pixels in the Ireland zone (Fig. 7a). The bloom extent in 2014 is below average in all 4 sub-areas (Fig. 7b), for the Southern (anomaly of $-1.1 \cdot 10^4$ pixels) and Northern Bay of Biscay ($-1.2 \cdot 10^4$ pixels), in the Celtic zone ($-1.8 \cdot 10^4$ pixels) and for the Ireland sub-area ($-2 \cdot 10^4$ pixels). Negative anomalies are also strong for 2015, except for the North of Biscay where a bloom is visible on Fig. 7a, earlier than climatologies. Some periodicities can be noticed for the South of Biscay where anomalies reach a positive maximum every 4 years in 2001, 2005, 2009 and 2013. For Ireland sub-area, there is a time-lag of 7 years between maximum positive anomalies reached in 2000 and 2007, and for minimum negative anomalies in 2004-2005 and 2011-2012.

The strong bloom observed in 2001 in all sub-areas is also visible in the full area (Fig. 8a), as the peak of the bloom reaches the highest value of the time-series (10^6 coccolith pixels) for this year. 2014 and 2015 show the lowest values of the time-series, with a signal about twice lower than the average ($1.07 \cdot 10^5$ pixels) for 2014. 2015 shows a number of coccolith pixels twice higher than in 2014 but which still remains lower than average. To check the effect of the number of cloud-free pixels on the coccolith budgets, the time-series of clear pixel numbers (Fig. 8b) was used to normalise the number of coccolith pixels (Fig. 8c). A maximum value is observed for the year 2001 (ratio= $3.6 \cdot 10^6$), similarly to what is observed in the series not weighted by the number of clear pixels. The lowest values of the time-series normalised are also observed in 2014 and 2015. Figure 8b shows a lower number of clear pixels for 2015 than for 2014, and the normalised time-series in 2015 shows a raise of 5.5 compared to the non-normalised time-series (Fig. 8a) whereas 2014 shows a raise of 4.5, which suggests a larger cover of clouds in 2015, and thus a lack of observation. Nevertheless, this cover of clouds remains close to the mean of the time-series which suggests low dynamics of blooms in 2015 as in 2014. On both time-series of coccolith pixels and normalised time-series, 2009 and 2013 show a double peak between April and August, which can be linked to a first early bloom in the South of Biscay (Fig. 7) followed by a second bloom in the Irish sub-area.

4 Discussion

4.1 Comparison to in situ data: application to an April case in the Bay of Biscay

A coccolithophore bloom of *Emiliana huxleyi* was reported from pigments and taxonomic data on April 24th 1998 during the Bio-Modycot 98 cruise in the Bay of Biscay (Lampert et al, 2002). 47 stations were sampled from 22 to 27 April 1998 (Fig. 9a) and coccolith and coccolithophore counts were performed in a restricted number (33) of samples.

The bloom is observed on the SPM_{fc} regional distribution of 27 April 1998, after applying the fuzzy method (Fig. 9b). The comparison between SPM_{fc} and in situ data was carried out between 24 April and 28 April. SPM_{fc} distribution (Fig. 9b) shows a bloom between 45°N/47°N and 1.5°W/5°W, where the concentration is higher than 4 mg L⁻¹. Moreover, Figure 9c shows for eight stations the quantitative comparison of the coccolith count (blue line), the coccosphere count (pink line) and SPM_{fc} from the fuzzy method (red line) between 22 April and 26 April. To compare the concentration calculated by the fuzzy method with insitu data, it is necessary to know the concentration in terms of mg which separately represent coccoliths and coccospheres. According to the hypothesis of a mean coccosphere weight of 60 (cf Beaufort et al., 2011) ± 10 pg and a coccolith weight of 15 ± 5 pg, the biomass of total coccolithophore ([cocco]) was estimated by Eq. (7) :

$$[cocco](mgL^{-1}) = (coccosphere \bar{m}_{coccosphere})1000 + (coccoliths \bar{m}_{coccoliths})1000 \quad (7)$$

with m_{cocco} the mean coccosphere weight and $m_{coccoliths}$ the mean coccolith weight.

