To the editors and reviewers

We greatly appreciate the constructive and helpful comments and criticisms by two anonymous reviewers. All comments were carefully considered and almost all suggestions will be incorporated in the revised version of the manuscript. We also thank the associate editor Naohiko Ohkouchi for handling the manuscript.
Moreover, we would like to add Thomas Laepple from the Alfred Wegener Institute, Helmholtz Center for Polar and Marine Research in Potsdam, Germany, to the list of co-authors. He provides the dual-carbon-isotope three end-member mxing mdoel suggested by reviewer 2 and contributes to the discussion of the results.
An overview of the changes made can be found in the attached pdf file (below). In those cases where we chose not to follow specific recommendations for alteration, we present further arguments supporting our approach.

Sincerely,
Maria Winterfeld

Biogeosciences Discussion, 11, 14413-14451, 2014

"Characterization of particulate organic matter in the Lena River Delta and adjacent nearshore zone, NE Siberia – Part 1: Radiocarbon inventories"

by Winterfeld et al.

Overview of revisions to the manuscript, and response to reviewer comments

As already mentioned in the reply to the review of the first manuscript (Winterfeld et al.) submitted as companion paper, we would like to ask the editors to consider a change of the order of the two submitted manuscripts and thus a change of the titles. As mentioned in the reply letter for the first manuscript, the paper on lignin phenols benefits from including carbon isotopic data from the second paper in the discussion and we refer to paper #2 several times throughout the manuscript. While we do not refer to lignin phenol paper that much in the discussion of the carbon isotopic data. Therefore it seems only consequent to treat paper on carbon isotopes as the background data providing "Part I" and the paper on lignin phenols as the second paper, i.e. "Part II".

Overall, the manuscript was restructered according to reviewer comments and also due to the additional dual-carbon-isotope three end-member mixing model as suggested by reviewer 2. In order to make the calculations of the soil fractions in the binary mixing models and new three end-member mixing model consistent and comparable (using Monte Carlo simulation) we chose end-member values and associated uncertainties, which are slightly different from the previous manuscript version. Yet, as shown in Fig. R2 at the end of this reply letter, the old and new (revised) calculated soil fractions for both binary mixing scenarios (based on POC:PN ratios and $\delta^{13}C$, respectively) are within the uncertainties of each model. An additional reason for differences in the soil fraction calculation of the POC:PN-based scenario is a mistake in the calculation of the soil end-member. It should have been 23.7 ± 11 (Table 4 in the manuscript), but was 20.2 due to missing rows in the respective excel sheet. Consequently, the calculated soil fractions and related soil $\Delta^{14}C$ estimates changed in Tabel 5 (previously Table 4). As noted above, the changes were within the model uncertainties (Fig. R2) and therefore did not change the outcome/interpretation of the data.

Overview of major changes:

- we added section 2.4 "End-member modeling" to the methods chapter
- previous sections 4.1.1 and 4.1.2 were combined and summarized them under 4.1 "Origin of organic matter in the Lena Delta
- we added section 4.3.3 "Comparison pf scenario 1 and 2
- we added section 4.3.4 Holocene soil versus Pleistocene Ice Complex deposits

- we added a table (now Table 4) with the end-member values used for each mixing model (binary and three end-member)
- we extended the supplementary information including several additional figures and three additional tables with individual values and their references used to calculate the soil, Holocene soil and Ice Complex end-members (POC:PN, $\delta^{13}C$, $\Delta^{14}C$)
- we added Fig. 4 showing the results of the dual carbon isotope three end-member mixing model
- Fig. S2 (POC vs. PN) was slightly changed, because the plot for 2010 did not include all samples for 2010 in the previous version

Additionally, the paragraph discussing the sorption of inorganic nitrogen to clay minerals and thereby lowering the POC:PN ratios was shifted from **page 14424 lines 8-12 to page 14423 line 23**. As noted below in comment 39 (reviewer 2, page 13 of this pdf file) the sentence paragraph "Using…" to "… in 2011" (page 14423 line 23 to page 14424 line 2 was deleted. Because we combined the sections 4.1.1 and 4.1.2, there is a rephrased paragraph on the $\delta^{13}C$ results following after the shifted paragraph.

Reviewer 1 general comments:

The authors consider two independent scenarios to estimate radiocarbon endmember of their interest. I could see wide range and slight difference in Δ14C estimates between POC:PN-based- and δ13C-based-scenarios. For more clarification, I wonder whether or not Keeling plot approach could be applied to the dataset. [POC] and radiocarbon data are available, and the authors have already assumed the other endmember (i.e., Δ14C of phytoplankton is 49‰). Therefore, y-intercept of the regression line obtained from a plot for Δ14C values (y) vs 1/[POC] (x), would indicate soil POM end-member. Further details on this approach may be found in e.g., "Mortazavi B, Chanton JP (2004) Use of Keeling plots to determine sources of dissolved organic carbon in nearshore and open ocean systems. Limnology and oceanography 49:102-108".

Reply: We did consider plotting 1/[POC] versus $\Delta^{14}C$. However, there is no linear relationship between 1/[POC] versus $\Delta^{14}C$ for our samples from 2009 and 2010. For 2011 the R^2 is ~0.6, however, based on only three samples. Please see Fig. R1 at the end of this reply letter for illustration.
Furthermore we decided to follow the suggestion from reviewer 2 to use a dual-carbon-isotope ($\delta^{13}C$ and $\Delta^{14}C$) three end-member model combined with a Monte Carlo Simulation to estimate the contributions from riverine phytoplankton, surface/modern soil, and Pleistocene soil/ice complex to the POM in our samples. This gives us the chance to distinguish not only between phytoplankton and terrestrial OM, but also

between the two terrestrial permafrost sources, namely the surface soil of Holocene age and the ice complex deposits of late Pleistocene age, which contribute to Lena POM.

The authors think that phytoplankton represents photosynthetic autotrophs in the Lena River. However, the study sites seem relatively shallow (water depth is 0.5m, Table 1) and I wonder there are any benthic primary producers (e.g., periphytic algae attached on reverbed substrate, or periphyton) contributing (suspended) POM to water column. If that is the case, the assumption used by authors (i.e., δ13C value of phytoplankton = –33‰ is questionable: in general, periphyton is more 13C-enriched than phytoplank- ton. For a study of similar setting (carbonate-weathering dominates the source of DIC) but different system (headwater stream), "Ishikawa NF, Uchida M, Shibata Y, Tayasu I (2012) Natural C-14 provides new data for stream food-web studies: a comparison with C-13 in multiple stream habitats. Marine and Freshwater Research 63:210-217" may provide some implications.

Reply: We have to apologize for the confusion resulting from the "water depth" column header in Table 1. It should have been "water depth sampled" and not just "water depth". In agreement with a comment from reviewer 2 we deleted **this particular column from Table 1** and mention the sampling depth for TSM/POM in the table caption as well as in section 2.2. Further, we **included the samples from 2011**, which were previously in Table S1 in the supplement into Table 1 of the manuscript.
We took our water samples of the Russian riverboat Puteyski 405. In the smaller and narrower channels particularly between the islands in the central delta we sampled from the middle of the channel. In the bigger delta channels, e.g. in the Bykovskaya channel, we did not sample directly in the middle of the channel as they can be >1km wide, but within the main river current far away from the riverbanks.
We appreciate the paper suggestion made by the reviewer. However, we do not think that there is any contribution from periphyton in our samples, because we sampled in water depth >0.5m.

The authors should carefully check terminology and δ- and Δ-notations throughout the text. For example, "Δ14C concentration" is not appropriate. Use "14C concentration" or "Δ14C value". Furthermore, "δ13C composition", "δ13C signature", "Δ14C composition" and "Δ14C signature" are often used in text, but some researchers do not accept these expressions. I recommend simply using "δ13C" or "Δ14C value".

