We appreciate the constructive comments by the two anonymous reviewers and think that the manuscript has improved accordingly from the inspired revisions. We have carefully considered all comments and suggestions. In particular, we have toned down the phytoplankton characterization which was criticized by both reviewers. Where requested, we have further elaborated our arguments with statistics (see Rev#1's comment) to reinforce our interpretations. Finally, we paid particular attention to rewriting those sections that were equivocal to better articulate the scope and results of our study.

We also apologize to the reviewers as some sentences looked odd due to the poor proofreading after that several people have worked on the same document with track change. These broken sentences were fixed and the manuscript has this time been proofread by a native English speaker.

The line number in the response to the referees' comments refers to the revised version. The following document has been organized such that first reviewer comments are given in italic directly followed by our detailed response in regular font (colored blue).

REV#1

Carbon geochemistry of plankton-dominated supra-1 micron samples in the Laptev and East Siberian shelves: contrasts in suspended particle composition.

General comments

This study want to improve the understanding of the chemical composition of plankton that dominates regions of the Arctic Ocean characterized by different sea-ice coverages and and in the ice-free Laptev Sea. The authors conclude that terrestrial carbon influence the POM in the Laptev Sea with higher influence of the River Lena. In the East Siberian Sea with ice-cover the influence of land is smaller. This is a valuable study in an important part of the ocean with a paucity of observations.

The methodology for 13C and 14C analyses seem adequate. However, this is not the case for the estimates of plankton diversity based on qualitative data from scanning electron microscopy. Either the authors have to convince us that this method is adequate and provide quantitative data analyzed by proper statistical tests. Otherwise markedly constrain conclusions regarding phytoplankton diversity or remove this entirely, perhaps using references to other studies instead.

Also the 13C and 14C analyses consistently lack objective statistical tests to support the conclusions made. Even if this data in general looks convincing this needs to be added.

The text is quite OK but there are some sentences which are not currently understandable. It does not appear that the last version has been checked by an English speaking person, which is recommended.

Provided that these remarks and important specific comments below are remedied I recommend that the manuscript is published after a major revision.

Specific comments

1) Title: Please revise the title replacing the "supra1-micron samples" term with "POM".

We have replaced the term *supra micron* with "particulate organic matter (>10µm)" throughout the text

2) r.28-36 I suggest to make the introductory paragraph shorter motivating the addressed question and the overall design of the study.

We have shortened the abstract as requested.

r.31-32 Unclear why "supra-" is used at this stage. Not a common term in my mind. Please just state "..the larger than 10 µm particulate organic matter (POM) fraction…" in the abstract. It´s clear from your definition what fraction that is covering.

Changed throughout the text consistent with the title (see above)

r. 37-41 These conclusions need to be better supported by statistical tests as specified in the result and discussion section.

We ran a two-tailed T-test (homoscedasticity) and a Welch T-test (heteroskedasticity) to assess whether the differences between Laptev Sea and East Siberian Sea were statistically significant (see Methods). The text was changed accordingly when discussing differences between regions (e.g. line 324, 326, 353, 360).

r. 42 "..communities via microorganisms". There are not reason to indicate several "loops". Please write concise and avoid unnecessary terms for clarity.

The term "loops" was removed according to the reviewer's suggestion

r. 44 The methodology does not seem adequate to assess the changes in diversity. Se comment in the result and discussion section. Also unclear what you mean by "…which is confirmed". Please rephrase.

We agree with this comment. We toned down the part that deals with the characterization of algal assemblages. This is no longer part of the abstract and conclusions. However, we would like to leave Fig. 5 and some brief discussion in the text just to provide qualitative information.

r. 45-46 Unclear what is meant by "…follows the general growth vs CO_2aq supply model…". Please present in a clearer way.

Rephrased to make it clearer (line 49-52).

r-48-50. What basis is this prediction based on? Please add a specification.

The destabilization of the Arctic cryosphere encompasses several aspects including sea-ice, glaciers and permafrost soil. Specific references were provided in the Introduction (line 63-84) which now contains even more details about the study region. Here in the abstract we can only report a summary of the future trajectories in response to the climate change.

r. 65 What uptake are you referring to? Please rephrase and clarify.

It refers to the CO2 uptake. Sentence rephrased (line 74).

r. 67 "…also project to water-…" Does not seem like proper English. Not understandable. Please rephrase.

The sentence was fixed (line 77).

r. 92-93 if the term supra- is not earlier defined and internationally agreed upon I see no reason to use it here. Just use POM and define the size fraction studies for simplicity.

We have replaced the term supra-micron fraction with POM throughout the text as suggested

r. 96. "..characterized by bulk…" If the MS has not been language checked by English speaking persons or dedicated companies please do so.

Again, this was one of those broken sentences resulting from poor text proof reading after several co-authors worked with track changes (line 107). All these odd sentences were corrected

r. 92-101. This paragraph should be moved to the methods section.

This is just a brief overview which we would like to keep in the Introduction. The Method section provides further and more specific details on the methods used.

r. 106 Please start with presenting the studied Sea areas and their characteristics. Please consider to use a map with sampling sites.

The map with cruise track and sampling site is already provided in the manuscript (Fig. 1). We added a section after the introduction to introduce the major features of the study region (section 2, line 114-129).

r. 112 Pleas add how the Falcon tubes were cleaned.

The method section was updated to include the cleaning procedure of the Falcon tubes (line 142-143).

r. 119 How long time after sampling was the analysis? Was samples frozen all the time to analysis?

The samples were kept frozen until lab analyses (line 141).

r. 166 How was samples taken and preserved? Were they concentrated in some way? Please add. Is there any reason why not other autotrophs like flagellates, picoplankton and cyanobacteria were included?

Samples were collected via large volume filtration of a nylon mesh. This was already well described in the text. We added further details about the samples collections (139-142). The 10um cut-off was chosen to avoid collecting fine terrigenous material in suspension.

r. 170 What was the number of cells counted and precision of counting per sample?

As we said this is a qualitative method which provides a snapshot of the dominant assemblages. It is not meant to be a statistical analysis. Thus, we agree with the previous comment and we have toned down this part.

r. 184 Please add the accuracy and precision of the measurements of CO_2 and $\delta^{13}C_{CO2}$.

We added the precision for both CO_2 and $\delta^{13}C$-CO_2. However, this section was moved to the supplementary material.

r. 229 Please present (for all variables) some confidence intervals or test, validating what are statistically significant differences between stations (i.e. accounting for spatial and short term temporal variability). E.g. if you want to claim differences between sea areas show by a proper statistical methods that they are different from each other.

We used the T-test between ice-free and ice-dominated regions while Pearson correlation was used to investigate the relationship among variables (see Methods section 3.5). However, we mainly focused on the new data presented in this study (dual-carbon isotope and biomarker data). The rest of the data regarding the surface water properties were elaborated, discussed and interpreted in other studies (Humborg et al., 2017; Salvado et al., 2016).

r. 236 The data referred to in Humborg et al. need either to be published before accepting this paper, or data presented in this paper.

Humborg et al has been just accepted in *Global Biogeochemical Cycles DOI: 10.1002/2017GB005656*

r. 243 How do you define depletion? Please be more specific and refer to comparative data or references. Similar for "low" at r. 245.

This sentence was modified according to this comment (line 286-289).

r. 255 Please specify what "margin" that is referred to. This sentence is not possible to understand. Please rephrase.

We have replaced "margin" with "continental margin" for clarity (line 300).

r. 259 It´s not obvious how the concentration of lignin or hydroxyl fatty acids will say anything about effects on the POM fraction. Do you mean the conc. of these compounds in the POM? What about many other effects on living POM like species composition and functionality like growth or edibility? Do assume that most POM is non-living? Please motivate you analyses better relative the aim.

As stated in the text, lignin phenols and cutin acids are <u>uniquely produced by terrestrial vegetation</u>. Therefore, these analyses were carried out to assess whether or not the samples were affected by land-derived material (i) directly supplied by rivers, (ii) trapped in the sea-ice and (iii) resuspended from the sediment. The concentrations of lignin phenols are close to the detection limit while cutin were not detectable. This implies that the material collected is primarily autochthonous marine POM.

Fig.3 Please add what error bars are showing. Please specify what the values are relative against (carbon or mass?).

This figure shows the CuO oxidation fingerprint of the samples. Upon Cuo oxidation organic matter releases different biomarkers whose composition provide information about the source. The percentage refers to the total CuO oxidation yields and the error bar reflects that natural variability observed in different carbon pools. For example, soils are rich in lignin phenols and cutin acids while phytoplankton batch cultures upon CuO oxidation don't yield these terrestrial biomarkers. By contrast, they produce a large amount of low molecular weight fatty acids consistent with the POM collected in this study. We added further details in the caption of Fig.3.

Samples

r. 276 Support you statement with a tests showing that these are different. What do a base the "high" and "low" assessments on (relative what)?

As previously mentioned, we used a T-test/Welch T-test to show that these differences are statistically significant ($p<0.01$) (line 324)

r. 275-287 SEM is not a proper method to asses diversity of phytoplankton (or present quantitative SEM data from sufficient number of samples?). That the diversity of phytoplankton is different between sea areas is therefore not sufficiently well demonstrated. Concentrations of at least major taxonomic groups is requested based on microscopy counting with adequate methodology (e.g. sediment chambers and reverse light microscopy) and statistical precision presented. Preferably also including flagellates, picoeucaryotes and cyanobacteria. Established diversity index should be used and tested for difference.

As previously mentioned, we have used this method to provide a general overview of the dominant taxa. We agree with the reviewer on this comment and we are aware that this procedure has only qualitative applications. We would still like to keep this part but make clear in the text that this is only qualitative information. Any reference in the abstract and conclusions about the phytoplankton taxa were removed from the revised text.

r- 288-289 Unclear how a line can detect a bloom. Please be more specific.

The sentence was corrected (line 336).

r.293-294 I don´t agree that the presented data convincingly show that dinoflagellates were dominating. How is the SEM preparation influencing different phytoplankton species? Is there a selections for robust dinoflagellate shells? Provide a reference or control experiment clarifying this.

ASK CHRISTOF FOR REFERENCES

r. 297-301 As presented isn´t IP25 then specifically indicating presence of sea-ice diatoms, not "..sampling of different plankton taxa…? Also can other sources of IP25 have contributed to the variability. Consider a re-interpretation. I suggest to calculate if the found conc. of IP25 could come from an expected conc. of diatoms in the sea ice and present that.

IP25 detects specific sea ice diatoms (Pleurosigma stuxbergii var. rhomboide, Haslea crucigeroides (and/or Haslea spicula) and Haslea kjellmanii, Brown et al., 2014b) which account for only a minor fraction of the sea-ice taxa. No other sources (e.g. from land) supply IP25 expect from these sea-ice taxa. To the best of our knowledge, the actual end-member doesn't exist and it likely varies depending on the concentration of these aforementioned species in the sea-ice

r. 309-310 Please provide a statistical test showing a significant trend of CO2 and δ13 CCO2.

p values were added in both linear correlations (Fig.7).

r. 316-317. On what basis is it assumed that the present dinoflagellates are hetero- or mixotrophic? That some dinoflagellates can eat bacteria is well shown in the literature. However, not that they are significant consumers of diatoms? Please provide a reference for this if so.

