UNIVERSITY OF Southampton

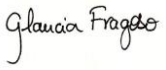
24th November, 2016 Editor, **Biogeosciences**

Dear Editor:

I, with my co-authors, would like to resubmit our article entitled "Spring phytoplankton communities of the Labrador Sea (2005-2014): pigment signatures, photophysiology and elemental ratios" for publication in **Biogeosciences**. The paper has been extensively revised in accordance with the suggestions of the reviewers, and a sheet detailing the changes made is included in this letter. A revised manuscript (with and without track changes) and new supplemental material are attached.

Thank you for your consideration, and we hope the paper is now acceptable to you.

Sincerely,



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Ms. Ref. No.: bg-2016-295
Title: Spring phytoplankton communities of the Labrador Sea (2005-2014): pigment signatures, photophysiology and elemental ratios. Resubmitted to Biogeosciences.

We thank the three reviewers for their comments and suggestions, which we feel have greatly improved the manuscript. Below we respond to each comment in detail. RC refers to "Reviewer's Comments" and AC to "Author's comments". We have enumerated the reviewer's comments to organise better our responses.

Reviewer #1:

RC1.1- General comments: This work provides information on the phytoplankton groups found in the surface waters of the Labrador Sea. Pigment signatures determined with HPLC were analyzed with CHEMTAX to obtain the contribution of the various algal groups to the total chlorophyll a concentration. The authors also related the phytoplankton biogeographic distribution to the properties of the various water masses and the photophysiology of cells during the late spring /early summer over a 10 year period. The use of CHEMTAX for this data set is a novel application, however, a previous publication by Fragoso et al. in 2016 described the phytoplankton communities linked to the various hydrographical areas of the Labrador Sea at depths less than 50 meters using microscopy. Although both microscopy and CHEMTAX analytical methods are critical to any biogeographic examination of phytoplankton, I feel the two methods should have been combined into a single manuscript as they complement one another.

AC1.1 - The first manuscript (Fragoso et al 2016, Progress in Oceanography) only focused on taxonomy of large phytoplankton (>4 µm), identifiable by light microscopy. Moreover, it only included data from 4 years (2011- 2014), and not all the stations sampled along the AR7W transect line. The current manuscript focuses on additional algal groups not considered in Fragoso et al (2016) through analysis of pigments. The new manuscript also covers a much larger dataset (10 years of data rather than only 4), many more stations from the AR7W line, and includes important biogeochemical aspects of the data not examined before. For these reasons we have kept the two papers separate and distinct, in order to ensure all aspects of phytoplankton (taxonomy, algal groups, biogeochemical and physiological aspects) are examined in full. This is now explained in line 71 and 628.

RC1.2 Therefore, although I consider this work to be of value in its contribution to our understanding of the dynamics of the biogeochemical characteristics of the Labrador Sea, I feel its content fails to merit publication in present form.

Key problems that I feel need to be addressed include: 1) the absence of the initial
CHEMTAX matrices and RMS errors

AC1.2 - CHEMTAX input matrices have now been inserted in the manuscript (Table 3). The output matrices and information about the range of RMS errors (in the legend of the Table S1) has now in the supplemental material.

RC1.3 2) the organization of the methodology section; it is not well structured, it includes CHEMTAX results and lacks information (see specific comments)

AC1.3 - The method section has now been modified according to the reviewer's suggestions. For specific changes, see the response to specific comments (methods, comments RC1.11 onwards) below.

RC1.4 - 3) the amount of information presented regarding taxonomy; species-specific information for the encountered groups of diatoms would have helped to understand differences on the photoprotective responses observed.

AC1.4 - We agree with the reviewer comment and have now added additional comments identifying the distinct diatom species influencing differences in the photoprotective response. See the line 564 in the revised manuscript.

RC1.5 - 4) of all the identified pigments (presented in Table 2) only the (DD+DT): chl a and DT:(DT+DD) are included for discussion on cell physiology. The authors should at least have included why they did not use the PPC:PSC, PPC:chl a or the pigment chlorophyllide a

AC1.5 - Although we appreciate the reviewer's suggestion, the ratios of PPC:PSC in this study varied according to phytoplankton community structure due to the inherent variations of PPC and PSC within different phytoplankton groups. For this reason, we decided to not include PPC:PSC ratios. We focused on AP:TChla ratios as a function of phytoplankton community structure. This relationship not been explored in detail previously. In contrast, the DD+DT:chla ratio only pertains to taxa which possess a xanthophyll cycle with a photoprotective (DD) and photosynthetic (DT) component, hence we have retained our focus on the DD and DT patterns.

RC1.6 - 6) the use of accepted and standardized abbreviations for the marker pigments and the phytoplankton groups in Tables 2 and 3 and throughout the text and finally

AC1.6 – We apologise for this oversight and have now updated and added the commonly accepted and standardized abbreviations for the marker pigments as suggested by the reviewer. See Tables 2 and 3 in the revised manuscript.

RC1.7 - 7) the correction of any incorrectly assigned references.

AC1.7 - References have now all been corrected.

RC1.8 - Specific comments
Introduction Line 54: change for Phaeocystis spp. colonies (> 100µm).

AC1.8 - This sentence has been removed as the introduction was rewritten following another reviewers comments.

RC1.9 Line 83: update references.

AC1.9 - This sentence has been removed.

RC1.10 - Line 84 what do you mean by "while the influence of phytoplankton composition on photophysiological patterns has not been investigated thoroughly?" please explain further.

AC 1.10 - This sentence has been removed.

RC1.11 - Methods In general this section is not well structured and needs clarification and more detail.

Sampling and analysis are combined throughout this section and need to be presented with more organization. I recommend organizing this section into separate Study Area, Sampling and Biogeochemical Analyses sub-sections and limiting relevant data to relevant sub-sections.

AC 1.11 An additional subsection named "Biogeochemical analysis" has been added including nutrient, POC:PON and chlorophyll *a* methodology.

RC1.12 - Line 138: please include the number of stations sampled before fixed stations (was it 28 as in the previous work?). The number of depths sampled at each station should appear in the text as well.

AC1.12 - Information about the number of stations has now been added to the manuscript (line 131). Samples for this manuscript were collected from the surface only and this information is now included in line 140.

RC1.13 - Line 141: please write the specifications of the Seabird CTD system.

AC 1.13 - Specifications of the Seabird CTD system (SBE 911) has been inserted (line 134).

RC1.14 - Lines 148-149: the description of how the total chl a was analyzed is presented before explaining how the collected samples for pigment analyses were filtered (probably on board?). Were samples for chl a fluorometric determination kept frozen at -20C until analyses or at -80C is a bit confusing. Was the extraction (90% acetone) performed by keeping the filters at -20C for 24 h? or rather the filters were kept at -20C until analysis (extraction for 24h with 90% acetone)? Was acidification of the samples performed?

AC 1.14 - We have now explained how chlorophyll a was sampled and analysed (see lines 143 and 153) and have cited the methodology of Holm-Hansen et al (1965) for chlorophyll determination by fluorescence, which includes acidification of the samples.

RC1.15 - Line 151: I recommend changing this line to "samples for detailed pigment analysis were filtered onto 25 mm Whatman GF/F filters".

AC 1.15 - We rewrote this sentence as: "…filtered onto 25 mm glass fibre filters (GF/F Whatman Inc., Clifton, New Jersey)" (see line 143).

RC1.16 - Lines 151-153 How much time passed between storage and analysis for the samples? Were the samples always analyzed in the same laboratory for every cruise over the 10-year period? Information on the maximum time of filtration is not provided and is very important for xanthophyll measurements. If too much passed while doing the filtration, the measurements of diatoxanthin are likely to be meaningless. This is also important for degradation pigment information, however the later data are not presented.

AC 1.16 – This information is now inserted in lines 143: "Samples for detailed pigment analysis were filtered onto 25 mm glass fibre filters (GF/F Whatman Inc., Clifton, New Jersey) and immediately flash frozen in liquid nitrogen, kept frozen in a freezer (at -80° C) until analysis in the BIO (2005-2013) or NOC (2014) laboratories within 2-3 months of collection. Volumes of water sampled for HPLC analysis were adjusted, such that samples were filtered as quickly as possible (< 10 mins)."

RC1.17 - Line 153: were the nutrient samples kept frozen or refrigerated until analysis?

AC 1.17 – Refrigerated (5°C and analyse at sea within 12 h of collection). We have now added this information in the text (line 147).

RC1.18 - Pigment analysis Line 166: Was calibration done with external pigment standards obtained from DHI? Was the precision of the instrument tested? Is there a variation coefficient? Do you have limits of detection? Please at least provide the limits of detection and quantification and how were they estimated and if the pigments with concentrations below this limit were reported or not. All this information is relevant and missing.

AC 1.18 - Information about the standards, calibration and quantification procedures are described in detail in Stuart and Head (2005) and Poulton et al (2006), which we have now cited in this section. We have also added information about precision, coefficient of variation and limits of detection (lines 158 - 165).

RC1.19 - Table 2: In this table and throughout the manuscript the authors should follow the abbreviations for phytoplankton pigments and pigment formulae suggested in the Scientific Council for Oceanic Research (SCOR), Jeffrey et al. 1997 or in Higgins et al. 2011 In: Roy S, Llewellyn CA, Egeland ES, Johnsen G (eds) Phyto- plankton pigments: char-acterization, chemotaxonomy and applications in oceanography. Cambridge University
Press, Cambridge, p 257

AC 1.19 - Pigment abbreviations in Table 2 were updated as suggested by the reviewer.

RC1.20 - This table should summarize the distribution of major taxonomically significant pigments found in the various algal groups during the study. This is poorly done in its current form. The authors should avoid ambiguity. For example when referring to 19'-hexanoyloxyfucocanthin (Hex-fuco), it should be mentioned that is a major pigment in haptophytes and dinoflagellates (Type-2, lacking peridinin), instead of "some dinoflagellates" or "various". This information –if provided here- would improve significantly the reading of the few next sections dealing with the marker pigments used for the CHEMTAX analysis. Only if the authors are more specific, the use of the references Jeffrey et al. 1997 or Higgins et al. 2011 make sense. Please delete the reference column of this table unless is useful (not the case in its present form).

AC 1.20 - This table has been updated. We have included the more specific information requested following Jeffrey et al (1997), Higgins et al (2011) and Vidussi et al (2004).

RC1.21 - Chlorophyll c1 + c2 should stay as Chlorophyll c1 + c2. Please avoid the use of CHLC12.

AC 1.21 - Abbreviations updated.

RC1.22- Zeaxanthin is a minor pigment present in various groups as cyanobacteria, however this group is supposed to be practically absent in polar waters. Although Blais et al. 2012 showed that cyanobacteria may be underestimated in polar regions (Beaufort Sea & Baffin Bay). Did the authors find presence of cyanobacteria using epifluorescence microscopy?

AC 1.22 - We did not count cyanobacteria but referred to information confirming the presence of *Synechococcus* in the Labrador Sea (Atlantic waters) from Li et al (2016) as stated in line 215.

RC1.23 - Also did the authors perform any correlation analyses between prasinoxanthin and zeaxanthin to prove that the zeaxanthin encountered did or did not correspond to a group of prasinophytes-containing zeaxanthin? Please provide this information.

AC 1.23 - We are not sure if we understand this point raised by the reviewer as a correlation between prasinoxanthin and zeaxanthin would not directly determine whether the zeaxanthin found belongs to prasinophyte-containing zeaxanthin or cyanobacteria. Zeaxanthin, in this study, represented not only prasinophytes type 2, but also chlorophytes and cyanobacteria. Moreover, species representing prasinophyte type 2, such as *Pyramimonas* and *M. pusilla* have been observed (qualitatively in our samples, although not directly counted due to difficulties in quantification) in the Labrador Sea from microscope observation of Lugols fixed samples. *M. pusilla* is abundant in the North Water Polynya in regions near the Labrador Sea as stated in line 213.

RC1.24 - Pigment interpretation There are major problems with this section. The title itself is more like the title of a results section. Actually the authors use the title "CHEMTAX interpretation" as a section included in the results. I suggest the authors change the title of the pigment interpretation section to HPLC pigment data or Clustering of HPLC data for CHEMTAX or CHEMTAX analysis or something similar-

AC 1.24 - The title of the section has been changed to "CHEMTAX analysis" as suggested (line 167).

RC1.25 - This section is not well structured and difficult to follow partially because the authors explain the use of the selected initial pigment ratios while presenting the output matrices after the CHEMTAX analyses (Table 3). This is confusing for the reader. The initial ratio matrices used to seed CHEMTAX are not presented or explained with detail. Instead ambiguous information is presented e.g. "diatoms were identified as containing high fucoxanthin to chl a ratios"

AC 1.25 - Initial pigment ratios (Table 3) have now been inserted in the new version of the manuscript and the output ratio information has been moved to the supplemental material. Explanation for the selected ratios are explained in lines 179 – 189 and we have now included a column in the initial pigment ratios table (Table 3) mentioning the source reference (*Ref) where the ratios were taken from to seed the initial CHEMTAX analysis.

RC1.26 - Line 171: change it for Mackey et al., 1996, version 1.95.

AC 1.26 – Changed.

RC1.27 - The following paragraph is not straightforward. The information on how CHEMTAX works in general and how version 1.95 works lacks clarity. This later version is a significant improvement on CHEMTAX application since the software sets up the multiple (60) initial pigment ratio matrices to obtain the more stable final values (as was recommended for example by Latasa 2007) and was actually used and described by Wright et al. 2009 and other authors before Coupel et al. 2015! Please add the references.

AC 1.27 - This paragraph has been rewritten (lines 168 - 177) and the earlier references have now been included.

RC1.28 - Line 179 to the end of the paragraph: please use the standardized abbreviations and you should at least explain why you decided to choose these particular marker pigments for the CHEMTAX analysis. Your microscopy results from the previous work should help here in a more detailed way.

AC 1.28 – We have now reorganised this paragraph. The explanation for the selection of the pigments are found in lines 182 – 189.

RC1.29 - Line 183: Again, please refer to Mackey et al. 1996 before more recent studies.

AC 1.29 - Reference now added (line 192).

RC1.30 - Line 191 to 197: Is figure 2 referring to the mean relative concentration of the main marker pigments to total accessory pigments (wt:wt) encountered or to chl a or total chl a or is based on the pigments absolute values? Unclear. It would have been helpful to include in this figure the biogeographical region linked with each cluster (as in figure 3).

AC 1.30 – Figure 2 has been moved to the supplemental material (now Fig S1a) and refers to the percentage contribution of each diagnostic pigment to the observed statistical Bray-Curtis similarity between samples (at the 60% level) after fourth root transformation (see explanation of the statistics in lines 197 - 201). Thus, it is the mean relative (%) fourth-root transformed concentration of each of the selected pigments to the total (selected) for each cluster (see

revised figure legend in supplemental material). We have now added to Figure S1 a biogeographical plot showing the cluster groups as depicted in Figure 3 (Fig. S1b).

RC1.31 - Line 198: you already explained this earlier (lines 173-74). I think this is not very well explained and this may be the reason why you mentioned it again here. Line 199-200:
"To satisfy this requirement, initial pigment ratios were carefully selected and applied to each cluster". This should actually be mentioned earlier in this section when you explain and justify why you use the selected pigment markers that best describe the phytoplankton community of your study area.

AC 1.31 - We have reorganised these sentences into the suggested order of explanation from the reviewer.

RC1.32- Line 204: The authors should justify why they have used the "high light" field ratios from
Higgins et al. 2011. Moreover, considering the importance on the photo-physiological results obtained in this study why is there not more information beside the irradiance of the experimental incubations? Was the PAR incident irradiance measured at the sampling sites?

AC 1.32 – "High light" field ratios were chosen because the samples were from surface waters (see explanation now in line 185). PAR incident irradiance was not measured at all sampling sites.

RC1.33 - Line 205: "Prasinophytes were separated into type 1 (containing prasinoxanthin) and type 2 (lacking prasinoxanthin)". Both genera were observed in light microscope counts
(Fragoso et al. 2016)" What do you mean? Fragoso et al. 2016 enumerated pico- phytoplankton (M. pusilla < 2 um)?

AC 1.33 - We apologise for the confusion. *Pyraminomas* and *M. pusilla* were observed qualitatively from microscope observations but not enumerated by Fragoso et al (2016). We have now changed the reference to (Fragoso, pers obs) in the text to avoid this confusion (line 213).

RC1.34 - Line 209: Did the authors detect by HPLC the unknown carotenoid that characterizes the unique pigment signature of *M. pusilla*? Did they detect the pigment micromonal in their samples? or micromonol?

AC 1.34 - The pigment micromonal was not identified as part of the HPLC analytical protocol followed (i.e. it was not a pigment peak listed for identification in the analysis).

RC1.35 - Line 211: "In addition to prasinophytes –type 2 (type 2A in Higgins et al. 2011- I assume), zea is also the major accessory pigment of cyanobacteria etc.. unclear para- graph.

AC 1.35 - The beginning of this sentence has been rewritten for clarification (line 214).

RC1.36 - Line 215: "Prasinophytes (type-1, Higgins et al. 2011) indeed contain chl b so do chlorophytes and they can be distinguished by their relative ratios of lutein to chl b
(Higgins et al. 2011). Was lutein detected with the HPLC analyses? Again correlations would have helped here.

AC 1.36 - The BIO method does not separate lutein and zeaxanthin so we have now renamed it as Zea + Lut.

RC1.37 - Line 218: I suggest the authors change Dino-2 class for Dino2 (dinoflagellates type-2).
Avoid the use of class, use what is suggested by Higgins et al. 2011. As mentioned before, this could have been nicely done in Table 2.

AC 1.37 - We have now rewritten this sentence using "… "dinoflagellate type 2" (Dino-2)" as was suggested by Higgins et al (2011) (line 221).

RC1.38 - Line 220: Why did the authors use the term Cryptophycea instead of cryptophytes?

AC 1.38 – We have now rephrased as cryptophytes rather than cryptophycea.

RC1.39 - Line 256: Please refer to algal groups or phytoplankton groups based on pigment composition instead of "class".

AC 1.39 - "Phytoplankton/algal class" has been changed to "phytoplankton/algal groups".

RC1.40 - Results Line 294-296: Where is cluster C1 mentioned in this section to explain Figure 4?

AC 1.40 - Cluster C1 has now been included in this sentence (line 309).

RC1.41 - Line 380: Why do you present saturation irradiances here as Wm-2 when in the methodology (line 237) you mentioned the 30 different irradiance levels is expressed as µmol quanta m-2s-1. Please use same units everywhere.

AC 1.4 - Irradiance units used throughout have been changed to "Wm^{-2}".

RC1.42 - Line 382: What was the % contribution of DD, DT and,-carotene to the total PPC for clusters C3b and C2?

AC 1.42 - The percentage contribution of DD, DT and,-carotene to the total PPC would vary according to the total amount of PPC (similar situation as comparing to DD+DT/Chl a, see comments below). Moreover, as mentioned previously, we feel that this information is difficult to interpret in a simple photo-physiological sense due to the influence of phytoplankton community structure in the overall PPC values.

RC1.43 - Line 381: DD+DT/Chl a; clusters C3b and C2 have also the lowest chl a concentration. However the level of deepoxidation is higher for these two cluster. How do your DDDT/chla and PPC/PSC ratios compare with other studies for the Arctic during spring/summer transition? Actually you don't present PPC/PSC, why?

AC 1.43 - Ratios of PPC:PSC would vary according to variability in phytoplankton community structure and not just cell photophysiology. For example, a community dominated by diatoms would have high PSC:PPC, while a community dominated by prasinophytes would have low PSC:PPC. Figure 7b shows that variability in pigment ratios is mainly driven by community structure. Thus, we believe that this information is insufficient and potentially misleading to discuss the photophysiology in mixed phytoplankton communities. However, as the reviewer pointed out, the level of de-epoxidation was high, which suggests that these communities were exposed to high light levels. We have now changed the sentence from line 619 - 623, and cited other studies (i.e., Alou-Font et al 2016) that show similar patterns.

RC1.44 - Legend of figure 3: would be better if each variable and parameter is related to the corresponding panel.

AC 1.44 - We have now revised the figure legend so that each variable and parameter is related to the corresponding panel (line 996).

RC1.45 - Discussion Very little information is discussed about spatial and temporal incident PAR irradiance variation.

AC 1.45 - Unfortunately we do not have PAR measurements from the ship for each site during the 10 years of cruise observations. However, we do discuss PAR indirectly through mixed layer depth, stratification index and progression of solar incidence from May to June throughout the discussion section.

RC1.46 - Line 405: Chlorophytes have also been associated with land-fast ice in the Arctic (e.g.
Palmer et al. 2011).

AC 1.46 - The suggested reference has now been added (line 450).

RC1.47 - Lines 524-529: I think this is a very interesting result and an interesting point for discussion. Here is where species identification for the diatom groups of Arctic and Atlantic waters would have been helpful. How do these results compare to other Arctic studies?

AC 1.47 - We have now added a discussion of the influence of distinct species in the variable AP:Tchla ratios (see comments above) (line 561 - 570).

RC1.48 - Lines 540 to 550: This paragraph deserves a better explanation with at least details on the microscopic most abundant genera for diatoms.

AC 1.48 - The distinct species of diatoms potentially influencing the AP:TChla have now been included (line 561 - 570).

RC1.49 - Lines 564 to 575: is more a repeated line of the introduction.

AC 1.49 - This whole paragraph has been removed.

RC1.50 - Lines 564 to the end: The resulting ratios of the final CHEMTAX analysis should have been discussed here, at least accordance/discrepancies with past studies in the polar environment. The interesting comparison among the carbon biomass- estimated from
CHEMTAX and the estimated by microscopic observations- should have been better structured and compared with other studies.

AC 1.50 - We have now included more references that compare the two methods of biomass estimations (CHEMTAX and microscopy) from polar environments (lines 637 – 654) to further clarify these points.

RC1.51 - Lines 987 to 993: please relate each variable to the corresponding panel.

AC 1.51 - We have now related each variable and parameter with the corresponding panel (line 996).

RC1.52 - References need further formatting review.
Latasa M (2007) Improving estimations of phytoplankton class abundances using CHEMTAX. Mar Ecol Prog Ser 329:13

Wright SW, Ishikawa A, Marchant HJ, Davidson AT, van den Enden RL, Nash G (2009)
Composition and significance of picophytoplankton in Antarctic waters. Polar Biol 32:797

AC 1.52 - References added.

Reviewer #2, Simon Wright:

RC2.1 - GENERAL COMMENTS:

This paper provides a decadal assessment of phytoplankton communities of the Labrador Sea using pigment markers and CHEMTAX analysis, as well as environmental parameters (T, S, nutrients, MLD, etc) and photosynthetic parameters. A single transect was sampled during each late spring – early summer for 10 years with high geographic resolution. The comprehensive suite of measurements makes this a valuable data set that should provide a useful reference for future cruises. I believe it is appropriate for Biogeosciences.

The analyses appear to have been competently performed and I have no worries about the data. Although the text itself is generally well written, at the broader level the manuscript itself unfortunately has two serious problems. First, it is not well structured – in particular, it lacks a clear Aim.

AC2.1 – We thank the reviewer for his comments have now rewritten large sections of the manuscript. The introduction now has a clear aim of the work identified and this has now been reinforced in the last paragraph.

RC2.2 -Secondly, and perhaps as a consequence, the authors have attempted to cover too much data in a single publication. They describe the entire data set rather than derive a clear story from it. As a result, key parts of the story are insufficiently described despite a huge volume of complex text, and the overall story is confusing. Three subplots are introduced (Accessory pigment:Chl_a ratios, POC:PON ratios, and photosynthetic parameters) that add little to (what I consider to be) the main story but add considerable verbiage and unnecessary confusion. There is possibly sufficient data here for a thesis, in which each of these subplots would warrant a separate chapter. Here they would be better relegated to separate publications, possibly followed by a review paper that integrates this study with previous work in the region. Due to lack of a coherent focus, the data and discussion are not well integrated.

AC2.2 - We have now reaffirmed the aims of the paper, which is to compare the biogeochemical and photo-physiological properties of phytoplankton communities from contrasting biogeographical regions in the Labrador Sea and to create a baseline of these trends which could be compared with in the future. These aims require that we comprehensively cover the various aspects ("sub-plots") of the data, and warrants that they are considered equally within the main theme of the paper: the analysis of the phytoplankton community composition from pigment analysis. Although we agree with the reviewer that this manuscript covers a lot of information, we feel that the results from sections 3.4 and 3.5 are directly linked to the information on phytoplankton groups and that writing a different paper about POC:PON ratios and photochemical aspects on their own would lead to them being watered down in importance or relevance to one another. We hope that the reviewer can appreciate our focus and that the restated aim and revised text address his concerns.

RC2.3 -Despite these problems, this is a very useful study that should be published, but the manuscript requires substantial revision.

STRUCTURAL COMMENTS:
Introduction:
This paper desperately needs a clear Aim to provide a basis for a narrative, to dictate what is included in (or excluded from) the paper, to provide a focus for the Results,
Discussion and Conclusions, and by which to judge the success of the project.