Reply: It seems that there was something mixed up regarding the δ- and Δ-notations in the editing process of the submitted manuscript and it slipped our attention in the proofreading process. We changed it accordingly.
Further, we follow the recommendations made about the appropriate use of words like "composition" and "signature" and only use "^{14}C concentration", "Δ^{14}C value", and "δ^{13}C value" in the manuscript.

Reviewer 1 specific comments:

1. P. 14414, L. 12, 20, 22: *“δ13C”, not “Δ13C”*

Changed.

2. P. 14415, L. 4, 7: *“Guo and MacDonald 2006”, not “Guo et al. 2006”. Check other references once again.*

Changed and remaining references checked.

3. P. 14418, L. 26: *Pore size of Whatman GF/F should be 0.7µm*

That’s correct, we changed it to 0.7µm.

4. P. 14419, *2.3 Laboratory analyses: Provide analytical precision/uncertainty*

Reply: We provided analytical precision and uncertainty, respectively.

5. P. 14422, L. 20: *“the lowest Δ14C values”, not “the most depleted Δ14C values”*

Changed.

6. P. 14425, L. 16-18: *Do you have any evidence of this statement? At least provide one reference otherwise delete the sentence.*

Reply: We deleted the sentence.

7. P. 14426, L. 25-26: *“indirect evaluations have to be considered estimates” is unclear. Do the author want to say that non-phytoplankton materials are potentially included in POM?*

Reply: We agree that this sentence is confusing and therefore rephrased and restructured it. We deleted the whole paragraph from **page 14426 line 23 to page 14427 line 6**, because the considerations and the dicussion about estimating the phytoplankton OM contribution and the phytoplankton $\Delta^{14}C$ end-member occurs again in section 4.4 “Implications of estimated soil derived POM $\Delta^{14}C$” on **page 14435 line 3**. Here, we added more information on possible DIC sources in the Lena watershed and factors possibly influencing the DIC $\Delta^{14}C$ of Lena water from **page 14435 line 11** onwards.

Surface water POM in rivers is heterogenous and can be distinguished in phytoplankton and terrestrial OM. Because we do not have data on chlorophyll-*a* contents and/or microscopic counts of phytoplankton species of our samples, which could be used to quantify the phytoplankton fraction, we wanted to emphasize the fact that our phytoplankton fractions based on the C/N ratio and $\delta^{13}C$ values are rough estimates compared to the above methods.

8. P. 14426, L. 29: *"Δ14C ~ 49‰´' not "Δ14C ~ 49‰ and"*

Changed. The new $\Delta^{14}C$ end-member value for modern phytoplankton for the binary mixing models and the three end-member mixing model is **41.9 ± 4.2‰**. This value is the average atmospheric CO_2 $\Delta^{14}C$ value for May-August 2009-2011 determined at the Schauinsland observators, Germany (Levin et al., 2013).

9. P. 14427, L. 1: *"although this might not be true" Why do you think so?*

Reply: We explained this in the sentences following this statement (P. 14426, L . 1-6). However, it seems we have not been clear enough and therefore rephased the paragraph and added more information to clarify the statement that the phytoplankton might be older than modern depending on the DIC ^{14}C of the Lena. **This sentence** is part of the deleted and shifted paragraph from reviewer comment 7 above. The detailed explaning paragraphs on this issue can be found on from **page 14435 line 11** following.

Because there exists no DIC ^{14}C value for the Lena, we assume it equals the current atmospheric CO_2 ^{14}C value (~41.9 ± 4.2‰) and we use this value as an end-member for our mass balance calculations. However, the DIC ^{14}C could be depleted compared to the modern atmospheric CO_2 ^{14}C value due to contribution of fossil carbon ($\Delta^{14}C = -1000‰$ by definition) from carbonate weathering to the total DIC pool. Further, Tank et al. (2012) found that the DIC yields are negatively correlated with continuous permafrost extent in watersheds of six large Arctic rivers including the Lena. This would imply that the contribution from carbonate weathering in the Lena catchment might not be so large afterall as 77% of the catchment are characterized by continuous permafrost (Tank et al., 2012).

Again, without any measured DIC ^{14}C values we cannot be sure. Under the assumption of a modern phytoplankton ^{14}C concentration the estimated ages of the soil-derived fraction have to be considered maximum.

10. P. 14427, L. 3: *"soils, both of which provide", not "soils, both, providing"*

Changed. As noted in the replies to **comments 7 and 9** above, this sentence was deleted from this part of the manuscript and is now discussed in section 4.4 of the manuscript.

11. P. 14427, L. 6: *"in other words, maximum", not "i.e. maximum*

Changed. As noted in the replies to **comments 7 and 9** above, this sentence was deleted from this part of the manuscript and is now discussed in section 4.4 of the manuscript.

12. P. 14428, L. 25: *“The calculated” not “The so calculated”*

Changed.

13. P.14429, L. 14: *“<11600 yrs BP” not “11600 yrs BP the oldest”*

Changed. Note, the new ^{14}C ages based on the modified soil and phytoplakton end-member are 17,250 years BP and fossil (>50,000 years BP).

14. P. 14430, L. 2: *“Hubberten, 1999). This is also reflected” not “Hubberten, 1999) also reflected”*

Changed.

15. P. 14430, L. 5: *“data suggest” not “data suggests”*

Changed. We rephrased this sentence.

16. P. 14430, L. 13-15: *Don’t you think that atmospheric CO2 is also important source for DIC? Your assumption was that modern C of phytoplankton came from atmosphere.*

Reply: Yes, indeed we think that atmospheric CO_2 is an important source for DIC and we forgot to mention this here. We added “atmospheric CO_2” as possible source.

17. P. 14431, L. 14: *“samples were” not “samples are”*

Changed. We deleted this sentence, because with the Monte Carlo simulation these samples could be considered in the model.

18. P. 14431, L. 15: *“values were” not “values are”*

Changed. As noted in comment 17, we deleted this sentence, because with the Monte Carlo simulation these samples could be considered in the model.

19. P. 14432, L. 17-18: *“considerably 14C-depleted” not “considerably depleted”*

Changed.

20. P. 14451, Fig. 3: *Additional plot for Δ14C vs sampling date may help understand seasonal variation.*

Reply: We agree, and will provide an additional plot. However, our samples were taken within one or two weeks during each sampling campaign as well as in same season for 2009 and 2010. There might not be a clear seasonal trend visible from our data.
We also included Lena discharge data for the three sampling years from the newly published Arctic Great Rivers Observatory (A-GRO data set 2, www.arcticgreatrivers.org, accessed: 15 January 2015), because the Lena discharge also has a very distinct seasonal discharge pattern.

We added **Fig. S1** to the supplement.

Reviewer 2 general comments:

Interpretation of river OM geochemistry can in my opinion not be done without information on hydrology (i.e. discharge). I do not see anything on this topic in the manuscript. The differences in radiocarbon age, POC concentrations, POC:PN values could very well be related to timing and intensity of the freshet (how "flushed out" is the system in August?), the discharge later in summer/early fall (are there precipitation- event related peaks? is there still enough "force" for active bank erosion), etc. It is also important to know whether 2009, 2010 and 2011 are anomalous or normal years with respect to river discharge. - The authors argue that the results are useful for dual- carbon-isotope simulation studies in the region focusing on unraveling source contri- butions. I agree with that. But, I think the authors miss out on a good opportunity: to do this themselves with the three end-members for the Lena River. As most of the POC data are from the delta and the river, I think the authors should ignore the marine end-member, and run simulations (preferably with Monte Carlo) with the three "fluvial" end-members: (1) Ice complex deposits, (2) surface soils, and (3) fluvial plankton. A sturdy assessment of the associated uncertainties would be needed though (e.g. this is now not done at all for the plankton end-member of 49 per mille). In the manuscript they now only make calculations with two end-members (page 14431). An attempt to assess the relative contributions of these three end-members for the Lena River would, despite the associated uncertainties, be very valuable. I feel that the authors can do more with the data than they currently present (all focused on source-apportionment), and this is only one suggestion.