Our hypothesis is based on the dual-carbon isotope fingerprint. Specifically, out of the different carbon source known in the region (Fig.6), the dual-carbon isotope signature of the POM is consistent only with the Lena river DOC (and rather different from the Lena river POM). Thus, in our hypothesis, dinoflagellates feed on bacteria that develop in the terrestrial DOC-rich plume of the Lena river.

r. 324-325 I suggest to rephrase to "…, supporting the importance of terrestrial DOC as a carbon source for the food web in the river plume….).

Text changed according to this suggestion (line 399-400).

REV#2

General comments

The objective of this study was to investigate the composition of the suspended particulate organic matter in ice-covered and ice-free waters over the Laptev and East Siberian shelves. The main problem of this study is to assume that these samples are plankton-dominated, as indicated by the title. There are no data to support the fact that phytoplankton dominated the suspended particulate matter and such a dominance would actually be quite surprising over the shallow Siberian shelves (a lot of particulate material is resuspended and/or transported with the ice). An effort should be made to quantify the phytoplankton contribution and composition before resubmitting this manuscript. If the authors somehow collected ice samples during this expedition, it would be relevant to compare the composition of the particulate matter in the ice with the composition of the suspended matter.

Another important problem is that too much of the current manuscript is based on another paper submitted elsewhere by many of the same authors that seems to be very similar to the current manuscript. This problem must be addressed. Overall, while the study had the potential to provide interesting results from a very rarely sampled region, the current results do not bring very interesting or new information. It is well-known that ice covered regions are productive and display high concentrations of particulate matter. The interpretation of the results must be reevaluated in this context. Also, please keep in mind and specify throughout the manuscript that these are late summer observations and that conditions may be quite different during the productive spring period. Finally, the manuscript is too long, often repetitive, and the text needs to be revised by a native English speaker.

Specific comments

Title

I have never heard the terms supra-micron or supra-POM and I don't think there is a need for it. Please remove the term supra- throughout the manuscript.

As mentioned earlier, we have replaced the *"supra-micron POM*" term with POM (>10µm)

Abstract

Lines 50-51: Comments like these are not informative. Always be specific.

We followed the suggestion. The paragraph ends with specific details about changes in the study region (line 53-57).

Introduction

Lines 54-55: Provide more recent references.

We have updated the references with recent studies dealing with the sea-ice retreat in the Arctic Ocean (line 64).

Material and methods

Some information on the dates of sampling are required.

Lines 110-113: Several steps are unclear. When it is mentioned that particulate material was kept frozen, it means the filters? In which state were the samples transferred into the centrifuge? Were the samples thawed first?

This part was edited to make it clearer (line 137-141).

Lines 115-116: Such information belongs in figure captions.

Removed

Lines 150 and 154: IP25 is a highly branched isoprenoid mono-unsaturated alkene. Introduce it properly and only once.

Text changed accordingly (line 180-184).

Section 2.3 Microscopic images of plankton This is probably the biggest shortcoming of the study. It is baffling that the authors use microscopic images as a qualitative tool but did not include a quantification of the different phytoplankton groups. This would definitely improve the quality of the study. *Always precise if it is phytoplankton or zooplankton. Plankton is not a term precise enough.*

Of course, we agree that quantitative information would have been more informative. Ours is essentially a biogeochemical study and these snapshots obtained via SEM and transmitted light microscope, provide only complementary information. For this study, we took several SEM images (in the paper we show just one magnified example) about 10 per station randomly sampled in the freeze-dried material. This, combined with traditional microscope analyses, allowed us to see major trends within the dataset, for example we could hardly see any diatoms in the Laptev Sea while further east diatoms were dominant. Thus, despite the limitation of our approach (which we acknowledge) we still believe that this is relevant information. We decided to tone down this part in acknowledgement to the reviewer's comment but still keep it in the discussion. In the revised manuscript, we removed any comment on phytoplankton taxa in both abstract and conclusion. However, we would still like to keep this part in the discussion making sure that the reader understands the qualitative applications of our approach.

The material and methods section is too long and often repetitive. Reduce.

We went through this section but couldn't find any redundant information. However, methods on CO2 measurements were moved to the supplementary materials.

Results/discussion

Section 3.1. Surface water conditions Most of this section does not belong in the paper. All the results (salinity, temperature, nutrients...) for which material and methods were not presented in the precedent section must be removed from the manuscript and the figures/tables as well. This is even more crucial considering that the same results are part of another submitted paper from the same authors. It is not appropriate to submit the same results twice and all the results that were submitted in Humborg et al. must be removed. Instead the authors should refer to these results in the discussion, which would be much stronger if or once the other paper is published. It would be more appropriate to start the discussion with section 3.2.

Humborg et al has been accepted in *Global Biogeochemical Cycles (and updated in our ref list)*. Following the reviewer's suggestion, the discussion now starts with section 4.1 and the former 3.1 section dealing with the surface water properties has become section 3 ("*Surface water conditions during the SWERUS-C3 expedition*).

Lines 220-222: You should never write sentences in this form: Figure 1 displays...

This sentence was changed according to the reviewer comment (line 262)

"Table 1 reports..." This is the type of mistakes made at the undergraduate level.

Specifically for the above comment, please refer to point 3 of the "General obligations for referees" document on the *Ocean Science* website. Thanks

Line 225 and others: All maps (figures 1, 2 and 4) should be switched with North towards the top to help with the description of the results. This is the usual and correct way to place a map and it is less confusing when looking for the westernmost stations.

We must disagree on this point. In this special issue, there are at least 8 manuscripts (Miller et al., Anderson et al., o'Regan et al, Björn et al, etc) with the exact same polar projection. In fact, this is the default ESRI Arcgis projection consistent with the IBCAO format (Jakobsson et al., 2012.GRL). We will keep this format.

Lines 230-232: DOC concentrations mirrored... 'Mirrored' does not mean the opposite, it means similar. Get an English speaker to review your paper. And please limit your use of the word 'thus'.

Of course, we did not mean similar in terms of absolute values. We were just commenting on the general spatial distribution as DOC is clearly affected by the river plume: low salinities are associated with high DOC concentrations. Sentence reformulated (line 273).

Line 252: It is late and unnecessary to introduce the term TerrOC at this point. Either you introduce it earlier or you use other terms for consistency.

TerrOC was removed and replaced with "terrestrial organic carbon"

Lines 284-287: These phytoplankton species are typically observed late in the season. This should be specified. Chaetoceros and Thalassiosira are pelagic species growing in water only while Fragilariopsis cylindrus and oceanica grow both in ice and water (they are not sea ice species necessarily). More information could be obtained through extensive and quantitative taxonomic analyses of the existing samples.

Again, we agree with the reviewer that this minor part of our ms has severe limitations but we believe it still provides some useful general information on the dominant trends within the study region (see previous comments). For example, we can clearly see a marked difference between the Laptev Sea (no diatoms with abundant dinoflagellates) and the East Siberian Sea (dominated by diatoms) regardless of the method used (SEM vs traditional microscope). Again, our analysis is based on several SEM images coupled with optical microscope slides.

Lines 304-306: ... captured the signal of the sea-ice retreat that occurred shortly before... Sea ice retreat actually took place weeks and months before so it is not appropriate to say shortly before. The fact that IP25 was still detectable would be more likely the result of advection or resuspension.

Corrected according to this suggestion. Resuspension would bring lignin and other wax lipids (cutins) which, however, were not detected in the POM samples. Advection from surface waters might be a reasonable hypothesis though. Text changed accordingly (line 355-356)

Lines 378-380: However, it would then remain elusive why such an aged land-derived influence was not visible in the river-dominated LS waters while it affected the sea-ice dominated region. Is it that elusive? It is puzzling that the authors did not consider that the presence of this land-derived material is likely the result of the release of material that was trapped in the ice during its formation on the shallow shelf. The trapped material is transported towards the outer shelves and released during ice melt, which was occurring at the time of sampling. This is an important and well-known process on the Siberian shelves. The interpretation must be improved to consider these ice- released particles. *How old? Be more specific.*

It's not puzzling at all as we specifically used organic biomarkers to trace the land-derived material. Figure 2 shows what type of CuO oxidation fingerprint you would get in a case of land-derived influence. For example, lignin phenols are clearly dominant in terrestrial soil samples while they are close to detection limit in our POM samples. Cutin derived products (waxes on plant leaves) were not even detected in the POM samples. As stated in the text, lignin phenols and cutins are exclusively produced on land and their negligible concentration in POM samples implies insignificant terrestrial influence. In other words, the samples are dominated by marine material. In fact, POM samples are consistent with the CuO oxidation fingerprint of algal batch cultures which mainly yield fatty acids, dicarboxylic acids, p-hydroxy phenols and benzoic acids (in this order of abundance) (Goni and Hedges, 1995).

Through this comment, we have realized that we did not mention the particulate transport by fast ice in the text. To provide a better picture regarding the sediment transport in the study region, we added a sentence about this mechanism (line 303).

Lines 394-396: Hence, results suggest a heterotrophic environment in the outer LS open waters where the river-derived DOC is transferred to relatively higher trophic levels via microbial incorporation (i.e, microbial loop). This sentence reflects a poor comprehension of the food web. Energy is not transferred to higher trophic levels through the microbial loop.

Here we refer to the terrestrial carbon being transferred from the DOC to the heterotrophic community via bacteria present in the Lena river plume. According to the radiocarbon signature (Fig. 6) of the Laptev samples, we infer that the DOC from the Lena is taken up by the microbial communities on which other heterotrophic communities (e.g. *Protoperidinium* spp;) feed on. Despite the fact that the samples have been collected in a region affected by the Lena plume (see salinity and DOC data, Fig 2), our modern 14C signature of POM largely differs from the particulate material supplied by the Lena river characterized by a 14C depleted signature (Fig. 6). In contrast, the POM signature seems to be more consistent with the Lena DOC fingerprint (Fig. 6). This would also explain the depleted stable carbon isotope composition despite the negligible terrestrial influence (biomarker results). It's also worth mentioning that DOC in the outer Laptev Sea is over one/two order of magnitude higher than the POC (Humborg et al., 2017; Salvado et al 2016).

Table 1 What is TN? Mean sea ice percentage is over which area?

TN is total nitrogen. Data are not discussed so they were removed in the new version

Table 2 This table does not belong in this manuscript.

As previously mentioned, these data are presented only with the intention of contextualizing our results. The discussion has been rearranged following the reviewer's suggestion. We now start the discussion from the new organic geochemistry data. Showing complementary data is a common procedure when studies are part of a multidisciplinary expedition during which research teams measured different parameters. Humborg et al has been recently accepted and properly cited in the manuscript.