SPM_{fc} concentration for the 8 stations fits better with coccolith data than coccosphere data (Fig. 9c). Indeed, SPM_{fc} concentration reaches a value of 1.5 mg L⁻¹ ± 0.6 mg L⁻¹ and coccolith in situ data a concentration of 1.3 mg L⁻¹ ± 0.5 mg L⁻¹, whereas coccospheres reach a lower concentration of 0.2 mg L⁻¹ ± 0.15 mg L⁻¹. The same pattern is observed for the other 7 stations (23, 24, 25 and 26 April). It is worth noticing that for a few stations (6, 14 and 39), there is a time-lag of 2 days between in situ data and SPM_{fc} pixels due to cloud coverage, but the comparison remains consistent as the bloom remains in the area until the 27 April (cf Lampert et al, 2002).

4.2 SPM concentration versus Calcite concentration from NASA algorithm

Coccolithophore blooms are commonly quantified from space through the PIC product provided by NASA (Fig. 1d). Two Calcite algorithms are currently applied. One is a Look-Up-Table (LUT) based on water-leaving radiance at 443 and 550 nm and is referred to as the 2-band algorithm (Balch et al., 2005). The second, the '3-band' method is a semi-analytical algorithm based on radiance in the red and Near-Infra-Red bands (670, 765 and 865 nm for SeaWiFS) to estimate backscattering at 546 nm (Gordon et al., 2001). The 3-band method is applied when the

2-band algorithm fails, generally in high turbidity. The 2-band method returns the backscattering coefficient $b_b[546]$ and the concentration of calcite is estimated by dividing $b_b[546]$, by an a priori-determined calcite-specific backscattering coefficient.

A comparison between mineral SPM and Calcite products was performed on 25 April 2013. Fig.10.a shows the scatterplot of SPM versus Calcite, applied to all valid ocean pixels without any restriction, corresponding to the MODIS image of April 25 2013 shown on Fig.1.b-c. SPM concentration was highly correlated to calcite concentrations with $r^2=0.92$. The ratio of SPM versus calcite is around 4 over the coccolithophore bloom (Fig10.b and 10.c). This ratio is also observed in many situations (not shown).

A part of the difference between both quantities is explained by the mass-specific backscattering coefficients used in the two methods.

- For Calcite, the backscattering coefficient of coccolithophore and detached coccoliths at 546 nm can be approached by the following formulation (Balch et al., 1999):

$$b_b(546) \text{ (m}^{-1}\text{)} = 1.63 \cdot (\text{Nb Moles of Calcite m}^{-3}\text{)} = 1.63 \cdot (\text{Calcite mass}/100.09) = 0.01623 \cdot \text{Calcite (in mg L}^{-1}\text{)} \quad (8)$$

- For SPM the backscattering coefficient is expressed by: $b_b(546) = 0.0074 \cdot \text{SPM (in mg L}^{-1}\text{)}$ (9)

From Eq. (8) and Eq. (9), a ratio of about 2.2 is expected between SPM and the Calcite mass to obtain equivalent backscattering coefficients. However, many approximations are made to come to such a conclusion. First of all, the mass-specific backscattering coefficients, 0.0074 and 0.0058 $\text{m}^2 \text{ g}^{-1}$ for the green and red bands respectively, used in the SPM algorithm are only intermediate parameters in the equations coming from the literature (only parameters α_0 and α_1 of equation have been adjusted by comparison to an in situ dataset). Secondly, the variability of the backscattering by coccospheres and coccoliths may be high (Balch et al., 1996). Fig.10.b shows the scatterplot for the fuzzy-selected coccolith pixels and Fig. 10c the scatterplot for the coccolith pixels corresponding to the full month of April 2013. Fig.10b-c shows the effect of the switch between SPM(555) for low turbidity to SPM(670) for moderate to high turbidity on coccolith-selected pixels. This switch, despite expected negative consequences in the continuity of the SPM retrievals, is justified by the fact that the reflectance at 555 nm saturates when turbidity increases (Gohin, 2011). Binti Jafar Sidik et al. (in revision) showed, from a large dataset of coastal measurements, that SPM(555) is adapted to summer turbidity when large aggregates, with low mass-specific backscattering coefficient, dominate, whereas SPM(670) is suited to winter mineral particles. Difficult to put in evidence from comparisons to coastal data observed in different water types and seasons, hence addressing different type of particles, the switch has a dramatic effect on SPM retrieved over a large area of homogeneous waters where coccolithophores at medium concentration govern the optical properties. However, it is clear from the good linearity of the relationship between SPM(550) and SPM(670) to Calcite concentration that a simple modification of the SPM algorithm with suppression of the switch (or displacement to much higher values) and an increase in the mass-specific backscattering coefficient could lead to SPM estimations on coccolithophores very similar to those derived from the Calcite algorithm. This needs a modification of the SPM product: which could be the scope of a future work. The SPM product has been designed