Reply: We agree that hydrology is important when interpreting riverine POM data. We had difficulties to get dischrage data for the Lena Delta and its channels for our sampling period (2009-2011). In the beginning of this year a new data set from the Arctic Great Rivers Observatory (A-GRO, www.arcticgreatrivers.org, accessed 15 January 2015) provides daily discharge values from the Kyusyur gauging station located about 200km upstream the delta entrance at Tit Ari Island. As already requested by reviewer 1 we will add a plot of POM ^{14}C concentration in relation to sampling date and discharge. However, when entering the delta the water gets distributed through the individual bigger and smaller delta channels and through the numeruos delta islands, which slows down the water flow and most likely results in different discharge values for different channels. The overall discharge pattern determined at Kyusyur, i.e. relatively higher and lower discharge values are reflected in the delta channels, but we cannot say anything about the actual discharge values during our sampling periods in the delta.

We appreciate the suggestion of running a dual-carbon-isotope simulation to estimate contribution from the three sources riverine phytoplankton, surface soil, and ice complex. Frankly, we did not consider this when writing the manuscript. We are currently working on this simulation with the help of Thomas Laepple from the Alfred Wegener Institute in Potsdam, Germany, and we will therefore add one section to the method chapter explaining the simulation and extent the results and discussion chapters explaining and interpreting the data.
As noted in comment 20 above, we added Fig. S1 to the supplement depicting the Lena River discharge determined at Kyusyur gauging station ca. 100km south of the delta and the POC content as well as δ^{13}C and Δ^{14}C values of our samples from 2009-2011.

We further added the method **section 2.4** on end-member modeling, the **section 4.3.4** dealing with the results of the dual-carbon isotope three end-member modeling, **Fig. 4** depicting individual fractions of phytoplankton OM, Holocene soil OM, and Pleistocene Ice Complex OM to each POM sample, and we added **Table S5** in the supplement displaying the fraction of the three end-member for each sample.

Reviewer 2 specific comments:

21. P. 14415, line 24: *There are in fact also a few 14C-POC values published from the Lena River: between -220 and -350 per mille in Vonk et al. Biogeosciences 7, 2010.*

Reply: We appreciate the information, because these two values slipped our attention as they are only mentioned in the text of the respective paper as unpublished data. Therefore we have no information on when and where these samples where taken in the Lena River and it is difficult to compare them with our data.

We **added the reference** of Vonk et al. (2010) to the sentence.

22. P.14418, line 17: *I find the information given on the soil profiles very minimal. Can you give a short description of the sites and the soils? Holocene/Ice Complex? Actively eroding sites or not? Also, you talk about "first delta". What does that mean?*

Reply: Indeed, the description on the soil profiles is lacking. We added information on the soil profiles in the text in section 2.2 (Sampling) as well as in the supplement with photos of the bluffs. All soil samples in the Lena Delta were of Holocene age.

Regarding the term “first delta”, we missed to explain the Lena Delta development and add this in the study site description (section 2.1). Briefly, according to Griegoriev (1993) and Schwamborn et al. (2002) the Lena Delta consists of three terraces. The first terrace including the active floodplains coves the eastern part of the delta and is referred to as the “active delta”. The second terrace is situated in the north western part of the delta consisting of sandy islands and the third terrace represents the late Pleistocene ice complex deposits predominantly found in the southern part of the delta.

We **added some sentences** describing the Lena Delta geomorphology and the first delta terrace in more detail on **page 14418 line 4** following.

23. P. 14418, line 21: *"... was removed with a spade for the total height of each bluff". You mean that you did this before you sampled the frozen soil that was behind the thawed material? Please elaborate a bit in the text.*

Reply: Yes, that is exactly what we did. We added some sentences to the sampling description in section 2.2, **page 14418 line 19** following.

24. P. 14418, line 21: *"peat"? Did you only sample peat?*

Reply: The term “peat” was used here as a generalization to describe the appearance of the whole soil sequence and it is not used correctly. As stated in the comment above we will be more detailed on the bluff sampling and refrain from using peat where it is not appropriate. We also samples layers with higher sand contributions and plant detritus, which are not peat.

25. P. 14418, line 26: *Glass fibre filters (GF/F) have a nominal pore size of 0.7 um, and not 0.45um.*

Reply: We changed it to 0.7µm.

26. P. 14419, line 22: *You write that you submitted the sampled unprocessed. I presume the inorganic carbon was removed at NOSAMS? It would be good to include that information.*

Reply: Yes, the inorganic carbon was removed as we specifically determined the ^{14}C concentration of organic matter. We added this information to the text.

27. Page 14421, line 17-18: *"and references therein" should be included in the brackets of the reference.*

Changed.

28. Section 4: *Discussion Can you think of a bit more descriptive titles of the different parts? It would be nice if the titles describe some of the main points instead of just using the measured parameter as the title?*

Reply: We will think more descriptive titles.

- we combined the previous sections 4.1.1 and 4.1.2 under section 4.1 "Origin of organic matter in the Lena Delta"
- section 4.2 was changed to "^{14}C and age heterogeneity of POM ($^{14}C_{POM}$) in the Lena Delta
- section 4.3 was changed to "Quantitative POM source determination and soil ^{14}C concentrations
- section 4.3.1 was changed to "Binary mixing model scenario 1" and section 4.3.2 was changed to "Binary mixing model scenario 2"
- section 4.3.3 "Comparison of scenario 1 and 2" was added
- section 4.3.4 "Holocene soil versus Pleistocene Ice Complex deposits" was added
- section 4.4 was not changed

29. Page 14423, line 9: *I suggest to write "(0-400m elevation)" or so to avoid confusion with depth of deposits.*

Changed.

30. Page 14425, line 16: *Both "permafrost" and "affected" say something about "water-sheds" so you should use a hyphen in between them: "permafrost-affected water- sheds". Same for "Pleistocene-aged OM" in this sentence.*

Reply: In agreement with the comment from reviewer 1 we deleted the whole sentence (comment 6 above).

31. Page 14425, line 18: *I suggest to write ".. might only have a minor effect".*

Reply: In agreement with the comment from reviewer 1 we deleted the whole sentence (comment 6 above).

32. Page 14426, line 29: *You give an algal-derived OM 14C signature of 49 per mille. Where is this based upon? You also write "and" in between the brackets, that should not be there I suppose. And, next page, "although this might not be true?" Can you clarify this part?*

Reply: Also in agreement with a comment from reviewer 1 we rephrased this paragraph and added more information on the ^{14}C value of 41.9 ± 4.2‰ (average atmospheric CO_2 Δ^{14}C May-August 2009-2011 from Schauinsland observatory, Germany; Levin et al. 2013) for phytoplankton and why we think that this might not be true. As noted in our

reply to comment 9 we deleted the paragraph from this section of the manuscript and dexplain and disuss the issue in **section 4.4** (**page 14435 line 11** following).

33. Page 14427, line 3: *";" should be removed.*

Changed.

34. Page 14428: Equation 1: *"POC:PNPOM" should be "POC:PNNEW" I think? Line 6: I presume you here mean "corrected value" instead of "measured value"? Also, I think it should be Table S2 instead of Table S1. Line 24: Doesn't it make more sense to write "or" instead of "and" here? Line 25: I suggest to remove "so". Where is Table 3 in the Supplementary Information?*

Reply: Yes, it should be POC:PN_{NEW}. We changed it and also followed the other suggestions made here. And it should be Table S4 in the supplement.