Table 3 This qualitative analysis is nearly useless. The authors should definitely invest in quantitative taxonomic analyses to support their results.

See comments above

Fig. 1 Switch North up.

See comments above

Fig. 2 Should be removed, presented in other submitted paper.

See comments above

Fig. 4 Patterns are often not as clear as described by the authors in the results/discussion.

Be careful when interpreting.

As just mentioned above, we have divided the study area into two sub-regions (Laptev Sea_open waters and East Siberian Sea-ice-dominated) and carried out a T-test as suggested by reviewer#1 to show whether or not the differences are statistically significant.

Fig. 6 The new results should also be presented as whisker boxes for consistency. In the caption: East Siberian Sea, not Eastern Siberian Sea.

Caption corrected. The whisker plots presented here are used to summarize large dataset. For example the ICD end-member is made of 301 radiocarbon data. However, we don't think we can do the same for the POM samples in each considering the limited number of radiocarbon values.

Fig. 7 Why only for East Siberian Sea?

Because only the East Siberian Sea is an autotrophic system where CO2 is actually consumed by biological activity (i.e., depletion compared to the atmospheric value) (Humborg et al., 2017)

Carbon geochemistry of plankton-dominated ~~supra-micron samples~~samples in the Laptev and East Siberian shelves: contrasts in suspended particle composition

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Abstract

Recent Arctic studies suggest that sea-ice decline and permafrost thawing will affect phytoplankton dynamics and stimulate ~~marine~~ heterotrophic communities. However, in what way the plankton composition will change as the warming proceeds remains elusive. Here we investigate the chemical signature ~~and~~ of the plankton-dominated ~~-speciation of the supra-micron (> 10 µm)~~ fraction of particulate organic matter (~~supra-~~POM, >10µm) ~~fraction~~ collected along the Siberian shelf. ~~Supra-~~POM (>10µm) samples were analysed ~~at~~ using molecular biomarkers (CuO oxidation and IP_{25}) and dual-carbon isotopes ~~bulk~~ ($\delta^{13}C$ and $\Delta^{14}C$) ~~and molecular level (CuO oxidation and IP_{25}) while plankton identification established the dominant taxa~~. In addition, surface water chemical properties were integrated with the ~~plankton~~ POM (>10µm) dataset to understand the link between plankton composition and environmental conditions.

~~The~~ $\delta^{13}C$ and $\Delta^{14}C$ ~~dual-carbon isotope fingerprint indicates~~exhibited a large variability in the ~~supra-POM~~POM (>10µm) distribution while terrestrial biomarkers ~~suggest~~showed a negligible ~~land-derived~~ input from terrestrial sources. In the Laptev Sea (LS) open-waters, ~~of the outer Laptev Sea (LS), heterotrophic plankton dominated the assemblages.~~ $\delta^{13}C$ and $\Delta^{14}C$ fingerprint of POM (>10µm) suggested that ~~modern terrestrial~~ dissolved organic carbon (DOC) from the Lena river ~~is~~ was the primary source of metabolizable carbon ~~which~~ indicating, thus, a~~is transferred to the~~ heterotrophic ~~communities~~environment ~~via microbial loops~~. Moving eastwards toward the sea-ice dominated East Siberian Sea (ESS), the system became progressively more autotrophic ~~and dominated by sea-ice and pelagic diatoms which is confirmed~~. Comparison between $\delta^{13}C$ of ~~supra-~~POM (>10µm) samples and CO_2aq concentrations ~~suggests~~ revealed that the carbon isotope fractionation increased moving toward the easternmost and ~~follows the general growth~~

~~*vs* CO2aq supply model with the highest $\delta^{13}C$ values found in the easternmost,~~ most productive stations.

In a warming scenario characterized by enhanced terrestrial DOC release (thawing permafrost) and ~~further~~ progressive sea-ice decline, heterotrophic conditions ~~fuelled by terrestrial DOC will~~might ~~likely~~ persist in the LS while the nutrient-rich Pacific inflow will likely stimulate greater ESS ~~might experience enhanced~~ primary productivity in the ESS. The contrasting trophic conditions will ~~This will~~ result in a sharp ~~compositional~~ gradient in $\delta^{13}C$ between the LS and ESS similar to what documented in our semi-synoptic study.

1. Introduction

The progressive reduction of sea-ice extent in the Arctic Ocean is indisputable evidence of modern global warming (Comiso et al., 2008; Ding et al., 2017; Kwok and Rothrock, 2009). The unprecedented decline of sea-ice is expected to alter several aspects of the Arctic marine ecology such as plankton abundance and its temporal distribution (Arrigo et al., 2008). For instance, recent studies suggest that the increase of solar irradiance will stimulate greater primary productivity in summer while the prolonged ice-free conditions will develop a second algal bloom in early fall, which is a distinctive feature of only lower latitudes (Ardyna et al., 2014; Lalande et al., 2009; Lalande et al., 2014). The phytoplankton communities are expected to profoundly change towards a higher contribution from open water phytoplankton at the expense of sea-ice assemblages (Fujiwara et al., 2014). Taken together, a greater productivity in the ice-free or marginal ice zone compare to ~~in the inner~~the multi-year ice system, is also expected to lead to greater carbon uptake and settling export of organic carbon from the surface to deeper strata of the Arctic Ocean (Gustafsson and Andersson, 2012).

Sea-ice decline will also ~~project~~ affect the~~to~~ water-air gas exchange, currents and river plume dispersion which, in turn, exert large control on the surface water chemical/physical properties (Aagaard and Carmack, 1989; Ardyna et al., 2014; Lalande et al., 2014). On top of this, destabilization of ~~several aspects of~~ permafrost soils and ~~hydrology~~ the terrestrial cryosphere ~~on land~~ will result~~s~~ in enhanced particulate and dissolved carbon input to the Arctic Ocean (Frey and Smith, 2005; Vonk et al., 2012). ~~Thus~~As a result, the geochemical signature of both autotrophic and heterotrophic plankton communities is also expected to change as the warming proceeds. However, how the ~~cryosphere destabilization~~warming will ultimately affect the marine geochemical signal is poorly understood. This study seeks a better understanding of the chemical composition of plankton that dominates regions of the Arctic Ocean characterized by different sea-ice coverages, nutrient availability and ~~terrestrial~~ riverine influence~~input~~. In particular, we focus on the carbon isotope fingerprint (i.e.~~,~~ $\delta^{13}C$ and $\Delta^{14}C$) of plankton that grows in ice-covered and ice-free Marginal Ice Zone (MIZ) regimes ~~in~~ on the ~~river-dominated~~ Siberian margin. The motivation behind investigating the chemical fingerprint of plankton from different ~~sea~~regimes ~~ice domains~~ is to provide a better understanding of the carbon signature for direct applications to carbon studies of both modern systems and paleo-reconstructions. In particular, the isotope composition of marine OC finds several applications in climate, ecology and carbon source apportionment studies. For example, stable carbon isotopes of marine phytoplankton are used for paleo-pCO_2 reconstructions over geological time scales (Hoins et al., 2015; Pagani et al., 1999; Popp et al., 1999; Rau, 1994). The $\delta^{13}C$ signature also provides a solid tool for marine food web and ecosystem structure investigations (Dunton et al., 2006; Iken et al., 2005; Kohlbach et al., 2016). Furthermore, dual-carbon isotope mixing models ($\delta^{13}C$ and $\Delta^{14}C$) are commonly used to quantify the relative proportion of marine and various allocht~~h~~onous sources (e.g., permafrost soil) in both contemporary and paleo-reconstructed carbon cycling of the Arctic (Karlsson et al., 2016; Tesi et al., 2016; Vonk et al., 2012; Vonk et al., 2014).

With this overarching goal in mind, here we investigate the >10 µm ~~(supra-micron)~~ fraction of particulate organic matter (~~supra-~~POM) in ice-covered and ice-free MIZ regimes of the Siberian Arctic Shelf during the SWERUS-C3 expedition (July-August 2014) (Fig. 1). The plankton-dominated ~~supraPOC~~ POM samples collected throughout the ca. 4,500 km long cruise track were characterized ~~at~~ using bulk parameters (OC, $\delta^{13}C$ and $\Delta^{14}C$) and biomarkers ~~level~~ (highly branched isoprenoids, IP_{25}; CuO oxidation products)~~ while plankton identification via microscope provided information about the dominant assemblages~~. In addition, continuous measurements of dissolved CO_2 (CO_{2aq}) and its stable carbon isotope composition ($\delta^{13}C_{CO2}$) were performed during the campaign (Humborg et al., 2017) and used for a direct comparison with the chemical composition of the ~~supra-~~POM fraction.

2. Study region

The Laptev Sea and the East Siberian Sea are shallow epicontinental seas in the Russian Arctic separated by the New Siberian Islands (Fig. 1). Sea-ice cover lasts for most part of the year over the shelf. Late spring/summer is characterized by the seasonal sea-ice retreat coupled with river freshet which supplies large amount of terrestrial carbon in the form of particulate and dissolved matters (Karlsson et al., 2016; Salvadó et al., 2016; Sánchez-García et al., 2011). The Lena (523 km3/y), Indigirka (54 km3/y), and Kolyma (48 km3/y) are the major rivers (Gordeev, 2006). During the ice-free season, the Lena plume can be traced in the outer-shelf of the Laptev Sea (Fichot et al., 2013; Salvadó et al., 2016; Sánchez-García et al., 2011) while Pacific inflow from the Be~~a~~ring strait affects further east the East Siberia margin (Semiletov et al., 2005). ~~The North~~ The Pacific inflow exerts control on the nutrient balance as it supplies nitrates and nitrites to an otherwise nutrient-depleted region (Anderson et al., 2011; Semiletov et al., 2005). Another important source of particulate material to the continental margin is the ~~Plesitocene~~Pleistocene Ice Complex Deposit (ICD) ~~which~~entering ~~enters~~ the ocean via coastal erosion (Lantuit et al., 2011; Vonk et al., 2012) which is the dominant carbon source between the Kolyma river and the Lena river (Vonk et al., 2012).

3~~2~~. Methods

3~~2~~.1 ~~Supra-micron~~POM (<10 μm) ~~fraction~~ sampling

Seawater was pumped from a stainless steel inlet on the hull of the icebreaker *Oden* positioned at 8 m below the sea surface. The inlet system is tested and further described in Sobek and Gustafsson (2004) and Gustafsson et al. (2005). Figure 1~~1~~a and 1b show~~s~~ the regions covered to harvest each ~~supra-~~POM (>10 μm) sample with their ~~whose~~ location~~ is~~ shown as ~~s.~~ time-averaged position. The particulate material was retained via a large volume filtration apparatus using a 10-μm Nitex® (nylon) mesh placed in a 29.3 cm filter holder. After collection, filtered particulate material was transferred in pre-clean HDPE tubes ~~filter~~by rinsing the ~~ed samples were rinsed~~ Nitex® filters with~~with~~ MilliQ water~~ and the particulate material (i.e., supra-micron fraction) was ~~. Samples were kept frozen throughout the expedition. In the lab, samples were transferred in pre-cleaned Falcon®~~-~~ tubes (rinsed with 0.1M HCl)~~and~~ and gently centrifuged to remove the supernatant. The residual particulate material was frozen and subsequently freeze-dried prior to biogeochemical analyses.