for forcing light through the water column for biochemical modelling, excluding from the satellite retrievals any residual effect of the bloom itself, (Gohin et al., 2005). Therefore, ideally, the coccolith signal should be suppressed from the SPM product.

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4.3 The fuzzy method versus PHYSAT

The fuzzy method can also be compared to another bio-optical algorithm such as PHYSAT (Alvain et al, 2008) that discriminates qualitatively 6 different classes of phytoplankton (diatoms, nanoeucaryotes, prochlorococcus, synechococcus-like/cyanobacteria, phaeocystis and coccolithophores) provided through monthly products. This method is based on the identification of specific optical signatures attributed to different phytoplankton groups, including coccolithophores, from SeaWiFS radiance data (between 412 and 550 nm). Figure 11 shows the area of the coccolithophore blooms estimated by the fuzzy method (red line) and PHYSAT (blue line) between 1998 and 2003, from April to August. The areas of coccolithophore blooms derived from PHYSAT and from the fuzzy method show the same evolution between 1998 and 2003. The largest area is observed in 2001 for both methods, with a bloom area of $7.85 \cdot 10^4 \text{ km}^2$ for May 2001 for PHYSAT and $1.11 \cdot 10^5 \text{ km}^2$ for the fuzzy method. The area estimated from the fuzzy method is 2.1 times higher than the PHYSAT area. The difference between these two methods may be due to the fact that PHYSAT aims to discern 6 phytoplankton groups by the dominant group classification and according by focuses on the peak of blooms. It must also be noted that PHYSAT makes use of L3 SeaWiFS level data with a high-reflectance mask which induces an underestimation of coccolith blooms (cf Alvain et al., 2008) whereas the fuzzy method uses L2 data. Despite a quantitative difference, both methods give similar results for assessing the interannual variability of the coccolithophore blooms.

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4.4 The fuzzy method applied to SPM: a relative index for monitoring the coccolithophore blooms

The selection of typical radiance spectra to build up the reference set is a major step in the fuzzy method. Without applying a complex methodology to discriminate coccoliths from other phytoplankton group; coccoliths are easily discerned by browsing the daily images of satellite-derived chlorophyll-a and non-algal SPM. One particularity of coccolithophore blooms is their low chlorophyll-a concentration (cf Tyrrell and Merico, 2004), thus pixels where both SPM and chl-a signatures are high are not selected for the assessment of the mean spectrum, as explained in Sect. 2.2. In the opposite case, a high signal of SPM and chlorophyll-a could be the artifact of a strong bloom generated by another class of phytoplankton as diatoms. The criterion here used, specifying that the radiance (nLw) value at 490 nm (or 488 nm) has to be greater than the radiance (nLw) value at 550 nm is also supported by the description of coccolith spectra in O'Reilly et al. (2000). Mineral

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SPM observed in the surface water of the continental shelf are also highly reflective. Shutler et al (2010) investigated the case of coccolithophore blooms within the coastal context of the Celtic Sea and the Western English Channel where mineral SPM in river plumes or resuspension are observed, particularly during the windy or rainy months of the end of winters. In the North-East Atlantic studied area, on the margin of the continental shelf, this issue is less critical, although it exists.