AC2.3 - Clear aims have now been added in the last paragraph of the introduction and the Results, Discussion and Conclusions all link to these aims.

RC2.4 -There is an implicit aim in the sampling regime – "What are the major determinants of phytoplankton composition and abundance in the Labrador Sea?" My comments hereafter will address this aim, and I leave the authors to judge how appropriate they are to the revised paper.

AC2.4 - The major determinants of phytoplankton composition and abundance have already been investigated in detail by Fragoso et al (2016). The uniqueness of the current manuscript is that it takes the subject matter a step further to focus on additional algal groups, including those not included in Fragoso et al (2016). Moreover, we investigate the biogeochemical (POC:PON) and photo-physiological signatures of these different communities and show that these all vary across the biogeochemical domains of the Labrador Sea. The current manuscript includes a much larger dataset (10 years) and more stations from the AR7W line than Fragoso et al. (2016). Following this reasoning we have decided not to follow the direct suggestions of the reviewer to focus only on hydrography, but rather we continue to examine other aspects (biogeochemistry and physiology) of the phytoplankton communities. We hope that this provides a more holistic understanding of phytoplankton dynamics in the Labrador Sea.

RC2.5 -The Introduction must provide sufficient information to provide the context for the Aim and to allow the reader to understand the significance of the results as they are presented. It must introduce all of the major topics covered in the paper, but nothing else.
Thus, the first two paragraphs (lines 42-65) are unnecessary; as is the paragraph on
CHEMTAX starting line 86 (which should be replaced by a brief outline on the approach taken to address the Aim).

AC2.5 – We retained the first two paragraphs as they refer to the impact of hydrography on community structure and photophysiology and to the impact of phytoplankton community structure on C:N ratios. However, we do agree with the reviewer that the introduction needs to better guide the reader and provide enough information for the stated aims of the paper. Thus, we have now radically shortened the whole introduction and reorganised it in attempt to make it clearer for the reader.

RC2.6 -The description of the study region is currently split between the Introduction (lines 66-
84), Methods (lines 114-132), and Discussion (lines 409 – 413). Given that the notional paper is now about the Labrador Sea, I suggest that all of this information should be amalgamated in the Intro, as should most of the description of the NAO (lines 425-430), and Figure 1.

AC2.6 – We believe that merging this information would result in a very long introduction and have decided to give only a brief overview of the regions of the Labrador Sea in the introduction (see paragraph 3), while we focus on the complex hydrography in the study area section (2.1) of the methods. The possible effects of the NAO are not investigated in depth in this manuscript and therefore we have retained it only in the discussion.

RC2.7 -I would specifically identify the main factors that may control phytoplankton – temp, salinity, mixed layer depth, light, nutrients, ice, meltwater. I also think that the Introduction should mention that the cruises occurred at different times of the Spring/Summer, introducing the notion of a temporal sequence, as this was the basis for one of the Conclusions (which surprised me on the first read!). Also that there were some cruises that deviated from the normal transect. I note that there was another publication by the same authors in the same region this year. I am surprised that there was not a specific reference to how this study relates to the previous one.

AC2.7 – The main factors that control phytoplankton bloom is now mentioned in line 65. Information about the seasonality of late spring/early phytoplankton communities is also inserted in the introduction (see third paragraph of the introduction), including findings from the previous study by Fragoso et al (2016) (line 65 - 71). We have also made it clear to the reader that there is (slight) temporal variability in the sampling times (line 128, Fig. 2b, Table 1). Line 420 – 431 also now includes a summary of the temporal progression of communities observed in this study.

RC2.8 -Method

The inclusion of results in section 2.4 surprised me at first, but I think that this section is peripheral to the main story and is appropriate here.

AC2.8 - We agree with the reviewer.

RC2.9 -Results:
I was frustrated by the fact that CHEMTAX results were presented only at the community level as defined through cluster analysis – but what was happening with the individual taxa that comprised these communities? Later I discovered that these results were (sort of) presented in the Discussion. I suggest that the distributions of individual taxa should be presented (with figures) before the distributions of communities.

AC2.9 - Information about individual taxa has now been inserted in the results section (lines 291 - 299, see also Fig. 3)

RC2.10 - I would like to see a more detailed analysis of the factors controlling phytoplankton in each water mass. Even though there was considerable data on photosynthetic properties, I didn't get a clear message on the role of light in controlling biomass.

AC2.10 – Unfortunately we do not have PAR measurements for each site during the 10 years of cruise observations. However, we do discuss PAR indirectly through mixed layer depth, stratification index and progression of solar incidence from May to June throughout the discussion section. Further analysis of nutrient and light variability across the Labrador Sea and its impact on phytoplankton composition is also discussed in Fragoso et al. (2016).

RC2.11 -The Results should include a specific section on the temporal sequence, possibly exploring the sequence of events in each region. I note in Fig 3 that the data for 2012 and 2014, which were sampled late in the season, differ from other years, particularly Chl and nutrients in the central region.

AC2.11 - Unfortunately, we do not feel that there is not enough information to provide a true temporal sequence of data or the succession of phytoplankton community composition. However, Fig. 2b does shows the temporal variability in sampling period.

RC2.12 -Discussion:
Much of the discussion about individual taxa in section 4.1 should be first described in the Results section.

AC2.12 - Information about individual taxa has now been added to the Results section (lines 291 – 299, Fig. 3)

RC2.13 -Most of sections 4.2 and 4.3 should be saved for another paper.

AC2.13 – Here we do not agree with the reviewer (see initial comment AC 2.2 and 2.4) and have retained them in the revised manuscript.

RC2.14 -The Discussion should focus specifically on the results of this paper in relation to the
Aim, only referring to other studies to provide context, generally in the style of "Our results match those of Smith and Jones…". Only then should the wider implications of the work be discussed, and there should be clear signals when the narrative extends beyond the current work. Much of this Discussion reads like a review. It was often difficult to determine whether the results being discussed were from this paper or from others.

AC2.14 - We agree with the reviewer and have now considerably revised the discussion to focus on our aims and our results, and have improved the interpretation of our data in comparison to other studies.

RC2.15 -Conclusions:

Most of the final paragraph seems more appropriate to the Introduction. The authors may also consider any further research questions that arise from this study.

AC2.15 - This last paragraph was removed from the conclusions.

RC2.16 -Abstract:
I think the first sentence is redundant and that the second sentence should be extended to include the Aim. The abstract will require revision in line with the changes to the rest of the manuscript.

AC2.16 - We have now changed the beginning of the abstract to reinforce the aims and the relevance of the study (line 14 – 18).

RC2.17 -SPECIFIC COMMENTS:
Line 186 and Table 3: Lutein not used for chlorophytes? (Does the BIO method separate ZEA & LUT?) If not, Table 3 ZEA must be ZEA+LUT

AC2.17 - The BIO method does not separate lutein and zeaxanthin so we have renamed it to Zea + Lut as suggested.

RC2.18 -Lines 192-200 and Figure 2: I note that two of the categories include Hex but no Chlc3 – I assume this is a simplification of the text and diagram as this combination does not exist to my knowledge. Figure 2 is unnecessary and should be replaced with a table including all pigments.

AC2.18 – This figure has now been moved to the supplemental material (Fig. S1). In this study, *Phaeocystis pouchetii* was not associated with 19-hex and was identified using Chl C3. This has previously been observed in the Labrador Sea (Stuart et al., 2000) and is stated in line 640.

RC2.19 -Section 3.2: Did the authors try further subdivision of group C3b? This group is by far the biggest, it is widest spread across the S-T diagram (Fig 5a), and its composition is "mixed", yet Fig 4a shows major divisions within the group. Would these subdivisions distinguish communities that were more coherent in composition and habitat?

AC2.19 - Cluster C3b had the highest level of internal Bray-Curtis similarity in terms of sample composition (i.e. samples in this group were more similar (73%) to one another than to other groups). Hence, we decided not to further divide it as we could in theory continue to subdivide until each subgroup contains very few samples.

RC2.20 -Line 316: change "Phaeocystis (cluster B)" to "A community dominated by diatoms and Phaeocystis (cluster B)". This is an important consideration throughout the document —- e.g. lines 328, 329 – there is not a careful distinction between the cluster groups (communities) and the taxa comprising them. I would invent an acronym or abbreviation for each community to avoid this confusion.

AC2.20 - Changes have now been added and updated here (line 331) and throughout the manuscript.

RC2.21 -Line 527: The possibility that "diatom species from both Arctic and Atlantic waters varied intrinsically in pigment composition" can be supported by consulting Table 3 of this paper, where we see that they do.

AC2.21 - This is true and we have cited the table showing the final matrix ratio, where fuco:chla varies among Artic and Atlantic diatoms (line 561). Diatom composition (polar versus Atlantic species) might be influencing these discrepancy and we have now added a line discussing this possibility (line 564 - 570).

RC2.22 -Line 551: "chlorophytes were present in high concentrations on the Labrador Shelf, which may explain the discrepancy between these results." Some more details are required to constitute an explanation.

AC2.22 - These sentences have been completely rewritten for clarification (line 605 – 610):

RC2.23 -Table 5: This table should be augmented by information on the region in which each cluster is found, and the major taxonomic components.

AC2.23 – We added the main taxonomic components to the table (See Table 5).

RC2.24 - Also expressing the values like Temperature with standard errors is inappropriate. The values are not based on repeat measurements of a single parameter –e.g. Cluster 3b is listed as 3.4+/-0.2 C, but the actual range is from about -1.3 to +8, the widest of any group. I would be surprised if the standard error given is correct. Even if is, it is meaningless. This table should list the range for each cluster instead.

AC2.24 – Presenting only the ranges for each cluster makes it very difficult to identify patterns of similarity between the environmental conditions associated with these clusters. To aid in interpretation we have now added standard deviations rather than standard errors to Table 5 and have also added a table with the parameter ranges (Table S2) to the Supplementary material.

RC2.25 -Also: I didn't see any reference to the data for DT:(DT+DD) in text (nor was there any reference to how long the filters were held between sample collection and freezing. This should be < 5-10 min for this parameter to be valid).

AC2.25 - We have now included information on the filtering time (<10 mins) in the methods (line 146).

RC2.26 -Results: I did not notice any indication that the raw pigment data were to be included in
Supplementary Material or an online databank. I would hope that this will be the case to increase the value of this data set.

AC2.26 - Data from Bedford Institute of Oceanography) are publically available online (http://www.dfo-mpo.gc.ca/science/data-donnees/biochem/index-eng.html). We are discussing with the co-authors the possibility of submitting additional data to PANGEA.

RC2.27 -TECHNICAL COMMENTS:
Line 67 and throughout: References should be cited in order of date – oldest to newest

AC2.27 - For Biogeosciences, the order can be based on relevance, as well as chronological or alphabetical listing, depending on the author's preference. This is state in the "Reference" tab in "Manuscript preparation guidelines for authors" (http://www.biogeosciences.net/for_authors/manuscript_preparation.html).

RC2.28 -Line 84: change "while" to "but"

AC2.28 - This sentence was removed.

RC2.29 -Line 118: inset "wide" after "km" (twice)

AC2.29 – Inserted (lines 102, 103).

RC2.30 -Line 123: change "fresh" to "low salinity". Rest of same paragraph: three water masses are described as "warm and salty" or "cold, low salinity" but other water masses lack these descriptions (parallel form required– see below). Also, is the warm arrow parallel to the Labrador Current in Fig 1 considered to be part of that current?

AC2.30 - This whole paragraph has changed, although we have described the water masses in an orderly manner as suggested by the reviewer (lines 105 - 110). The light red arrow in Figure 1 represents the extended branch of the IC, which is a modified (cooled and freshened) water mass caused by lateral and vertical mixing along the Labrador slope. We have now clarified this information in the Figure legend (lines 992).

RC2.31 -Line 177: The correct reference for the method ascribed to "Coupel et al. (2015)" is
Higgins et al (2011).

AC2.31 - We have actually updated this reference to Wright et al., (2009) (line 176).

RC2.32 -Line 316: Add "respectively" after "(IC)"?

AC2.32 - Both communities were related to the Irminger Current (an Atlantic water mass) so it is unsuitable to add "respectively" as suggested. However, we have rewritten the end of this sentence for further clarification (line 330).

RC2.33 -Line 325: Replace "respond strongly to" with "are associated with" and "spatial aspects of the data" with "environmental parameters"

AC2.33 – Changed (line 352).

RC2.34 -Line 331: The description of Fig 5b could hardly be more obscure: "In Atlantic waters, temporal aspects of the data were also observed (upper and lower right quadrants (Fig. 5b))." There is nothing in that figure that implies a temporal sequence. It was only when the Conclusions mentioned clear temporal differences that I searched the document for "temporal" to find what I had missed and came back to this figure. After some cross-referencing I realised that the description should have read, "In Atlantic waters (upper and lower right quadrants (Fig. 5b)), the phytoplankton community was composed of mixed taxa during May (orange circles), but became dominated by diatoms and dinoflagellates during the bloom in June (red circles), showing a clear temporal succession in these waters". More generally, the authors must not rely on the reader to discern what is in a figure. The reader is not familiar with the data and may not see what the author sees, or they may see something different. Whatever story exists in the figure, it must be stated clearly in text as part of the narrative. The figure supports the narrative, it does not replace it.

AC2.34 - We have now clarified the temporal succession of the spring bloom in the Labrador Sea in the text and changed this sentence following the suggestion of the reviewer (lines 358 – 361).

RC2.35 -Line 368: Replace "lower accessory pigments to TChla ratio" with "lower ratio of accessory pigments to Tchla"

AC2.35 – Changed (line 397).

RC2.36 -Line 369: Replace "(Fig. 7b). Furthermore, communities from warmer waters (Irminger
Current from Atlantic origin), particularly those co-dominated by diatoms and dinoflagellates had " with "(Fig. 7b) than communities from warmer waters (Irminger Current from Atlantic origin), particularly those co-dominated by diatoms and dinoflagellates which had"

AC2.36 – Changed (line 395 – 399).

RC2.37 - Line 376: Replace "µg C µg Chla h-1W m-2" with "µg C µg Chla h-1 W-1 m2" or " (Wm-2)-1 " Also line 378

AC2.37 – All changed.

RC2.38 -Lines 375 to 386. Sentences should be rearranged to "parallel form" i.e. talk about the same things in the same order for each case cited

AC2.38 – We have rewritten this whole paragraph (see lines 404 – 416).

RC2.39 -Line 392: Insert "Atlantic," before "Labrador"

AC2.39 – We are not sure what the reviewer is referring to here.

RC2.40 -Lines 437 – 450: Reads like a review. Note also that the paragraph starts with "Phaeo-
cystis and diatoms… (Fragoso etal 2016)" but by line 441 it's "PRESUMABLY of
Phaeocystis and diatoms (Fragoso etal 2016)". Also is "eastern central Labrador Sea" (line 437) equivalent to "West Greenland Current" (line 440)?

AC2.40 - We have now changed the beginning of this paragraph. See lines 472 - 478.

RC2.41 -Line 598: Add reference e.g Gieskes and Kraay (1983) Mar. Biol. 75, 179-185.

AC 2.41 - Suggested reference added (line 653).

RC2.42 -Line 886: remove "et al" ; page numbers = 78 – 80

AC 2.42 – Changed (line 887).

RC2.43 -Figure 2 is unnecessary and should be replaced with a table including all pigments.

AC 2.43 - We believe that Figure 2 is key to the CHEMTAX analysis. However, we have now moved it to the supplemental material.

RC2.44 -Figure 4b. The colours of the sectors would be much more easily interpreted if they made sense to a phycologist! Surely cyanobacteria = Cyan, chlorophytes = Dk Green,
Prasinophytes = Lt Green, Phaeocystis = Brown, etc. (Leave diatoms white)

AC 2.44 - Although we appreciate the colour selection of the reviewer, we have retained the original pastel colours in 4b as the colours suggested are already used elsewhere in Figure 4 (4a, 4c) and this could confuse the reader.

RC2.45 -Figure 4c. The single circle as a scale is ambiguous. Does the biomass relate to the diameter or the area of the circle? In any case it's difficult to judge. There should be a range of circles representing a biomass scale (if circles are to be used). Also I estimate that about 20% of the data points are hidden in this diagram as they underlie another circle. This could be solved by increasing the breadth of the figure or using vertical bars instead of circles. Could the fronts be marked for each year by dotted lines?

AC 2.45 – We have now increased the breadth of Fig 4c and added a scale for the bubbles. Physical fronts are already discernible through sharp changes in phytoplankton community composition, as mentioned in line 319, whereas dotted lines would become confusing between adjacent years.

RC2.46 -Figure 5: It would be good to see individual taxa plotted in such diagrams.

AC 2.46 –We could add the "arrows" of individual taxa in the figure 5b, however, we decided to leave the figure unchanged as adding information on taxa would be confusing to interpret the main message of the figure, which is the

effect of environmental factors on distinct phytoplankton communities. That is because, diatoms, for example, are the dominant taxa in all communities (except cluster C3b), so the "diatom arrow" in just one direction could bias the interpretation. We are focused on the whole community rather than individual taxa.

RC2.47 -Table 2 is unnecessary. The individual pigments are not part of the story – simply quote the references.

AC 2.47 - We believe that this Table is important for the CHEMTAX analysis so we have it in the manuscript.

RC2.48 -Table 3: The legend doesn't make it clear that the references cited provided the starting ratios from which these data were calculated. Cyanobacteria is misspelt.

AC 2.48 - We have now clarified the legend of Table 3. "Cyanobacteria" has now been spelt correctly.

RC2.49 -Table 4: The formatting is strange. It looks as if it should be split into A & B, horizontally.

AC 2.49 - Table 4 has now been split horizontally into a) and b) for better clarity.

Reviewer #3:

RC3.1 - The manuscript "Spring phytoplankton communities of the Labrador Sea (2005-2014): pigments signatures, photophysiology and elemental ratios" present a time series of pigments and nutrients data in the Labrador Sea from 2005 to 2014. The authors use the CHEMTAX method to interpret the pigment dataset in term of phytoplankton groups and then to describe the distribution of these phytoplankton groups. Oceanographic provinces of the Labrador Sea are identified using on physical and biogeochemical parameters as well as phytoplankton diversity. Several statistical approaches based on clustering, ordination plot and regression were used to link the distribution in time and space of the phytoplankton with the environmental parameters. Finally, several physiological parameters related to the phytoplankton communities were measured (P curves, POC/PON, POC/POC Chla) or extract from the pigments distribution (AP/Chla, photoprotective pigments). The physiological information is used to go further in the explanation of the link between the phytoplankton community's distribution and the environmental conditions.

General comments:
The introduction is not well structured and full of too heavy and unclear sentence.

AC3.1 - We have now completely rewritten and reduced the introduction to provide a better focus.

RC3.2 - But, the manuscript goes better in the result and discussion section. The results section is clear with a good choice of graph. Sometimes, it was difficult to get the point of the use of methods and the information that sort from some data.

AC3.2 - We have now changed and improved the methods section for better clarification by adding further explanation on the use of the different methods to examine the data.

RC3.3 - Finally, the discussion put together in a clear way all the information in the results section and brings interesting information to parameters that were of unclear utility in the result section. The authors highlight the specificity of the species and explained their success in the different regions and use well the comparison with the literature. I recommend important

change in the introduction to make it more fluent, to better extract the key information and topics of each sub-paragraph. The sentences are generally way too long and confusing.
Most of them could be cut in two parts. There are several mistakes on the use of superlative in the results section. The discussion is well conducted and uses interestingly the results

AC3.3 – We thank the reviewer for their helpful comments and suggestions. The introduction has been shortened and sentences are now condensed throughout.

RC3.4 - Specific comments by section
Introduction
L51: better to use "structure"

AC3.4 – Changed (line 52).

RC3.5 - L51: change the order to "functional role in the community"

AC3.5 - We have removed "in the community" to avoid redundancy (line 52).

RC3.6 - L 54 to 59: there is some redundancy with the lines 51-53

AC3.6 - We have now changed/reduced the introduction and shortened the sentences, so this redundancy does not exist anymore.

RC3.7 - L59 to 64: Unclear about the conservation or not of the stoichiometry. You said the "stoichiometry is consistent phylogenetically" and latter you mentioned, "they may vary (…) phenotypically within species". Be more precise on when the ratios are conserved or not.

AC3.7 - This sentence has been rewritten (lines 56).

RC3.8 - L70 "shelves and the basin"

AC3.8 - This sentence has been removed.

RC3.9 - L75-76: I don't think the interest to study the phytoplankton is to use it as an index of waters masses since simple parameters as temperature and salinity did a good job. It appears to me more important to highlight the possible importance of the biogeography on the biological pump, carbon export or the energy transfer to upper trophic level.

AC3.9 - We agree with the reviewer and have rewritten these lines to focus on ocean biogeochemistry and marine ecosystems (lines 70).

RC3.10 - L78-84: The same idea is repeated. Please reduce the size of the sentence, too much utilization of the conjunction "and".

AC3.10 - This paragraph has been removed to shorten the introduction overall.

RC3.11 -L82: could simplify "high-latitude Arctic/Atlantic waters" by "polar waters".

AC3.11 - This paragraph has been removed.

RC3.12 -L100: redundancy with the line 88-90

AC3.12 - Lines 77 - 79 refers to analysis of pigments using the HPLC while line 85 refers to CHEMTAX analysis of pigment data; hence we do not see them as redundant.

RC3.13 -L93: Please precise the concept of "functional cell size"

AC3.13 - This sentence has been removed from the introduction.

RC3.14 -L94-95: "assemblage dominance": wrong, it's the dominance of phytoplankton groups and not assemblages

AC3.14 - This sentence has been removed from the introduction.

RC3.15 -L95: remove "however"

AC3.15 – Changed (line 79, 80).

RC3.16 -L99: remove the comma.

AC3.16 – Changed (line 84).

RC3.17 -L107: "comprehensively understand" is a pleonasm.

AC3.17 - The word "comprehensively" was removed and this last paragraph of the introduction has been rewritten.

RC3.18 -L108-L111: you repeat the same information than the line 106-108.

AC3.18 - This paragraph (last of introduction) has been reduced.

RC3.19 -Methods

There is some confusion on the water composition of the Labrador Sea. Moreover the authors depicted as well deep and shallow currents and water masses. The authors should focus on the surface and sub-surface water-masses and circulation since the pigment dataset presented here concerned only the upper 10m.

AC3.19 - We believe that the reviewer is referring to the Irminger Current (IC). The IC is described as a surface current (see Hauser et al., 2015; Yashayaev and Seidov, 2015), however the Western Greenland Current may occasionally "slide" over the IC in the central-eastern part of the Labrador Sea and form a "tongue" of fresh, cold and less dense water. The lateral advection of this tongue (i.e. how offshore it goes) varies inter-annually during spring. We have used a T-S diagram to discern these water masses (IC, Labrador Current and WGC, see Figure 5a). As the reviewer has noted there were some relatively warm (> 3°C) and salty (> 34) water found at the surface. We refer to this as part of the IC, although it might have been slightly modified due to the dynamic features of surface waters, which includes the influence of precipitation/evaporation, meltwater, riverine input and mesoscale eddies. Although the IC is "conserved" at mid-depth waters (200-600 m), it does reach the surface, however it becomes "modified" due to the factors mentioned.

RC3.20 - L115: "transition zone between the Arctic and …"

AC3.20 – Changed (lines 97, 98).

RC3.21 - L115: Newfoundland is not really the southern boundary. The North Atlantic is the southern boundary.

AC3.21 - We have now defined the limits of the Labrador Sea according to the International Hydrography Organization (line 100).

RC3.22 - L119: The lower limit of the Greenland Shelf (ie 2500m) sounds very deep to characterize a shelf! I think you characterize the extension of the Greenland Current here.

AC3.22 - We apologise for the confusion. We were referring to the Greenland shelf and slope and not just the shelf. We have corrected this now (line 102).

RC3.23 - L122: remove "mostly"

AC3.23 – This whole paragraph has changed (lines 105 – 110).

RC3.24 - L122: The Irminger current is not the main water masses of the Labrador Basin since this current it is confined on the east and west borders of Labrador Basin at a mid-depth (200-600m). The Labrador Sea Water composes the water of the basin and their characteristics are mainly influenced by the winder convection with the deeper water masses (see the work of Yashayaev et al.).

AC3.24 - We apologise for the confusion in this section. We were referring to surface hydrography only. As discussed above (AC 3.19), the WGC often "slides" over the IC, creating a broad and thin layer of fresh and cold water, usually observed in the central-eastern section of the AR7W transects. On the western part of the section the IC intrudes into upper waters. This is observed in the T-S diagrams when salty (> 34) and warm (> 3°C) waters of Atlantic origin are found at the surface. We have rewritten this paragraph for clarification (lines 112 – 123).

RC3.25 - L123: There is no evidence than the cold fresh after originated from Arctic contribute substantially to the deep basin since the front between the basin and the shelf is very strong. Part of the VITALS program using gliders is actually studying the exchange between the basin and the Labrador Shelf (B. De young, J. Palter et al.).

AC3.25 - We have changed this paragraph to aid clarification. We now refer to the upper Labrador Sea layers (< 200 m) that are comprised of waters originating from the North Atlantic (IC) and the Arctic (LC and WGC) (line 105). See the response above (AC3.23).