~~In all figures, sample location of the supra-POM samples refers to its time-averaged position as shown in Figure 1.~~

~~2~~3.2 Bulk carbon isotopes and biomarker analyses

Organic carbon (OC) and stable carbon isotope ($\delta^{13}C$) analyses were carried out on acidified samples (Ag capsules, HCl, 1.5M) to remove the carbonate fraction (Nieuwenhuize et al., 1994). Analyses were performed using a Thermo Electron mass spectrometer directly coupled to a Carlo Erba NC2500 Elemental Analyzer via a Conflo III (Department of Geological Sciences, Stockholm University). OC values are reported as weight percent (%d.w.) whereas stable isotope data are reported in the conventional $\delta^{13}C$ notation (‰). The analytical error for $\delta^{13}C$ was lower than ±0.1‰ based on replicates. Acidified (HCl, 1.5 M) samples for radiocarbon abundance were analysed at the US-NSF National Ocean Science Accelerator Mass Spectrometry (NOSAMS) facility (Woods Hole Oceanographic Institution, Woods Hole, USA). Radiocarbon data are reported in the standard $\Delta^{14}C$ notation (‰).

Alkaline CuO oxidations were carried out using an UltraWAVE Milestone microwave as described in Tesi et al. (2014). Briefly, about 2 mg of OC was oxidised using CuO under alkaline (2N NaOH) and oxygen-free conditions at 150 °C for 90 min in teflon tubes. After the oxidation, known amounts of recovery standards (trans-cinnamic acid and ethylvanillin) were added to the solution. The NaOH solutions were then acidified to pH 1 with concentrated HCl and extracted with ethyl acetate. Extracts were dried and redissolved in pyridine. CuO oxidation products were quantified by GC-MS in full scan mode (50-650 m/z). Before GC analyses, the CuO oxidation products were derivatized with bis(trimethylsilyl) trifluoroacetamide+1% trimethylchlorosilane at 60°C for 30 min. The compounds were separated chromatographically in a 30m×250 µm DB5ms (0.25 µm thick film) capillary GC column, using an initial temperature of 100°C, a temperature ramp of 4°C/min and a final temperature of 300°C. Lignin phenols (terrestrial biomarkers) were quantified using the response factors of commercially available standards (Sigma-Aldrich) whereas the rest of the CuO oxidation products were quantified by comparing the response factor of trans-cinnamic acid. Lignin-derived reaction products include vanillyl phenols (V=vanillin, acetovanillone, vanillic acid), syringyl phenols (S=syringealdehyde, acetosyringone, syringic acid) and cinnamyl phenols (C=p-coumaric acid, ferulic acid). In addition to lignin, cutin-derived products (hydroxyl fatty acids) were used to trace the land-derived input (Goñi and Hedges, 1990; Tesi et al., 2010). Other CuO oxidation products include para-hydroxybenzene monomers (P-series), benzoic acids (B-series) and short-chain fatty acids (FA-series) which can have both terrestrial and marine origin (Goñi and Hedges, 1995; Tesi et al., 2010).

The sSea-ice proxy IP_{25} (mono-unsaturated highly branched isoprenoid (HBI) alkenemonounsaturated highly branched isoprenoid) was quantified according to Belt et al. (2012). IP_{25} producers are a minor (<5%) fraction of the total sea-ice taxa which are, however, ubiquitous in pan-Arctic sea-ice. Species include *Pleurosigma stuxbergii var. rhomboide*, *Haslea crucigeroides* (and/or *Haslea spicula*) and *Haslea kjellmanii* (Brown et al., 2014a). Briefly, lipids were extracted via sonication using a dichloromethane/methanol solution (2:1 v/v × 3). Prior to the extraction, two internal standards (7-hexylnonadecane, 7-HND and 9- octylheptadecene, 9-OHD) were added to permit quantification of IP_{25} (monounsaturated highly branched isoprenoid) following analysis via GC-MS. Total lipid extracts (TLEs) were dried under N_2 after removing the water excess with anhydrous $NaSO_4$. Dry TLEs were redissolved in dichloromethane and the non-polar hydrocarbon fraction was purified using open column chromatography (deactivated SiO_2) and hexane as eluent. Saturated and unsaturated n-alkanes were further separated using 10% $AgNO_3$ coated silica gel using hexane and dichloromethane, respectively.

Quantification of IP_{25} was carried out in SIM mode (*m/z* 350.3) as described in Belt et al. (2012). The GC was fitted with a 30m×250 µm DB5ms (0.25 µm thick film) capillary GC column. Initial GC oven temperature was set to 60°C followed by a 10°C/min ramp until a final temperature of 310°C (hold time 10 min).

23.3. Microscope images of plankton

High resolution digital images were taken with an Environmental Scanning Electron Microscope (ESEM) Philips XL30 FEG in high voltage (15kV) and magnification 250X. Samples were further studied for identification of diatoms and dinoflagellates using a transmitted light microscope (Leitz Laborlux 12 Pol) equipped with differential interference contrast optics at 1000X magnification. Microscope slides were prepared using settling chambers to achieve an even distribution of particles on the cover glass, regardless of size and shape Warnock and Sherer (2014).

2.4 WEGAS measurements of CO_2aq

Cavity ring-down spectrometer (CRDS) measurements were used to continuously monitor CO_2 concentrations and $\delta^{13}C_{CO2}$ composition of gas stripped via headspace equilibration from the water column using the Water Equilibration Gas Analyser System (WEGAS) (Thornton et al. 2016). It consists of three major components:

a) Water handling system including i) showerhead equilibrator (head space volume 1 L) fed by the sea water intake described above, ii) continuous pH measurements by E&H electrode probe and iii) T and salinity measurements by Seabird TSG 45.

b) CRDS gas analyzers for CO_2 stable carbon isotopes (model G2131-i, Picarro Inc., Sunnyvale, CA) and CO_2 concentrations (model G2301, Picarro Inc., Sunnyvale, CA).

c) Gas handling system with circulation pumps for headspace and ambient air from meteorological tower.

Continuous measurements of surface water CO_2 and $\delta^{13}C_{CO2}$, were thus performed using IB/Oden's seawater intake. Water was pumped through spray nozzles into the open headspace equilibrator at ~4.5 L min^{-1}. By creating a fine spray of droplets, the exchange surface between headspace and water is maximized and an optimal equilibration is achieved. The gas of the headspace was analysed using two different CRDS (cavity ring-down) analysers. The second analyser was operated in parallel and its flow (~25 mL min^{-1}) was not fed back into the closed cycle. Thus, it created a defined vent flow. This vent flow is compensated by a flow of ambient air (AA) taken from the top inlet of the meteorological tower (20 m height). To be able to correct the data for the vent flow, the concentration of CO_2 and $\delta^{13}C_{CO2}$ in AA is monitored by frequent switching. During the SWERUS-C3 expedition, continuous CO_2 and $\delta^{13}C$-CO_2 measurements in the surface waters have been performed in the period 10 July - 9 August resulting in a total of 238 864 data points.

23.45 Sea-ice data

Daily AMSR2 sea-ice extent and concentration maps were provided by the Institute of Environmental Physics, University of Bremen, Germany (Spreen et al., 2008) as GeoTIFF files (ftp://seaice.uni-bremen.de).

3.5 Statistics

We used two-tailed T-test (homoscedasticity) and Welch T-test (heteroskedasticity) to assess whether the differences between open waters and sea-ice dominated waters were statistically significant. For this study, significance level (alpha) was set at 0.01.

3. Results and discussion

34..1 Surface water conditions during the SWERUS-C3 expedition

Before discussing the chemical composition of the supra-POM (>10 µm)fraction, here we briefly introduce the different environmental conditions encountered throughout the cruise track. The surface water data presented in this section were pulled together from previous studies which provide an in-depth analysis of the surface water properties during the SWERUS-C3 expedition in 2014 (Humborg et al., 2017; Salvadó et al., 2016) (Table 2). For this study, continuous CO_2aq and $\delta^{13}C_{CO2}$ data (Humborg et al., 2017) acquired throughout the expedition were averagedorganized to match the water sampling stations allowing for a direct comparison with DOC and salinity data (Fig. 2) (Supplementary Material).

Summer 2014 was consistent with the long-term downward trend in Arctic sea-ice extent. The strongest anomalies were observed in the LS which experienced the most northerly sea-ice shift since satellite observations began in 1979 (National Snow and Sea Ice Data Center, NSIDC. http://nsidc.org/data). Unpublished data). In general, sea-ice displayed a strong gradient over the study region going from ice-free conditions in the outer LS to ice-dominated waters in the outer ESS (Fig.1.) Three snapshots of the sea-ice extent and concentrations (-Figure 1 displays sea-ice extent and concentration.i.e. at the beginning, in the middle and at the end of the sampling) is shown in Fig.1. Furthermore, Table 1 reports the averaged sea-ice concentrations encountered during the collection offor each sample.

The surface water salinity exhibited a longitudinal trend characterized by low values in the outer LS while the sea-ice dominated ESS waters showed relatively higher values (Fig. 2a; Table 2). However, the highest salinity values were measured in the westernmost stations resulting in a sharp gradient in the LS. The low surface water salinities in the outer LS are most likely the result of both Lena river input and sea-ice thawing (Humborg et al., 2017) that started in late May (Janout et al., 2016).

The highest DOC concentrations were measured in the mid-outer LS in the surface water plume affected by Lena River runoff (Fig.2b; Table 2). Overall, DOC concentrations

~~mirrored~~ followed the plume dispersion ~~salinity distribution~~ with high DOC concentrations corresponding to low salinities (Fig. 2). Carbon stable isotopes ($\delta^{13}C$) and terrestrial biomarkers (of the solid-phase extracted DOC fraction; Salvado et al., 2016) further confirmed the influence ~~of terrestrial~~of terrestrial DOC in the outer LS, while the ~~terrestrial~~ land-derived input progressively~~imprint~~ decreased moving eastward.

CO_2aq concentrations exhibited a typical estuarine pattern over the study region (Humborg et al., 2017) (Fig. 2d; Table 2). Low salinity waters in the outer LS showed above atmospheric CO_2 concentrations (i.e., oversaturation) while surface waters below sea-ice exhibited undersaturated concentrations. The most depleted $\delta^{13}C_{CO2}$ values were measured off the Lena river mouth ~~(Humborg et al., 2017)~~ (Fig. 2e; Table 2). Being relatively rich in land-derived material, it is likely that respired terrestrial OC within the Lena river plume exerted control on the CO_2 isotopic signature and concentration (Humborg et al., 2017).