35. Page 14429: Line 5-6: *Remove "theses". Line 8: Do you mean "POC:PNNEW" or is "POC:PNcorr" something else? Line 19: Insert "of" in between "estimation" and "soil".*

Reply: *Changed* as suggested. Also, our terminology is inconsistent regarding newly calculated POC:PN ratios. It should be POC:PN_{NEW} throughout the manuscript and we changed this accordingly.

36. Page 14430, lines 16-19: *How certain are you about these fractionation factors?*

Reply: We took this fractionation factor from the literature and forgot citation in the submitted mannuscript. The reference of Mook & Tan (1992) will be added. Furthermore, we re-phrased parts of this paragraph to elaborate more on the $\delta^{13}C$ and ^{14}C values of the phytoplankton end-member, particularly in regard to the dual-carbon-isotope three end-member simulation that we are adding to the manuscript. In summary, Mook & Tan (1992) give a range from -20 to -25‰ as fraction factors between bicarbonate and primary production. The lower value under warm temperatures and higher fractionation factor under cold temperatures. We took the value of -25‰ for cold temperatures for our study area. However, because the Lena water is coming from the south, where it is much warmer in the summer season resulting in water temperatures of >10°C (July to Sept.; A-GRO data set 2), it seems more appropriate to use a mean fractionation factor of -22.5 ± 2.5‰ to account for some uncertainty. Further, Galimov et al. (2006) published values for bicarbonate and plankton $\delta^{13}C$ in the Ob' and Yenisey estuaries. Both rivers have a comparable watershed size to the Lena and also drain several climate zones. The southernmost samples taken in both estuaries, which were 100% riverine according to the phytoplankton species distribution had bicarbonate $\delta^{13}C$ values of for example -9.1 to -

14.8 ‰ for the Yenisey with corresponding plankton $\delta^{13}C$ of -31.9 to -36.2‰. Similar values were observed for the Ob' River by Galimov et al. (2006). Our assumption of the pyhtoplankton $\delta^{13}C$ end-member seems to within the possible range of large Arctic rivers.

For the Monte Carlo Simulation of the binary mixing model (scenario 2) we chose a phytoplankton $\delta^{13}C$ end-member of –30.5 ± 2.5‰ and added a short explanation and the reference of **Mook and Tan (1991)** on **page 14430 line 16** following.

37. Page 14431 Line 9: *Can you give uncertainties for the soil and plankton end-members? Lines 3-13: I am not convinced that the seasonal "aging" of POC is (only) due to active layer deepening (like it is for DOC). POC and DOC seem, age-wise, very decoupled and POC is also much strongly affected by erosion. Before making conclusions on why POC is sometimes older than other times, I think the data need to be correlated/associated with discharge. Related to this, it would be very interesting if you could plot for example a figure of 14C-POC age (y-axis) against julian day (x-axis) for all samples (for all years in the same graph), or, 14C-POC age (y-axis) against km from delta head (x-axis) to see patterns in potential aging (?) when travelling through the delta?*

Reply: See also **comment 36** above. We reconsidered to take a $\delta^{13}C$ fractionation factor for phytoplankton of -22.5±2.5‰, which results in an $\delta^{13}C$ end-member for phytoplankton of – 30.5±2.5‰ (-8‰ for DIC $\delta^{13}C$). Accordingly, we will change the calculation.
Furthermore, as also suggested by reviewer 1 we added a plot Julian day versus POC ^{14}C and discharge data for the sampling period from the new A-GRO dataset 2 (see also our comment to reviewer 1 on page 6 of this pdf file).

38. Page 14433, line 3: *it is not entirely clear what you mean by "here".*

Reply: It should refer the southern boreal hinterland of the Lena. We clarified this to avoid an further confusion.

39. Page 14434 Line 8: *here the end-member has a 13C value of -26.6 per mille, but on page 14431, line 9 it is -26.9 per mille. This is a bit confusing, could you be a bit more clear/insightful on how you get to this number(s)? Line 6: so do you use published Alaskan soil values to calculate the Lena River soil OM end-member?* ***Line 15:*** *Selective degradation is a very important issue. This point, together with the lowering of POC:PN ratios by the contribution of inorganic nitrogen, should in my opinion already be discussed earlier in the manuscript: preferably when first introducing scenario 1.* ***Line 18:*** *"fairly well constrained"? I do not completely agree, because (i) the readers need more information on where the plankton end-member value comes from, and (ii) how you calculate the soil-derived 13C end-member value is also not so transparent (it is based on your data and data from literature, right? could you for example include all the values in a table?)*

Reply: First of all, the δ^{13}C value should be -26.6 ± 1‰. The other value is a typing mistake. We agree that we not explained the very clear where this value comes from. We followed the suggestion and added a table to the supplement with all the values used for end-member calculation including the used literature values. And yes, we used Alaskan soil values as well as they were from similar environment as the tundra in the Lena catchment. But even if we leave these values out and only use Siberian δ^{13}C values we get a value of -26.6‰.

Line 15: We adopted the suggestion of discussing the selective degradation and associated lowering of POC:PN ratios earlier (**page 14423 line 23 following**). Note, **page 14423 line 23 to page 14424 line 2** were deleted ("Using…." to "…in 2011"), because the discussion on end-member mixing was completely shifted to section 4.3.

Line 18: The reviewer has a point. As mentioned above (also as reply to reviewer 1) we will provide a table with data used for end-member calculation and elaborate on that subject in more detail in the text.

We added the **Tables S1, S2, and S3** to the supplement showing the individual values used to calculate the soil OC:TN end-member, the soil δ^{13}C end-member, and the Holcene soil Δ^{14}C end-member. Further, we added this information when discussing the mixing models in section 4.3.

40. Page 14436, line 13: *here and at more places in the manuscript: please write 14C (or 13C) concentrations and not "D14C concentrations". Also, note that the delta symbol for 14C (not stable) is different than for 13C (stable); this is not correct in the abstract.*

Reply: An notet in our reply to the third general comment to reviewer 1 (page 4 of this pdf file) this happened during the typesetting process and slipped our attention in the proof reading process. .

41. Table 1: - *Could you write ◦N and ◦E with the coordinates? And also explain somewhere that "dec." means "decimal degrees". - Could you add "soils" and "SPM" or something like that to the first two lines and the rest, respectively? - is it an idea to just say that all SPM samples were collected at 0.5m depth instead of listing the same number many times? Same for bluff height (maybe just mention that somewhere else?)*

Reply: *Changed* everything as suggested.

42. Table 2: - *In 2009 the differences between mean and median values are quite high. So are there relatively many extreme values this year? Maybe something to elaborate on?*

Reply: We will add some plots with Julian Day versus POC content, POC:PN, etc. including the discharge data from A-GRO to the supplement. However, the delta is very dynamic and POC samples represent only spatially very limited snapshots of the surface water. It is difficult to relate the difference between the mean and the median values to any particular process/event. We do not recall any distinct event, such as heavy rain event during the sampling periods.

43. Table 3: *"Lena Delta Aug 2009" is all TSM right? Could you add that?*

Reply: "TSM" added.

44. Table 2-3-4: *Could you make the headings of the different parts of the tables more consistent? (e.g. now Table 3 "Lena Delta TSM late May 2011" and Table 4 "late May 2011").*

Reply: We made the headers consistent.