RC3.26 - L134: "Data used in this study"

AC3.26 – Changed (line 126).

RC3.27 - L134: remove "from stations" and "repeat".

AC3.27 – Changed (line 126).

RC3.28 - L146: Choose between "surface" or "near-surface" and stick to it all along the manuscript.

AC3.28 - We have chosen to refer to *surface* waters throughout the entire manuscript.

RC3.29 - L155: Maybe add the underline word "Back in the laboratory, POC/PON samples…"

AC3.29 – Changed (line 153).

RC3.30 - L171: I think the good way to describe the CHEMTAX output is "relative abundance" instead of "ratios of abundance"

AC3.30 – Changed (line 168).

RC3.31 - L173: not clear if all the pigments ratios are from the literature.

AC3.31 – Line 183 indicates that all pigment ratios are from the literature. We have now added a sentence to the legend of Table 3 mentioning the exact references for each pigment ratio.

RC3.32 - L174: Please indicate how the algal groups present in the study area are identified.

AC3.32 - The identification is described in full in Fragoso et al. (2016). We have now included this reference in this sentence of the manuscript (line 182). See below.

RC3.33 - L187: remove "that"

AC3.33 – Changed (line 196).

RC3.34 - L190: explain here the purpose of the fourth-root transforamation.

AC3.34 - An explanation has now been included (lines 197 - 201): "Due to the high abundance of diatoms in the data, we have decided to apply a fourth-root transformation to increase the importance of less abundant groups, which would allow us to better discerning the spatial-temporal patterns of the phytoplankton communities in the Labrador Sea."

RC3.35 - L195: "higher" than what? Be careful to compare with something when you use a superlative.

AC3.35 - This word was changed to "*high*" (line 206).

RC3.36 - Results
L277: "less well stratified"…"at those stations where"

AC3.36 – Words "less" and "those" were removed. (line 281).

RC3.37 - L278: replace "during" by "in"

AC3.37 – Changed (line 282).

RC3.38 - L279: "more highly stratify": pleonasm again…

AC3.38 - We have removed the word "*more*" from the sentence (line 284).

RC3.39 - L281: "higher": then superlative to be compared with something.

AC3.39 - We have removed the parenthesis in this sentence so it has changed to: "…POC:PON ratios were also higher > 8…" (line 286).

RC3.40 - L288: Not clear if the "pairwise analysis" you mentioned refer to the ANISOM one-way pairwise?

AC3.40 - We have changed the sentence to: "*Pairwise one-way analysis* of similarity (*ANOSIM*) between clusters…" (line 302).

RC3.41 - L289: too long sentence, please reduce or cut in two parts. Parentheses are at the wrong place.

AC3.41 - We have now split the sentence into two (line 302 - 305).

RC3.42 - L298: "especially" is useless here. In general, there is an over utilisation of adverbs in the text (mostly/especially…).

AC3.42 - The word "especially" was removed from this sentence (line 312).

RC3.43 - L313: superlative!! No subject of comparison…

AC3.43 - We have rewritten this whole paragraph. See comment below (AC.3.44).

RC3.44 - L315: superlative again. Wrong use.

AC3.44 - We have rewritten this paragraph (lines 326 – 332).

RC3.45 - L321-324: Too long sentence make it confusing. Separate in two sentences?

AC3.45 - This sentence was split into two (lines 336 – 340).

RC3.46 - L340: The table 4 is difficult to understand and could earn a better presentation.

AC3.46 - We have now reorganised Table 4, separating it into Table 4a and Table 4b. Further explanation is given in lines 336 – 350 and in the revised legend of Table 4 (Line 962).

RC3.47 - L345: there is a problem, the title is the same than 3.3!!

AC3.47 - The title has now been updated to "Phytoplankton distributions and elemental stoichiometry".

RC3.48 - L344-352: Please present the POC-PON relationships somewhere.

AC3.48 - We are not sure what the reviewer means by this comment, but POC:PON relationships are shown in Figure 6a and has been referred to in line 372.

RC3.49 - L354: Please quickly explain the purpose of calculating the relationships between POCphyto and POC:PON.

AC3.49 - We have now added a short explanation of the purpose of studying the relationships between POCphyto and POC:PON (lines 375 – 377).

RC3.50 - L359: I would say, "…contribute for a high proportion…"

AC3.50 – Changed (line 382).

RC3.51 - L362: superlative lower (use low or compare to something).

AC3.51 - We have now included an object of comparison in this sentence (lines 385).

RC3.52 - Discussion

L392: as noted earlier in the manuscript, the surface phytoplankton didn't growth in the Irminger water since this water mass is observed only the slope and at great depth.

AC3.52 - In the central-eastern part of the Labrador Sea, the IC is found below the WGC "tongue", as the reviewer mentioned. However, in the central-western region the IC is found at the surface so phytoplankton do grow in these different water masses (IC, LC, WGC). Phytoplankton species found in the IC are usually found in Atlantic waters, while polar species are found in the LC and WGC (see Fragoso et al 2016).

RC3.53 - L396-397: Here the concept of ecological succession should be better presented. Is the variation between a deep and shallow mixed layer associated to the season or the two conditions (shallow/deep mixed layer) can be observed at the same time of the year?

AC3.53 – This whole paragraph has been rewritten to clarify the seasonal and temporal patterns of phytoplankton communities (lines 420 – 431).

RC3.54 - L401-403: A link is missing between this information and the above sentence.

AC3.54 - This sentence has been rewritten for clarification (lines 433 – 438).

RC3.55 L406: "often" and "as well" mean the same here. Please remove one of the two.

AC3.55 -We have changed the word "often" to "occasionally" to clarify the sentence (lines 441).

RC3.56 L470: I would prefer to use the mean POCphyto rather than POC>…The latter formulation is not really comparable since we don't know the dispersion of the data.

AC3.56 -The spread of the data of POCphyto/total POC and POC:PON ratios are shown in Figure 6c and 6b, respectively. We have now specifically referred to the figure in the text (lines 505 – 508).

RC3.57 - L475: were also abundant

AC3.57 - We are not sure what the reviewer is referring to here.

RC3.58 - L512-519: It should be interesting to explain the meaning of the AP/TChla ratio in term of strategy for the adaptation to light regime.

AC3.58 - Few studies have examined this in any depth and hence we can conclude very little in the present study. AP/TChla ratio varied according to community composition and species adaptation to light environments, mixing regimes, competition for light with other dissolved substances (etc) could explain the observed trend. Further in depth physiological work is really needed to full elucideate the meaning of the variability in AP/TChla. We have extended the discussion a little bit in the paper in attempt to explain why such trend is observed (lines 571-574).

RC3.59 - L522-523: Conufsing because you introduce "two parameters" and after you cite three parameters (Nitrate, Silicate and SI).

AC3.59 - The word "two" has been removed from this sentence (lines 558).

RC3.60 - L540-552: You show interesting difference in the photophysiological characteristics of phytoplankton, especially between the west and east communities. Near Greenland, the communities is composed of species resistant to high light while on the Labrador Shelf, the species are less resistant to photo-inhibition. Is the light conditions are so different between east

and west to explain these different adaptations to light? It could be interesting to describe these difference in the light regimes between the two side of the Labrador Sea. The latter melting of the ice cover on the Labrador Shelf could be an explanation?

AC3.60 - We have now improved the discussion about the influence of PAR in separating the polar phytoplankton communities observed. See the rewritten paragraphs (lines 586 – 610).

RC3.61 - L555 to 558: The sentence is confusing. It takes time for me to understand that dinoflagellates bloom in May to avoid higher light levels. Please rephrase or separate in two sentences to improve the clarity.

AC3.61 - The beginning of this paragraph has been rewritten for better elucidation (lines 614 – 617).

Hauser, T., Demirov, E., Zhu, J., Yashayaev, I., 2015. North Atlantic atmospheric and ocean inter-annual variability over the past fifty years – Dominant patterns and decadal shifts. Prog. Oceanogr. 132, 197–219. doi:10.1016/j.pocean.2014.10.008

Yashayaev, I., 2007. Hydrographic changes in the Labrador Sea, 1960-2005. Prog. Oceanogr. 73, 242–276. doi:10.1016/j.pocean.2007.04.015

Yashayaev, I., Seidov, D., 2015. The role of the Atlantic Water in multidecadal ocean variability in the Nordic and Barents Seas. Prog. Oceanogr. 132, 68–127. doi:10.1016/j.pocean.2014.11.009

Spring phytoplankton communities of the Labrador Sea (2005-2014): pigment signatures, photophysiology and elemental ratios

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Abstract. The Labrador Sea is an ideal region to study the biogeographical, physiological and biogeochemical implications of phytoplankton communities due to sharp transitions between distinct water masses across its shelves and central basin. The aim of this study is to provide a baseline description of the distributions and biogeochemical traits of phytoplankton communities from distinct biogeographical regions of the Labrador Sea. We have investigated the multi-year (2005-2014) distributions of late spring and early summer (May to June) phytoplankton communities in the various hydrographic settings of the Labrador Sea. Our analysis is based on pigment markers (using CHEMTAX analysis), and photophysiological and biogeochemical characteristics associated with the communities present in the different water masses of the Labrador Sea. ~~The Labrador Sea is an ideal region to study the biogeographical, physiological and biogeochemical implications of phytoplankton communities due to sharp transitions of distinct water masses across its shelves and the central basin, intense nutrient delivery due to deep vertical mixing during winters and continual inflow of Arctic, Greenland melt and Atlantic waters. In this study, we provide a decadal assessment (2005-2014) of late spring/early summer phytoplankton communities from surface waters of the Labrador Sea based on pigment markers and CHEMTAX analysis, and their physiological and biogeochemical signatures.~~ Diatoms were the most abundant group, blooming first in shallow mixed layers of haline-stratified Arctic shelf waters. Along with diatoms, chlorophytes co-dominated at the western end of the section (particularly in the polar waters of the Labrador Current (LC)), whilst *Phaeocystis* co-dominated in the east (modified polar waters of the West Greenland Current (WGC)). Pre-bloom conditions occurred in deeper mixed layers of the central Labrador Sea in May, where a mixed assemblage of flagellates (dinoflagellates, prasinophytes, prymnesiophytes, particularly coccolithophores, and chrysophytes/pelagophytes) occurred in low chlorophyll areas, succeeding to blooms of diatoms and dinoflagellates in thermally-stratified Atlantic waters in June. Light-saturated photosynthetic rates and saturation irradiance levels were higher at stations where diatoms were the dominant phytoplankton group (> 70 %), as opposed to stations where flagellates were more abundant (from 40 % up to 70 %). Phytoplankton communities from the WGC (*Phaeocystis* and diatoms) had lower light-limited photosynthetic rates, with little evidence of photo-inhibition, indicating greater tolerance to a high light environment. By contrast, communities from the central Labrador Sea (dinoflagellates and diatoms), which bloomed later in the season (June), appeared to be more sensitive to high light levels. Ratios of accessory pigments (AP) to total chlorophyll *a* (TChl*a*) varied according to phytoplankton community composition, with polar phytoplankton (cold-water related) having lower AP to TChl*a* ratios. Phytoplankton communities associated with polar waters (LC and WGC) also had higher and more variable particulate organic carbon (POC) to particulate organic nitrogen (PON) ratios, suggesting the influence of detritus from freshwater input, derived from riverine, glacial and/or sea-ice meltwater. Long-term observational shifts in phytoplankton communities were not assessed in this study due to the short temporal frame (May to June) of the data. Nevertheless, these results have provided a baseline of current distributions and an evaluation of the biogeochemical role of spring phytoplankton communities in the Labrador Sea, which will improve our understanding of potential long-term responses of phytoplankton communities in high-latitude oceans to a changing climate.

Keywords

Phytoplankton communities, CHEMTAX, hydrography, photophysiology, biogeochemistry, Labrador Sea

1. Introduction

Marine phytoplankton form a taxonomically and functionally diverse group, where communities are structured by a variety of factors, including nutrient and light availability, predation and competition for resources (Litchman and Klausmeier, 2008). Environmental heterogeneity, thus, creates biogeographical patterns of abundance, composition, traits and diversity of phytoplankton communities in the global ocean (Barton et al., 2013; Follows et al., 2007; Hays et al., 2005). Phytoplankton communities within a biogeographical region are subject to similar environmental conditions, such as temperature (Bouman et al., 2003), nutrient concentration (Browning et al., 2014) and irradiance (Arrigo et al., 2010). These environmental factors, along with phytoplankton composition itself (Bouman et al., 2005), affect the overall photophysiological response and bulk primary productivity of the phytoplankton community.

Biogeography of phytoplankton communities and their photophysiological characteristics, consequently, impact the structure of marine ecosystems due to their functional role in biogeochemical cycling and transfer of energy to higher trophic levels. For example, distinct phytoplankton assemblages have been reported to influence differently particulate (Martiny et al., 2013a, 2013b; Smith and Asper, 2001) and dissolved elemental stoichiometry (C:N:P)(Weber and Deutsch, 2010), the drawdown of gases (Arrigo, 1999; Tortell et al., 2002) and the efficiency of carbon export (Guidi et al., 2009; Le Moigne et al., 2015). Patterns of phytoplankton stoichiometry may be consistent phylogenetically and within higher taxonomic levels (Ho et al., 2003; Quigg et al., 2003). However, phytoplankton stoichiometry has also been reported to vary according to nutrient supply ratios (Bertilsson et al., 2003; Rhee, 1978), as well as phenotypically within species across the same population (Finkel et al., 2006).

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The sub-Arctic North Atlantic is a complex system with contrasting hydrography that structures plankton communities within distinct biogeographical provinces (Fragoso et al., 2016; Head et al., 2003; Li and Harrison, 2001; Platt et al., 2005; Sathyendranath et al., 2009, 1995). Biogeographical regions of the Labrador Sea shape phytoplankton community composition (Fragoso et al., 2016), bio-optical properties (Cota, 2003; Lutz et al., 2003; Platt et al., 2005; Sathyendranath et al., 2004; Stuart et al., 2000) and the seasonality of phytoplankton blooms (Frajka-Williams and Rhines, 2010; Lacour et al., 2015; Wu et al., 2008, 2007). Phytoplankton blooms, for example, occur first (April to early May) in the shelves due to haline-driven stratification driven by the input of Arctic-related waters, in addition to rapid sea ice melt in the Labrador Shelf near Canada (Frajka-Williams and Rhines, 2010; Wu et al., 2007). The central Labrador bloom occurs later in the season (late May to June) as result of thermal stratification (Frajka-Williams and Rhines, 2010). Fragoso et al. (2016) showed that the biogeography of phytoplankton communities in the Labrador Sea during spring and early summer is shaped by distinct species found Atlantic or Arctic waters, which may have distinct impact on the biogeochemical cycles and transfer of energy to upper trophic level. However, these authors focused in taxonomy and investigated only larger phytoplankton (> 4µm). The photophysiological and

biochemical signatures, such as stoichiometry (C:N ratio) of these distinct spring phytoplankton communities occurring in distinct sectors of the Labrador Sea has not been previously investigated.

Quantification of marine phytoplankton community composition, for large numbers of samples, is challenging because small cells (< 5µm) are difficult to identify and count using light microscopy, in addition to being a time-consuming method. To overcome these problems, quantification and analyses of phytoplankton pigments by high performance liquid chromatography (HPLC) has been widely used to monitor phytoplankton community distributions over large temporal and spatial scales (e.g., Aiken et al., 2009; Peloquin et al., 2013; Platt et al., 2005). The interpretation of the pigment data is not always straightforward, since some pigments are shared by several algal groups and can change according to local nutrient and light conditions (DiTullio et al., 2007; van Leeuwe and Stefels, 2007, 1998). The chemotaxonomic tool, CHEMTAX (CHEMical TAXonomy), provides a valuable approach to estimate phytoplankton group abundances when used in conjunction with microscopic information (Irigoien et al., 2004; Mackey et al., 1996; Wright et al., 1996). CHEMTAX has the advantage of providing more information about phytoplankton groups than individual diagnostic pigments or ratios and has been used widely to investigate phytoplankton biogeography on regional scales (Muylaert et al., 2006; Wright and Van den Enden, 2000) and globally (Swan et al., 2015).

The aim of this study is to provide a baseline description of the current distributions and biogeochemical traits of phytoplankton communities from distinct biogeographical regions of the Labrador Sea. For this purpose, we investigate the multi-year (2005-2014) distributions of late spring and early summer (May to June) phytoplankton communities in the various hydrographic settings across the shelves, slopes and deep basin of the Labrador Sea based on phytoplankton pigments. In addition, we examine the overall photophysiological and biogeochemical traits associated with these phytoplankton communities. We believe that the results presented here will provide important information about the current condition of phytoplankton communities in the Labrador Sea and improve our understanding of potential long-term changes in high-latitude oceans.

~~Marine phytoplankton form a taxonomically and functionally diverse group, with different requirements and modes of acquisition of light and nutrients, as well as strategies for resource competition and predation defence (Acevedo-Trejos et al., 2015; Bonachela et al., 2015; Falkowski, 2004). Thus, marine phytoplankton communities are structured by the overall fitness of individuals within species assemblages with respect to a variety of factors, including the physical setting, nutrient and light availability, dispersal, predation and competition for resources (Litchman and Klausmeier, 2008). Over large scales, environmental heterogeneity selects for phytoplankton assemblages, which creates biogeographical patterns of abundance, composition, traits and diversity distributions of phytoplankton communities in the global ocean (Barton et al., 2013; Follows et al., 2007; Hays et al., 2005).~~

~~Phytoplankton communities also impact the structuring of marine ecosystems due to their functional community role in biogeochemical cycling, efficiency of carbon transport to deeper waters, palatability and transfer of energy to higher trophi c~~

levels. Diatoms, for example, are assumed to be the major contributor to the biological pump (Smetacek et al., 2004), large *Phaeocystis* spp. (> 100 µm) colonies are apparently not grazed as efficiently as diatoms due to the exudation of mucilage (Haberman et al., 2003) and some cyanobacteria are able to fix nitrogen, which can provide a significant amount of nitrogen to the oligotrophic regions of the ocean (Barton et al., 2013; Tyrrell, 1999). Distinct phytoplankton assemblages have been reported to influence differently particulate (Martiny et al., 2013a, 2013b; Smith and Asper, 2001) and dissolved elemental stoichiometry (C:N:P)(Weber and Deutsch, 2010), the drawdown of gases (Arrigo, 1999; Tortell et al., 2002) and the efficiency of carbon export (Guidi et al., 2009; Le Moigne et al., 2015). Patterns of phytoplankton stoichiometry appear to be consistent phylogenetically and within higher taxonomic levels (Ho et al., 2003; Quigg et al., 2003), although they may vary according to nutrient supply ratios (Bertilsson et al., 2003; Rhee, 1978), as well as phenotypically within species across the same population (Finkel et al., 2006). However, detritus and dead plankton material also influence overall particulate C:N:P ratios in the ocean, which complicates the interpretation of *in situ* observations of phytoplankton elemental stoichiometry (Martiny et al., 2013a).

The sub-Arctic North Atlantic is a complex system with contrasting hydrography that structures plankton communities within distinct biogeographical provinces (Fragoso et al., 2016; Head et al., 2003; Li and Harrison, 2001; Platt et al., 2005; Sathyendranath et al., 1995, 2009). The Labrador Sea is a particularly interesting region to study the biogeographical and biogeochemical implications of phytoplankton communities due to the sharp transitions of distinct water masses across its shelves and basin (Yashayaev, 2007). Biogeographical regions of the Labrador Sea shape zooplankton (Head et al., 2000, 2003) and phytoplankton community composition (Fragoso et al., 2016), cell size (Platt et al., 2005) and bio-optical properties (Cota, 2003; Lutz et al., 2003; Platt et al., 2005; Sathyendranath et al., 2004; Stuart et al., 2000), as well as the seasonality of phytoplankton blooms (Frajka-Williams and Rhines, 2010; Lacour et al., 2015; Wu et al., 2007, 2008). More recently, Fragoso et al. (2016) showed that the biogeography of phytoplankton communities in the Labrador Sea is shaped by specific species as indicators of Atlantic or Arctic waters, emphasising the potential importance of using phytoplankton composition as indicators of water masses.

Phytoplankton communities within a biogeographical region are subject to similar environmental conditions, such as temperature (Bouman et al., 2003), nutrient concentration (Browning et al., 2014) and irradiance (Arrigo et al., 2010), and these, along with community composition (Bouman et al., 2005), affect their overall photophysiological response. It has been suggested that irradiance (light levels and day length) and temperature are the primary factors that influence phytoplankton photophysiology in high-latitude Arctic/Atlantic waters, including the Labrador and Barents seas and Baffin Bay (Platt et al., 1982; Rey, 1991; Subba Rao and Platt, 1984), while the influence of phytoplankton composition on photophysiological patterns has not been investigated thoroughly.

~~Quantification of marine phytoplankton community composition, for large numbers of samples, is challenging if the time-consuming methods of microscopic identification and enumeration are employed. Moreover, small cells (< 5µm) are difficult to identify and count using light microscopy. To overcome these problems, quantification and analyses of phytoplankton pigments by high performance liquid chromatography (HPLC) has been widely used to monitor phytoplankton community distributions over large temporal and spatial scales (e.g., Aiken et al., 2009; Peloquin et al., 2013; Platt et al., 2005). Photosynthetic pigments (chlorophylls and carotenoids) are frequently used to identify taxonomic and functional groups. Pigment-based chemotaxonomy can be used to determine phytoplankton classes (Coupel et al., 2012, 2015; Gonçalves-Araujo et al., 2012), functional cell sizes (Aiken et al., 2009; Platt et al., 2005; Poulton et al., 2006) and assemblage dominance using accessory pigment ratios (Fragoso and Smith, 2012). The interpretation of the pigment data is not always straightforward, however, since some pigments are shared by several algal groups and can change according to local nutrient and light conditions (DiTullio et al., 2007; van Leeuwe and Stefels, 1998, 2007). The chemotaxonomic tool, CHEMTAX (CHEMical TAXonomy), provides a valuable approach to estimate phytoplankton class abundances when used in conjunction with microscopic information (Irigoien et al., 2004; Mackey et al., 1996; Wright et al., 1996). CHEMTAX has the advantage of providing more information about phytoplankton classes than individual diagnostic pigments or ratios, and has been used widely to investigate phytoplankton biogeography on regional scales (Muylaert et al., 2006; Wright and Van den Enden, 2000) and globally (Swan et al., 2015).~~

~~In this study, we investigate the multi-year (2005-2014) distributions of late spring and early summer (May to June) phytoplankton communities in the various hydrographic settings across the shelves, slopes and deep basin of the Labrador Sea based on phytoplankton pigments. In addition, we examine the overall photophysiological and biogeochemical traits associated with these phytoplankton communities. Long-term analyses of phytoplankton communities and their potential biogeochemical and physiological signatures are needed to comprehensively understand current conditions and to project possible responses of these communities to climate change. The results presented here will improve our understanding of potential long-term changes of phytoplankton communities in high-latitude oceans and provide a baseline description of the current distributions and biogeochemical traits of phytoplankton communities in the Labrador Sea with which future observations can be compared.~~

2. Methods

2.1 Study area

The Labrador Sea is a high latitude marginal sea located in the northwestern part of the Atlantic Ocean and is a transition zone ~~of~~ between the Arctic and sub-Arctic ecosystems. It is bounded by Davis Strait to the north, a line from Cape St. Francis in Newfoundland (47°45' N, 52°27'W) to Cape Farewell (southern tip of Greenland) to the southeast and the coast of Labrador and Newfoundland to the west (Fig. 1) (International Hydrographic Organization, 1953). ~~It is bounded by Davis Strait to the~~

~~north, Newfoundland to the south, the Labrador Coast of Canada to the west and Greenland to the east (Fig 1).~~ The bathymetry of the Labrador Sea can be subdivided into the wide continental shelf and relatively gentle continental slope on its western side (the Labrador Shelf, > 500 km wide and < 250 m deep) and narrow shelf and very steep continental slope on the eastern side (the Greenland Shelf and Slope, < 100 km wide and < 2500 m deep). T~~he bathymetry of the Labrador Sea can be subdivided into the wide continental shelf and relatively gentle continental slope on its western side (the Labrador Shelf, > 500 km and < 250 m deep), narrow shelf and very steep continental slope on the eastern side (the Greenland Shelf, < 100 km and < 2500 m deep) and the deep basin (> 3000 m deep) confined by the continental slopes (Fragoso et al., 2016).~~

The upper Labrador Sea (< 200 m) is comprised of waters originating from the North Atlantic and the Arctic (Yashayaev, 2009). Atlantic-influenced waters occur mostly in the central Labrador Sea, where waters are relatively warm, salty and mainly identified as the Irminger Current (IC). Cold, low salinity waters originate from the Arctic *via* the surrounding shelves and are mainly identified as the Labrador Current (LC) and the West Greenland Current (WGC) (Fig 1). Circulation in the central basin of the Labrador Sea is complex, often showing a gyre-like flow system that alternates in direction (Palter et al. 2016, Wang et al, 2016).~~The central deep basin (> 3000 m) of the Labrador Sea contains a counter-clockwise flow and is comprised of a mixture of, mostly, relatively warm and salty waters originating from the Atlantic, which is mainly identified as the Irminger Current (IC) and cold fresh water, originating from the Arctic *via* the surrounding shelves (Fig 1).~~

The inshore branch of the LC overlies the Labrador Shelf and includes Arctic waters originating from Baffin Bay and the Canadian Arctic Archipelago *via* Davis Strait and from Hudson Bay *via* Hudson Strait, together with inputs of melting sea ice, which originate locally or from farther north. The main branch of the LC flows along the Labrador slope from north to south and is centered around the 1000 m depth contour. It is composed of a mixture of Arctic water from Baffin Bay *via* Davis Strait and the branch of the WGC that flows west across the mouth of Davis Strait. The WGC, which flows from south to north over the Greenland shelf and along the adjacent slope, is a mixture of cold, low salinity Arctic water exiting the Nordic Seas with the East Greenland Current (EGC) (Yashayaev, 2007), together with sea ice and glacial melt water (Fig 1). The WGC often spreads westwards, forming a "tongue" of buoyant fresher water, with the accumulation of low salinity waters, driven by high eddy kinetic activity in the central eastern Labrador Sea during spring (Frajka-Williams and Rhines, 2010). The WGC often floats over the IC in the central-eastern part of the Labrador Sea, however, the IC is usually observed in surface waters of the central-western Labrador Sea during spring. More detailed descriptions of the hydrography of the Labrador Sea can be found elsewhere (Fragoso et al., Head et al. 2013, Yashayaev and Seidov, Yashayaev 2007)."