Finally, nutrient distribution revealed nitrate (NO_3) and nitrite (NO_2) depletion in surface waters throughout the cruise track (Humborg et al., 2017) in comparison with the Arctic Ocean gateways such as the Bering strait. Here, ~~(Torres-Valdés et al., 2013)~~nutrient concentrations in surface waters are two-order of magnitude higher compared the our study region (Torres-Valdés et al., 2013).~~.~~ Phosphate (PO_4) exhibited rather low concentrations in the outer LS and relatively higher concentrations below the sea-ice in the outer ESS (Humborg et al., 2017) likely reflecting the ~~Pacific~~ inflow of nutrient-rich Pacific waters (Anderson et al., 2011; Semiletov et al., 2005; Torres-Valdés et al., 2013).

5. Results and discussion

5.1~~2~~ Source of the ~~supra-micron~~ POM (>10 µm) fraction

The Arctic Ocean off northern Siberia receives large quantities of dissolved and particulate terrestrial organic carbon ~~(TerrOC)~~ via continental runoff and coastal erosion

(Alling et al., 2010; Dittmar and Kattner, 2003; McClelland et al., 2016; Sánchez-García et al., 2011; Semiletov et al., 2013; Vonk et al., 2012). The land-derived material that does not settle in the coastal zone further travels across the continental margin reaching out to the outer-shelf region resuspended within the benthic nepheloid layer or in suspension within the surface river plume (Fichot et al., 2013; Sánchez-García et al., 2011; Wegner et al., 2003). Another fraction of terrestrial material can travel across the Siberian margin trapped in fast ice (Dethleff, 2005). Considering the potential allochthonous contribution,~~Thus,~~ we addressed to what extent ~~TerrOC~~ terrestrial organic material affects the ~~supra-~~POM (>10µm) fraction by quantifying the concentration of lignin phenols and C16-18 hydroxy fatty acids (cutin-derived products). These biomarkers are exclusively formed by terrestrial vegetation and, thus, serve as tracers of ~~TerrOC~~ land-derived material in the marine ~~Arctic~~ environment (Amon et al., 2012; Bröder et al., 2016b; Feng et al., 2015).

Upon CuO alkaline oxidation the ~~supra-~~POM (>10µm) samples yielded only traces of lignin phenols while the cutin-derived products were not detected (Fig. 3). Other oxidation products in high abundance included saturated and mono-unsaturated short chain fatty acids (C12-18FA), para-hydroxy phenols, benzoic acids and dicarboxylic acids. These other reaction products are ubiquitous in both marine and terrestrial environments but they are predominant in plankton-derived material, especially short-chain fatty acids (Goñi and Hedges, 1995)~~.~~. When compared with active-layer permafrost soils and ice-complex deposits (Tesi et al., 2014), ~~supra-~~POM (>10µm) –samples displayed a distinct CuO fingerprint dominated by short chain fatty acids (Fig. 3), consistent with the typical CuO ~~fingerprints~~ products yields by~~of~~ phytoplankton batch cultures upon CuO alkaline oxidation (Goñi and Hedges, 1995). SEM images further corroborated the abundance of marine plankton detritus in the ~~supra-~~POM (>10µm) fraction while lithogenic particles (clastic material) appeared to be sporadic in all samples.

The OC content (% d.w.) of the ~~supra~~ POM (>10um) fraction decreased eastwards showing high concentrations in the LS and relatively low values in the ESS (Table 1; *p*<0.01 T-test). However, in terms of absolute concentration in the water column (μC/l), the highest levels were generally observed in the sea-ice covered region (Table 1; Fig. 4a; *p*<0.01 T-test). Qualitative analyses by SEM and transmitted-light microscopy highlight important differences in plankton assemblages which reflect different timing of the plankton blooms which can explain these differences in concentration. Specifically, the open-water LS stations exhibited a low degree of plankton diversity and were largely dominated by a bloom of heterotrophic dinoflagellate cysts (*Protoperidinium* spp) (Fig. 5a; Table 3). Moving towards the ice-dominated regions, diatoms become the prevailing species. Dominant diatom genera include *Chaetoceros spp.* (dominant diatom in several stations), *Thalassiosira spp.*, *Rhizosolenia spp.*, *Coscinodiscus spp.*, *Asteromphalus spp.*, *Navicula spp.* as well as sea-ice species such as *Fragilariopsis cylindrus* and *Fragilariopsis oceanica* (Fig. 5b,c; Table 3).

~~A m~~Moored ~~line~~ optical sensors deployed in the LS shelf recorded the sea-ice retreat in 2014 and found no sign of pelagic under-ice blooms despite available nutrients while high chlorophyll concentrations were detected immediately after the ice retreated in late May (Janout et al., 2016). The ice-edge blooms lasted for about 2 weeks according to the high resolution chlorophyll time-series (Janout et al., 2016). Thus, our post-bloom sampling in the LS essentially captured an oligotrophic environment ~~(Gustafsson et al., 2016)~~ dominated by heterotrophic dinoflagellate cysts (i.e, Protoperidinium spp) which likely fed on phytodetritus and river-derived organic material. Such conditions are fairly consistent with the relatively low carbon contents (μgC/L) observed in LS waters (Fig. 4a).

The Arctic sea-ice biomarker IP25 (Fig. 4b) ~~further~~ further highlight the different regimes observed in ~~corroborated the sampling of different plankton taxa in~~ ice-free and ice-dominated surface waters. IP25 is a proxy of sea-ice based on a highly branched mono-unsaturated isoprenoid alkene found in some sea-ice diatoms which, however, generally account for 5% of the total sea-ice taxa (Belt et al., 2007; Brown et al., 2014b). The IP25 concentrations varied by several orders of magnitude over the study area showing low concentrations in the open-water western region while the sea-ice dominated surface waters to the east exhibited high concentrations especially at station 31b (Fig. 4b; Table 1) ; $p<0.01$ Welch T-test). The fact that IP25 was still detectable throughout the ice-free outer LS suggests that the proxy captured the signal of the sea-ice retreat that occurred shortly before the sampling at the end of May/early June (Janout et al., 2016). Alternatively, the IP25 could have been advected from nearby sea-ice dominated regions.

~~3~~5.2~~3~~. Dual carbon isotopes: $\delta^{13}C$ and $\Delta^{14}C$ ~~of the supra-micron POM fraction~~

$\delta^{13}C$ and $\Delta^{14}C$ of the ~~supra-~~POM (>10µm) ~~fraction~~ samples ~~also~~ exhibited a distinctive longitudinal trend across the study area between LS and ESS (Fig. 4c,d) ($p<0.01$ T-test) .

Depleted $\delta^{13}C$ values characterized the LS open waters ranging from -28.1 to -24.7‰ (Fig. 4c). Although within the range of terrestrially-derived material, our CuO oxidation data (i.e. trace of lignin phenols and absence of cutin-derived products) suggest that the "light" isotopic composition in the LS might instead reflect the plankton assemblage dominated by heterotrophic dinoflagellate cysts as previously described (e.g., *Protoperidinium* spp; Fig. 5a). More specifically, heterotrophic dinoflagellates can adapt their metabolism depending on the substrate available (e.g., diatoms and bacteria). Several studies have shown that terrestrial DOC greatly promotes bacteria biomass production which in turn stimulates the growth of heterotrophic dinoflagellates (Carlsson et al., 1995; Purina et al., 2004; Wikner and Andersson, 2012). Thus, in these conditions, allochthonous terrestrial DOC is actively recycled by bacteria and transferred to dinoflagellates which explains, thus, the depleted $\delta^{13}C$ values observed in the river-dominated samples (Carlsson et al., 1995).

The modern radiocarbon fingerprint of the Lena DOC discharge is consistent with $\Delta^{14}C$ signature of the supra-POM (>10μm) fraction in the LS (up to +99 ‰), supporting the importance of terrestrial DOC as a carbon source for the food web in the river plumesupporting, thus, the DOC microbial loop within the river plume (Fig. 4d and 6). By contrast, comparison with other potential carbon sources which include the Lena river particulate organic carbon, surface sediments, Pleistocene coastal Ice-Complex Deposit and Pacific DIC inflow reveals a different (more depleted) radiocarbon fingerprint (Fig. 6). -It is also import to highlight that the DOC within the Lena plume is one/two-order of magnitude higher than the particulate carbon pool supporting, thus, our hypothesis (Humborg et al., 2017; Salvadó et al., 2016).

Moving towards the ice-dominated ESS, surface waters progressively become more autotrophic and productive (Humborg et al., 2017) while the supra-POM (>10μm) exhibited a wide $\delta^{13}C$ signature ranging from -28.6 to -21.2‰ (Fig. 4c). The most depleted values were observed across the transition zone between open-waters and sea-ice. Visual inspections of these samples revealed large abundance of the centric diatom *Chaetoceros* spp. (spores and vegetative cells; St22, Fig. 5b) while lignin and cutin data indicated, a negligible input of land-derived material. Primary factors determining the fractionation of stable carbon isotopes in phytoplankton are several and include CO_2aq concentration, δ^{13}Caq, growth rate, cell size, cell shape, light and nutrient availability (Gervais and Riebesell, 2001; Laws et al., 1997a; Popp et al., 1998; Rau et al., 1996). Our understanding about isotopic fractionation has been historically achieved via laboratory experiments designed to test each factor under controlled conditions. In natural environments, however, different factors can compete with each other, sometimes in opposite directions. Yet, the existing knowledge about surface water properties during the expedition (Humborg et al., 2017) can provide important constraints for the isotopic signal interpretation.

For example, comparison with continuous $\delta^{13}C$-CO_2aq and CO_2aq data measured throughout the cruise track - time-averaged to match the large volume filtration along the cruise track (Table 1) - suggested a negligible role exerted by $\delta^{13}C$-CO_2aq (Fig. 7b) while CO_2aq concentration correlated with the $\delta^{13}C$ of the ~~supra-~~POM (>10µm) fraction (r^2=0.72; *p*<0.01) (Fig. 7a). Such a relationship fits with the general model according to which a low demand (i.e., low growth rate) and high supply (i.e., abundant CO_2aq) favour high fractionation and vice versa (Laws et al., 1997b; Laws et al., 1995; Wolf-Gladrow et al., 1999).

During the expedition, surface water properties (i.e. O_2 and CO_2, Table 2) (Humborg et al., 2017) suggest that the productivity in the outer ESS increases moving eastward, as commonly observed, likely due to the Pacific inflow (Björk et al., 2011; Semiletov et al., 2005). ~~Thus~~As a result, the wide range of plankton $\delta^{13}C$ over the ESS can be explained in terms of two different regimes: (a) in the transition zone between open waters and sea-ice, the productivity was low but CO_2aq was oversaturated while (b) in the easternmost ESS, productivity was high but CO_2aq was depleted (Fig. 7b). The former regime favours fractionation while the latter does not (Fig. 7b). Different diatom assemblages can also be another factor to consider although the phytoplankton diversity observed over ESS can be considered rather small (e.g. *Chaetoceros spp.* dominant in most of the samples) compared to the wide range of $\delta^{13}C$ observed (i.e., from -28.8 to -21.6) (Table 3).