5 The fuzzy method enables an overview of the seasonal and interannual variability of the coccolithophore blooms and, applied to SPM images, it provides a good proxy for coccolith abundance. The yearly coccolith budget and the seasonal evolution of the bloom from south northwards correspond well to the known scheme (Tyrrell and Merico, 2004). It is worth noticing that, as any other remote-sensing application, this study focuses on a particular moment of the life of the coccolithophore blooms when detached coccoliths are present in the surface water. Due to the threshold in the fuzzy indice
10 our method selects only the most significant blooms, excluding the low concentration events. Conversely, it excludes most of the mineral particles on the continental shelf (resuspension and river plumes) and there is no residual signal of mineral particles on the monthly averages shown on Figure 5.

The quasi absence of coccolithophores in 2014 is intriguing. What could be the cause of this absence of coccolithophores that corresponds to the exceptional development of large patches of gelatinous filter-feeding salps reported in the
15 PELGAS2014 cruise (IFREMER). Salps mucus also impaired the hake fishery of the Bay of Biscay in May and June by clogging the fishing nets. It is quite possible that the salps and other gelatinous plankton trapped coccoliths in their mucus net, gathering them into fecal pellets at an unprecedented level in 2014. As these pellets are large, their sinking rates may reach hundreds of meters per day (Iseki, 1981), much higher than that of detached coccoliths and hence could have favoured the cleaning of the surface mixed layer. As mentioned in Olson and Strom (2002), grazing by microzooplankton is an
20 important factor in the formation and persistence of coccolithophore blooms. Another hypothesis for the 2014 anomaly is that coccolithophores, gelatinous organisms, and zooplankton in general, react identically to a change in their environment, a change that has to be determined. This supposes further investigations about environmental (ligh, stratification, turbulence, nutrients) and biological parameters (competition, grazing...) to explain the interannual and spatial variabilities of coccolith blooms.

25 5 Conclusion

The fuzzy method applied to SeaWiFS and MODIS spectral radiance gave results in agreement with in situ data obtained in 1998 in the Bay of Biscay and with Calcite and PHYSAT products for abundance and identification respectively. The seasonal and interannual variability of the coccolith blooms in the vicinity of the continental shelf break from the Bay of Biscay to Southern Ireland between 1998 and 2015 has been described in detail. Seasonal climatologies suggested a
30 northward progression of the coccoliths blooms from the Bay of Biscay in April to the Celtic Sea in June. A remarkable conclusion is that the budget of coccolithophores (SPM_{fc}) is twice as strong in the Celtic Sea sub-area than in the Southern

and Northern Bay of Biscay sub-areas. Moreover, in the Northern and Southern Bay of Biscay and in Southern Ireland, the maximum bloom development corresponds to the period of maximum bloom extent; but in the Celtic Sea, the bloom intensity reaches its maximum value two weeks before the maximum bloom extent.

At interannual time scales, the discrimination method showed differences in the development of the coccolith blooms between sub-areas over 18 years punctuated by particular dynamics in 2014 when the signal of the bloom dropped to the lowest level of the time-series and in 2001 when the bloom reached its highest level. Although it has been shown that a small mixed layer depth, strong irradiance and high temperatures are factors which enhance coccolithophore development, other environmental and biological factors must be taken into account for a better understanding of the temporal (seasonal and interannual) variability of the coccolithophore blooms detected by remote-sensing.

The fact that SPM_{fc} reproduced better the variability of coccoliths than coccospheres suggests that the fuzzy method applied to the SPM predominantly detects detached coccoliths. The timing of the seasonal and interannual variability is similar to that obtained by CALCITE NASA and PHYSAT but for this latter, owing to a daily identification in the fuzzy method, the apparent intensity of the blooms is stronger.

This study shows that the fuzzy method can be applied with success to screen coccolith pixels on SeaWiFS and MODIS SPM images in the margin of the European shelf. It would be more difficult to identify spectrally coccolithophore blooms occurring in the Western English Channel at the end of June or in July, sometimes at high levels of concentration with imbedded patches of *Karenia mikimotoi* (Smyth et al., 2002). However at that time of the year, the surrounding waters are clear enough to enable an easy separation of the coccolith signal from the mineral SPM reflectance. One possibility could be to exploit further the concept advanced by Shutler et al. (2010) that the dynamics of coccolithophores and mineral SPM are different. To this purpose it is possible to consider that mineral SPM responds quickly to waves and tides (Rivier et al., 2012; Gohin et al., 2015) while coccolithophore blooms respond to more complex environmental factors. The seasonal and interannual variability obtained in this work could be used in future studies to analyse the impact of the climatic and environmental changes in the North East Atlantic which might considerably affect coccolithophore blooms. The investigation of other phytoplankton groups with the same remote-sensing scheme could also contribute to a better understanding of the chronology of the different phytoplankton populations before and after the coccolithophore blooms.