~~The inshore branch of the Labrador Current (LC) overlies the Labrador Shelf and includes Arctic water originating from Baffin Bay and the Canadian Arctic Archipelago *via* Davis Strait and from Hudson Bay *via* Hudson Strait, together with inputs of melting sea ice, which originate locally or from farther north. The main branch of the Labrador Current flows along the Labrador slope from north to south and is centred at the 1000 m contour. It is composed of a mixture of Arctic water from Baffin Bay *via* Davis Strait and the branch of the West Greenland Current that flows west across the mouth of Davis Strait. The West Greenland Current (WGC), which flows from south to north on the Greenland shelf and along the adjacent slope, is~~

~~a mixture of cold, low salinity Arctic water exiting the Nordic Seas with the East Greenland Current (EGC) (Yashayaev, 2007), together with sea ice and glacial melt waters (Fig 1). More detailed descriptions of the hydrography of the Labrador Sea can be found elsewhere (Fragoso et al. 2016, Head et al.2013, Yashayaev and Seidov 2015, Yashayaev 2007).~~

2.2 Sampling

Data used for this study were obtained ~~from stations~~ along the AR7W Labrador Sea ~~repeat~~ hydrography line (World Ocean Circulation Experiment Atlantic Repeat 7-West section, for details see Fragoso et al., 2016), which runs between Misery Point on the Labrador coast (through Hamilton Bank on the Labrador Shelf) and Cape Desolation on the Greenland coast. Stations were sampled during late spring and/or early summer, varying mostly within a 6 week window (see sampling dates in Table 1) over a 10 year period (2005-2014) by scientists from the Canadian Department of Fisheries and Oceans. Fixed stations (total of 28), as well as some additional non-standard stations, were sampled across shelves and in the deep central basin on the AR7W section or slightly north or south of this transect (Fig. 1). ~~Stations were sampled for over a decade (2005-2014) by scientists from the Canadian Department of Fisheries and Oceans during late spring and/or early summer (Table 1).~~ Fixed stations (total of 28), as well as some additional non-standard stations, were sampled across shelves and in the deep central basin on the AR7W section or slightly north or south of this transect (Fig. 1)~~Fixed stations were sampled on the AR7W section, across shelves and in the deep central basin, as well as some additional non-standard stations (Fig. 1)~~.

Vertical profiles of temperature and salinity were measured with a Seabird CTD system **(SBE 911)**. Water samples were collected using 10-L Niskin bottles mounted on the rosette frame. Mixed layer depths were calculated from the vertical density (σ_Θ) distribution and defined as the depth where
σ_Θ changes by 0.03 kg m^{-3} from a stable surface value (~10 m) (Weller and Plueddemann, 1996). A stratification index (SI) was calculated as the seawater density difference (between 10 m to 60 m) normalised to the equivalent difference in depth.

Water samples from the surface ~~(near-surface)~~ layer (< 10 m) were collected (0.5 L–1.5 L) for the determination of chlorophyll *a*, accessory pigments, nutrients, particulate organic carbon (POC) and nitrogen (PON) analysis, and for primary production measurements. Filters for chlorophyll a measurements were immediately put in scintillation vials containing 10 ml of 90% acetone, which were placed into a -20oC freezer and extracted for 24 h.~~Bulk chlorophyll *a* concentration was measured after extraction from filters in 90 % acetone at -20°C for approximately 24 hours and fluorescence was determined using a Turner Designs fluorometer (Holm-Hansen et al., 1965)~~. Samples for detailed pigment analysis were filtered onto 25 mm glass fibre filters (GF/F Whatman Inc., Clifton, New Jersey) and immediately flash frozen in liquid nitrogen and kept frozen in a freezer (at -80° C) until analysis in the BIO (2005-2013) or NOC (2014) laboratories within 2-3 months of collection. Volumes of water sampled for HPLC analysis were adjusted such that samples took less than 10 mins to filter.~~Samples for detailed pigment analysis were filtered onto 25 mm glass fibre filters (GF/F Whatman Inc., Clifton, New Jersey) and immediately flash frozen in liquid nitrogen and kept frozen in a freezer (at -80° C) until analysis in the laboratory.~~ Nutrient samples were kept

refrigerated at 5°C and analysed at sea (within 12 h of collection) on a SEAL AutoAnalyser III. ~~Nutrient samples were analysed at sea (within 12 h of collection) on the SEAL AutoAnalyser III.~~ Samples for POC and PON were filtered (0.25–1 L) onto 25 mm pre-combusted GF/F filters and frozen (-20° C) and returned to the laboratory for later analysis. ~~In the laboratory samples were oven-dried (60 °C) for 8-12 hours, stored in a dessicator, pelletelised in pre-combusted tin foil cups and analysed using a Perkin Elmer 2400 Series CHNS/O analyser as described in Pepin and Head (2009).~~

2.3 ~~Pigment~~ Biogeochemical analysis

For chlorophyll *a* samples, fluorescence was determined on board after 24 h of extraction using a Turner Designs fluorometer (Holm-Hansen et al., 1965). Fluorometric analysis of chlorophyll and phaeo-pigments, using the Turner fluorometer, was always within 48 h. For POC and PON analysis, ~~In the laboratory~~ samples were oven-dried (60 °C) for 8-12 hours, stored in a dessicator, pelletelised in pre-combusted tin foil cups and analysed using a Perkin Elmer 2400 Series CHNS/O analyser in the laboratory as described in Pepin and Head (2009).

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2.4 Pigment analysis

Pigments (chlorophyll *a* and accessory pigments) were quantified using reverse-phase, High-Performance Liquid Chromatography (HPLC). Methods for 2005-2013 (Hudson cruises), including information about the standards, calibration and quantification procedures are described in detail in Stuart and Head (2005), known as the "BIO method". Methods for samples collected in 2014 (JR302 cruise) are described in Poulton et al (2006). Quality control of both methods was applied according to Aiken et al (2009). Precision of the instruments was tested by running samples and standards and the coefficient of variation for pigments were < 10% of the mean. Limits of detection were ~0.01 and 0.002 mg m^{-3} for carotenoids and chlorins, respectively (Head, pers. comm, Poulton et al 2006). Pigments concentrations below detection limits were not reported. ~~Pigments (chlorophyll *a* and accessory pigments) were quantified using reverse-phase (Beckman C18, 3 µm Ultrasphere column), High-Performance Liquid Chromatography (HPLC) according to the procedure described in Stuart and Head (2005), known as the "BIO method". Prior to analysis, pigments were extracted by homogenizing the frozen filters in 1.5 mL 95 % acetone, grinding the filters using a motorized grinder, centrifuging to remove the solids and taking an aliquot of the supernatant, which was buffered by dilution with 0.5 M aqueous ammonium acetate at a ratio of 2:1 before injection on to the column. The samples were run using a gradient elution method, with methanol, aqueous ammonium acetate and ethyl acetate as solvents (Stuart and Head 2005). Pigment peaks were identified and quantified by their retention times and absorbance or fluorescence signals, by comparison with those of pure pigments (Stuart and Head, 2005).~~ A list of pigments identified and quantified for this study is included in Table 2.

2.4 ~~Pigment interpretation~~CHEMTAX analysis

The CHEMTAX software (Mackey et al., 1996) was used to estimate the relative abundance of distinct micro-algal groups to total chlorophyll *a* from *in situ* pigment measurements. The software utilises a factorization program that uses "best guess" ratios of accessory pigments to chlorophyll *a* that are derived for different groups from the literature available and marker pigment concentrations of algal groups that are known to be present in the study area as reported in Fragoso et al (2016). The program uses the steepest descent algorithm to obtain the best fit to the data based on assumed pigment to chlorophyll *a* ratios (for more detail, see Mackey et al 1996). Because CHEMTAX is sensitive to the seed values of the initial ratio matrix (Latasa et al 2007), we used a later version (1.95) to obtain the more stable output matrices. In this CHEMTAX version, the initial matrices are optimized by generating 60 further pigment ratio tables using a random function (RAND in Microsoft Excel) as described in Wright et al., (2009). The results of the six best output matrices (with the smallest residuals, equivalent to 10 % of all matrices) were used to calculate the averages of the abundance estimates and final pigment ratios. ~~The CHEMTAX software (version 1.95, Mackey et al., 1996) was used to estimate ratios of abundance of distinct micro-algal classes to total chlorophyll *a* from *in situ* pigment measurements. The software utilises a factorization program that uses "best guess" ratios of accessory pigments to chlorophyll *a* that are derived for different classes from the literature and marker pigment concentrations of algal groups that are known to be present in the study area. This program uses the steepest descent algorithm to obtain the best fit to the data based on assumed pigment to chlorophyll *a* ratios. The initial matrices are optimized by generating 60 further pigment ratio tables using a random function (RAND in Microsoft Excel) as described in Coupel et al. (2015). The results of the six best output matrices (with the smallest residuals, equivalent to 10 % of all matrices) were used to calculate the averages of the abundance estimates and final pigment ratios.~~

One of the main assumptions of the ~~The other main requirement of the~~ CHEMTAX method is that information about the phytoplankton taxonomy is used to assure that the pigment ratios are applied and interpreted correctly (Irigoien et al., 2004). To satisfy this requirement, initial pigment ratios were carefully selected and applied to each cluster to adjust the pigments to the appropriate ~~classes~~groups according to microscopic observations (Fragoso et al., 2016) and literature information (see Table 3). Pigment ratio tables were based on the literature in waters having comparable characteristics to the Labrador Sea, such as Baffin Bay (Vidussi et al., 2004), the Beaufort Sea (Coupel et al., 2015) and the North Sea (Antajan et al., 2004; Muylaert et al., 2006) or from surface (high light) field data (Higgins et al., 2011) (Table 3). High light field ratios were chosen because samples were collected from surface waters during May and June (average monthly irradiance >30 mol m^{-2} d^{-1}, Harrison et al 2013). The following pigment chosen for CHEMTAX analysis were: 19-butanoyloxyfucoxanthin (~~BUT19~~But-fuco), 19-hexanoyloxyfucoxanthin (~~HEX19~~Hex-fuco), alloxanthin (~~ALLOX~~Allo),- chlorophyll *a* (~~CHLA~~Chl *a*), chlorophyll *b* (~~CHLB~~Chl *b*), chlorophyll *c*3 (Chl c_3~~CHLC3~~), fucoxanthin (~~FUCOX~~Fuco), peridinin (~~PERID~~Peri), prasinoxanthin (~~PRASINO~~Pras) and zeaxanthin + lutein (~~ZEA~~Zea + Lut).

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The other main requirement of the ~~One of the main assumptions of~~ the CHEMTAX method is that pigment ratios remain constant across the subset of samples that are being analysed (Mackey et al 1996) ~~(Swan et al., 2015)~~. To satisfy this assumption, *a priori* analysis was performed, where pigment data were sub-divided into groups using cluster analysis (Bray-Curtis similarity) and each group was processed separately by the CHEMTAX program (Table 3; for the final ratio matrix, see supplemental material). This approach was used because distinct phytoplankton communities have been observed in the Labrador Sea (Fragoso et al., 2016) so ~~that~~ the ratio of accessory pigment to chlorophyll *a* probably varies within different water masses across the Labrador Sea (LC, IC and WGC). Pigment concentration data (~~BUT19~~But-fuco, Hex-fuco~~HEX19~~, ~~ALLOX~~Allo, Chl *a*~~CHLA~~, Chl *b*~~CHLB~~, Chl c_3~~CHLC3~~, Fuco~~FUCOX~~, ~~PERID~~Peri, Pras~~PRASINO~~ and Zea + Lut~~ZEA~~) were standardized and fourth-root transformed before being analysed. Due to the high abundance of diatoms in the data, we have decided to apply a fourth-root transformation to increase the importance of less abundant groups, which would allow us to better discerning the spatial-temporal patterns of the phytoplankton communities in the Labrador Sea.~~were standardized and fourth-root transformed before being analysed.~~

A first cluster analysis on transformed pigment data identified five major groups having 60 % similarity between samples. Clusters included stations partially located: 1) on the shelves, where Fuco~~FUCOX~~ dominated at a few stations (I), 2) in the eastern part of the Labrador Sea, where most stations had high relative concentrations of Fuco~~FUCOX~~ and Chl c_3~~CHLC3~~ (II), 3) in the central Labrador Sea, where a few stations had high~~er~~ proportions of Fuco~~FUCOX~~, Hex-fuco~~HEX19~~ and Peri~~PERID~~ (III), 4) on the western part of the section, where Chl *b*~~CHLB~~ and Fuco~~FUCOX~~ were the main pigments at most stations (IV) and 5) in the central Labrador Sea, where most stations had a mixture of pigments (Fuco~~FUCOX~~, Chl c_3~~CHLC3~~, Hex-fuco~~HEX19~~, Chl *b*~~CHLB~~, Peri~~PERID~~ and others) (V) (Fig 2). ~~The other main requirement of the CHEMTAX method is that information about the phytoplankton taxonomy is used to assure that the pigment ratios are applied and interpreted correctly (Irigoien et al., 2004). To satisfy this requirement, initial pigment ratios were carefully selected and applied to each cluster to adjust the pigments to the appropriate classes according to microscopic observations (Fragoso et al., 2016) and literature information (see Table 3). Pigment ratio tables were based on the literature in waters having comparable characteristics to the Labrador Sea, such as Baffin Bay (Vidussi et al., 2004), the Beaufort Sea (Coupel et al., 2015) and the North Sea (Antajan et al., 2004; Muylaert et al., 2006) or from surface (high light) field data (Higgins et al., 2011) (Table 3).~~

Prasinophytes were separated into "prasinophyte type 1", which contains Pras~~PRASINO~~ and "prasinophyte type 2", such as *Pyramimonas* and *Micromonas*, with the latter previously found lacking Pras~~PRASINO~~ but containing Zea + Lut~~ZEA~~ in North Water Polynya (Canadian Arctic) (see Vidussi et al., 2004). Both genera were observed in light microscope counts in Labrador Sea samples (Fragoso, pers. obs.) ~~(Fragoso et al., 2016)~~ and *M. pusilla* has been observed in the Beaufort Sea (Coupel et al., 2015), and was found to be one of the main pico-eukaryotes in the North Water Polynya (Canadian Arctic) from April to July of 1998 (Lovejoy et al., 2002). Zea + Lut is not only found in prasinophytes –type 2, but is also the major accessory pigment of cyanobacteria~~In addition to prasinophytes type 2, ZEA is also the major accessory pigment of cyanobacteria~~, such as

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Synechococcus spp., which has been previously observed in the Labrador Sea, particularly in Atlantic waters (Li et al., 2006), and which is also a minor pigment in chlorophytes (Vidussi et al., 2004). Because of its association with the warmer Atlantic waters, it was assumed that cyanobacteria were absent from very cold waters, such as the Labrador Current (Fragoso et al., 2016). Prasinophytes contain Chl *b*~~CHLB~~, but so do chlorophytes (Vidussi et al., 2004), which were observed in large numbers with the microscope. Dinoflagellates were separated into those species that contain Peri~~PERID~~, such as *Heterocapsa* sp. and *Amphidium* (Coupel et al., 2015; Higgins et al., 2011) and those that do not, such as *Gymnodinium* spp. (herein defined as dinoflagellates type-2 (DINO-2) according to Higgins et al (2011)~~here defined as Dino-2 class as in Higgins et al. (2011)~~), but which may contain Chl c_3~~CHLC3~~, But-fuco~~BUT19~~, Hex-fuco~~HEX19~~ and Fuco~~FUCOX~~. Dinoflagellates were observed in lower concentrations in the eastern Labrador Sea, so that Dino-2 was assumed absent from this area (clusters I & II in Table 3). Cryptophytes (~~Cryptophycea in~~ Table 3) are the only group to contain Allo~~ALLOX~~.

Prymnesiophytes were divided into three groups: 1) *Phaeocystis pouchetii*, which was observed in high concentrations in the eastern Labrador Sea (Fragoso et al., 2016) (clusters I & II, Table 3); 2) Prymnesiophyte HAPTO-7 (as in Higgins et al. (2011)), associated with *Chrysocromulina* spp. previously observed in the western Labrador Sea (in the Labrador Current, this study) (cluster IV, Table 3) and HAPTO-6 (as in Higgins et al. (2011)), which included the coccolithophores, particularly *E. huxleyi* associated with Atlantic waters (central-eastern region of the Labrador Sea) (clusters I, II, III and V, Table 3). *Phaeocystis pouchetii* occurred in waters having low Hex-fuco~~HEX19~~ and But-fuco~~BUT19~~ concentrations and high Chl c_3~~CHLC3~~ and Fuco~~FUCOX~~ concentrations (cluster II, Fig. ~~2~~S1, supplemental material). Similar pigment compositions were found in *Phaeocystis globosa* blooms in Belgian Waters (Antajan et al., 2004; Muylaert et al., 2006) and high ratios of Chl c_3~~CHLC3~~ to Chl *a*~~CHLA~~ were, previously, used to identify *Phaeocystis pouchetii* in the Labrador Sea (Stuart et al., 2000). Th~~u~~us, Chl c_3~~CHLC3~~ and Fuco~~FUCOX~~ were the only pigments that could be used to represent this species. In addition to Chl c_3~~CHLC3~~ and Fuco~~FUCOX~~, HAPTO-7 included Hex-fuco~~HEX19~~ and HAPTO-6 included Hex-fuco~~HEX19~~ and But-fuco~~BUT19~~ as in Higgins et al. (2011). Chrysophytes and pelagophytes, such as *Dictyocha speculum* have high ratios of But-fuco ~~BUT19~~ to Chl *a*~~CHLA~~ (Coupel et al., 2015; Fragoso and Smith, 2012). Finally, diatoms were identified as containing high Fuco~~FUCOX~~: Chl *a*~~CHLA~~ ratios (Vidussi et al., 2004) (Table 3).

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2.5 Photosynthesis versus irradiance incubations

Water samples were spiked with ^{14}C-bicarbonate and incubated in a light box under 30 different irradiance levels (from ~~5-2000 µmol quanta~~1 - 600 W m^{-2} ~~s^{-1}~~) at *in situ* temperature for 2 to 3 hours to measure parameters derived from photosynthesis versus irradiance (P-E) curves as described by Stuart et al. (2000). Measurements were fitted to the equation of Harrison and Platt (1986) to determine the initial slope of the P-E curve, also known as the photosynthetic efficiency (α^B), the maximum photosynthetic rate normalized to chlorophyll biomass (P_m^B), the light intensity approximating the onset of saturation (E_k), the saturation irradiance (E_s) and the photo-inhibition parameter (β).

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2.6 Statistical analysis

2.6.1 Phytoplankton-derived POC estimation

Fragoso et al. (2016) found a significant linear relationship between phytoplankton carbon calculated from phytoplankton cell counts and POC data using results from 2011-2014 surveys in the Labrador Sea ($POC = 1.01POC_{phyto} + 240.92$; $r^2 = 0.47$; $n = 44$; $p < 0.0001$). To estimate phytoplankton-derived carbon (POC_{phyto}) concentration (as opposed to total POC, which includes detritus and heterotrophic organisms), regression analysis was performed ($POC_{phyto} = 38.9Chl\textit{a}$; $r^2 = 0.9$; $n = 41$; $p < 0.0001$) using the carbon calculated from cell counts (derived from Fragoso et al., 2016) and measurements of total chlorophyll *a*: this expression was then applied to estimate POC_{phyto} for stations where phytoplankton cell counts were not available (2005-2010).

2.6.2 Phytoplankton community structure

Phytoplankton community structure derived from pigment concentrations was investigated using PRIMER-E (v7) software (Clarke and Warwick, 2001). Chlorophyll *a* concentrations derived for each algal ~~class~~ group resulting from CHEMTAX analysis were standardized (converted to percentage values) to obtain their relative proportions, which were fourth-root transformed to allow the least abundant groups to contribute to the analysis. Similarity matrices were generated from Bray-Curtis similarity for cluster analysis. A SIMPER (SIMilarity PERcentages) routine with a cut off of 90 % cumulative contribution to the similarity was used to reveal the contributions of each ~~class~~ group to the overall similarity within clusters. One-way ANOSIM was also applied to determine whether taxonomic compositions of the clusters were significantly different.

A redundancy analysis (RDA) using the CANOCO 4.5 software (CANOCO, Microcomputer Power, Ithaca, NY) was performed to analyse the effects of environmental factors on the Labrador Sea phytoplankton community structure as described in Fragoso et al. (2016). Data were log-transformed and forward-selection (*a posteriori* analysis) identified the subset of environmental variables that significantly explained the taxonomic distribution and community structure when analysed individually (λ_1, marginal effects) or when included in a model where other forward-selected variables were analysed together (λa, conditional effects). A Monte Carlo permutation test (n=999, reduced model) was applied to test the statistical significance ($p < 0.05$) of each of the forward-selected variables.

3. Results

3.1 Environmental variables

Environmental parameters, as well as chlorophyll *a* (Chl*a*) concentrations varied noticeably along the southwest-northeast section of the Labrador Sea (Fig. 2~~3~~). The shelf and slope regions (LSh, LSl, GSl, GSh) had colder and fresher waters (< 3 °C and < 33.5, respectively) compared to the central basin (CB), where surface waters were saltier (> 33.5) and warmer (> 3 °C), particularly in 2005, 2006, 2012 and 2014 (> 5 °C) (Fig. 2~~3~~c~~b~~, d~~e~~). Shelf waters that were colder and fresher were the most highly stratified (> 5×10^{-3} kg m^{-4}), particularly on the Labrador Shelf (> 15×10^{-3} kg m^{-4}), whereas waters from the CB were less ~~well~~ stratified (< 5×10^{-3} kg m^{-4}), except at ~~those~~ stations where waters were slightly warmer than usual (> 5°C) in~~during~~ 2005, 2012 and 2014 (Fig. 2~~3~~e~~d~~). Chl*a* concentrations were higher (> 4 mg Chl*a* m^{-3}) at stations where waters were ~~more~~ highly stratified, particularly on the shelves (Fig. 2~~3~~f~~e~~). Nitrate, phosphate and silicate concentrations were inversely related to Chl*a* concentration, being lower (< 5, 0.5, 3 µmol L^{-1}, respectively) on the shelves and, during some years, in the CB (e.g. 2012), where blooms formed (Fig. 2~~3~~f~~e~~-i~~h~~). POC:PON ratios were also ~~higher (~~> 8~~)~~ at most stations in shelf and slope waters and at a few stations in the CB during 2009 and 2011 (Fig. 2~~3~~j~~i~~). Shelf waters mostly had higher silicate:nitrate ($Si(OH)_4$:NO_3^-) ratios (> 1) than the CB, particularly the LSh (Fig. 2~~3~~k~~j~~). Labrador Sea surface waters usually had nitrate:phosphate (NO_3^-:PO_4^{3-}) < 16, although NO_3^-:PO_4^{3-} were higher in the CB than in shelf regions (> 10) (Fig. 2~~3~~l~~k~~).

3.2 CHEMTAX interpretation and group distributions

Diatoms were the most abundant phytoplankton group found in the Labrador Sea, particularly at the shelves where they dominated almost 100% of the total phytoplankton community (Fig. 3a). Chlorophytes and prasinophytes were common in the center-western part (Fig. 3b,c), whereas *Phaeocystis* was abundant at the eastern part of the Labrador Sea (Fig. 3d). Dinoflagellates were abundant in the center region of the Labrador Sea (Fig. 3e). Other prymnesiophytes, including coccolithophores and *Chrysochromulina* were also common at the center part of the Labrador Sea (Fig. 3f). Overall, chrysophytes and pelagophytes were found in low abundance in the Labrador Sea, except at the center region of the Labrador Sea during 2011 (Fig. 3g). Cyanobacteria was more abundant at the Labrador Slope and Greenland Shelf and during some years (2005 and 2012) at the center Labrador Sea (Fig. 3f). Cryptophytes comprised less than 10% of total phytoplankton chlorophyll concentrations (data not shown).