The ~~supra-~~POM (>10µm) fraction in the sea-ice dominated ESS exhibited slightly - but consistently - depleted $\Delta^{14}C$ values ranging from -62 to -49 ‰ (Fig. 4d). This region is affected by the inflow of Pacific waters whose DIC exhibits, however, a modern $\Delta^{14}C$ signature (Griffith et al., 2012) (Fig. 6). By contrast, these results suggest the influence from an aged carbon pool. As the ESS remains covered by sea-ice for most of the year, it is possible that the sea-ice hampers the gas exchange with the atmosphere and acts as a lid by trapping CO_2 which derives from the breakdown of sedimentary organic material (Anderson et al., 2009; Semiletov et al., 2016), which might have such ages (Bröder et al., 2016a; Vonk et al., 2012). In these conditions, the pre-aged CO_2 accumulates underneath the sea-ice and is subsequently incorporated during carbon fixation by the phytoplankton. While oversaturated bottom waters were extensively documented in the region with important consequences on the local DIC (Anderson et al., 2009; Pipko et al., 2009), more work is clearly needed to understand if early diagenesis in sediments can also affect the radiocarbon signature of the CO_2aq underneath the sea-ice. Alternatively, the slightly depleted radiocarbon signature might indicate the presence of pre-aged terrestrial organic carbon (Fig. 6) in the ~~supra-~~POM (>10μm) samples, not reflected in the lignin and cutin tracers (Fig. 3). However, it would then remain elusive why such an aged land-derived influence was not visible in the river-dominated LS waters while it affected the sea-ice dominated region.

Taken together, our results indicate that the dual-carbon isotope fingerprint is highly affected by the trophic conditions (heterotrophic *vs* autotrophic) as well as the extent of primary productivity. In a warming scenario characterized by sea-ice retreat (Arrigo et al., 2008; Comiso et al., 2008) and enhanced terrestrial input from land as result of hydrology and permafrost destabilization (Frey and Smith, 2005; Vonk et al., 2012), the geochemical composition of plankton will likely change as the warming proceeds.

6. Conclusions

Analyses of large-volume filtrations of plankton-dominated >10 μm particle samples revealed a high degree of heterogeneity in ~~geochemical~~ the dual carbon isotope signature ($\delta^{13}C$ and $\Delta^{14}C$) ~~and plankton composition with an in~~ between ice-free waters (Laptev Sea) and the ice-covered region (East Siberian Sea).

~~Thus, the dual-carbon isotope fingerprint of the plankton-dominated fraction reflects a contemporary terrestrial DOC signature (i.e., depleted $\delta^{13}C$ and modern $\Delta^{14}C$ fingerprint). Heterotrophic dinoflagellates dominated the plankton assemblages in this ice-free region.~~ Our ~~Hence,~~ results suggest a heterotrophic environment in the outer LS open waters where the $\delta^{13}C$ depleted river ~~-derived~~ DOC is transferred to relatively higher trophic levels via microbial incorporation in the river plume ~~(i.e, microbial loop)~~.

Moving eastwards towards the ice-dominated outer ESS, surface waters became progressively more autotrophic~~e and largely dominated by diatoms~~. Here, the isotopic fractionation appears to follow the phytoplankton growth *vs* CO_2 demand model according to which carbon fractionation ~~increases~~ decreases at ~~low~~ high growth and ~~high~~ low CO_2 ~~supply~~concentrations. As a result, the transition between open-waters and sea-ice exhibited more depleted $\delta^{13}C$ values compared to the productive easternmost stations. Radiocarbon signatures were slightly depleted over the whole sea-ice dominated area. This raises the question whether the sea-ice hampers the gas exchange with the atmosphere and trap the CO_2 sourced from reactive sedimentary carbon pools.

In a warming scenario, it is likely that the oligotrophic ice-free LS will be dominated by heterotrophic metabolism fuelled by terrestrially-derived organic material (i.e., Lena input). In these conditions, the dual-carbon isotope signature of the heterotrophic plankton will essentially reflect the terrestrial ~~input~~fingerprint. In the ESS, which receives the inflow of the nutrient-rich Pacific waters, ice-free conditions will ~~promote~~ enhance~~the~~ light penetration. This in turn might further stimulate phytoplankton growth with important implications in terms of CO_2 depletion and resulting low isotope fractionation. Altogether, ~~It is likely that~~ this will result in a sharp compositional gradient (e.g. $\delta^{13}C$) between LS and ESS similar to what ~~has already been~~ captured in our semi-synoptic study.

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Table 1. Chemical composition of the Supra-micron POM (>10µm) (supraPOM) compositionfraction and continuous CO_2aq measurements*

ID	Time averaged latitude (N)	Time averaged longitude (E)	Mean sea-ice percentage (%)	~~Supra-micron~~ POM (>10μm) concentration (mg/l)	OC (d.w.)	$\delta^{13}C$ (‰)	$\Delta^{14}C$ (‰)	IP25 (ng/gOC)	averege CO_2aq (ppm)*	average $\delta^{13}C-CO_2aq$ (‰)*
ST4	81.68	105.96	98.4	6	18.2	-26.7	n.d.	n.d.	323	-10.9
ST5	80.47	114.07	98.7	15	42.6	-27.6	n.d.	n.d.	322	-11.0
ST6	78.86	125.22	82.2	1	51.7	-26.6	99	n.d.	325	-10.8
ST7	77.88	126.62	0.0	11	43.1	-25.7	n.d.	88	350	-10.7
ST8	77.16	127.32	0.0	17	30.9	-26.7	41	n.d.	391	-10.5
ST9	76.78	125.83	0.0	3	31.5	-27.9	30	48	385	-10.5
ST10	76.90	127.81	0.0	11	40.9	-24.7	n.d.	n.d.	349	-11.0
ST11	77.12	126.66	0.0	13	29.6	-28.1	27	13	428	-10.7
ST22	77.67	144.63	0.0	20	11.3	-28.8	n.d.	95	394	-11.0
ST23	76.43	147.53	0.0	6	7.6	-28.5	-50	n.d.	394	-11.2
ST24	76.42	149.84	34.4	19	11.9	-26.8	-62	368	374	-11.1
ST25	76.62	152.03	96.7	23	19.5	-25.7	-31	465	263	-10.8
ST26	76.14	157.85	96.2	109	30.8	-24.2	-30	217	316	-10.9
ST27	75.00	161.03	91.5	41	23.3	-23.0	n.d.	256	299	-11.1
ST28	74.63	161.98	86.3	28	15.5	-23.8	n.d.	n.d.	214	-11.3
ST29	73.61	169.72	79.3	31	14.7	-23.2	-50	518	184	-11.3
ST30	75.61	174.01	66.7	43	22.6	-27.0	n.d.	n.d.	304	-10.5
ST31A	75.85	174.41	75.6	30	10.9	-21.6	-62	1911	182	-10.6
ST31B	74.26	173.74	63.5	15	4.6	-23.3	n.d.	783	n.d.	n.d.
ST32	73.56	176.06	51.8	21	11.3	-24.5	-58	131	n.d.	n.d.
ST33	72.35	-175.14	0.0	20	15.5	-23.5	n.d.	473	n.d.	n.d.
ST34	73.28	-173.05	28.7	76	13.4	-21.6	-52	970	n.d.	n.d.
ST35	75.21	-172.05	53.9	24	14.3	-24.2	n.d.	268	n.d.	n.d.

n.d = not determined

*Humborg et al. (2017)

Table 2. Surface water (0-20 m) chemical and physical properties during the SWERUS-C3 expedition*

	Salinity	Temperature	DIC	DOC	POC	$\delta^{13}C$-DIC	NO_2 - NO_3	PO_4	O_2
		ºC	µmol kg^{-1}	µmol kg^{-1}	µmol kg^{-1}	‰	µmol kg^{-1}	µmol kg^{-1}	µmol kg^{-1}
	median	**median**	**median**	**median**	**median**	**median**	**median**	**median**	**median**
Outer LS shelf (0-20 m)	32.87	3.84	2139	149.1	7.9	0.75	0.21	0.27	323.0
LS shelf break (0-20 m)	33.56	0.57	2114	91.5	10.1	1.10	0.26	0.15	364.9
Outer ESS shelf (0-20 m)	29.45	-1.33	1969	84.2	10.7	1.14	0.25	0.97	381.5
ESS shelf break (0-20 m)	28.23	-1.32	1979	73.7	4.6	1.47	0.11	0.59	394.1
	mean	**mean**	**mean**	**mean**	**mean**	**mean**	**mean**	**mean**	**mean**
Outer LS shelf (0-20 m)	31.17	3.40	2119	179.8	7.9	0.58	0.60	0.29	327.0
LS shelf break (0-20 m)	33.42	0.96	2111	97.5	10.0	1.10	0.61	0.16	358.1
Outer ESS shelf (0-20 m)	28.95	-0.05	1949	95.8	11.9	1.26	0.26	0.95	386.8
ESS shelf break (0-20 m)	28.27	-1.31	1975	72.0	4.6	1.49	0.12	0.60	397.0
	s.d.	**s.d.**	**s.d.**	**s.d.**	**s.d.**	**s.d.**	**s.d.**	**s.d.**	**s.d.**
Outer LS shelf (0-20 m)	3.22	2.38	89	66.3	1.7	0.50	0.91	0.11	14.6
LS shelf break (0-20 m)	0.70	2.07	23	21.2	1.7	0.11	0.74	0.06	22.5
Outer ESS shelf (0-20 m)	1.41	2.28	75	30.2	4.6	0.49	0.12	0.19	32.2
ESS shelf break (0-20 m)	0.53	0.04	49	3.2	0.3	0.08	0.03	0.02	8.3

*data from Humborg et al. (2017) and Salvadó et al. (2016)

Table 3. Qualitative plankton characterization of selected supra-micron POMC (>10μm) samples

ID	Region	Diatoms	Dinoflagellates	Other species
ST6	LS	Few *Coscinodiscus*	None observed	
ST9	LS	None observed	Few *Protoperidinium*	
ST11	LS	None observed	Abundant *Protoperidinium*	
ST22	LS-ESS	Abundant *Chaetoceros*, few *Rhizosolenia*, *Thalassiosira*	None observed	
ST25	LS-ESS	High diversity. Abundant *Chaetoceros*, *few Rhizosolenia*, *Coscinodiscus*, *Thallasiosira*, *Asteromphalus*, *Navicula*	None observed	Silicoflagellate
ST31A	ESS	High diversity. Abundant *Chaetoceros*, *few Rhizosolenia*, *Thallasiosira*, *Bacterosira*, *Navicula*	None observed	
ST31B	ESS	High diversity. Few *Chaetoceros*, *Thallasiosira*, *Fragilariopsis*	Few *Protoperidinium*	
ST34	ESS	Abundant *Chaetoceros*, few *Thalassiosira*, *Navicula*	Few *Protoperidinium*	