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REFERENCES

Alvain, S., Moulin, C., Dandonneau, Y., Loisel, H.: Seasonal distribution and succession of dominant phytoplankton groups in the global ocean: A satellite view. Global Biogeochem. Cycles 22, GB3001. doi:10.1029/2007GB003154, 2008.

- Balch, W.M., Kilpatrick, K., Holligan, P.M., Harbour, D., Fernandez, E.: The 1991 coccolithophore bloom in the central north Atlantic II: Relating optics to coccolith concentration, *Limnol. Oceanogr.* 41, 1684–1696, 1996.
- Balch, W.M., Drapeau D.T., Cucci T.L., Vaillancourt R.D., Kilpatrick K.A., Fritz J.J.: Optical backscattering by calcifying algae: separating the contribution of particulate inorganic and organic carbon fractions. *J Geophys Res*, 104:1541–1558, 1999.
- Balch, W.M., Gordon, H.R., Bowler, B.C., Drapeau, D.T., Booth, E.S.: Calcium carbonate measurements in the surface global ocean based on Moderate-Resolution Imaging Spectroradiometer data. *Journal of Geophysical Research: Oceans* 110, n/a–n/a. doi:10.1029/2004JC002560, 2005.
- Beaufort, L., Probert, I., de Garidel-Thoron, T., Bendif, E.M., Ruiz-Pino, D., Metzl, N., Goyet, C., Buchet, N., Coupel, P., Grelaud, M., Rost, B., Rickaby, R.E.M., de Vargas, C.: Sensitivity of coccolithophores to carbonate chemistry and ocean acidification. *Nature* 476, 80–83. doi:10.1038/nature1029, 2011.
- Binti Jafar Sidik, M., Gohin, F., Bowers, D., Howarth, J., Hull, T.: Relationship between Turbidity and Suspended Particulate Matter at a mooring station in a coastal environment: consequences for remote-sensing, submitted.
- Brown, C.W., Yoder, J.A.: Coccolithophorid blooms in the global ocean. *J. Geophys. Res.* 99, 7467–7482. doi:10.1029/93JC02156, 1994.
- Capuzzo, E., Painting, S. J., Forster, R. M., Greenwood, N., Stephens, D. T., Mikkelsen, O. A.: Variability in the sub-surface light climate at ecohydrodynamically distinct sites in the North Sea. *Biogeochemistry* 113:1-3, 85-103, 2013.
- Doney, S.C., Fabry, V.J., Feely, R.A., Kleypas, J.A.: Ocean Acidification: The Other CO₂ Problem. *Annual Review of Marine Science* 1, 169–192. doi:10.1146/annurev.marine.010908.163834, 2009.
- Garcia-Soto, C; Pingree, R.D.: Spring and summer blooms of phytoplankton (SeaWiFS/MODIS) along a ferry line in the Bay of Biscay and western English Channel. *Continental Shelf Research*, 29. 1111 - 1122. 2009.
- Gohin, F., Druon, J.N., Lampert, L.: A five channel chlorophyll concentration algorithm applied to SeaWiFS data processed by SeaDAS in coastal waters. *International Journal of Remote Sensing* 23, 1639–1661. doi:10.1080/01431160110071879, 2002.
- Gohin, F., Loyer, S., Lunven, M., Labry, C., Froidefond, J.-M., Delmas, D., Huret, M., Herbland, A.: Satellite-derived parameters for biological modelling in coastal waters: Illustration over the eastern continental shelf of the Bay of Biscay. *Remote Sensing of Environment* 95, 29–46. doi:10.1016/j.rse.2004.11.007, 2005.
- Gohin, F.: Annual cycles of chlorophyll-a, non-algal suspended particulate matter, and turbidity observed from space and in-situ in coastal waters. *Ocean Sci.* 7, 705–732. doi:10.5194/os-7-705-2011, 2011.
- Gohin, F., Bryère, P., Griffiths, J.W.: The exceptional surface turbidity of the North-West European shelf seas during the stormy 2013–2014 winter: Consequences for the initiation of the phytoplankton blooms? *Journal of Marine Systems* 148, 70–85. doi:10.1016/j.jmarsys.2015.02.001, 2015.
- Gordon, H.R., Boynton, G.C., Balch, W.M., Groom, S.B., Harbour, D.S., Smyth, T.J. : Retrieval of coccolithophore calcite concentration from SeaWiFS Imagery. *Geophys. Res. Lett.* 28, 1587–1590. doi:10.1029/2000GL012025, 2001