45. Figure 1: *The scale of panel (b) is in my opinion much too small. Can you "zoom in" even more? (i.e. only north of 71 degrN and only west of 134 degrE). And make the figure larger? It is very difficult to exactly see the delta channel patterns and also the sampling points.*

Reply: The quality of the underlying image does not allow to zoom more in. Furthermore, the GPS coordinates in these high latitudes are not accurate enough to provide a realistic location within the individual channels (despite the decimal places in the coordinates. However, we **added a description** of our general sampling locations in smaller delta channels (sampled in the middle) and the main delta channels (not in the middle, but far off the shore, in water depth >10m) to **section 2.2 "Sampling"**.

46. Figure 2: *- what does "a" mean as the superscript of TCM ? - I suggest to remove "surface water total suspended matter" above every row of graphs and instead include it in the caption. - here too, could you "zoom in" even more? The actual area of the figures where the colored data points are can be much larger. - it might be an idea to adjust the color scale when there is one data point that is much higher or lower than he rest. For example, for TSM in late May 2011: the extreme value of this sample makes interpreting the color scale on panel A, B and D rather difficult.*

Reply: The "a" was meant as a citation for Winterfeld et al. (2014), submitted as companion paper, but it is redundant with the figure caption where this is explained as well. We deleted "a".

47. Figure 3: *- what do the "a" and "b" mean as the superscripts of "ratios" (panel A, purple and green diamonds), and at the proposed end-member (superscript "c")? - I suggest to leave out the ice complex end-member proposed by Karlsson et al. 2011 as the Vonk et al. (2012) ice complex end-member is based on about 900 datapoints and much more solid than the one from Karlsson et al. - Why (this also relates to the manuscript text) do you choose your Lena River soil OM end-member to be close to the 13C- derived estimates (in the figure it even looks more enriched than these estimates?!) instead of the POC:PN-derived estimates? You claim that the 13C-derived estimates are more robust, I* *believe, but I am not sure I agree with this, as you essentially base your 13C-estimate on 13C values you choose yourself to be the end-member.*

Reply: 1) The “a”, “b”, and “c” are redundant and were deleted from the figure.

2) We show the Karlsson et al. (2011) ice complex end-member in the figure, because we are also showing the surface soil/riverine and marine end-member to illustrate the different end-members used in the past. We agree that the Vonk et al. (2012) ice complex end-member is based on a very solid data set (n=300) and we will use this end-member for our dual-carbon-isotope simulation.

3) The comment regarding the δ^{13}C of the soil end-member relates also to a comment from above. Generally, we provided three tables in the supplement (**Tables S1, S2, S3**) giving the values that comprise our chosen end-members including the literature values. Further, the δ^{13}C end-member used to estimate the contribution of phytoplankton- and soil-derived OM to our POM samples should be the same as the proposed Lena River soil end-member, but there was a mistake in the Excel-sheet Fig. 3 is based on. We corrected this in **Fig. 3** and only show the end-members used by Vonk et al. (2012).

48. Supplementary information: Table S1: *in caption you have ".." in the end. And, you here write "shown in Figure 2D", but there is no Figure 2D.*

Reply: Table S1 was included into Table 1 from the manuscript to avoid confusion. The new Table S1 showes the soil OC:TN end-member values.

49. Table S2: *you only report TSM for the June/July 2011 samples, but since you both have POC (mg/L) and POC in %weight, you can also back-calculate TSM for all the other samples. I do not understand why you do not do this.*

Reply: We apologize for creating such confusion here. We did not make it clear enough how our two companion manuscripts submitted to Biogeosciences Discussion are interrelated. Of course, we have the TSM data for our samples. They are shown for example in Fig. 2, panels A-D, but they are taken from the first manuscript (Winterfeld et al., 2015: Characterization of particulate organic matter … Part 2: Lignin-derived phenol composition). We **added the TSM** values from the companion manuscript to **Tabel S4** (previously Table S2) to give a comprehensive overview of our data.

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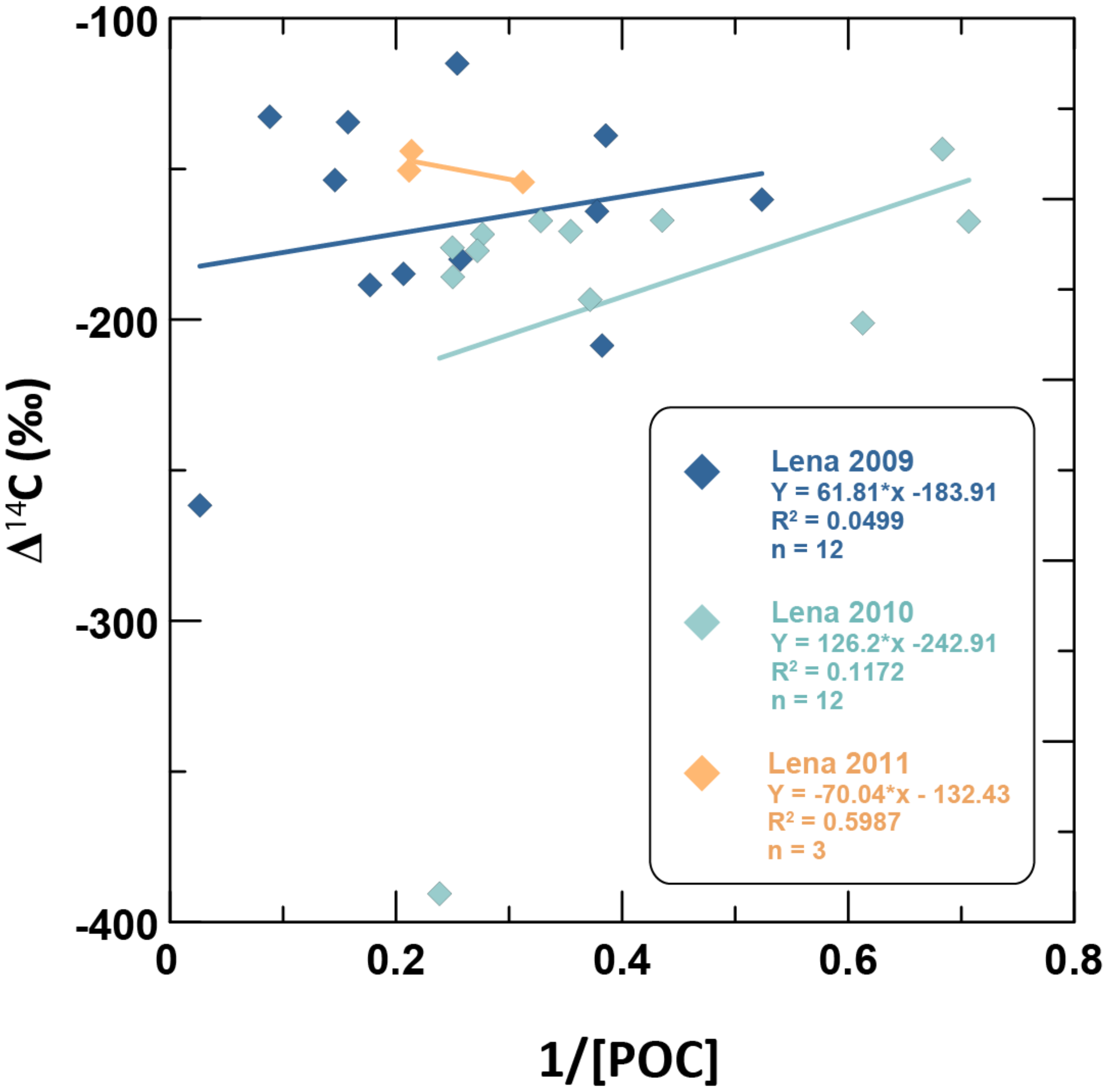

Fig. R1. Relationship between the POM bulk $\Delta^{14}C$ and 1/POC content for the samples from 2009, 2010, and 2011. There is no correlation for the years 2009 and 2010 and the R^2 of 0.6 for 2011 is based on only three samples.