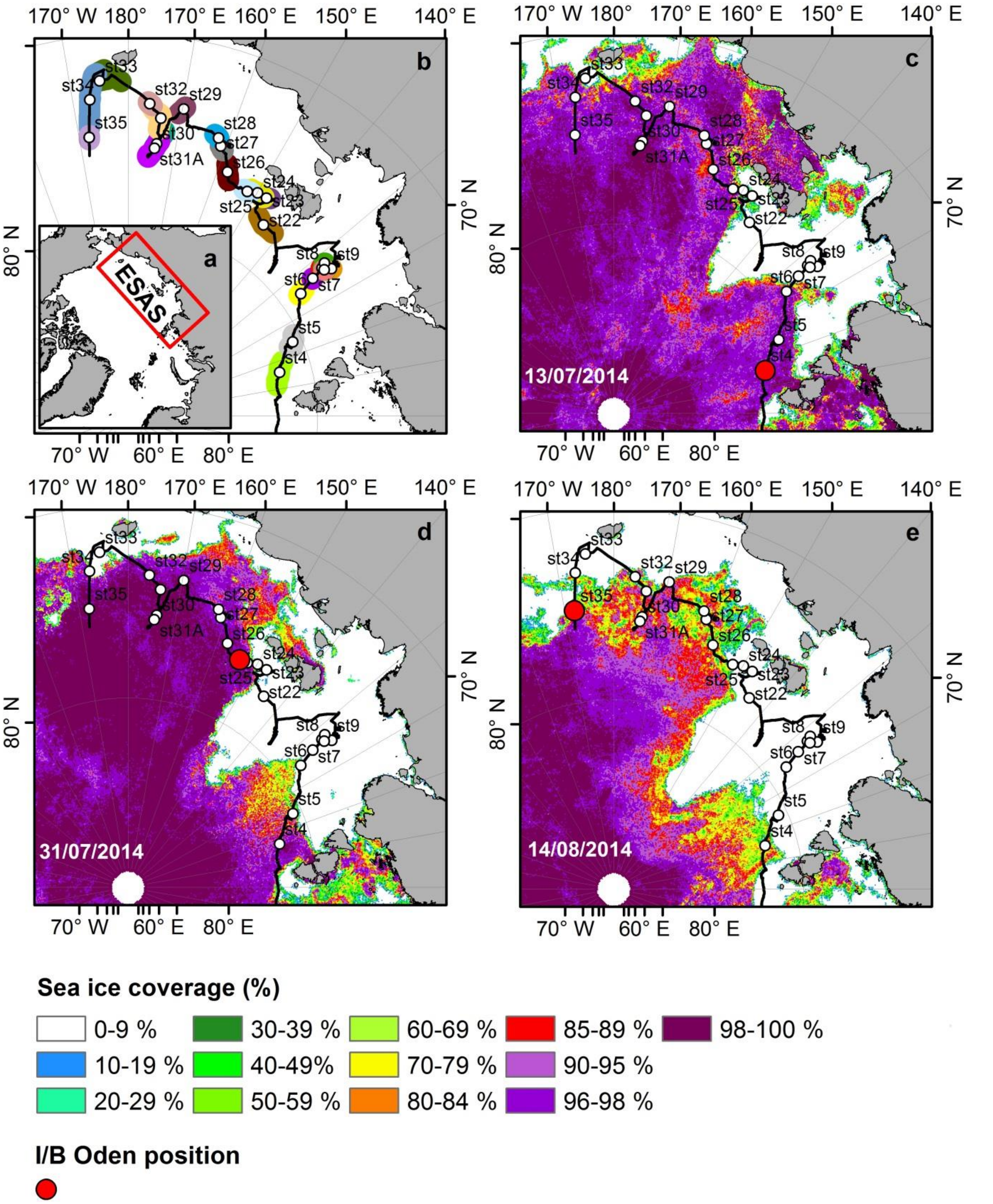

Fig. 1 (a) The study area in the East Siberian Arctic Shelf. (b) Time-averaged position during the large-volume filtration (circles) of the ~~supra-~~POM (>10µm) samples. Shaded coloured areas show the sampling area covered to harvest each ~~supra-~~POM (>10µm) sample. Sea-ice extent and concentration at the beginning (c), in the middle (d) and at the end (e) of the sampling campaign. The ship position is shown by a filled red circle.

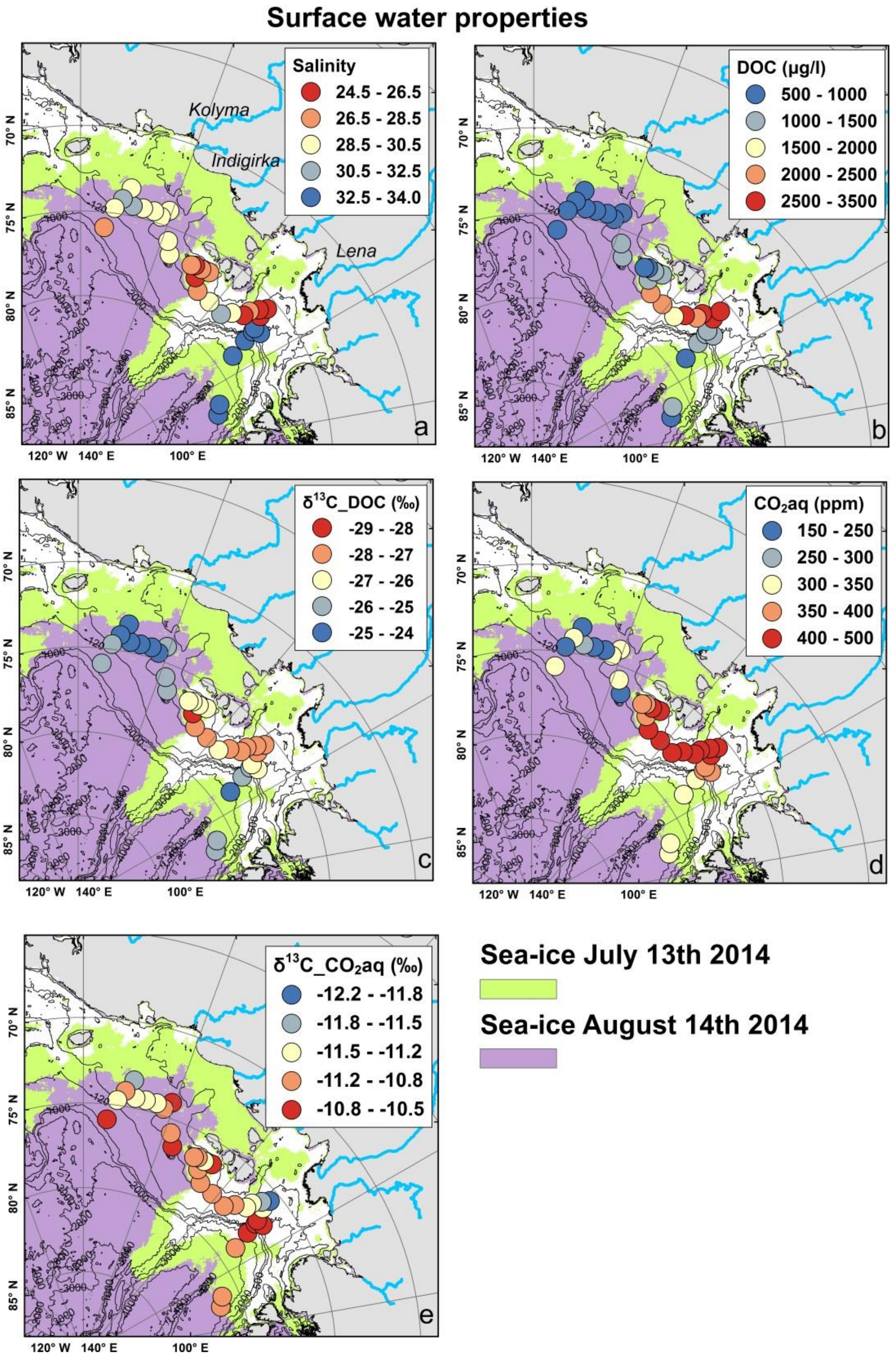

Fig.2 Surface water properties. (a) Salinity. (b) DOC. (c) δ^{13}C-DOC. (d) CO_2aq. (e) δ^{13}C-CO_2aq. Shaded areas show the sea-ice extent at the beginning (13/07/2014) and at the end of the sampling campaign (14/08/2014) (Humborg et al., 2017; Salvadó et al., 2016).

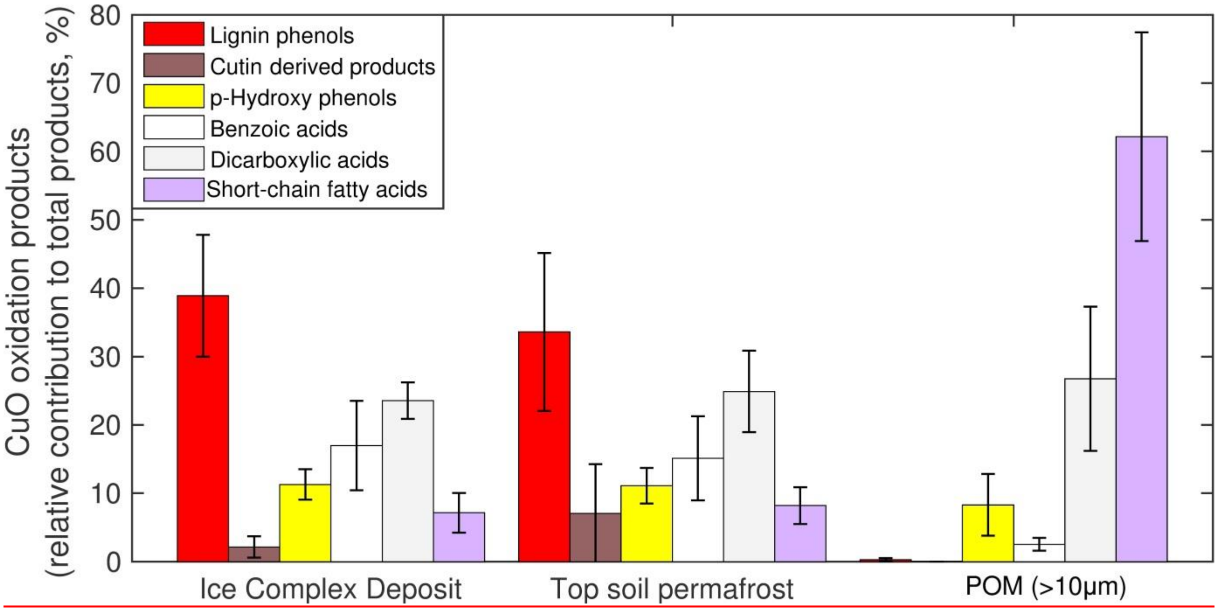

Fig.3 Alkaline CuO fingerprint of top-soil permafrost samples (Tesi et al., 2014), Pleistocene Ice Complex Deposit (Tesi et al., 2014) and ~~supra P~~POM (>10µm) fraction (this study). The plot displays the relative proportion ~~of the CuO oxidation~~ products yield upon alkaline CuO oxidation. The error bar refers to the natural variability of each dataset~~.~~