- Harlay J, Borges, A.V., Van der Zee, C., Delille, B., Godoi, R.H.M., Schiettecatte, L.S., Roevros, N., Aerts, K., Lapernat, P.E., Rebreanu, L., Groom, S., Daro, M.H., Van Grieken, R., Chou, L.: Biogeochemical study of a coccolithophorid bloom in the northern Bay of Biscay (NEW Atlantic Ocean) in June 2004. *Prog Oceanogr.* 86(3-4), 317–336, 2010.
- 5 Holligan, P.M., Viollier, M., Harbour, D.S., Camus, P., Champagne-Philippe, M.: Satellite and ship studies of coccolithophore production along a continental shelf edge. *Nature* 304, 339–342. doi:10.1038/304339a0, 1983.
- Iseki, K.: Particulate organic matter transport to the deep sea by salp fecal pellets. *Mar. Ecol. Prog. Ser.* 5:55–60, doi:10.3354/meps005055, 1981.
- 10 Lampert, L., Quéguiner, B., Labasque, T., Pichon, A., Lebreton, N.: Spatial variability of phytoplankton composition and biomass on the eastern continental shelf of the Bay of Biscay (north-east Atlantic Ocean). Evidence for a bloom of *Emiliana huxleyi* (Prymnesiophyceae) in spring 1998. *Continental Shelf Research* 22, 1225–1247, 2002.
- 15 Moore, T.S., Campbell, J.W., Dowell, M.D.: A class-based approach to characterizing and mapping the uncertainty of the MODIS ocean chlorophyll product. *Remote Sensing of Environment* 113, 2424–2430. doi:10.1016/j.rse.2009.07.016, 4 2009.
- Moore, T.S., Dowell, M.D., Franz, B.A.: Detection of coccolithophore blooms in ocean color satellite imagery: A generalized approach for use with multiple sensors. *Remote Sensing of Environment* 117, 249–263. doi:10.1016/j.rse.2011.10.001, 2012.
- 20 Morozov, E., Pozdnyakov, D., Smyth, T., Sychev, V., Grassl, H.: Space-borne study of seasonal, multi-year, and decadal phytoplankton dynamics in the Bay of Biscay. *International Journal of Remote Sensing* 34, 1297–1331. doi:10.1080/01431161.2012.718462, 2013.
- 25 Olson, M.B., Strom, S.L.: Phytoplankton growth, microzooplankton herbivory and community structure in the southeast Bering Sea: insight into the formation and temporal persistence of an *Emiliana huxleyi* bloom. *Deep Sea Research Part II: Topical Studies in Oceanography, Ecology of the SE Bering Sea* 49, 5969–5990. doi:10.1016/S0967-0645(02)00329-6, 2002.
- 30 O'Reilly, J. E., et al.: Ocean color chlorophyll-a algorithms for SeaWiFS, OC2 and OC4: Version 4, SeaWiFS Postlaunch Technical Report Series, Vol. 11, Part 3, NASA/TM-2000-206892, 2000.
- Orr, J.C., Fabry, V.J., Aumont, O., Bopp, L., Doney, S.C., Feely, R.A., Gnanadesikan, A., Gruber, N., Ishida, A., Joos, F., Key, R.M., Lindsay, K., Maier-Reimer, E., Matear, R., Monfray, P., Mouchet, A., Najjar, R.G., Plattner, G.-K., Rodgers, K.B., Sabine, C.L., Sarmiento, J.L., Schlitzer, R., Slater, R.D., Totterdell, I.J., Weirig, M.-F., Yamanaka, Y., Yool, A.: Anthropogenic ocean acidification over the twenty-first century and its impact on calcifying organisms. *Nature* 437, 681–686. doi:10.1038/nature04095, 2005.
- 35 Poulton, A.J., Stinchcombe, M.C., Achterberg, E.P., Bakker, D.C.E., Dumousseaud, C., Lawson, H.E., Lee, G.A., Richier, S., Suggett, D.J., Young, J.R., 2014. Coccolithophores on the north-west European shelf: calcification rates and environmental controls. *Biogeosciences* 11, 3919–3940. doi:10.5194/bg-11-3919, 2014
- 40 Rencher, Alvin C.: *Methods of Multivariate Analysis*. Wiley, N.Y., 1995.
- 45 Rivero-Calle, S., Gnanadesikan, A., Castillo, C.E.D., Balch, W.M., Guikema, S.D.: Multidecadal increase in North Atlantic coccolithophores and the potential role of rising CO₂. *Science* 350, 1533–1537. doi:10.1126/science.aaa8026, 2015.
- Rivier, A., Gohin, F., Bryère, P., Petus, C., Guillou, N., Chapalain, G.: Observed vs. predicted variability in non-algal suspended particulate matter concentration in the English Channel in relation to tides and waves. *Geo-Mar Lett* 32, 139–151. doi:10.1007/s00367-011-0271-x, 2012.
- 50