A cluster analysis of algal ~~classes~~ groups derived from CHEMTAX results revealed clusters of stations at various similarity levels (Fig. 4). *Pairwise one-way analysis* of similarity (*ANOSIM*) between clusters suggested that they were significantly different in terms of algal pigment composition (p = 0.001). However, pairwise analysis of clusters C3a and C3b showed that these groups were more similar in composition (R statistic = 0.33) than other clusters (R statistic values approached 1) (see Clarke and Warwick, 2001).~~ANOSIM one-way pairwise analysis between clusters suggested that they were significantly different in pigment algal composition (p = 0.001), although pairwise analysis of clusters C3a and C3b showed that these~~

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groups overlapped (more similar composition, R statistic = 0.33) than other clusters, which were clearly separated (R statistic values approached 1) (see Clarke and Warwick, 2001). The first division occurred at 61 %, separating three main clusters (A, B and C) (Fig. 4a). Cluster C was subdivided at 65 % resulting in clusters C1, C2 and C3 (Fig. 4a). A third division (similarity of 73 %) occurred at cluster C3 resulting in two other clusters C3a and C3b (Fig. 4a). Overall, six functional clusters (A, B, C1, C2, C3a and C3b) represented the distinct phytoplankton communities occurring in the Labrador Sea (Fig. 4a). These communities generally occupied different regions of the Labrador Sea, namely the Labrador Shelf/Slope (west, Cluster C1 and, mainly, Cluster C3a), Central Basin (middle, mainly Clusters C2 or C3b) and the Greenland Shelf/Slope (east, mainly Clusters C3a, A, B) (Fig. 4b,c).

Chl*a* concentrations were higher at stations where diatoms were especially dominant (Fig. 4b,c). Diatoms were the most abundant phytoplankton group in Labrador Sea waters, particularly at stations on the shelves, where communities were sometimes composed of almost 100 % diatoms (clusters A and C1) (Fig. 4b,c). Diatoms were also abundant at (or near to) the Greenland Shelf, where *Phaeocystis* was co-dominant (cluster B) and at (or near to) the Labrador Shelf in the west section, where chlorophytes were the second most abundant group (cluster C3a). Likewise, diatoms were dominant in the central Labrador Sea in some years (2008, 2012 and 2014, cluster C2), where dinoflagellates were also dominant (Fig. 4b,c). Most stations in the central basin had low Chl*a* concentrations and high diversity of algal groups (cluster C3b), with mixed assemblages of diatoms, dinoflagellates and other flagellates (Fig. 4b,c). The positions of fronts, usually characterised by sharp transitions in phytoplankton communities varied from year to year, but were generally located near the continental slopes (Fig. 4c).

3.3 Phytoplankton distributions and environmental controls

Distributions of surface phytoplankton communities in the Labrador Sea during spring and early summer (2005-2014) varied according to the water mass distributions across the shelves and central basin of the Labrador Sea. Potential temperatures and salinities also varied among these water masses (Fig. 5a). In general, a community dominated by chlorophytes and diatoms (cluster C3a) were associated with the inshore branch of the Labrador Current (LC), on the Labrador Shelf. Surface waters from the LC were the coldest (temperature < 2°C) and least saline with the lowest density ($\sigma\Theta$ of most stations approximately < 26.5 kg m^{-3}) of all the surface water masses of the Labrador Sea (Fig. 5a). Mixed assemblages (cluster C3b), as well as blooms (chlorophyll average = 4 mg Chl*a* m^{-3}) of dinoflagellates and diatoms (cluster C2) were associated with the Atlantic water mass, and the Irminger Current (IC) (Fig. 5a). These were the warmest (temperature > 3°C), saltiest (salinity > 34) and densest ($\sigma\Theta$ of most 315 stations < 27 kg m^{-3}) surface waters of the Labrador Sea (Fig. 5a). In general, chlorophytes and diatoms (cluster C3a) were associated with the inshore branch of the Labrador Current (LC), on the Labrador Shelf, where the surface waters were fresher (salinity < 33.5), colder (temperature < 2°C) and least dense (σ_θ of most stations approximately < 26.5 kg m^{-3}) (Fig. 5a). Mixed assemblages (cluster C3b), as well as blooms (chlorophyll average = 4 mg Chl*a* m^{-3}) of dinoflagellates and diatoms (cluster C2) were associated with the warmer (temperature > 3°C), saltier (salinity > 34) and denser (σ_θ of most

~~stations < 27 kg m^{-3}) Atlantic water mass, and the Irminger Current (IC) (Fig. 5a).~~ A community dominated by diatom and *Phaeocystis* (cluster B) ~~dominated~~ occurred in waters of the West Greenland Current (WGC), which had intermediate temperatures (mostly 0-4°C) and salinities (33-34.5) when compared to those of the LC and IC (Fig. 5a).

Redundancy analysis (RDA) was used to investigate the hydrographic variables that explained the variance (explanatory variables) in the phytoplankton communities based on pigment analyses. The ordination diagram revealed that stations from each distinct clusters are concentrated in different quadrants (Fig. 5b). The arrows in the ordination diagram represent the environmental variables. Positive or negative correlations indicate that the arrows are orientated parallel to the distribution of cluster stations with the strength of the correlation proportional to the arrow length.~~The ordination diagram (Fig. 5b) revealed that most stations from distinct clusters were concentrated within one quadrant, where arrows representing environmental variables in the same or opposite directions of the clusters of stations suggest positive or negative correlations proportional to the length of the arrow.~~ Table 4a indicates that the first axis (x-axis) of the redundancy analysis explained most of the variance (83.5 % of species-environment relationship; taxa-environmental correlation = 0.68). Summed, the canonical axes explained 99.8 % of the variance (axis 1, p = 0.002; all axes, p = 0.002) (Table 4a), which indicates that the environmental variables included in this analysis explained almost 100 % of the variability. Forward selection showed that five of the six environmental factors (silicate, temperature, salinity, nitrate and phosphate) included in the analysis best explained the variance in phytoplankton community composition when analysed together ($p<0.05$, Table 4b). When all variables were analysed together (conditional effects, referred to as λ_a in Table 4b), silicate was the most significant explanatory variable ($\lambda_a = 0.2$, p = 0.001), followed by temperature ($\lambda_a = 0.05$, p = 0.001), salinity ($\lambda_a = 0.02$, p = 0.002), nitrate concentration ($\lambda_a = 0.01$, p = 0.016) and phosphate concentration ($\lambda_a = 0.02$, p = 0.002) (Table 4). Stratification Index (SI) was the only explanatory variable that had no statistical significance in explaining the distribution of phytoplankton communities (Table 4b).

The first axis (x-axis) of the analysis, which explained most of the variance ~~(eigen value = 25.7 % of species data and 83.5 % of species-environment relation, Table 4),~~ clearly shows that the phytoplankton communities ~~respond strongly to spatial aspects of the data~~ are associated with environmental parameters (Fig. 5b). Thus, stations in Arctic waters were to the left of the y-axis (low nutrient concentrations, temperature and salinity values), while stations located in Atlantic waters were to the right (opposite trend, Fig. 5b). A community dominated by d~~D~~iatoms and chlorophytes (cluster C3a, upper left quadrant of Fig. 5b) were associated with lower salinities and temperatures and highly stratified waters. Another community dominated by *Phaeocystis* and diatoms (cluster B, lower left quadrant of Fig. 5b) were associated with waters where nutrient concentrations (mainly nitrate, but also phosphate and silicate) were relatively low (average nitrate concentration from cluster B < 3 µM, Table 5). In Atlantic waters (upper and lower right quadrants (Fig. 5b)), the phytoplankton community was composed of mixed taxa during May (orange circles), but became dominated by diatoms and dinoflagellates during the bloom in June (red circles), showing a clear temporal succession in these waters". ~~In Atlantic waters, temporal aspects of the data were also observed (upper and lower right quadrants (Fig. 5b)).~~ Thus, mixed assemblages (cluster C3b) were associated with higher nutrient concentrations (pre-bloom conditions in Atlantic waters, upper right quadrant), whereas dinoflagellates and

diatoms (cluster C2) were associated with warmer and saltier waters, resembling blooming conditions in Atlantic waters induced by thermal stratification (lower right quadrant of Figure 5b). Summed, the canonical axes explained 99.8 % of the variance (axis 1, p = 0.002; all axes, p = 0.002) (Table 4), which means that the environmental variables included in this analysis explained almost 100 % of the variability.

~~Forward selection showed that five of the six environmental factors (silicate, temperature, salinity, nitrate and phosphate) included in the analysis best explained the variance in the phytoplankton community distributions when analysed together (Table 4). When all variables were analysed together (conditional effects, referred to as λ_a in Table 4), silicate was the most significant explanatory variable (λ_a = 0.2, p = 0.001), followed by temperature (λ_a = 0.05, p = 0.001), salinity (λ_a = 0.02, p = 0.002), nitrate concentration (λ_a = 0.01, p = 0.016) and phosphate (λ_a = 0.02, p = 0.002) (Table 4). SI was the only explanatory variable that had no significance in explaining the distribution of phytoplankton communities (Table 4).~~

3.4 Phytoplankton distributions and elemental stoichiometry~~environmental controls~~

Particulate organic carbon (POC) collected on filters can include organic carbon from a variety of sources, such as phytoplankton, bacteria, zooplankton, viruses and detritus (Sathyendranath et al., 2009). Assuming that phytoplankton associated organic carbon, as estimated from phytoplankton cell volumes (POC_{phyto}) is strongly correlated with Chl*a* values, the proportion of POC_{phyto} should increase in eutrophic waters, which usually occurs with high Chl*a* and POC concentrations, and that it should be lower in oligotrophic waters. Indeed, our results showed higher proportions of POC_{phyto} (> 60 %) in waters with higher POC concentrations (Fig. 6a). However, there were stations where POC levels were high and where the contribution of POC_{phyto} was low, suggesting that there may have been other sources of POC (e.g. detritus).

To investigate the influence of phytoplankton community structure on the stoichiometry of particulate organic material of surface Labrador Sea waters, the relationships between POC_{phyto} (the estimated proportion of POC from phytoplankton) and the ratio of POC to PON were examined. In general, different phytoplankton communities had distinct relationships between POC_{phyto} and POC:PON.~~The relationships between POC_{phyto} and POC:PON also varied among the different phytoplankton community types (Fig. 6).~~ ~~In general, s~~Stations in shelf regions, which have higher inputs of Arctic and glacial melt waters (lower salinity values), where diatoms co-dominated with chlorophytes in the west and east (cluster C3a) or with *Phaeocystis* in the east (cluster B), had higher and more variable values for POC:PON ratios than did stations influenced by Atlantic water (Fig. 6b). Some shelf stations had relatively high proportions of POC_{phyto} to total POC, suggesting that phytoplankton community growth dominated by diatoms and chlorophytes (cluster C3a) contributed ~~more to~~to a high proportion of the total POC (most stations from cluster C3a had POC_{phyto} > 50 %) (Fig. 6b). On the other hand, some shelf stations, particularly the one dominated by a community composed of diatoms and *Phaeocystis* (cluster B) had high POC:PON ratios (> 10), with low POC_{phyto} contributions, suggesting an increased contribution of detritus to the total POC (Fig. 6c). Stations influenced by Atlantic waters had generally lower contributions of POCphyto compared to Arctic-related waters, with most stations having

POC:PON ratios < 6.6 (Fig. 6c).~~Stations influenced by Atlantic waters had generally lower contributions of POC_{phyto}, with most stations having POC:PON ratios < 6.6 (Fig. 6c)~~.

3.5 Physiological patterns

Accessory pigments (AP) versus total chlorophyll *a* (TChl*a*) scatterplot from surface waters of the Labrador Sea showed a log-log linear relationship (Fig. 7). The slopes of these relationships varied within temperature (Fig. 7a) and among the distinct phytoplankton communities (Fig. 7b). Phytoplankton communities in cold waters (of Arctic origin), such as those co-dominated by diatoms and *Phaeocystis* in the east and diatoms and chlorophytes in the west, had a lower ratio of accessory pigments to TChl*a* ~~ratio~~ (logAP:logTChl*a*) (slope = 0.86 and 0.89, respectively) ~~(Fig. 7b)~~ than communities from warmer waters (Irminger Current from Atlantic origin), particularly those co-dominated by diatoms and dinoflagellates (~~. Furthermore, communities from warmer waters (Irminger Current from Atlantic origin), particularly those co-dominated by diatoms and dinoflagellates had higher ratios of~~ logAP:logChl*a*, ~~(~~slope = 1.03) (Fig. 7b). Slopes of the logAP to logTChl*a* relationships were not statistically different among the different communities (ANCOVA, $p > 0.05$), except for those communities co-dominated by diatoms and *Phaeocystis* (cluster B), which had a slope that was statistically different from the others (ANCOVA, p= 0.016).

Photosynthetic parameters differed among the different phytoplankton communities. *Phaeocystis* and diatom communities near Greenland (cluster B) had the lowest photosynthetic efficiencies (average $\alpha^B = 6.8 \times 10^{-2}$ mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$) with relatively high onset saturation irradiances (average $E_k = 60 \pm 33$ W m^{-2}) and little photo-inhibition ($\beta = 4 \times 10^{-4}$ mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$) (Table 5). By contrast, phytoplankton communities dominated by diatoms and chlorophytes typically found in the Labrador Current (cluster C3a) were highly susceptible to photo-inhibition ($\beta = 16 \times 10^{-4}$ mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$), had lower onset saturation irradiances ($E_k = 29$ W m^{-2}) and higher photosynthetic efficiencies ($\alpha^B = 9.2 \times 10^{-2}$ mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$) (Table 5). Phytoplankton communities in Atlantic waters (clusters C3b and C2) had the highest levels of photoprotective pigments, such as those used in the xanthophyll cycle (diadinoxanthin (DD) + diatoxanthin (DT)):Chl*a* > 0.07), particularly those communities co-dominated by diatoms and dinoflagellates (cluster C2) from stratified Atlantic waters (Table 5). These communities were the most susceptible to photo-inhibition ($\beta = 29 \times 10^{-4}$ mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$), had the highest ratios of photoprotective pigments to Chl*a* ((DD+DT):Chl*a* = 0.12 ± 0.01), and the highest maximum photosynthetic rates ($P_m^B = 3.3 \pm 0.7$ mg C [mg Chl*a*] h^{-1}) (Table 5).

4. Discussion

4.1 Biogeography of phytoplankton communities in the Labrador Sea

In this study, our assessment of phytoplankton pigments from surface waters of the Labrador Sea during spring/early summer are based on a decade of observations and show that the distribution of phytoplankton communities varied primarily with distinct waters masses (Labrador, Irminger and Greenland Currents). However, a temporal succession of phytoplankton communities from the central region of the Labrador Sea was observed as waters became thermally stratified from May to June. Major blooms (Chl*a* concentrations > 3 mg Chl*a* m^{-3}) occurred on or near the shelves in shallower mixed layers (< 33 m, Table 5). Diatoms were abundant in these blooms, however they co-dominated with 1) chlorophytes in the west (mostly in the Labrador Current) and 2) *Phaeocystis* in the east in the West Greenland Current. A more diverse community with low chlorophyll values (average Chl*a* concentrations ~2 mg Chl*a* m^{-3}, Table 5) was found earlier in the season (May) in deeper mixed layers (> 59 m, Table 5) of the central basin. Once these waters of the central basin became thermally-stratified (June), a third bloom co-dominated by diatoms and dinoflagellates occurred, revealing an ecological succession from mixed flagellate communities. These patterns are similar to those seen in other shelf and basin regions of Arctic/subarctic waters (Coupel et al., 2015; Fujiwara et al., 2014; Hill et al., 2005). ~~In this study, our assessment of phytoplankton pigments from surface waters during spring/early summer of the Labrador Sea based on a decade of observations showed that the distribution of phytoplankton communities varied primarily with the distinct waters masses (Labrador, Irminger and Greenland Currents). There were three regions where major blooms (Chl*a* concentrations > 3 mg Chl*a* m^{-3}) occurred. For all three blooms, diatoms were predominant; however, they co-dominated with 1) chlorophytes in the west (mostly in the Labrador Current), 2) *Phaeocystis* in the east in the West Greenland Current and 3) dinoflagellates in the central basin of the Labrador Sea, once waters were thermally stratified. While diatoms bloomed in shallower mixed layers (< 33 m, Table 5), a more diverse community was found in most years in deeper mixed layers (> 59 m) in the central basin, resembling pre-bloom conditions. These patterns are similar to those seen in other shelf and basin regions of Arctic/subarctic waters (Coupel et al., 2015; Fujiwara et al., 2014; Hill et al., 2005).~~

It is well known that diatoms tend to dominate in high-nutrient regions of the ocean due to their high growth rates, while their low surface area to volume ratios mean that they do not do as well as smaller nano- or picoplankton in low nutrient conditions (Gregg et al., 400 2003; Sarthou et al., 2005). The Labrador Sea is a high-nutrient region during early spring due to the deep winter mixing (200 – 2300 m) that provides nutrients to the surface layers. High nutrient concentration supports phytoplankton spring blooms, particularly those dominated by diatoms, once light becomes available (Fragoso et al., 2016; Harrison et al., 2013; Yashayaev and Loder, 2009). ~~It is well known that diatoms tend to dominate in high-nutrient regions of the ocean due to their high growth rates, while their low surface area to volume ratios mean that they do not do as well as nano- or picoplankton in low nutrient conditions (Gregg et al., 2003; Sarthou et al., 2005). In the Labrador Sea, deep winter mixing (200 – 2300 m)~~

~~provides nutrients to the near surface layers, which supports phytoplankton spring blooms, particularly of diatoms once light becomes available (Fragoso et al., 2016; Harrison et al., 2013; Yashayaev and Loder, 2009).~~

Chlorophytes were the second most abundant phytoplankton group, particularly in the central-western part of the Labrador Sea, but ~~often~~ occasionally occurring in the east as well. Chlorophytes are thought to contribute 1-13 % of total Chl*a* in the global ocean (Swan et al., 2015) and to inhabit transitional regions, where nutrient concentrations become limiting for diatoms, but are not persistently low enough to prevent growth due to nutrient limitation, as occurs in the oligotrophic gyres (Gregg and Casey, 2007; Gregg et al., 2003; Ondrusek et al., 1991). The Labrador Shelf is a dynamic region during springtime, where melting sea ice in May provides a local freshwater input (Head et al., 2003). Melting sea ice provides intense stratification and shallow mixed layers for the phytoplankton and thus access to light, which promotes rapid growth of cold Arctic/ice-related phytoplankton near the sea ice shelf (Fragoso et al., 2016), and which likely stimulates the succession from large diatoms to smaller phytoplankton forms, such as chlorophytes, as nutrients become exhausted. Chlorophytes, as well as Prasinophytes such as *Pyramimonas*, a genus found in high abundances in surface Labrador Shelf waters, might also be associated with melting sea ice and land-fast ice (Palmer et al 2011), given that they have been found blooming (chlorophyll concentration ~ 30 mg Chl*a* m^{-3}) in low salinity melt waters (salinity = 9.1) under the Arctic pack-ice (Gradinger, 1996).

Dinoflagellates were associated with the Irminger Current, where they were occasionally found blooming with diatoms in the warmer, stratified Atlantic waters of the central basin. These blooms dominated by dinoflagellates and Atlantic diatom species, such as *Ephemera planamembranacea* and *Fragilariopsis atlantica*, start later in the season (end of May or June) as thermal stratification develops in the central Labrador Sea (Frajka-Williams and Rhines, 2010, Fragoso et al., 2016). Transition from diatoms to dinoflagellates has been well-documented in the North Atlantic between spring and summer, which occurs because dinoflagellates can use mixotrophic strategies to alleviate nutrient limitation as waters become warmer, highly stratified and nutrient-depleted (Barton et al., 2013; Head and Pepin, 2010; Head et al., 2000; Henson et al., 2012; Leterme et al., 2005). The North Atlantic Oscillation index (NAO) and sea surface temperature (Zhai et al., 2013) appear to influence the relative proportions of diatoms and dinoflagellates as well as the variability in the start date of the North Atlantic bloom. A negative winter phase of NAO is associated with weaker northwest winds over the Labrador Sea and reductions in the depth of winter mixing and supply of nutrients to the upper layers (Drinkwater and Belgrano, 2003). Vertical stability, thermal stratification and the initiation of the spring bloom tend to occur earlier under negative NAO conditions and the proportion of dinoflagellates in the warmer, more nutrient-limited waters may be higher (Zhai et al., 2013). Unfortunately, it was not possible to investigate the influence of NAO on the relative contribution of dinoflagellates and diatoms in the Labrador Sea section of the North Atlantic in this study, given that the sampling period varied from early/mid-May to mid/late-June. On the other hand, abundances of dinoflagellates appeared to be higher in warmer waters (> 5°C), suggesting that the communities were shifting from diatoms to dinoflagellates as the water became stratified and nutrient concentrations decreased.

In this study, a community dominated by *Phaeocystis* and diatoms were observed blooming together in waters of the WGC, in the eastern central part of the Labrador Sea. The occurrence of *Phaeocystis* in these waters has been observed before by several authors (Fragoso et al., 2016; Frajka-Williams and Rhines, 2010; Harrison et al., 2013; Head et al., 2000; Stuart et al., 2000; Wolfe et al., 2000). The eastern part of the Labrador Sea is a region with high eddy kinetic energy during spring (Chanut et al., 2008; Frajka-Williams et al., 2009; Lacour et al., 2015), which causes the accumulation of low-salinity surface waters from the West Greenland Current. This buoyant freshwater layer contains elevated levels of biomass of both *Phaeocystis* and diatoms (this study, Fragoso et al., 2016). ~~*Phaeocystis* and diatoms bloom together in the eastern central Labrador Sea (Fragoso et al., 2016; Frajka-Williams and Rhines, 2010; Harrison et al., 2013; Head et al., 2000; Stuart et al., 2000; Wolfe et al., 2000). This is a region with high eddy kinetic energy during spring (Chanut et al., 2008; Frajka-Williams et al., 2009; Lacour et al., 2015), which causes the accumulation of low-salinity surface waters from the West Greenland Current and confines elevated levels of phytoplankton biomass, presumably of *Phaeocystis* and diatoms, in buoyant freshwater layers (Fragoso et al., 2016).~~ Mesoscale eddies may stimulate growth of *Phaeocystis* and diatoms by inducing partial stratification at irradiance levels that are optimal for their growth, but too low for their competitors (blooms in these eddies usually start in April). Lacour et al. (2015) showed that irradiance levels estimated from satellite-derived photosynthetically active irradiance (PAR) and mixed layer depth climatologies are similar for thermally and haline-stratified spring blooms in the Labrador Sea. Nonetheless, these authors recognise the need for *in situ* measurements to confirm whether Labrador Sea spring blooms, presumably composed of distinctive phytoplankton communities, respond in the same manner to light-mixing regimes. The ability of *Phaeocystis* to grow under dynamic light irradiances explains why they are often found in deeper mixed layers, such as those found in Antarctic polynyas (Arrigo, 1999; Goffart et al., 2000), although this genus can also occur in shallow mixed layers, such as those found close to ice edges (Fragoso and Smith, 2012; Le Moigne et al., 2015).

Mesoscale eddies are also often associated with elevated zooplankton abundances (Frajka-Williams et al., 2009; Yebra et al., 2009). In the Labrador Sea, lower grazing rates have been observed in blooms dominated/co-dominated by colonial *Phaeocystis*, which are often located in these eddies and which may, in turn, explain why this species is dominant (Head and Harris, 1996; Wolfe et al., 2000). Although the exact mechanism that facilitates *Phaeocystis* growth in the north-eastern region of the Labrador Sea is not clear, it is evident that blooms of this species are tightly linked to mesoscale eddies, and that this relationship needs further investigation to better explain their regular reoccurrence in these waters.

4.2 Phytoplankton composition and related biogeochemistry

Particulate organic carbon (POC) and nitrogen (PON) concentrations, as well as the molar ratio of POC:PON varied within distinct hydrographic zones, indicating the presence of different biogeochemical provinces in the Labrador Sea. A canonical Redfield ratio of 6.6 for POC:PON appears to represent the global average (Redfield, 1958), although regional variations on the order of 15 to 20 % have also been reported (Martiny et al., 2013a). The POC:PON appears to be closer to the Redfield ratio of 6.6 in productive sub-Arctic/Arctic waters, such as the northern Baffin Bay (Mei et al., 2005), the north-eastern

Greenland shelf (Daly et al., 1999), and in Fram Strait and the Barents Sea (Tamelander et al., 2012). Crawford et al. (2015), however, recently reported very low POC:PON ratios in oligotrophic Arctic waters of the Beaufort Sea and Canada Basin, where depth-integrated values of the POC:PON ratio were ~ 2.65, much lower than those in more productive domains, such as the sub-Arctic central Labrador Sea (POC:PON ~ 4).