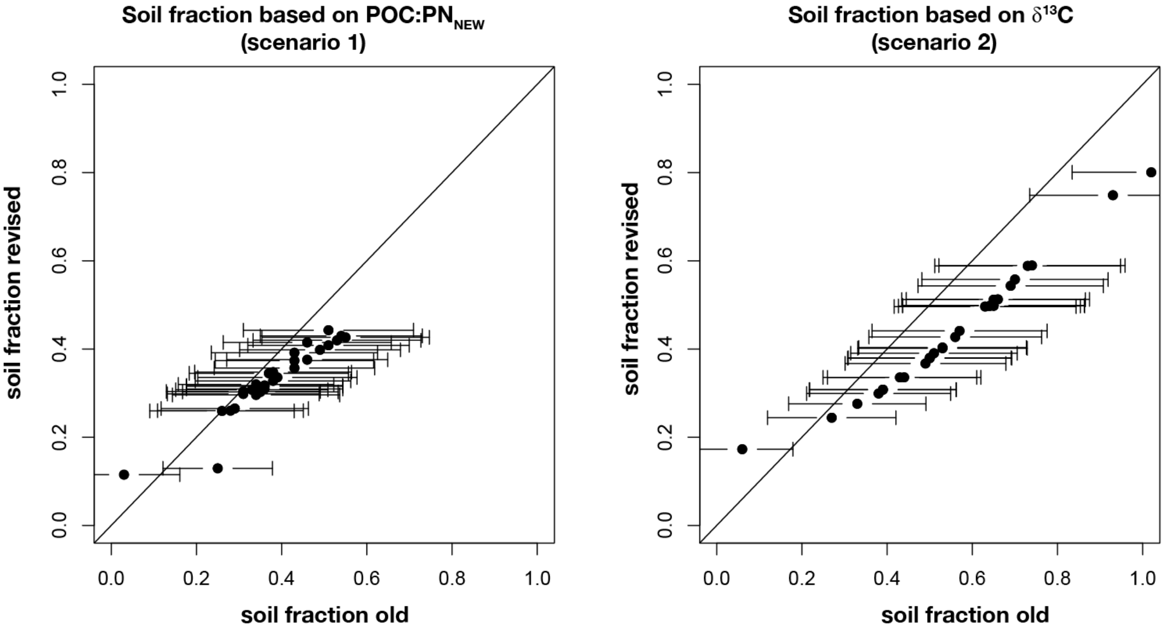

Fig. R2. The old calculated soil fractions versus the newly calculated soil fractions using modified end-member values and Monte Carlo Simulation for both binary mixing models.

Characterization of particulate organic matter in the Lena River Delta and adjacent nearshore zone, NE Siberia. Part I~~I~~: Radiocarbon inventories

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Abstract

Particulate organic matter (POM) derived from permafrost soils and transported by the Lena River represents a quantitatively important terrestrial carbon pool exported to Laptev Sea sediments (next to POM derived from coastal erosion). Its fate in a future warming Arctic, i.e. its remobilization and remineralization after permafrost thawing as well as its transport pathways to and sequestration in marine sediments is currently under debate. We present one of the first radiocarbon (^{14}C) data sets for~~of~~ surface water POM within the Lena Delta sampled in summers 2009-2010 and spring 2011 (n=30 samples). The bulk Δ^{14}C values~~concentrations~~ varied from –55 to –391‰ translating into ^{14}C ages of 395 to 3920 years BP. We further estimated the fraction of phytoplankton-derived POM to our samples based on 1) particulate organic carbon to particulate nitrogen ratios (POC:PN) and 2) on the stable carbon isotope (δ^{13}C) composition of our samples. Assuming that this phytoplankton POM has a modern ^{14}C concentration~~signature~~ we inferred the ~~Δ~~^{14}C concentrations of the soil-derived POM fractions. The results ranged from –322~~258~~ to –~~768~~ 884 ‰ (i.e. ~~2340~~ 3.060 to ~~11700~~ 17,250 ^{14}C years BP) for the POC:PN-based scenario and from –261~~191~~ to –~~704~~ 944 ‰ (i.e. ~~1640~~ 2,370 to ~~9720~~ 23,100 ^{14}C years BP). Despite the limitations of our approach, the estimated Δ^{14}C ~~concentrations~~ values of the soil-derived POM fractions seem to

reflect the heterogeneous ^{14}C concentrations~~signal~~ of the Lena River catchment soils covering a range from Holocene to Pleistocene ages better than the bulk POM Δ^{14}C values. We further used a dual-carbon isotope three end-member mixing model to distinguish between POM contributions from Holocene soils and Pleistocene Ice Complex deposits to our soil-derived POM fraction. Ice Complex contributions are comparatively low (mean of 0.14) compared to Holocene soils (mean of 0.32) and riverine phytoplankton (mean of 0.55), which could be explained with the restricted spatial distribution of Ice Complex deposits within the Lena catchment. Based on our newly calculated soil-derived POM Δ^{14}C values, ~~We~~ we~~therefore~~ propose an ~~typical~~ isotopic range~~signature~~ ~~of~~ for the riverine soil-derived POM end-member with Δ^{14}C of –495 ± 153‰ deduced from our δ^{13}C-based binary mixing model and ~~a~~ δ^{13}C of –26.6 ± 1.~~1~~ ‰ deduced from our data of Lena Delta soils and literature~~published~~ values.~~s, and a Δ^{14}C concentration of –362 ± 123‰ deduced from our δ^{13}C-based estimates.~~ These estimates~~data~~ can help to improve the dual-carbon-isotope simulations used to quantify contributions from riverine soil POM, Pleistocene ice complex POM from coastal erosion, and marine POM in Siberian shelf sediments.

1 Introduction

Huge amounts of soil organic carbon are currently stored frozen in permafrost soils of the high northern latitudes (e.g. Tarnocai et al., 2009; Zimov et al., 2009) and excluded from biogeochemical cycling. Due to recent observed and projected amplified warming of the Arctic (ACIA, 2005; Serreze et al., 2000) carbon cycling and the fate of organic carbon released from permafrost soils have received growing attention (e.g. Guo et al., 2007; McGuire et al., 2009; Schuur et al., 2008; 2009; Zimov et al., 2006).

Increasing permafrost temperatures, increasing thaw depth in summer (active layer depth), increasing river runoff (Boike et al., 2013; McClelland et al., 2012; Peterson et al., 2002), and increasing length of the open water season (Markus et al., 2009) affecting coastal erosion of permafrost deposits will likely lead to enhanced mobilization and export of old, previously frozen organic matter (OM) to the Arctic shelf seas. The understanding of the different terrestrial OM sources (e.g. fresh vegetation, surface soils, Pleistocene ice complex), their age and quality has significantly improved over the last decade (e.g. Guo et al., 2007; Vonk et al., 2010). The use of carbon isotopes (δ^{13}C, Δ^{14}C) of dissolved and particulate organic matter as well as individual biomarkers has helped characterizing and distinguishing these different carbon pools, e.g. ~~the~~ old, yet little degraded Pleistocene ice complex-derived OM and comparatively younger and more degraded fluvial OM reaching the Siberian shelf seas (Feng et al., 2013; Guo et al., 2004; Gustafsson et al., 2011; Karlsson et al., 2011; Vonk et al., 2012; 2010).