- Sabine, C.L., Feely, R.A., Gruber, N., Key, R.M., Lee, K., Bullister, J.L., Wanninkhof, R., Wong, C.S., Wallace, D.W.R., Tilbrook, B., Millero, F.J., Peng, T.-H., Kozyr, A., Ono, T., Rios, A.F.: The Oceanic Sink for Anthropogenic CO₂. *Science* 305, 367–371. doi:10.1126/science.1097403, 2004.
- 5 Shutler, J.D., Grant, M.G., Miller, P.I., Rushton, E., Anderson, K.: Coccolithophore bloom detection in the north east Atlantic using SeaWiFS: Algorithm description, application and sensitivity analysis. *Remote Sensing of Environment* 114, 1008–1016. doi:10.1016/j.rse.2009.12.024, 2010.
- 10 Sourisseau, M., Jegou, K., Lunven, M., Quere, J., Gohin, F., Bryere, P.: Distribution and dynamics of two Dinophyceae producing high biomass blooms over the French Atlantic Shelf. In press in *Harmful Algae*, doi.org/10.1016/hal.2015.11.016, 2016
- 15 Smyth, T.J., Moore, G.F., Groom, S.B., Land, P.E., Tyrrell, T.: Optical Modeling and Measurements of a Coccolithophore Bloom. *Applied Optics* 41, 7679. doi:10.1364/AO.41.007679, 2002.
- Sykes, P.A., Barciela, R.M.: Assessment and development of a sediment model within an operational system. *J. Geophys. Res.* 117, C04036. doi:10.1029/2011JC007420, 2012.
- 20 Thierstein, Hans R., et Jeremy R. Young. *Coccolithophores: From Molecular Processes to Global Impact*. Springer Science & Business Media, 2004.
- 25 Thomas Y., Pouvreau S., Alunno-Bruscia M., Barillé L., Gohin F., Bryère P., Gernez P.: Global change and climate-driven invasion of the Pacific oyster (*Crassostrea gigas*) along European coasts: a bioenergetics modelling approach. *Journal of Biogeography*, 43, 568–579, 2016.
- Tyrell, T., Merico, A. *Emiliania huxleyi*: bloom observations and the conditions that induce them, in: Thierstein, H.R., Young, J.R. (Eds.), *Coccolithophores: From Molecular Processes to Global Impact*. Springer, pp. 75–97, 2004.,
- 30 Van Oostende, N. Harlay, J., Vanellander, B., Chou, L., Vyverman, W., Sabbe, K.: Phytoplankton community dynamics during late spring coccolithophore blooms at the continental margin of the Celtic Sea (North East Atlantic, 2006–2008). *Prog. Oceanogr.* 104: 1–16, 2012.
- 35 Winter, A., Henderiks, J., Beaufort, L., Rickaby, R.E.M., Brown, C.W.: Poleward expansion of the coccolithophore *Emiliania huxleyi*. *J. Plankton Res.* 36, 316–325, 2014.