In this study, highly productive surface waters of Arctic origin (near or over the shelves) had higher phytoplankton-derived particulate organic carbon (POCphyto > 43 % of total POC, Fig. 6c), as well as higher and more variable POC:PON ratios (average > 6.9, Fig. 6b) compared with stations influenced by Atlantic water (average POC:PON < 6.3, POCphyto > 35 %, Fig. 6b). ~~In this study, highly productive surface waters of Arctic origin (near or on the shelves) had higher phytoplankton-derived particulate organic carbon (POC_{phyto} > 43 % from total POC), as well as higher and more variable POC:PON ratios (average > 6.9) compared with stations influenced by Atlantic water (average POC:PON < 6.3, POC_{phyto} > 35 %).~~ Diatoms have been suggested to contribute to larger phytoplankton-derived POC in Arctic/sub-Arctic waters (Crawford et al., 2015). The Labrador Shelf region, where blooms are generally dominated by large Arctic/ice-related diatoms (Fragoso et al., 2016), had relatively high contributions of POC_{phyto} (> 50 %) to the total POC, even though smaller phytoplankton forms, such as chlorophytes, were also abundant. Low POC:PON ratios, as well as low POC_{phyto} concentrations were associated with Atlantic waters, which had greater contributions of flagellates (particularly before bloom initiation). Similar findings were reported by Crawford et al. (2015), where low POC_{phyto} was associated with larger contributions of flagellates (< 8 μm) in oligotrophic Arctic waters, such as the Beaufort Sea and Canada Basin. Crawford et al. (2015) also considered that POC:PON ratios might have been reduced by the presence of heterotrophic microbes (bacteria, flagellates and ciliates) since these microorganisms have POC:PON ratios lower than the canonical Redfield ratio of 6.6 (Lee and Fuhrman, 1987; Vrede et al., 2002). Bacteria and other heterotrophic organisms were not quantified in our study, although Li and Harrison (2001) showed that bacterial biomass from surface waters was 62 % greater (average from 1989 to 1998 =13.8 mg C m^{-3}) in the central region than in shelf areas of the Labrador Sea.

Changes in POC:PON may be related to the physiological status of phytoplankton and/or community structure. In the North Water Polynya (Baffin Bay), POC:PON ratios during phytoplankton blooms increased between spring (5.8) and summer (8.9) as phytoplankton responded to nitrate starvation by producing N-poor photo-protective pigments (Mei et al., 2005). Daly et al. (1999) also found high POC:PON ratios (~8.9) in Arctic surface waters dominated by diatoms on the north-eastern Greenland shelf, which were attributed to nutrient limitation. Atlantic waters appear to have an excess of nitrate compared with Arctic waters (Harrison et al., 2013), which could explain why phytoplankton from Atlantic Waters had lower POC:PON ratios. Conversely, Arctic-influenced waters on or near the shelves had higher $Si(OH)_4$:NO_3^- and lower NO_3^-:PO_4^{3-} than those in the central basin in this study, which could also have contributed to the observed high POC:PON ratios.

A few stations in shelf waters of the Labrador Sea also had remarkably high POC:PON ratios (> 10), and low POC_{phyto} contributions, suggesting high contributions of detritus. These waters probably receive higher inputs of Arctic and glacial ice melt, which could introduce POC from external sources. Hood et al. (2015) showed that POC export from glaciers is large, particularly from the Greenland Ice Sheet and it occurs in suspended sediments derived from glacier meltwater. High POC:PON ratios (> 10), particularly in waters where *Phaeocystis* were abundant, may also be linked to the mucilaginous matrix of the *Phaeocystis* colonies (Palmisano et al., 1986). The mucopolysaccharide appears to contain excess carbon, particularly when nutrients start to become depleted and colonies become senescent (Alderkamp et al., 2007; Wassmann et al., 1990).

4.3 Physiological parameters of distinct phytoplankton communities

Accessories pigments (AP) are assumed to have a ubiquitous, global, log-log linear relationship with chlorophyll *a* in aquatic environments (Trees et al., 2000). This linear relationship is often used as an index of quality-control in pigment analysis, which are required due to uncertainties of the quantitative comparability of data among surveys, related to differences in analytical procedures and sample storage methods used in different laboratories. In the current study, the slope of AP to total chlorophyll *a* (TChl*a*) on a logarithm scale (Fig. 7) passed the quality control criteria of slopes ranging from 0.7 to 1.4 and r^2 > 0.90 as applied in previous studies (e.g., Aiken et al., 2009; Peloquin et al., 2013; Thompson et al., 2011) and were within the range observed throughout worldwide aquatic systems (slope from 0.8 to 1.3 compared to 0.86 to 1.03 observed in our study) (Trees et al., 2000). An interesting trend was found where phytoplankton pigment ratios varied clearly within distinct communities in the Labrador Sea. According to our data, phytoplankton communities found in colder waters (of Arctic origin) had lower accessory pigments ratios to total chlorophyll *a* ratio (logAP:logTChl*a*) (slope = 0.86) when compared to communities from warmer waters (Irminger Current from Atlantic origin) (slope = 1.03). Changes in the ratios of logAP:logTChl*a* as a function of phytoplankton community composition has been previously observed by Stramska et al. (2006). These authors showed a higher slope of logAP:logTChl*a* when dinoflagellates were dominant during summer in northern polar Atlantic waters as opposed to lower ratios associated with flagellates in spring. ~~Changes in logAP:logTChl*a* as a function of phytoplankton community composition has been observed before, when Stramska et al. (2006) related a higher slope of logAP:logTChl*a* to dinoflagellates dominating during summer in northern polar Atlantic waters as opposed to lower ratios of flagellates occurring in spring.~~ Trees et al. (2000) and Aiken et al. (2009) also reported lower logAP:logTChl*a* (slope < 1.00) in oligotrophic waters dominated by picoplankton as opposed to higher ratios in upwelling waters where microplankton, particularly diatoms, were dominant.

Environmental parameters, such as nutrients and light conditions, have also been suggested to influence logAP:logTChl*a* regardless of community composition (Trees et al., 2000). Nonetheless, in our study, these ~~two~~ parameters, analysed as nitrate and silicate concentrations and Stratification Index, did not vary with logAP:logTChl*a* (data not shown) as opposed to temperature. Phytoplankton community distributions varied clearly according to temperature with *Phaeocystis* occurring in

colder Arctic waters and dinoflagellates in warmer Atlantic waters. Although both communities were co-dominated by diatoms (relative abundance > 70 % of total chlorophyll), the ratio logAP:logTChl*a* varied considerably, suggesting that either 1) diatom species from both Arctic and Atlantic waters varied intrinsically in pigment composition, or 2) temperature had a physiological effect on the logAP:logTChl*a* ratio. Fragoso et al (2016) has previously observed that the diatom species from Arctic and Atlantic waters of the Labrador Sea during spring varied in terms of species composition. According to the study by Fragoso et al. (2016), the diatoms *Ephemera planamembranacea* and *Fragilariopsis atlantica* were typically found in Atlantic waters, whereas polar diatoms, including *Thalassiosira* species (*T. hyalina*, *T. nordenskioeldii*, for example), in addition to *Bacterosira bathyomphala*, *Fossula arctica*, *Nitzschia frigida* and *Fragilariopsis cylindrus* were all found in Arctic-influenced waters. It is possible that the distinct composition of diatoms from these biogeographical regions might have influenced the pigment composition in these waters. Despite the observed trend of logAP:logTChl*a* varying with temperature, a direct physiological temperature-induced effect in logAP:logTChl*a* is currently unknown."~~Although both communities were co-dominated by diatoms (relative abundance > 70 %), logAP:logTChl*a* varied considerably, suggesting that either 1) diatom species from both Arctic and Atlantic waters varied intrinsically in pigment composition, or 2) temperature had a physiological effect on the logAP:logTChl*a* ratio. Despite the observed trend of logAP:logTChl*a* varying with temperature, a direct temperature-induced effect in logAP:logTChl*a* is unknown~~.

The variation in photosynthetic parameters in the distinct phytoplankton biogeographical provinces demonstrated how each phytoplankton community responds to environmental conditions. Harrison and Platt (1986) found that the photophysiology of phytoplankton from the Labrador Sea is influenced by temperature and irradiance. Nonetheless, phytoplankton composition may also influence the values of the photosynthetic parameters. Light-saturated photosynthetic rates and saturation irradiances, for instance, were higher at stations where diatoms were dominant (> 70 %), as opposed to stations where flagellates were more abundant (from 40 % up to 70 %). Similar findings were reported by (Huot et al., 2013), who observed that light-saturated photosynthetic rates in the Beaufort Sea (Arctic Ocean) were higher for communities composed of large cells, presumably diatoms, compared to smaller flagellates.

Polar phytoplankton communities from shelf waters (east *versus* west) observed in this study had distinctive photo-physiological characteristics. Comparing these blooms, diatom/chlorophyte communities (west) had higher photosynthetic efficiency ($\alpha^B = 9.2 \times 10^{-2}$ mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$), lower onset light-saturation irradiance ($E_k = 29$ W m^{-2}) and higher photo-inhibition ($\beta = 16 \times 10^{-4}$ mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$) than communities from the east. This suggests that the community located in the Labrador Shelf waters (west) was more light-stressed compared to the community observed in the east (diatom/*Phaeocystis*). Haline-stratification due to the influence of Arctic waters occur in both regions during spring, contributing to the shallow mixed layer depth (<33 m) observed (Table 5). However, waters from the Labrador Shelf (west, Cluster C3a) were more stratified than the Greenland Shelf (cluster B, see stratification index (SI) values, Table 5) because of the local sea ice melt observed in this area, which contributes to increased stratification in this region. The diatom species

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observed on the Labrador Shelf were mostly sea-ice related (*Fragilariopsis cylindrus*, *Fossula arctica*, *Nitzschia frigida*) compared to pelagic species observed in the Greenland Shelf waters (*Thalassiosira gravida*, for example) (Fragoso et al., 2016). Sensitivity of sea-ice related diatoms to irradiance > 15 µmol photons m^{-2} s^{-1} has been reported (Alou-Font et al., 2016), which can help explaining why phytoplankton communities from the west were photo-inhibited.

The community dominated by *Phaeocystis*/diatoms located near Greenland (east) had the inverse pattern: low photosynthetic efficiency (average α^B = 6.8 × 10^{-2} mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$) and high onset light-saturation irradiances (E_k = 60 W m^{-2}). This pattern in diatom/*Phaeocystis*-dominated communities mean that photosynthetic rates were relatively low at high light intensities, although photo-inhibition was low (β = 4 × 10^{-4} mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$). *Phaeocystis antarctica*, widespread in Antarctic waters, *relies* heavily on photo-damage recovery, such as D1 protein repair (Kropuenske et al., 2009), which could explain how these communities overcome photo-inhibition. Stuart et al. (2000), however, found a high photosynthetic efficiency (α^B) for a population dominated by *Phaeocystis* near Greenland and attributed this to the small cell size of *Phaeocystis*. However, in addition to the exposure of ice-related diatoms to high light levels due to increased stratification, the high concentration of chlorophytes and prasinophytes, which are also small in cell size, might also explain the higher α^B observed in the Labrador Shelf waters (west, cluster C3a) when compared to values from a community dominated by diatom/*Phaeocystis* blooms (east, cluster B).

~~Polar phytoplankton communities from shelf waters (east *versus* west) observed in this study had distinctive photo-physiological characteristics. Diatom/*Phaeocystis* dominated communities from waters located near Greenland (east) had low photosynthetic efficiency (average α^B = 6.8 × 10^{-2} mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$) and high onset light-saturation irradiances (E_k = 60 W m^{-2}), while diatom/chlorophyte dominated communities on or near the Labrador Shelf (west) had the reverse (E_k = 29 W m^{-2}, α^B = 9.2 × 10^{-2} mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$). Low photosynthetic efficiency and high light saturation irradiance in diatom/*Phaeocystis* dominated communities mean that photosynthetic rates were relatively low at high light intensities, although photo-inhibition was low (β = 4 × 10^{-4} mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$). *Phaeocystis antarctica*, widespread in Antarctic waters, relies heavily on photo-damage recovery, such as D1 protein repair (Kropuenske et al., 2009), which could explain how these communities overcome photo-inhibition. These results were inconsistent with those reported by Stuart et al. (2000); however, who found a higher photosynthetic efficiency (α^B) for a population dominated by *Phaeocystis* near Greenland compared with that of a diatom dominated population near the Labrador coast. Stuart et al. (2000) attributed the higher α^B to the smaller cell size of *Phaeocystis*. In the current study, however, chlorophytes were present in high concentrations on the Labrador Shelf, which may explain the discrepancy between these results.~~

Phytoplankton communities from Atlantic waters (co-dominated by diatoms and dinoflagellates) were highly susceptible to photo-inhibition (β = 29 × 10^{-4} mg C [mg Chl*a*] h^{-1} [W m^{-2}]$^{-1}$) compared with the other communities in the Labrador Sea. Days are longer and solar incidence is higher in June compared to May at these latitudes (Harrison et al., 2013). Dinoflagellates

were found to bloom in the central Labrador Sea in June as a consequence of increased thermal stratification. To cope with high light levels and potential photo-damage, this phytoplankton community appeared~~Days are longer and solar incidence is higher in June as compared to May at these latitudes (Harrison et al., 2013), which, in this study, was the time when dinoflagellates bloomed in the central Labrador Sea as a consequence of thermal stratification, which explains the sensitivity of this community to high light levels. To cope with photo-damage, this phytoplankton community appeared~~ to increase the levels of photoprotective pigments, such as those used in the xanthophyll cycle (diadinoxanthin (DD) + diatoxanthin (DT)). These communities also had high diatoxanthin levels compared with the other phytoplankton communities in this study, suggesting that the community was experiencing higher light intensities (Moisan et al., 1998). Increases in photoprotective pigments, including (DD+DT)/Chla, have been reported to occur in Arctic phytoplankton communities from spring to summer presumably as a response to higher irradiance (Alou-Front et al 2016). Thus, photoprotective capacity can be a key determinant for phytoplankton survival and may also be related to the taxonomic segregation observed in Arctic and Atlantic phytoplankton communities.~~These communities also had high diatoxanthin levels compared with the polar phytoplankton communities, suggesting that the community was experiencing higher light intensities (Moisan et al., 1998)~~.

4.4 Phytoplankton communities assessed by HPLC and CHEMTAX methods

~~Phytoplankton pigments and CHEMTAX methods provide information about phytoplankton community structure, and are especially powerful when used in conjunction with microscopic analysis (light and high resolution scanning electron microscopy) (Coupel et al., 2012, 2015; Eker-develi et al., 2012; Muylaert et al., 2006), cytometry (Devilla et al., 2005; Fujiki et al., 2009) and molecular techniques (Not et al., 2007; Piquet et al., 2014; Zhang et al., 2015). However, choosing the pigment markers in the CHEMTAX analysis and interpreting the results are not always straightforward and, therefore, conclusions need to be drawn with caution. Many environmental factors (primarily light and nutrients) (DiTullio et al., 2007; Henriksen et al., 2002; van Leeuwe and Stefels, 1998, 2007), in addition to natural variability among species from the same classes or even strains of the same species (Zapata et al., 2004) affect accessory pigment levels and ratios to chlorophyll *a*, which could introduce some uncertainties when applying CHEMTAX. Thus, phytoplankton abundances determined using CHEMTAX represent approximations based on pigment distributions. These limitations can, however, be lessened when this technique is combined with existing knowledge of main phytoplankton groups occurring in the samples through microscopic identification.~~

A number of studies have used CHEMTAX methods to determine phytoplankton community structure in Arctic/subarctic waters (Coupel et al., 2012, 2015; Lovejoy et al., 2007; Piquet et al., 2014; Vidussi et al., 2004; Zhang et al., 2015). Spring phytoplankton communities from the Labrador Sea have already been investigated in detail (Fragoso et al., 2016), although the analysis did not include most nano- and pico-flagellates (except cryptophytes and *Phaeocystis pouchetii*) and were done over only four years (2011-2014) at selected stations along the L3 (=AR7W) transect. Here, we have combined phytoplankton information from Fragoso et al. (2016) with additional pigment analyses. Although cross comparison among these two

techniques (carbon biomass estimated from microscopic counts *versus* algal ~~class~~ group chlorophyll *a* estimated from CHEMTAX) should not be expected to give exactly equivalent results, given that most flagellates observed in the pigment analysis were not counted under the microscope, some comparability should be possible, at least for the larger cells (e.g. diatoms).

Phaeocystis (r^2 = 0.79) and diatom (r^2 = 0.74) biomasses were well correlated when carbon biomasses estimated from microscopic counts when compared with CHEMTAX-derived algal chlorophyll *a* biomass (data not shown). Diatoms are the group that usually show the best agreement between the two methods of biomass estimations (Vidussi et al. 2004, Coupel et al 2015, Mendes et al 2012). For *Phaeocystis*, a positive relationship between the two methods of biomass estimation (CHEMTAX and microscopy) confirms that using chlorophyll *c3* was appropriate for detecting and quantifying *Phaeocystis* biomass in the Labrador Sea. Similar associations have been observed for *Phaeocystis* from boreal waters (e.g. *P. pouchetii* and *P. globosa* ; Antajan et al., 2004; Muylaert et al., 2006; Stuart et al., 2000; Wassmann et al., 1990), while other pigment markers have been used elsewhere, e.g. 19- hexanoyloxyfucoxanthin, which is characteristic of *Phaeocystis antarctica* in austral polar waters (Arrigo et al., 2010, 2014; Fragoso and Smith, 2012; Fragoso, 2009). Dinoflagellates gave a poor correlation between biomass estimates made using the two methods (r^2 = 0.12, data not shown). A lack of or weak relationship between both biomass estimations for dinoflagellates has been previously reported in Artic waters (Vidussi et al 2004 Coupel et al 2005). The argument for this inconsistency is that some heterotrophic dinoflagellates, which usually lack photosynthetic pigments unless they ingest a prey that contains them, might have been included in the microscopic counts, and it is possible that the same occurred in Fragoso et al. (2016). Cryptophyte biomass estimates from both methods were not related (data not shown), likely as the biomass of this group was underestimated in microscopic counts. Inconsistences between CHEMTAX and microscopy methods of estimating biomasses have also been observed in nanoflagellates and this is assumed to be because of the low accuracy of visual microscopic counts (Gieskes and Kraay, 1983; Coupel et al 2015).

~~*Phaeocystis* (r^2 = 0.79) and diatom (r^2 = 0.74) biomasses were well correlated when carbon biomasses estimated from microscopic counts were compared with CHEMTAX-derived algal chlorophyll *a* biomass (data not shown). This confirms that using chlorophyll *c3* was appropriate for detecting and quantifying *Phaeocystis* biomass in the Labrador Sea. Similar associations have been observed in *Phaeocystis* from boreal waters (e.g. *P. pouchetii* but *P. globosa* as well; Antajan et al., 2004; Muylaert et al., 2006; Stuart et al., 2000; Wassmann et al., 1990), while other pigment markers have been used elsewhere, e.g. 19- hexanoyloxyfucoxanthin, which is characteristic of *Phaeocystis antarctica* in austral polar waters (Arrigo et al., 2010, 2014; Fragoso and Smith, 2012; Fragoso, 2009). Dinoflagellates gave a poor correlation between biomass estimates made using the two methods (r^2 = 0.12, data not shown) possibly because some heterotrophic dinoflagellates, which lack photosynthetic pigments, might have been included in the microscopic counts from Fragoso et al. (2016). Cryptophyte biomass estimates were not related (data not shown), likely because their biomass was underestimated in microscopic counts.~~

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5. Conclusions

In this study, we have provided a geographical description of phytoplankton community structure in spring and early summer surface waters of the Labrador Sea based on pigment data from over a decade of sampling (2005-2014). Phytoplankton communities and their photophysiological and biogeochemical signatures were assessed using CHEMTAX, so that a geographical baseline of the major phytoplankton groups has been provided for the central Labrador Sea and its adjacent continental shelves. In spite of interannual variability (due to differences in survey dates and natural variability), spring phytoplankton communities showed distinct spatial variations from east to west and there were clear temporal differences between May and June. The main conclusions of our study are that: 1) diatoms contributed most to the chlorophyll *a* in waters where phytoplankton blooms were observed (> 3 mg Chl*a* m^{-3}); while other groups (chlorophytes, dinoflagellates and *Phaeocystis*) were geographically segregated within distinct hydrographical zones; 2) a diverse mixed assemblage dominated by flagellates from several groups occurred in low chlorophyll, pre-bloom conditions in the central Labrador Sea; and 3) different phytoplankton communities had different ratios of accessory pigments to total chlorophyll *a*; and 4) POC:PON ratios were influenced by phytoplankton community composition, as well as freshwater input of allochthonous carbon in shelf waters which have nearby sources (e.g. melting glacial and sea- ice and river outflows).

~~Marine phytoplankton respond rapidly to changes in the ocean, and their responses directly impact local marine food webs and global biogeochemical cycles. Climate-driven processes modify the factors, including light availability, nutrient input and grazing pressure that shape phytoplankton physiological traits and alter community structure (Litchman et al., 2012; Montes-Hugo et al., 2009). High latitude seas, particularly the Labrador Sea, are regions that are extremely vulnerable to climate change and often show similar patterns of variability on interannual and decadal scales across the entire domain (Yashayaev and Seidov, 2015; Yashayaev et al., 2015) and they could, therefore, be subject to rapid shifts in phytoplankton biomass, size and species composition. Although climate-induced responses of phytoplankton communities in vulnerable regions are difficult to predict, the long-term observations of these communities reported here and the analysis of their biogeochemical and physiological signatures are important in order to create a baseline for evaluation of changes that will occur in the future, as greenhouse gas-driven warming continues in this and other regions of the global ocean.~~

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organic carbon and nitrogen and hydrographic data are also acknowledged. We would like to thank Simon Wright (Australian Antarctic Division) for providing us with a copy of the CHEMTAX software v.1.95. We are grateful to the reviewers who offered useful suggestions to improve the manuscript. G.M.F. was funded by a Brazilian PhD studentship, Science without Borders (CNPq, 201449/2012-9). This research was also partially funded by UK Ocean Acidification, a National Environment Research Council grant (NE/H017097/1) through an added value award to AJP.

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Table 1. Research cruises, sampling dates and number of samples per cruise (n) where pigment data were collected in the Labrador Sea during early spring and late summer (2005-2014).

Cruise	Dates	Year	n
HUD-2005-16	29 May - 3 June	2005	25
HUD-2006-019	23 May - 31 May	2006	12
HUD-2007-011	11 May - 21 May	2007	32
HUD-2008-009	22 May - 29 May	2008	25
HUD-2009-015	18 May - 23 May	2009	26
HUD-2010-014	14 May - 24 May	2010	27
HUD-2011-009	11 May - 17 May	2011	33
HUD-2012-001	3 June - 11 June	2012	30
HUD-2013-008	9 May - 21 May	2013	27
JR302	10 June - 24 June	2014	16

Table 2. List of phytoplankton pigments and their distributions in algae groups, abbreviations and formulas.

Abbreviation	Name	Characteristic of the pigment	Present in/ Index of/Formula
PSC	Photosynthetic carotenoid	Light harvesting	All algae
PPC	Photoprotective carotenoid	Photoprotection	All algae
PPP	Photosynthetic pigment	Light harvesting	All algae
But-fuco	19'-butanoyloxyfucoxanthin	PSC	Prymnesiophytes, crysophytes and dinoflagellates Type 2* (lacking Peridin)
Hex-fuco	19'-hexanoyloxyfucoxanthin	PSC	Major in prymesiophytes and dinoflagellates Type 2* (lacking Peridin)
Allo	Alloxanthin	PPC	Cryptophytes
α-Car	α-carotene	PPC	Dominant in prochlorophytes, rhodophyte and cryptophyte
β-Car	β-carotene	PPC	Dominant in cyanobacteria, prochlorophytes, chlorophytes, prasinophytes, euglenophytes and diatoms
Chl *b*	Chlorophyll *b*	PPP	Chlorophytes, prasinophytes, euglenophytes
Chl $c_1 + c_2$	Chlorophyll $c_1 + c_2$	PPP	Diatoms, prymnesiophytes, dinoflagellates, cryptophytes, chrysophytes and raphidophytes
Chl c_3	Chlorophyll c_3	PPP	Prymnesiophytes, chrysophytes and dinoflagellates Type 2* (lacking Peridin)
Chlide *α*	Chlorophyllide *a*	Degradation product of Chl *a*	Senescent phytoplankton
DD	Diadinoxanthin	PPC	Diatoms, prymnesiophytes, dinoflagellates, chrysophytes and raphidophytes
DT	Diatoxanthin	PPC	Diatoms, prymnesiophytes, dinoflagellates, chrysophytes and raphidophytes
Fuco	Fucoxanthin	PSC	Diatoms, prymnesiophytes, chysophytes, pelagophytes and dinoflagellates Type 2* (lacking Peridin)
Chl *a*	Chlorophyll *a*	PPP	All phytoplankton except *Prochlorococcus*
Peri	Peridinin	PSC	Dinoflagellates Type 1*
Pras	Prasinoxanthin	PPC	Prasinophytes Type 1**
Viola	Violaxanthin	PPC	Chlorophytes, prasinophytes and eustigmatophytes
Zea + Lut	Zeaxanthin + Lutein	PPC	Cyanobacteria, *Prochlorococcus*, chlorophytes and prasinophytes Type 2**
TChl*a*	Total chlorophyll *a*		Chl *a* + Chlide *α*
TC	Total carotenoids	Include all carotenoids	But-fuco + Hex-fuco + Allo + α-Car + β-Car + DD + DT + Fuco + Peri + Pras + Viola + Zea + Lut
AP	Accessory pigments	Include all pigments except Chl *a*	TC + Chl *b* + Chl $c_1 + c_2$ + Chl c_3

According to Jeffrey et al (1997) or *Higgins et al (2011) or **Vidussi et al (2004).