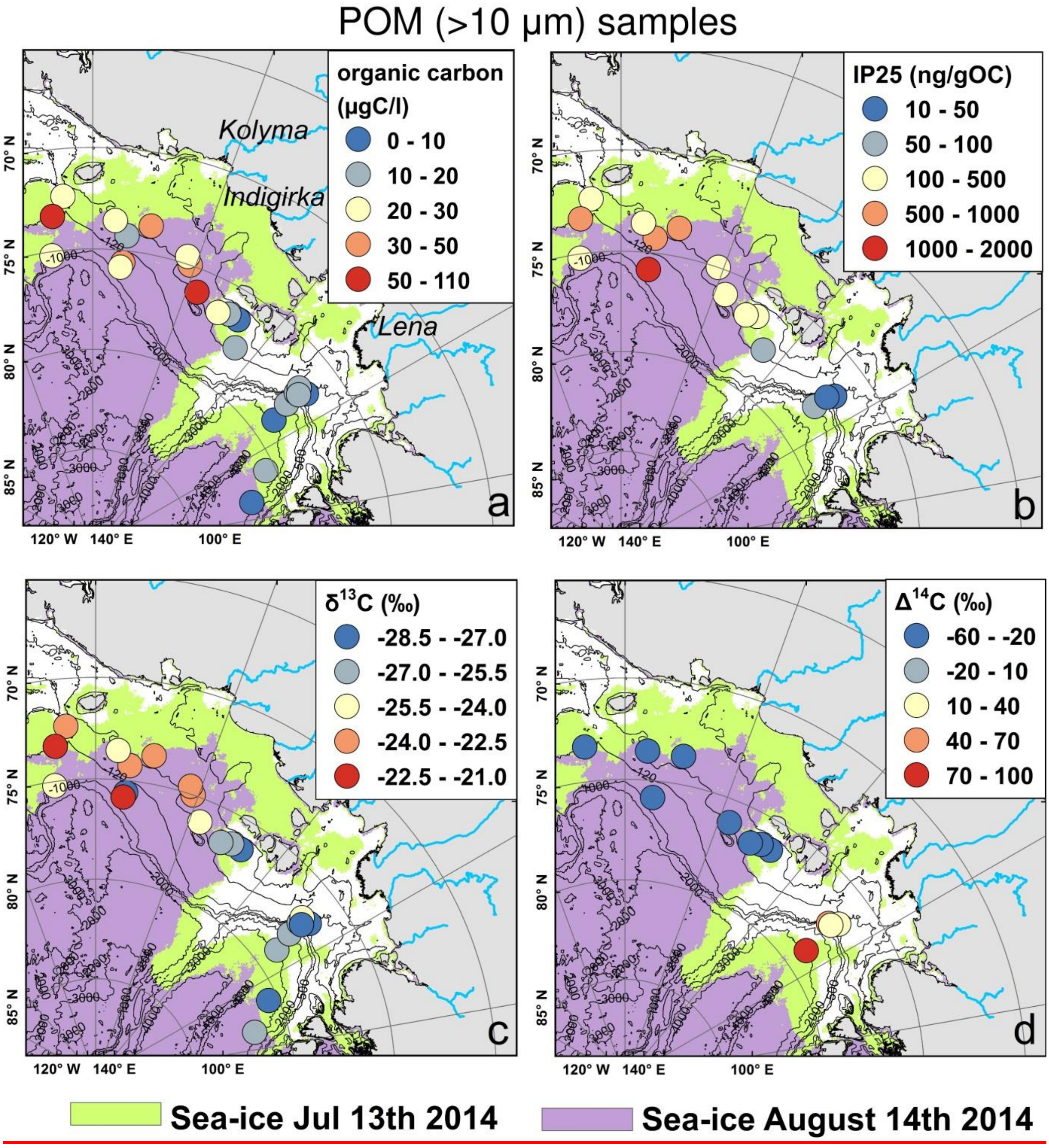

Fig. 4 ~~Supra~~ POM (>10µm) composition~~.~~ (a) Organic carbon concentration. (b) IP25 (mono-unsaturated highly branched isoprenoid. (c) $\delta^{13}C$. (d) $\Delta^{14}C$. Shaded areas show the sea-ice extent at the beginning (13/07/2014) and at the end of the sampling campaign (14/08/2014).

Fig. 5 SEM images. (a) ST-11: Dinoflagellates (*Protoperidinium* spp.) in open-waters of the Laptev Sea. (b) ST22: Diatoms, mostly spines (setae) of *Chaetoceros* spp. in the transition between Laptev Sea and East Siberian Sea. (c) ST-34: Diatoms from sea-ice dominated waters in the East Siberian Sea

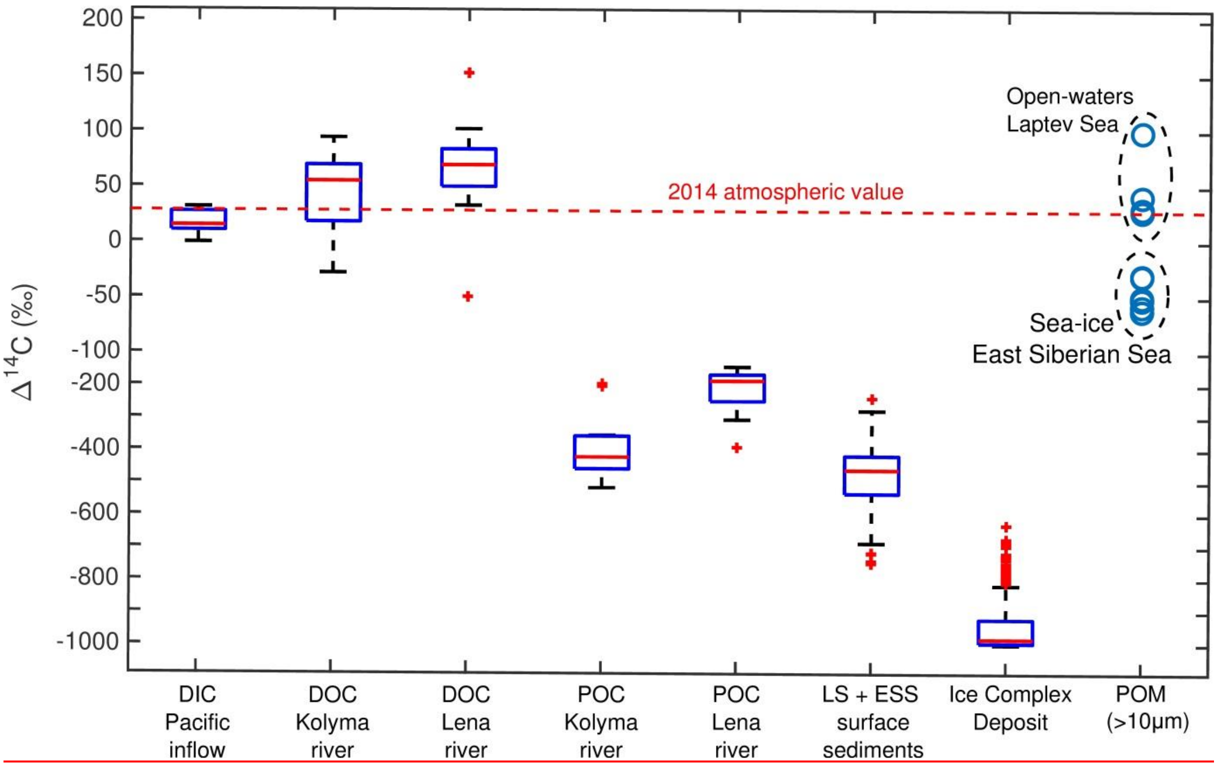

Fig. 6 Radiocarbon signature of inorganic and organic carbon pools. Whisker plots of radiocarbon values for different inorganic and organic carbon sources from the literature, compared to the outer Laptev Sea and outer East Siberian Sea (blue circles, this study). Solid lines show the median, the box limits display the 25th and 75th percentiles while the crosses show the outliers. Source: DIC (Griffith et al., 2012), DOC-Kolyma (2009-2014), DOC-Lena (2009-2014), POC-Kolyma (2009-2011), POC-Lena (2009-2011) (www.arcticgreatrivers.org), Laptev Sea and Eastern Siberia Sea surface sediments (Salvadó et al., 2016; Vonk et al., 2012) and Ice Complex Deposit (Vonk et al., 2012).

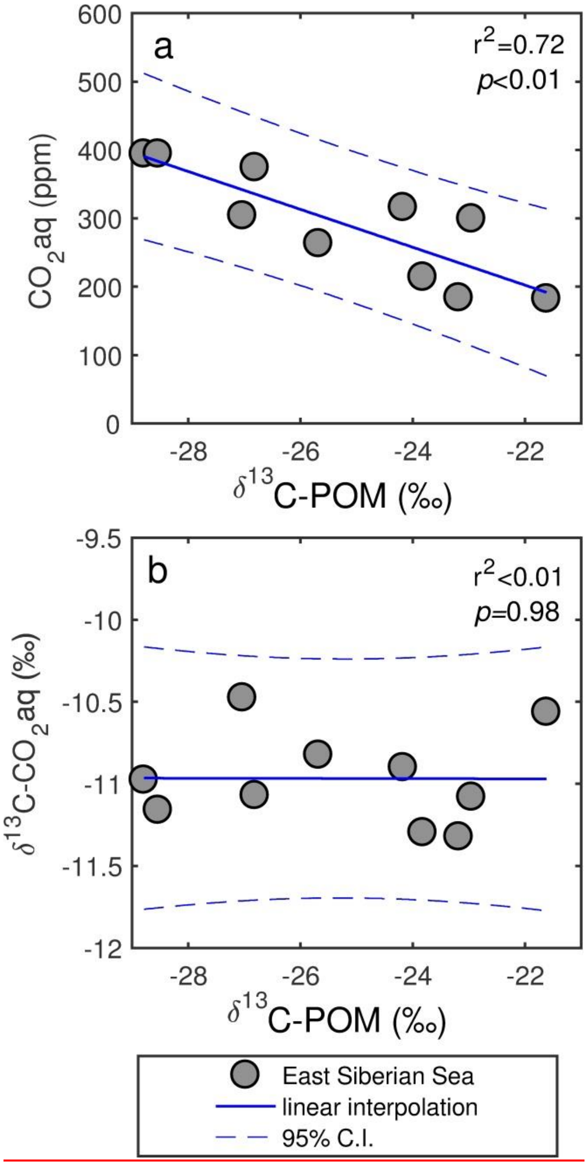

Fig. 7 Correlations (a) CO_2aq vs δ^{13}C (~~supra-micron~~ POM (>10μm) fraction) and (b) δ^{13}C-CO_2aq vs δ^{13}C in the East Siberian Sea (filled circles). The solid line shows the linear interpolation while the dashed line shows the 95% confidence intervals.

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