Figure 1. The study area and a coccolithophore bloom on 25 April 2013 observed through 3 different satellite products: RGB (Red-Green-Blue) image, non-algal SPM and calcite. (a) Studied area and the four subregions: Ireland zone (A: 52° N, 53° N; -12° W, -11° W), Celtic Sea (B: 48.5° N, 49.5° N; -8.5° W, -7.5° W), Northern Bay of Biscay (C: 46° N, 47° N; -4° W, -3° W), Southern Bay of Biscay (D: 44.5° N, 45.5° N; -3.5° W, -2.5° W)); On April 25 2013: (b) RGB image from MODIS; (c) non-algal SPM concentration; (d) calcite concentration.

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Figure 2. SeaWiFS and MODIS reference spectra (nLw) for applying the fuzzy method. (a) nLw spectra selected (reference spectrum in red) for SeaWiFS; (b) and for MODIS.

Figure 3. Application of the fuzzy method on 25 April 2013: (a) Fuzzy index for MODIS, with index between 0 (0 % coccolith signal) and 1 (100 % coccolith signal); (b) SPM concentration for pixels where fuzzy index is greater than 0.4.

Figure 4. Sensitivity test on the number of coccolith pixels on 25 April 2013: (a) effect of the fuzzy index level in selecting coccolith pixels; (b) effect of the number of spectra in the reference database.

Figure 5. Monthly averages of coccolith concentration from April to June: (a-c) averages on the 1998-2014 period; (d-f) year 2013; (g-i) year 2014.

Figure 6. Average number of coccolith pixels identified on the different areas (1998-2014): (a) Number of coccolith pixels in the 4 sub-areas (Ireland, Celtic sea, North Bay and South Bay of Biscay); (b) Coccolith budget in the 4 sub-areas and (c) Number of coccolith pixels in the entire zone.

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Figure 7. Biweekly interannual variability (1998-2015) of the number of coccolith pixels from April to August and anomalies for the sub-regions: (a) Northern Bay of Biscay (zone C in Fig.1a), Southern Bay of Biscay (zone D in Fig.1a), Celtic sea (zone B in Fig.1a), Southern Ireland (zone A in Fig.1a); (b) Anomaly of sum of coccolith pixels.

Figure 8. Interannual variability of coccolith pixels and clear pixels on the full area (1998-2015): (a) number of coccolith pixels with the mean and mean plus 2 standard deviation; (b) number of clear pixels; (c) number of coccolith pixels normalised by number of clear pixels, with the mean values of time-series.

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Figure 9. Comparison of the coccolith abundance derived from the fuzzy method (SPM_{fc} in mg L⁻¹) with in situ data (liths ml⁻¹ and coccospheres ml⁻¹) in April 1998 in the Bay of Biscay: (a) Sampling stations of Modycot (22-27 April 1998); (b) SPM_{fc} concentration of coccolith pixels; (c) SPM_{fc} concentration on selected pixels for 8 stations (red spots) between 22 and 26 April, in situ concentration of coccoliths (blue line), and in situ concentration of coccospheres (pink line).

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Figure 10. Scatterplots of mineral SPM versus Calcite concentration in April 2013: (a) for all pixels of the entire area on 25 April 2013 ($r^2=0.92$); (b) for coccolith pixels (SPM_{fc}) on 25 April 2013 ($r^2=0.69$); (c) for coccolith pixels (SPM_{fc}) for the month of April 2013 ($r^2=0.73$). Sampling is made on a grid of 10*10 pixels for (b) and 90*90 pixels for (c).

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Figure 11. Comparison of coccolith areas obtained from the fuzzy method and from PHYSAT between 1998 and 2003.

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