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Table 2. List of phytoplankton pigments and their distributions in algae classes, abbreviations and formulas.

Abbreviation	Name	Characteristic of the pigment	Present in/ Index of/Formula	Ref.
PSC	Photosynthetic carotenoid	Light harvesting	All algae	
PPC	Photoprotective carotenoid	Photoprotection	All algae	
PPP	Photosynthetic pigment	Light harvesting	All algae	
BUT19	19'-butanoyloxyfucoxanthin	PSC	Prymnesiophytes and crysophytes	1
HEX19	19'-hexanoyloxyfucoxanthin	PSC	Diatoms, prymnesiophytes and some dinoflagellates	2
ALLOX	Alloxanthin	PPC	Cryptophytes	1
ACAROT	α-carotene	PPC	Various	1
BCAROT	β-carotene	PPC	Various	1
CHLB	Chlorophyll *b*	PPP	Chlorophytes, prasinophytes, euglenophytes	1
CHLC12	Chlorophyll c_1+ c_2	PPP	Diatoms, prymnesiophytes, dinoflagellates, chrysophytes and raphidophytes	1
CHLC3	Chlorophyll c_3	PPP	Prymnesiophytes, chrysophytes	1
CHLIDEA	Chlorophyllide *a*	Degradation product of CHLA	Senescent phytoplankton	
DIADINOX	Diadinoxanthin	PPC	Diatoms, prymnesiophytes, dinoflagellates, chrysophytes and raphidophytes	1
DIATOX	Diatoxanthin	PPC	Diatoms, prymnesiophytes, dinoflagellates, chrysophytes and raphidophytes	1
FUCOX	Fucoxanthin	PSC	Diatoms, prymnesiophytes, raphidophytes and some dinoflagellates	1
CHLA	Chlorophyll *a*	PPP	All phytoplankton except *Prochlorococcus*	1
PERID	Peridinin	PSC	Some dinoflagellates	1
PRASINO	Prasinoxanthin	PPC	Some prasinophytes	1
VIOLAX	Violaxanthin	PPC	Chlorophytes, prasinophytes and eustigmatophytes	1
ZEA	Zeaxanthin	PPC	Cyanobacteria, *Prochlorococcus*, chlorophytes	1
TChl*a*	Total chlorophyll *a*		CHLA + CHLIDEA	
TC	Total carotenoids	Include all carotenoids	BUT19 + HEX19 + ALLOX + ACAROT + BCAROT + DIADINOX + DIATOX + FUCOX + PERID + PRASINO + VIOLAX + ZEA	
AP	Accessory pigments	Include all pigments except CHLA	TC + CHLB + CHLC12 + CHLC3	
FUCOX/AP	Fucoxanthin to accessory pigments ratio	-	FUCOX/AP	-

[1](Jeffrey et al., 1997), [2](Higgins et al., 2011).

Table 3 - Initial ratio matrix of accessory pigment to chlorophyll *a* for distinct algal groups for each cluster group. *Ref refers to the literature where the pigment ratios were extracted. See explanation of each group in the methods section.

Region	I & II (Eastern Labrador Sea)										
Group / Pigment	Chl *b*	Chl c_3	Fuco	Peri	Zea + Lut	Allo	But-fuco	Hex-fuco	Pras	Chl *a*	*Ref
Prasinophyte 1	0.512	0	0	0	0	0	0	0	0.075	1	2
Prasinophyte 2	0.738	0	0	0	0.008	0	0	0	0	1	2
CHLORO-1	0.339	0	0	0	0.047	0	0	0	0	1	4
Dinoflagellates	0	0	0	0.600	0	0	0	0	0	1	5
Cryptophytes	0	0	0	0	0	0.673	0	0	0	1	2
Phaeocystis	0	0.208	0.350	0	0	0	0	0	0	1	1
HAPTO-6	0	0.155	0.195	0	0	0	0.019	1.054	0	1	4
Chryso/Pelagophyte	0	0.114	0.398	0	0	0	0.595	0	0	1	2
Cyanobacteria	0	0	0	0	0.232	0	0	0	0	1	3
Diatoms	0	0	1.229	0	0	0	0	0	0	1	2
Region	III & V (Central Labrador Sea)										
Group / Pigment	Chl *b*	Chl c_3	Fuco	Peri	Zea + Lut	Allo	But-fuco	Hex-fuco	Pras	Chl *a*	*Ref
Prasinophyte 1	0.512	0	0	0	0	0	0	0	0.075	1	2
Prasinophyte 2	0.738	0	0	0	0.008	0	0	0	0	1	2
CHLORO-1	0.339	0	0	0	0.047	0	0	0	0	1	4
Dinoflagellates	0	0	0	0.600	0	0	0	0	0	1	5
Dino-2	0	0.179	0.300	0	0	0	0.081	0.194	0	1	4
Cryptophytes	0	0	0	0	0	0.673	0	0	0	1	2
HAPTO-6	0	0.155	0.195	0	0	0	0.019	1.054	0	1	4
Chryso/Pelagophyte	0	0.114	0.398	0	0	0	0.595	0	0	1	2
Cyanobacteria	0	0	0	0	0.232	0	0	0	0	1	3
Diatoms	0	0	1.229	0	0	0	0	0	0	1	2
Region	IV (Western Labrador Sea)										
Group / Pigment	Chl *b*	Chl c_3	Fuco	Peri	Zea + Lut	Allo	But-fuco	Hex-fuco	Pras	Chl *a*	*Ref
Prasinophyte 1	0.512	0	0	0	0	0	0	0	0.075	1	2
Prasinophyte 2	0.738	0	0	0	0.008	0	0	0	0	1	2
CHLORO-1	0.339	0	0	0	0.047	0	0	0	0	1	4
Dino-2	0	0.179	0.300	0	0	0	0.081	0.194	0	1	4
Dinoflagellates	0	0	0	0.600	0	0	0	0	0	1	5
Cryptophytes	0	0	0	0	0	0.673	0	0	0	1	2
Prymnesiophyte 1	0	0.038	0.416	0	0	0	0	1.108	0	1	2
Chryso/Pelagophyte	0	0.114	0.398	0	0	0	0.595	0	0	1	2
Diatoms	0	0	1.229	0	0	0	0	0	0	1	2

[1](Antajan et al., 2004), [2](Vidussi et al., 2004), [3](Muylaert et al., 2006), [4](Higgins et al., 2011), [5](Coupel et al., 2015)

Table 3. Final ratio matrix of accessory pigment to chlorophyll *a* for distinct algal classes for each cluster group.

Region	I & II (Eastern Labrador Sea)										
Class / Pigment	CHLB	CHLC3	FUCOX	PERID	ZEA	ALLOX	BUT19	HEX19	PRASINO	CHLA	Ref
Prasinophyte 1	0.459	0	0	0	0	0	0	0	0.075	1	2
Prasinophyte 2	0.650	0	0	0	0.008	0	0	0	0	1	2
Chlorophyte	0.168	0	0	0	0.040	0	0	0	0	1	2
Dinoflagellates	0	0	0	0.609	0	0	0	0	0	1	2,5
Cryptophycea	0	0	0	0	0	0.785	0	0	0	1	2
Phaeocystis	0	0.167	0.188	0	0	0	0	0	0	1	1
HAPTO 6	0	0.199	0.270	0	0	0	0.021	1.261	0	1	4
Chryso/Pelagophyte	0	0.120	0.454	0	0	0	0.589	0	0	1	2
Cyanobateria	0	0	0	0	0.262	0	0	0	0	1	3
Diatoms	0	0	0.328	0	0	0	0	0	0	1	2
Region	III & V (Central Labrador Sea)										
Class / Pigment	CHLB	CHLC3	FUCOX	PERID	ZEA	ALLOX	BUT19	HEX19	PRASINO	CHLA	Ref
Prasinophyte 1	0.316	0	0	0	0	0	0	0	0.129	1	2
Prasinophyte 2	0.716	0	0	0	0.008	0	0	0	0	1	2
Chlorophyte	0.171	0	0	0	0.025	0	0	0	0	1	2
Dinoflagellates	0	0	0	0.681	0	0	0	0	0	1	2,5
Dino-2	0	0.290	0.348	0	0	0	0.060	0.168	0	1	4
Cryptophycea	0	0	0	0	0	0.674	0	0	0	1	2
HAPTO 6	0	0.081	0.202	0	0	0	0.018	1.549	0	1	4
Chryso/Pelagophyte	0	0.049	0.184	0	0	0	0.264	0	0	1	2
Cyanobateria	0	0	0	0	0.142	0	0	0	0	1	3
Diatoms	0	0	0.512	0	0	0	0	0	0	1	2
Region	IV (Western Labrador Sea)										
Class / Pigment	CHLB	CHLC3	FUCOX	PERID	ZEA	ALLOX	BUT19	HEX19	PRASINO	CHLA	Ref
Prasinophyte 1	0.216	0	0	0	0	0	0	0	0.078	1	2
Prasinophyte 2	1.081	0	0	0	0.012	0	0	0	0	1	2
Chlorophyte	0.113	0	0	0	0.045	0	0	0	0	1	2
Dinoflagellates	0	0	0	0.785	0	0	0	0	0	1	2,5
Dino-2	0	0.028	0.049	0	0	0	0.018	0.040	0	1	4
Cryptophycea	0	0	0	0	0	0.703	0	0	0	1	2
HAPTO 7	0	0.030	0.389	0	0	0	0	1.218	0	1	4
Chryso/Pelagophyte	0	0.056	0.470	0	0	0	0.613	0	0	1	2
Diatoms	0	0	0.343	0	0	0	0	0	0	1	2

[1](Antajan et al., 2004), [2](Vidussi et al., 2004), [3](Muylaert et al., 2006), [4](Higgins et al., 2011), [5](Coupel et al., 2015)

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Table 4 – Results of the Redundancy Analyses (RDA) with the eigen-values, taxa-environmental correlations and percentages of variance explained used in the analysis (a). Automatic forward selection (a *posteriori* analysis) was used to determine the environmental variable(s) that best explain the variance of the data (b). The subset of environmental variable(s) that significantly explained phytoplankton distribution are referred to marginal effects (λ_1) when analysed individually, or conditional effects (λa) when analysed additively in the model (b). Explanatory variables are temperature (°C), salinity, nitrate (NO_3^-; µmol L^{-1}), phosphate (PO_4^{3-}; µmol L^{-1}), silicate ($Si(OH)_4$; µmol L^{-1}) and Stratification Index (SI) (kg m^{-4}). Significant p-values ($p < 0.05$) represents the variables that explain the variation in the analyses. ~~Results of the Redundancy Analyses (RDA) with the effects, eigenvalues and percentages of variance explained used in the analysis. Marginal (λ_1) and conditional effects (λ_a) refers to the absolute and additional effects, respectively, of the environmental variable (s) used in the RDA analysis after the automatic forward selection. Explanatory variables are temperature (°C), salinity, nitrate (NO_3^-), phosphate (PO_4^{3-}), silicate ($Si(OH)_4$) (µmol L^{-1}) and Stratification Index (SI) (kg m^{-4}). Significant p-values ($p < 0.05$) represents the variables that significantly explains the variation in the analyses.~~

a) Axes	1	2	3	4	Total variance
Eigen-values	0.26	0.04	0.005	0	1
Taxa-environment correlations	0.68	0.4	0.321	0.25	
Cumulative percentage variance					
of species data	25.7	29.9	30.3	30.7	
of species-environment relation	83.5	97.2	98.8	99.8	
Sum of all eigenvalues					1
Sum of all canonical eigenvalues					0.31

b) Marginal Effects		Conditional Effects			
Variable	λ_1	Variable	λ_a	*P*	*F*
$Si(OH)_4$	0.2	$Si(OH)_4$	0.2	0.001	61.7
NO_3^-	0.19	Temperature	0.05	0.001	17.3
PO_4^{3-}	0.17	Salinity	0.02	0.002	6.94
Salinity	0.09	NO_3^-	0.01	0.016	4.31
Temperature	0.07	PO_4^{3-}	0.02	0.002	7.22
SI	0.06	SI	0.01	0.153	1.72

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Marginal Effects			Conditional Effects			
Variable	λ_1		Variable	λ_a	*P*	*F*
$Si(OH)_4$	0.2		$Si(OH)_4$	0.2	0.001	61.65
NO_3^-	0.19		Temperature	0.05	0.001	17.3
PO_4^{3-}	0.17		Salinity	0.02	0.002	6.94
Salinity	0.09		NO_3^-	0.01	0.016	4.31
Temperature	0.07		PO_4^{3-}	0.02	0.002	7.22
SI	0.06		SI	0.01	0.153	1.72
Axes	1	2	3	4		Total variance
Eigen-values	0.257	0.042	0.005	0.003		1
Taxa-environment correlations	0.676	0.404	0.321	0.245		
Cumulative percentage variance						
—of species data	25.7	29.9	30.3	30.7		
—of species-environment relation	83.5	97.2	98.8	99.8		
Sum of all eigenvalues						1
Sum of all canonical eigenvalues						0.307

Test of significance of first canonical axis: eigen-value = 0.257; F-ratio = 84.938; P-value = 0.002.

Test of significance of all canonical axis: trace = 0.307; F-ratio = 18.184; P-value = 0.002.

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Table 5 – Average, standard errors and number of observations (in parenthesis) of environmental and biological variables of each cluster group. MLD = mixed layer depth, SI= Stratification index, NO_3^- = nitrate, PO_4^{3-} = phosphate, $Si(OH)_4$ = silicate, DT= diatoxanthin, DD= diadinoxanthin, POC= particulate organic carbon, PON= particulate organic nitrogen, POC_{phyto} = phytoplankton-derived particulate organic carbon, α^B = initial slope of the photosynthesis-irradiance curve, P_m^B = maximum normalised photosynthesis, E_k = half-saturation irradiance, Es = saturation irradiance.

	Cluster A		Cluster B		Cluster C3a		Cluster C3b		Cluster C2		Cluster C1	
	DIAT (> 99%)		DIAT + PHAEO		DIAT + CHLORO		MIXED		DIATO + DINO		DIAT (> 93%)	
Temperature (°C)	2.8 ± 2.4	(17)	2.0 ± 1.8	(46)	1.6 ± 1.9	(62)	3.4 ± 1.9	(92)	4.8 ± 1.5	(32)	1.4 ± 1.7	(4)
Salinity	33.4 ± 1.5	(17)	33.7 ± 0.8	(46)	33.1 ± 1.2	(62)	34.1 ± 1.0	(92)	34.4 ± 0.5	(32)	33.0 ± 1.6	(4)
MLD (m)	32.2±43.8	(17)	32.6 ± 23.4	(46)	31.2 ± 28.5	(62)	59 ± 71.1	(92)	29.8 ± 17.0	(32)	16.0 ± 4.2	(4)
SI × 10^{-3} (kg m^{-4})	9.1 ± 6.3	(17)	6.3 ± 5.7	(46)	10.7 ± 8.5	(62)	5.0 ± 6.8	(92)	6.1 ± 4.5	(31)	6.6 ± 8.5	(4)
NO_3^- (µmol L^{-1})	2.9 ± 4.7	(17)	2.7 ± 3.5	(46)	3.4 ± 4.3	(58)	8.4 ± 4.1	(83)	3.7 ± 3.9	(32)	3.8 ± 6.8	(4)
$Si(OH)_4$ (µmol L^{-1})	2.2 ± 2.7	(17)	2.8 ± 2.1	(46)	3.5 ± 2.4	(58)	5.4 ± 2.2	(83)	3.0 ± 2.2	(32)	2.3 ± 3.4	(4)
PO_4^{3-} (µmol L^{-1})	0.3 ± 0.3	(17)	0.3 ± 0.2	(45)	0.4 ± 0.2	(55)	0.7 ± 0.2	(79)	0.3 ± 0.2	(32)	0.4 ± 0.3	(4)
$Si(OH)_4$:NO_3^-	6.0 ± 11.8	(14)	3.6 ± 7.9	(37)	8.5 ± 18.2	(54)	1.1 ± 1.5	(82)	1.6 ± 1.8	(32)	3.9 ± 4.4	(4)
NO_3^-:PO_4^{3-}	8.2 ± 6.7	(11)	5.2 ± 5.0	(45)	5.9 ± 5.8	(55)	11.4 ± 4.1	(79)	8.7 ± 4.6	(32)	5.5 ± 7.1	(4)
Chlorophyll *a* (mg Chl*a* m^{-3})	3.8 ± 4.7	(17)	5.5 ± 4.8	(45)	7.7 ± 5.6	(59)	2.0 ± 1.7	(91)	4.0 ± 1.8	(31)	8.8 ± 9.6	(4)
DT:(DT+DD)	0.01±0.03	(16)	0.02±0.05	(44)	0.04±0.05	(62)	0.10±0.01	(92)	0.08±0.07	(32)	0.02±0.04	(4)
(DD+DT):Chl*a*	0.08±0.07	(17)	0.03±0.03	(46)	0.04±0.02	(62)	0.07±0.03	(92)	0.12±0.03	(32)	0.07±0.04	(4)
POC (mg C m^{-3})	245 ± 90	(4)	498 ± 198	(27)	533 ± 198	(45)	234 ± 145	(63)	512 ± 179	(15)	393 ± 418	(2)
PON (mg N m^{-3})	39 ± 16	(4)	65 ± 23	(27)	74 ± 30	(45)	38 ± 26	(64)	83 ± 33	(15)	42 ± 41	(2)
POC_{phyto} (%)	23.0 ± 5.2	(4)	49.2 ± 29.5	(26)	60.9 ± 25.6	(44)	33.3 ± 10.1	(64)	36.0 ± 11.4	(15)	37.8 ± 1.3	(2)
POC:PON	6.5 ± 1.2	(4)	7.8 ± 2.1	(27)	7.5 ± 2.1	(45)	6.6 ± 1.3	(64)	6.2 ± 0.9	(15)	8.6 ± 1.6	(2)
α^B × 10^{-2} (mgC[mgChl*a*]h^{-1}[Wm^{-2}]$^{-1}$)	-		6.8 ± 6	(9)	9.2 ± 10	(10)	7.1 ± 4	(18)	7.1 ± 1.5	(4)	-	
P_m^B (mgC[mgChl*a*]h^{-1})	-		3.0 ± 1.2	(9)	2.3 ± 0.8	(10)	2.3 ± 0.6	(18)	3.3 ± 0.7	(4)	-	
E_k (W m^{-2})	-		60 ± 33	(9)	29 ± 13	(10)	39 ± 14	(18)	46 ± 5	(4)	-	
Es (W m^{-2})	-		62 ± 32	(9)	35 ± 18	(10)	43 ± 18	(18)	56 ± 8	(4)	-	
β × 10^{-4} (mgC[mgChl*a*]h^{-1}[Wm^{-2}]$^{-1}$)	-		4 ± 7	(9)	16 ± 23	(10)	10 ± 16	(18)	29 ± 24	(4)	-	

	Cluster A		Cluster B		Cluster C3a		Cluster C3b		Cluster C2		Cluster C1	
Temperature (°C)	2.8 ± 0.6	(17)	2.0 ± 0.3	(46)	1.6 ± 0.2	(62)	3.4 ± 0.2	(92)	4.8 ± 0.3	(32)	1.4 ± 0.9	(4)
Salinity	33.4 ± 0.4	(17)	33.7 ± 0.1	(46)	33.1 ± 0.2	(62)	34.1 ± 0.1	(92)	34.4 ± 0.1	(32)	33.0 ± 0.8	(4)
MLD (m)	32.2 ± 10.6	(17)	32.6 ± 3.4	(46)	31.2 ± 3.6	(62)	59 ± 7.4	(92)	29.8 ± 3.0	(32)	16.0 ± 2.1	(4)
SI × 10^{-3} ($kg\ m^{-4}$)	9.1 ± 1.5	(17)	6.3 ± 0.8	(46)	10.7 ± 1.1	(62)	5.0 ± 0.7	(92)	6.1 ± 0.8	(31)	6.6 ± 4.3	(4)
NO_3^- ($\mu mol\ L^{-1}$)	2.9 ± 1.1	(17)	2.7 ± 0.5	(46)	3.4 ± 0.6	(58)	8.4 ± 0.5	(83)	3.7 ± 0.7	(32)	3.8 ± 3.4	(4)
$Si(OH)_4$ ($\mu mol\ L^{-1}$)	2.2 ± 0.7	(17)	2.8 ± 0.3	(46)	3.5 ± 0.3	(58)	5.4 ± 0.2	(83)	3.0 ± 0.4	(32)	2.3 ± 1.7	(4)
PO_4^{3-} ($\mu mol\ L^{-1}$)	0.3 ± 0.1	(17)	0.3 ± 0	(45)	0.4 ± 0	(55)	0.7 ± 0	(79)	0.3 ± 0	(32)	0.4 ± 0.2	(4)
$Si(OH)_4$:NO_3^-	6.0 ± 3.2	(14)	3.6 ± 1.3	(37)	8.5 ± 2.5	(54)	1.1 ± 0.2	(82)	1.6 ± 0.3	(32)	3.9 ± 2.2	(4)
NO_3^-:PO_4^{3-}	8.2 ± 2.0	(11)	5.2 ± 0.7	(45)	5.9 ± 0.8	(55)	11.4 ± 0.5	(79)	8.7 ± 0.8	(32)	5.5 ± 3.5	(4)
Chlorophyll *a* ($mg\ Chla\ m^{-3}$)	3.8 ± 1.1	(17)	5.5 ± 0.7	(45)	7.7 ± 0.7	(59)	2.0 ± 0.2	(91)	4.0 ± 0.3	(31)	8.8 ± 4.8	(4)
DT:(DT+DD)	0.01±0.006	(16)	0.02±0.01	(44)	0.04±0.01	(62)	0.10±0.01	(92)	0.08±0.01	(32)	0.02±0.02	(4)
(DD+DT):Chla	0.08±0.02	(17)	0.03±0.004	(46)	0.04±0.003	(62)	0.07±0.004	(92)	0.12±0.01	(32)	0.07±0.02	(4)
POC ($mg\ C\ m^{-3}$)	245 ± 45	(4)	498 ± 38	(27)	533 ± 30	(45)	234 ± 18	(63)	512 ± 46	(15)	393 ± 296	(2)
PON ($mg\ N\ m^{-3}$)	39 ± 8	(4)	65 ± 4	(27)	74 ± 4	(45)	38 ± 3	(64)	83 ± 9	(15)	42 ± 29	(2)
POC_{phyto} (%)	23.0 ± 2.6	(4)	49.2 ± 5.8	(26)	60.9 ± 3.9	(44)	33.3 ± 1.3	(64)	36.0 ± 3.0	(15)	37.8 ± 0.9	(2)
POC:PON	6.5 ± 0.6	(4)	7.8 ± 0.4	(27)	7.5 ± 0.3	(45)	6.6 ± 0.2	(64)	6.2 ± 2.0	(15)	8.6 ± 1.1	(2)
α^B × 10^{-2} ($\mu g\ C\ \mu g\ Chla^{-1}\ h^{-1}\ W\ m^{-2}$)	-	-	6.8 ± 2	(9)	9.2 ± 2	(10)	7.1 ± 1	(18)	7.1 ± 1	(4)	-	-
P_m^B ($\mu g\ C\ \mu g\ Chla\ h^{-1}\ W\ m^{-2}$)	-	-	2.9 ± 0.4	(9)	2.3 ± 0.3	(10)	2.3 ± 0.1	(18)	3.2 ± 0.4	(4)	-	-
E_k ($W\ m^{-2}$)	-	-	60 ± 11	(9)	29 ± 4	(10)	38 ± 3	(18)	46 ± 3	(4)	-	-
E_5 ($W\ m^{-2}$)	-	-	62 ± 11	(9)	35 ± 6	(10)	43 ± 4	(18)	56 ± 4	(4)	-	-
β × 10^{-4} ($\mu g\ C\ \mu g\ Chla\ h^{-1}\ W\ m^{-2}$) x$10^{-}$	-	-	4 ± 2	(9)	16 ± 7	(10)	10 ± 4	(18)	29 ± 10	(4)	-	-

FIGURE LEGENDS

Figure 1- Map showing stations along the AR7W transect and additional stations sampled during late spring and early summer (2005–2014). The station positions are superimposed on a composite image of sea surface temperature for the last three weeks of May 2006 collected by the NOAA satellite (AVHRR). White patches represent ice (Labrador and Greenland coasts).