However, particularly in Siberia ^{14}C data on riverine suspended particulate organic matter (POM) are~~is~~ only ~~very~~ sparsely available. A recently published dataset from the Arctic Great Rivers Observatory (A-GRO, www.arcticgreatrivers.org, published 10 January 2015) provides POM ^{14}C data for the Lena River from Zhigansk, approximately 900km upstream of the delta. To our knowledge the~~re~~ ~~are~~ only~~no~~ additional POM ^{14}C concentrations ~~published which were~~ sampled directly in Siberian rivers ~~besides a few samples~~are from the Lena River (unpublished in Vonk et al., 2010) and the Kolyma River (supplementary data of Vonk et al., 2012). Available POM ^{14}C concentrations for Siberia are from offshore the deltas of the Lena (Karlsson et al., 2011) and Kolyma Rivers (Vonk et al., 2010) and inferred from a sediment core taken in a floodplain lake of the Ob' River (Dickens et al., 2011). Due to settling of POM and marine primary production fueled by the riverine nutrients, the ^{14}C concentrations of samples taken off the river mouths are already altered from the "original" river signal. In contrast to Siberia, there are detailed studies on riverine POM ^{14}C concentrations in the Yukon and Mackenzie Rivers in the North American Arctic (Goñi et al., 2005; Guo et al., 2007; Guo and Macdonald, 2006). Here, the POM was found to be significantly older than the dissolved organic matter (DOM) of these rivers and interpreted to be derived from riverbank erosion and thawing permafrost soils compared to a modern vegetation source of DOM (Guo et al., 2006; 2007). Against the backdrop of a warming Arctic (IPCC 2013, ACIA, 2005) and the projected release of old OM in the future presumably as POM rather than DOM (Guo et al., 2007), it is important to assess the age heterogeneity carried by riverine POM and sequestered in the nearshore zone today. This will be an important benchmark to distinguish catchment related changes caused by increasing temperatures from the natural variability of the river system.

In contrast to sedimentary OM, POM provides limited spatial and temporal snapshots of its OM properties. However, it has been shown that riverine POM in arctic rivers carries an integrated signal from their permafrost watersheds ~~and~~ representing~~s~~ the watershed environmental characteristics (e.g. Goñi et al., 2000; Lobbes et al., 2000; Vonk and Gustafsson, 2009; Winterfeld et al., 2015). ~~See also Winterfeld et al. 2014 submitted as companion paper dealing with lignin phenol compositions of POM.~~ Despite the fact that Lena River POM flux is an order of magnitude smaller than its DOM flux it is more likely to transports the climate change signal from permafrost soils of the river catchment (Guo et al., 2007). Also, it has been proposed that the POM pool could be as important as the DOM pool for arctic carbon cycling, because of its possibly high degradation rates in the water column compared to DOM (Sanchéz-García et al., 2011; van Dongen et al., 2008).

Here we present the first~~second~~ part of a study on particulate OM in the Lena Delta, Siberia (Winterfeld et al. 2015~~4~~, ~~submitted as~~ companion paper). Our POM samples taken in three consecutive years (2009-2011) in the spring and summer seasons add up to the existing data on elemental composition,~~and~~ stable carbon isotopes ($\delta^{13}C$), and radiocarbon (^{14}C) values~~composition~~ as well as provide a first POM ^{14}C data set covering three consecutive years for the Lena Delta. Because riverine and marine POM concentrations are usually too low for source specific biomarker ^{14}C analysis, the available ^{14}C data is from bulk OM. This could result in a considerable age bias depending on the contribution of phytoplankton OM with a rather enriched (modern) ^{14}C concentration~~signature~~ to the individual samples. We used different approaches to estimate the fraction of soil and plankton-derived OM in our POM samples and estimated~~corrected~~ the ^{14}C concentration for the soil-derived fraction of POM transported by the Lena River in summer and spring. Further, we used a dual-carbon isotope three end-member mixing model according to Vonk et al. (2010, 2012) to distinguish not only between plankton- and soil-derived OM, but also between OM from Holocene soils and Pleistocene Ice Complex deposits. The ~~corrected~~ estimated ^{14}C concentration~~composition~~ of soil-derived POM will further help to define the typical isotopic signature of river POM more accurately for modeling riverine OM contributions to Laptev Sea shelf sediments.

2 Material and Methods

2.1 Study Area

A detailed description of the Lena River watershed and Lena Delta can for example be found in the companion~~first~~ paper of this study dealing with the lignin composition of OM in the Lena Delta (Winterfeld et al., 2015~~4, submitted as companion paper~~).

In short, the Lena River is one of the largest Russian Arctic rivers draining a watershed of ~2.46 × 10^6 km^2 in central Siberia into the Laptev Sea. Continuous permafrost makes up about 72-80 % of the drainage area (Amon et al., 2012; Zhang et al., 2005) storing huge amounts of old OM. The permafrost acts as a water impermeable layer and thus affects the regional hydrology and hydrochemistry. The Lena River water discharge and related dissolved and particulate load discharge are highest during spring ice-breakup and snow melt in late May to June while summer and winter discharges are lower (Rachold et al., 2004). The mean annual water discharge is ~588 km^3 for the years 1999 to 2008 (Holmes et al., 2012). Corresponding annual sediment, dissolved organic carbon (DOC), and particulate organic carbon (POC) fluxes are 20.7 Tg/yr (Holmes et al., 2002), 5.7 Tg/yr (Holmes et al., 2012), and 1.2 Tg/yr respectively (Rachold and Hubberten, 1999). A second major source for terrigenous OM delivered to the Laptev Sea is the thermal erosion of ice- and OM-rich Pleistocene ice complex deposits along the coast (see Gustafsson et al., 2011; Mueller-Lupp et al., 2000; Rachold and Hubberten, 1999; Rachold et al., 2004). Recently, it has been shown that the annual supply of total organic carbon from ice complex deposits along the Laptev Sea coast by erosion is ~0.66 Tg/yr (Günther et al., 2013).

The Lena River Delta is the largest A~~a~~rctic delta (~32,000 km^2). It is characterized by a polygonal tundra landscape with active floodplains and it can be divided into three geomorphological terraces (Grigoriev, 1993; Schwamborn et al., 2002). The first terrace includes active floodplains that were formed during the Holocene and makes up about 55% of the total delta area (Morgenstern et al., 2008) covering the central and eastern part. Within the first delta terrace remains of a Pleistocene accumulation plain, also called Ice Complex or Yedoma deposits, form the third terrace covering about 6% of the total delta area (Morgenstern et al., 2008). Sandy islands forming the second terrace cover the rest of the delta in the west. Water and sediment discharge are not equally distributed through the several delta channels. Approximately 80-90 % of the total water and up to 85% of the sediment discharge are delivered through the three main eastern channels to the Buor Khaya Bay east of the delta (Fig. 1B), i.e. through the Sardakhsko-Trofimovskaya channel system (60-75% water, 70% sediment) and the Bykovskaya channel (20-25 % water, 15 %sediment). Only a minor portion is discharged to the north and west through the Tumatskaya and Olenyokskaya channels (5-10% water, 10% sediment) (Charkin et al., 2011 and references therein; Ivanov and Piskun, 1999).

2.2 Sampling

The sampling sites presented in this study are located in the eastern part of the Lena Delta and adjacent Buor Khaya Bay (Table 1, Fig. 1B) in and along the channels of highest discharge. Permafrost soil samples from the first delta and surface water total suspended matter (TSM) were collected during three expeditions in August 2009, July/August 2010, and in late May and late June/early July 2011. All soil samples were taken from riverbank bluffs of the first delta terrace (Holocene formation), which is elevated (5-16m) over the active floodplains. The soil profiles vary strongly in sediment composition and OM content. Within the profiles sandy layers derived from extreme flooding events (Schwamborn et al., 2002) and aeolian input (Kutzbach et al., 2004; Sanders, 2011) alternate with buried surface soil layers and peat layers rich in fibrous plant and root detritus in different stages of decomposition. The peat layers are either of autochthonous or of allochthonous origin. Allochthonous material is eroded from river banks further upstream and re-deposited in the delta. In order to obtain samples that reflect the original state of the frozen permafrost soils, we removed the thawed soil material from each riverbank bluff ~~was removed with a spade~~ for its~~the~~ total height ~~of each bluff~~with a spade. Frozen pieces of soil~~peat~~ were then excavated at different depths using hatchet and hammer.