~~**Figure 2. Percentage contribution of each pigment to the similarity of sampled stations in different clusters (I-V). Pigment abbreviations are described in Table 2.**~~

Figure 2~~3~~ – Map with sampling stations and distances from a fixed reference position (Northeast Gulf of St Lawrence) in the x-axis shown by the star (a). Values are given at individual stations sampled between 2005 and 2014 (y-axis) for the following variables: date of sample collection (b), temperature (c), salinity (d), stratification index (SI) (e), chlorophyll *a* (f), nitrate (NO_3^-) (g), phosphate (PO_4^{3-}) (h), silicate ($Si(OH)_4$) concentrations (i), ratios of particulate organic carbon (POC) to particulate organic nitrogen (PON) (j), silicate to nitrate ($Si(OH)_4$:NO_3^-) ratios (k), and nitrate to phosphate (NO_3^-:PO_4^{3-}) ratios (l). LSh = Labrador Shelf, LSl = Labrador Slope, CB = Central Basin, GSl = Greenland Slope, GSh = Greenland Shelf.~~**Values for environmental variables (temperature, salinity, stratification index (SI)), concentrations of nutrients (nitrate (NO_3^-), silicate ($Si(OH)_4$), phosphate (PO_4^{3-})), chlorophyll *a* and ratios between nutrients and for particulate organic carbon (POC) to particulate organic nitrogen (PON) at individual stations sampled between 2005 and 2014 (y-axis) and distances from a fixed reference position in the Northeast Gulf of St Lawrence shown by the star in Figure 3a (x-axis). LSh = Labrador Shelf, LSl = Labrador Slope, CB = Central Basin, GSl = Greenland Slope, GSh = Greenland Shelf.**~~

Figure 3. Relative contribution (%) of chlorophyll a from distinct phytoplankton classes at each station from 2005 to 2014 along the section distance from Labrador coast represented in Figure 3a (star symbol in a). LSh = Labrador Shelf, LSl = Labrador Slope, CB = Center Basin, GSl = Greenland Slope, GSh = Greenland Shelf. Note the distinct scales for each group.

Figure 4. Dendrogram showing clustering of samples (a) and the proportion of chlorophyll *a* contributed by each phytoplankton ~~class~~ group for each cluster (b). Spatial distribution of distinct phytoplankton communities (cluster groups) along the section, showing the distance from the star in Fig 3a) (c). Bubble size in (c) represents total chlorophyll *a* biomass (minimum = 0.3 mg Chl*a* m^{-3} and maximum = 25 mg Chl*a* m^{-3}).

Figure 5- Positions of individual stations in relation to temperature (°C) and salinity (a) and redundancy analysis (RDA) ordination plot (b). The stations are colour-coded according to the cluster groups (see details in Figure 4). The TS plot (a) shows the approximate ranges of potential temperature (°C) and salinity of the Labrador Current (LC), the West Greenland Current (WGC) and the Irminger Current (IC). Arrows in (b) show the explanatory (environmental) variables used in the analysis.

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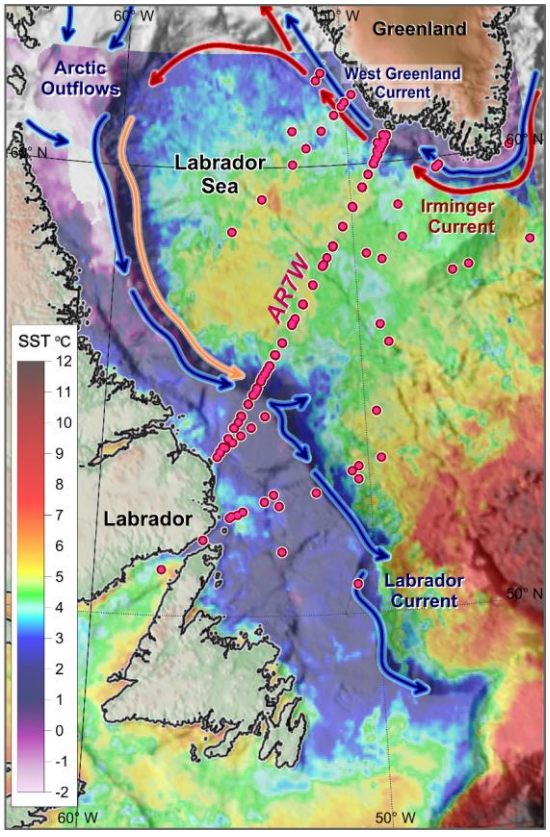
Figure 6- Relationship between particulate organic carbon (POC) and particulate organic nitrogen (PON) in a logarithmic scale, with the points (stations) as a function of phytoplankton-derived organic carbon content (POC_{phyto}/POC, %) (a), POC:PON *versus* salinity (b), phytoplankton-derived organic carbon content (POC_{phyto}/POC, %) *versus* the POC:PON ratio (c). The points (stations) in (b) and (c) are colour-coded according to the cluster groups (see details in Figure 4). Solid lines in (b) and (c) show the C:N Redfield ratio of 6.6 and the dashed line in (c) shows where POC_{phyto} contributes 50 % of the total POC.

Figure 7. Relationship between total accessory pigments (mg AP m^{-3}) and total chlorophyll (mg TChl*a* m^{-3}) on a logarithmic scale, with the points (stations) according to temperature (a) and colour-coded according to phytoplankton community cluster group (see details in Figure 4) (b).

Figure 1



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~~Figure 2~~

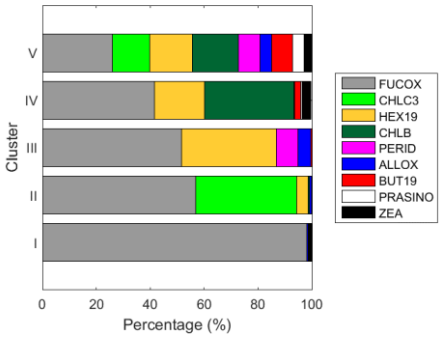

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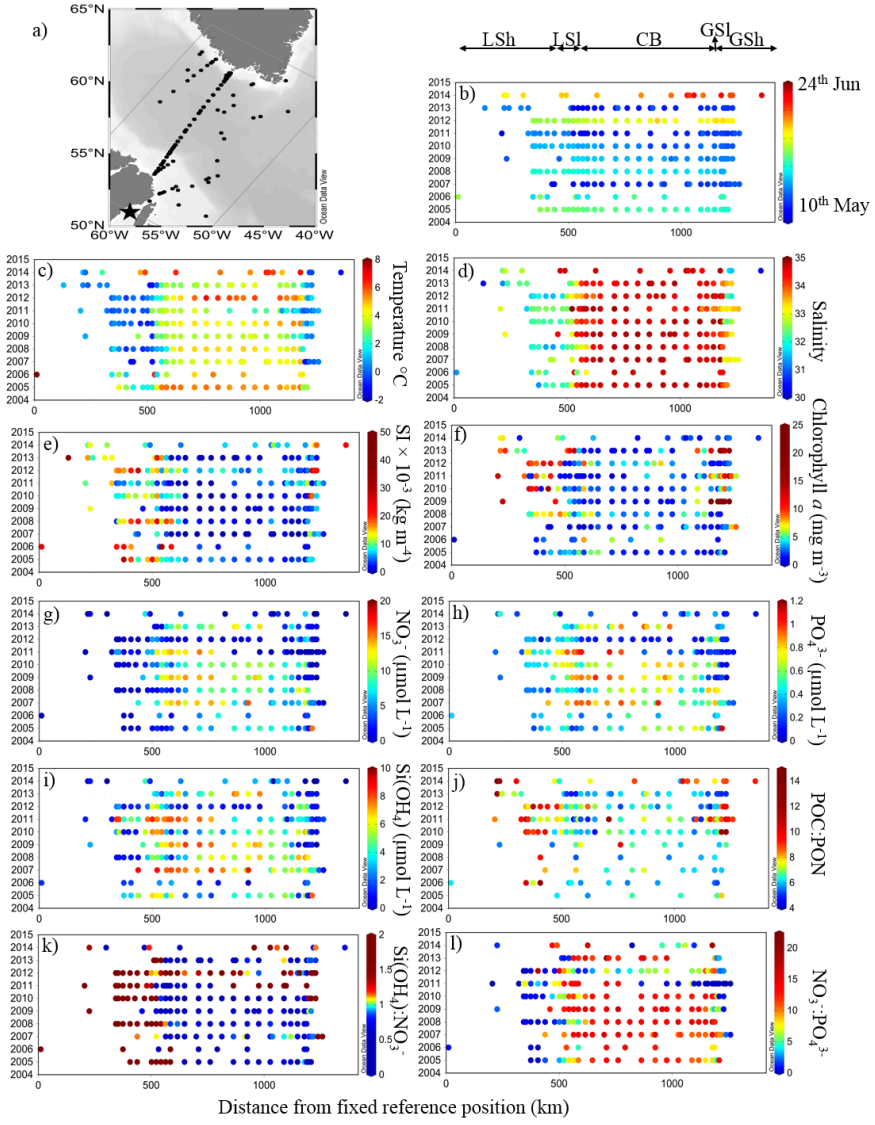
Figure 23


a)
65°N
60°N
55°N
50°N
60°W 55°W 50°W 45°W 40°W
LSh
LSl
CB
GSl
GSh
b)
24th Jun
10th May
c)
Temperature °C
d)
Salinity
e)
SI × 10³ (kg m⁻⁴)
f)
Chlorophyll a (mg m⁻³)
g)
NO₃⁻ (µmol L⁻¹)
h)
PO₄³⁻ (µmol L⁻¹)
i)
Si(OH)₄ (µmol L⁻¹)
j)
POC:PON
k)
Si(OH)₄:NO₃⁻
l)
NO₃⁻:PO₄³⁻
Distance from fixed reference position (km)

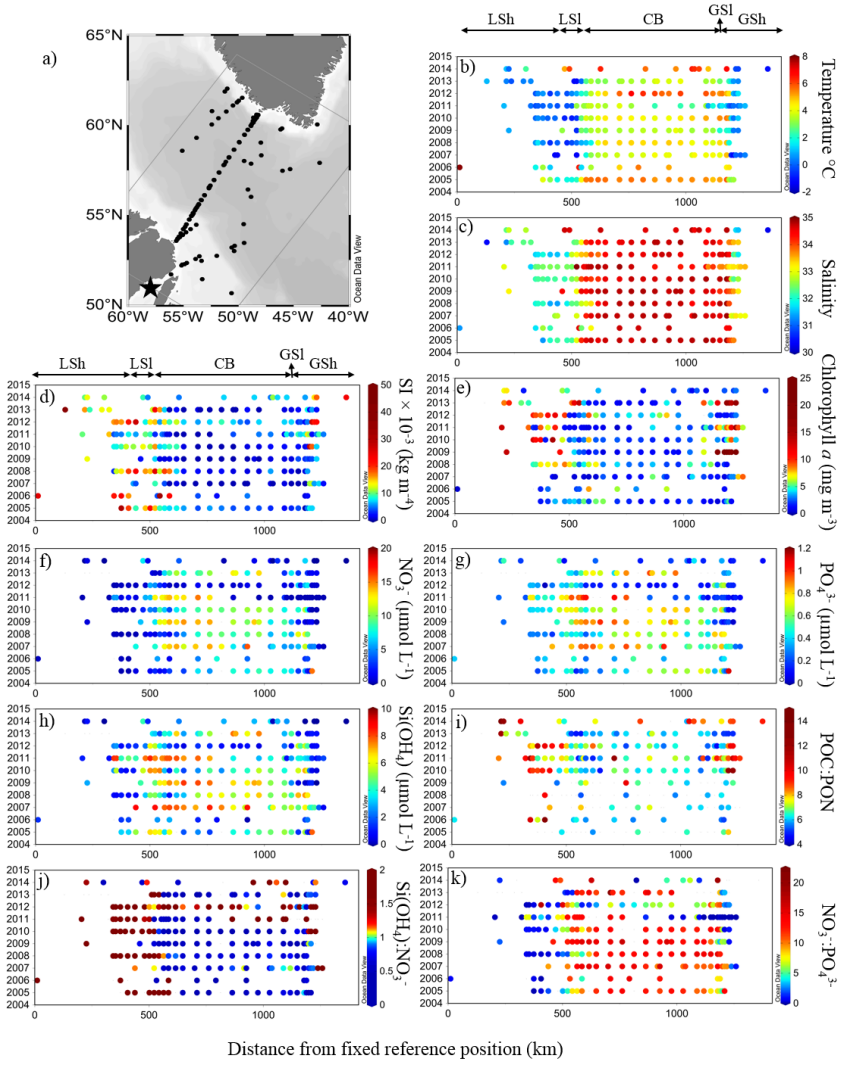


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Figure 3

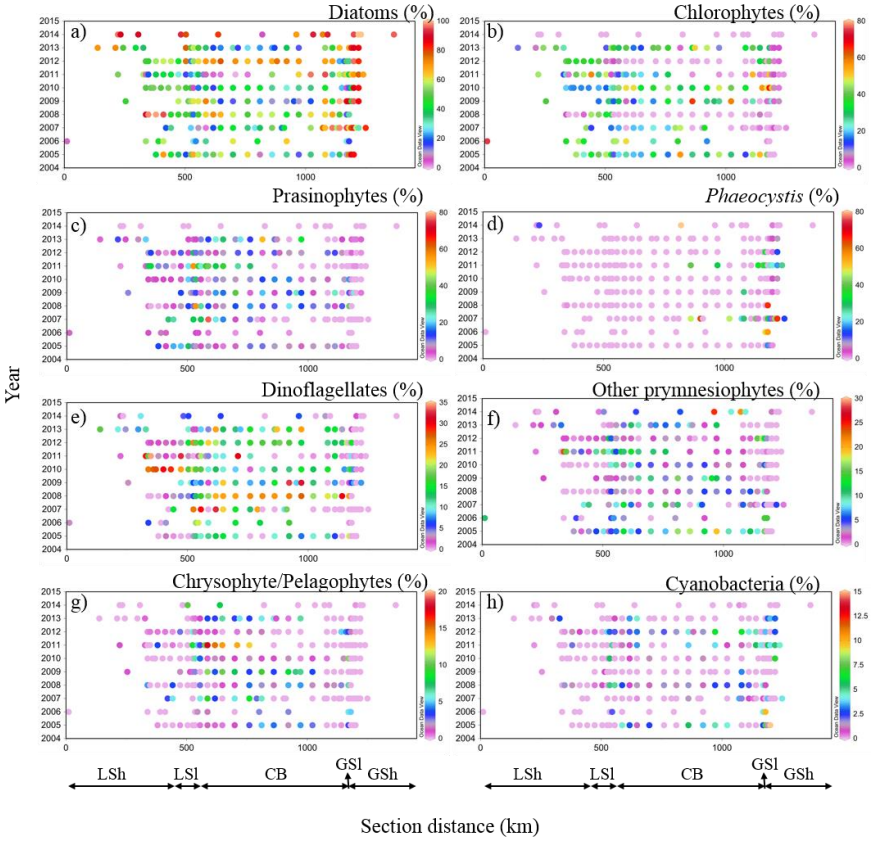

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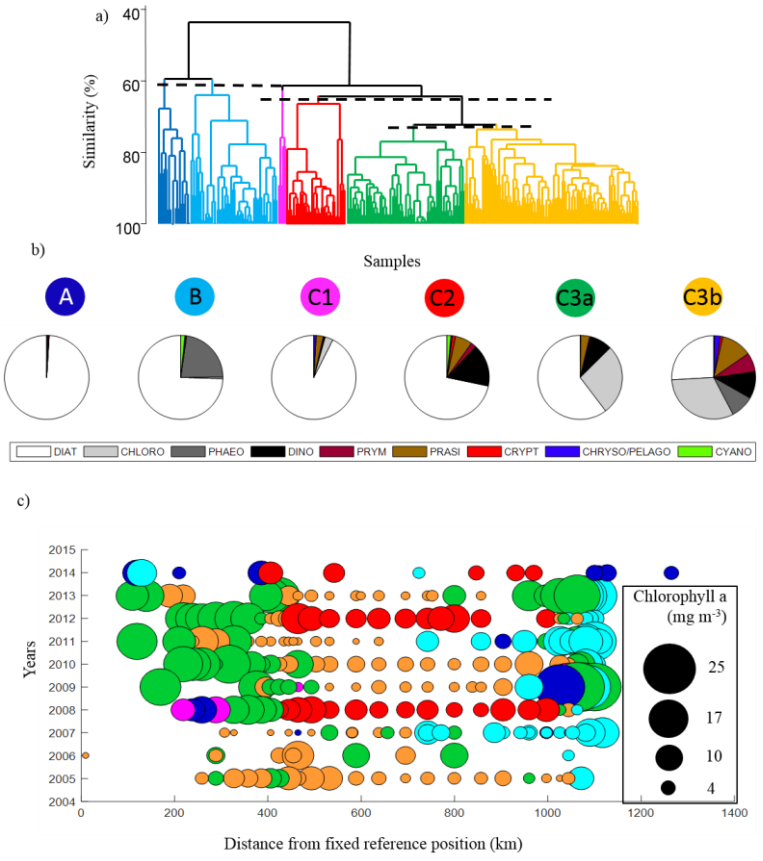
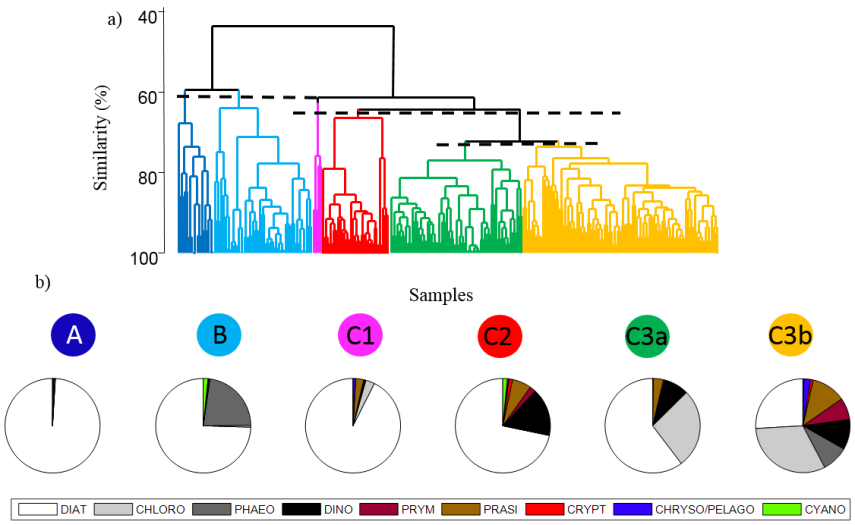
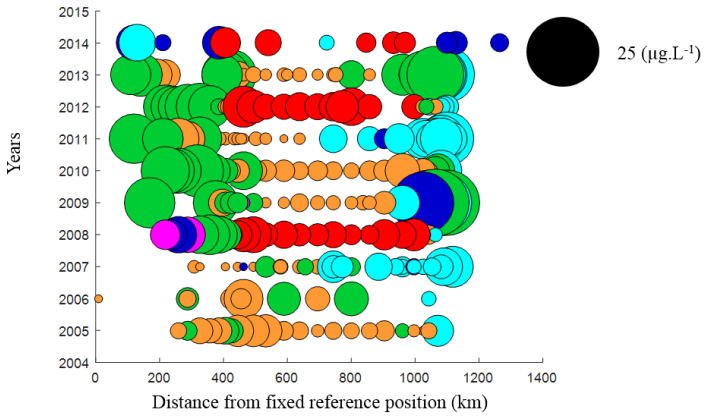
Figure 4


a)
Similarity (%)
40
60
80
100
b)
Samples
A
B
C1
C2
C3a
C3b
DIAT
CHLORO
PHAEO
DINO
PRYM
PRASI
CRYPT
CHRYSO/PELAGO
CYANO
c)
Years
2015
2014
2013
2012
2011
2010
2009
2008
2007
2006
2005
2004
0
200
400
600
800
1000
1200
1400
Distance from fixed reference position (km)
Chlorophyll a
(mg m-3)
25
17
10
4





Formatted: Caption, Left

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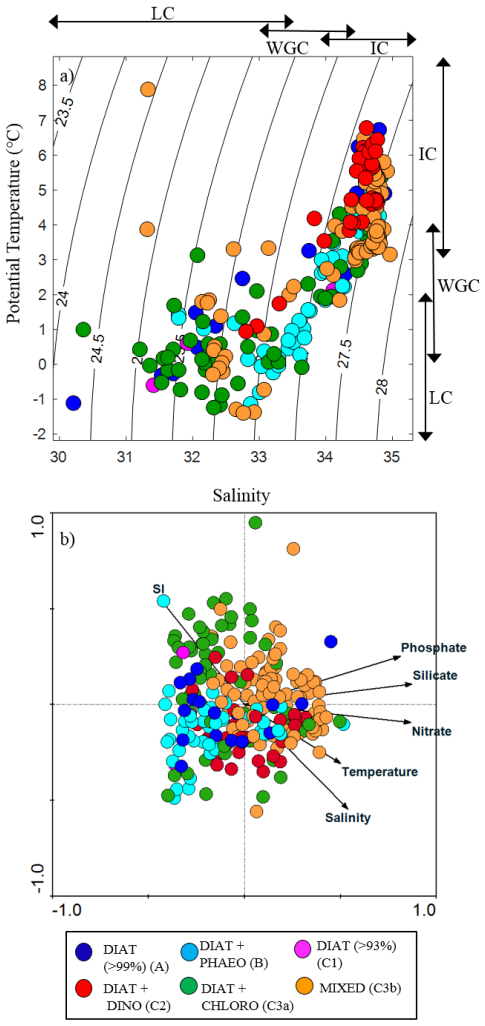
Figure 5


LC
WGC
IC
a)
Potential Temperature (°C)
Salinity
23.5
24
24.5
27.5
28
IC
WGC
LC
b)
SI
Phosphate
Silicate
Nitrate
Temperature
Salinity
DIAT (>99%) (A)
DIAT + PHAEO (B)
DIAT (>93%) (C1)
DIAT + DINO (C2)
DIAT + CHLORO (C3a)
MIXED (C3b)

1355 **Figure 6**

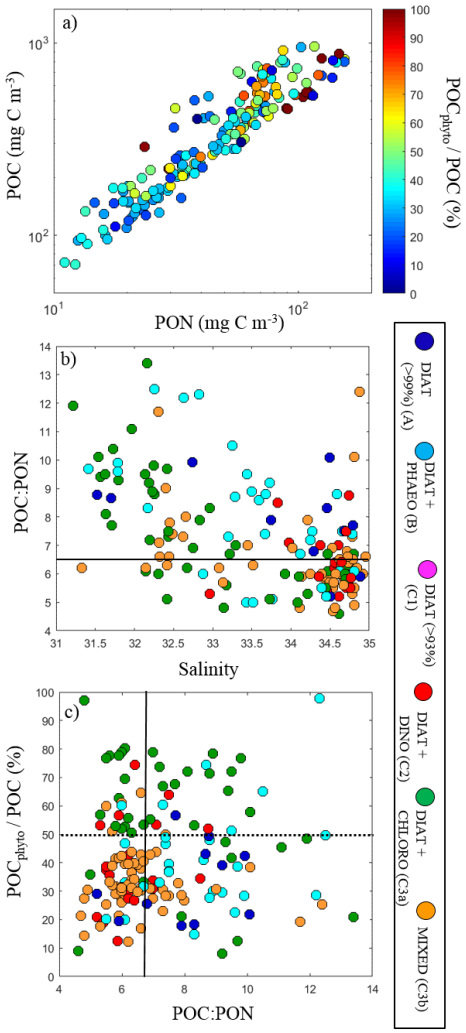
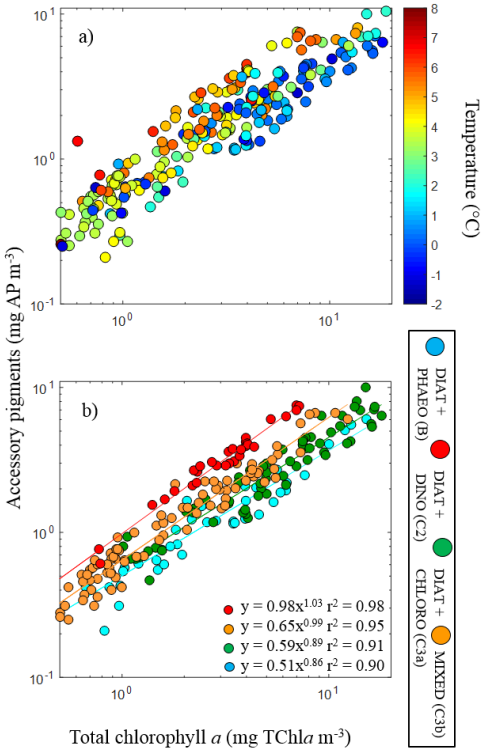


Figure 7



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