Suspended particulate matter of Lena River surface water was sampled at different stations in the main river channels of the delta on the Russian vessel Puteyski 405 (Fig. 1B, Table 1) from the upper 0.5m of the water column. In the smaller delta channels the samples were taken approximately in the middle of the channel. Because the main delta channels can be >1km in width, we sampled a couple of hundred meters off the shore with water depth of >10m, but not in the middle of the channel. Three samples (19, 35, 36; Fig. 1B) were taken outside the delta in shallow water depth. We discuss these samples as “delta samples”, because the surface water at these sampling locations is outflowing Lena River water as shown by temperature and salinity data by Semiletov et al. (2011) and Kraberg et al. (2013). Between 1 and 30 L of water were filtered on pre-combusted (4.5h at 450°C) and pre-weighed glass fiber filters (GF/F Whatman, 0.7~~45~~µm membrane) for particulate organic carbon (POC) and nitrogen (PN) analysis as well as carbon isotope analysis. Additionally, one water sample of 20 L from the spring freshet in 2011 was stored cooled in opaque canisters for several days to allow for the suspended matter to settle. Before decanting the supernatant water it was filtered on pre-combusted and pre-weighed GF/F filters to check for the SPM remaining in suspension. For this sample~~e sample presented here~~ (sample ID 37) the SPM of the supernatant water represented 0.1 % of the settled material on a dry weight basis and therefore the loss of material in suspension can be neglected.

The soil samples were stored in pre-combusted glass jars (4.5h at 450°C) and GF/F filters were either stored in pre-combusted petri dishes (Ø 47mm) or wrapped in pre-combusted aluminum foil. All samples were kept frozen at –20°C during storage and transport until analysis.

2.3 Laboratory analyses

Soil and sediment samples were freeze-dried, homogenized, and subsampled for elemental analysis. All filters were oven-dried at 40°C for 24h.

2.3.1 Elemental analyses

Weight percent organic carbon (OC) and total nitrogen (TN) content of soil samples were determined by high temperature combustion after removal of carbonates as described by Goñi et al. (2003). TSM samples were analyzed for OC and TN at the Alfred Wegener Institute in Bremerhaven, Germany, using the same protocol. Every 10 samples control standards were analyzed to constrain the analytical uncertainty of 0.1%.

2.3.2 Carbon isotope analysis ($\Delta^{14}C$, $\delta^{13}C$)

Samples were radiocarbon-dated at the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS) facility at Woods Hole Oceanographic Institution, USA. Bulk sediment samples and filters with TSM were submitted unprocessed and inorganic carbon was removed during sample preparation at NOSAMS. The r~~R~~adiocarbon analyse~~i~~s at NOSAMS were carried out using standard methods (McNichol et al., 1994). Results are reported as $\Delta^{14}C$, conventional radiocarbon ages (years BP), and fraction modern carbon (*fMC*) including the correction for isotope fractionation (Stuiver and Polach, 1977).

The stable carbon isotope composition ($\delta^{13}C$) was measured on splits of the CO_2 gas of the samples generated prior to graphite reduction at NOSAMS using a VG Optima IRMS. Results are reported in per mil (‰) relative to VPDB.

2.4 End-member modeling

The relative source contributions (phytoplankton and soil) to the POM samples are estimated by solving linear two end-member (using POC:PN and $\delta^{13}C$ as parameters) and dual-carbon isotope~~s~~ ($\delta^{13}C$, $\Delta^{14}C$) three end-member mixing models with end-members determined from own data and the literature. The end-member values used in the different modeling approaches are given in Table 4. Source data for the calculation of the POC:PN soil end-member as well as the $\delta^{13}C$ and $\Delta^{14}C$ end-members for Holocene soil are given in Tables S1-3 in the supplement.

To account for the uncertainty of the measured values, we assume independent, normally distributed uncertainties for the observed values with a standard deviation characterizing the measurement uncertainty. For the end-members we assume a normal distribution using the standard deviations from the observed samples on which the end-members are based on.

Uncertainty in the resulting contributions of phytoplankton and soil are estimated in a Monte Carlo approach similar to Vonk et al. (2010, 2012) and Karlsson et al. (2011). The source fractions are estimated 100,000 times, each time drawing the end-members as well as the observed values from their distributions. Solutions with negative fractions are omitted and the mean and the standard deviation are reported.

3 Results

3.1 Elemental composition

The organic carbon (OC) and total nitrogen (TN) concentrations of the first terrace soil samples can be found in T~~t~~ables 3 and S3 in the companion paper Winterfeld et al. (2015~~4~~)~~, submitted as companion paper~~.

The surface water POC concentrations within the delta showed a high spatial and interannual variability similar to the TSM concentrations of the respective samples. The mean POC concentration for August 2009 was 1.21 mg/L (n=21, range 0.35-7.24 mg/L) and corresponding POC content was 7.2 wt% (range 1.9-37.7 wt%, Table 2 and Table S1). The POC contents for July/August 2010 and June/July 2011 were lower than in 2009 with mean concentrations of 0.57 mg/L (n= 13, 0.15-1.30 mg/L) and 3.1 wt% (1.4-4.7 wt%) as well as 0.74 mg/L (n=9, 0.29-1.51 mg/L) and 4.3 wt% (3.2-5.0 wt%), respectively. The single sample from late May 2011 (sample ID 37) showed highest POC concentration per liter (8.2 mg/L) of all presented samples with a related POC content of 1.7 wt%. Our POC data are well within the range of values reported for the Lena Delta before (Cauwet and Sidorov, 1996; Rachold and Hubberten, 1999; Semiletov et al., 2011). The two samples from the Buor Khaya Bay surface waters showed the lowest POC concentrations per liter, i.e. 0.37 mg/L (sample 35) and 0.15 mg/L (sample 36) with corresponding 4.4 and 1.5 wt% POC, respectively (Table 2 and S4 supplement~~1~~). The considerable drop of POC (and TSM) concentrations offshore the delta is due to flocculation and settling of TSM in the zone of fresh and salt water mixing (e.g. Lisitsyn, 1995) and was also observed during other years of sampling (e.g. Cauwet and Sidorov, 1996; Semiletov et al., 2011).

The atomic particulate organic carbon (POC) to particulate nitrogen (PN) ratios (POC:PN) of samples taken in summer 2009 were slightly higher than for summer 2010 samples with mean values of 9.6 (n=20, 6.8-19.3) and of 8.0 (n=13, 5.0-10.3, Table 2 and S4~~1~~), respectively. The POC:PN ratios of samples taken in June/July 2011 were rather similar to the July/August samples with a mean of 7.8 (n=9, 5.9-9.7). The sample from late May 2011 had a POC:PN ratio of 7.5.

3.2 Carbon isotope inventories

The stable carbon (δ^{13}C) and radiocarbon (^{14}C) results are shown in Table 3. The radiocarbon data presented here will predominantly be discussed in terms of Δ^{14}C in per mil (‰). Additionally, the fraction modern carbon (*fMC*) and ^{14}C ages in years before present (yrs BP) are given in Table 3.

3.2.1 Stable carbon isotope composition (δ^{13}C)

The two soil profiles from the first delta terrace showed δ^{13}C values between –27.0 ‰ and –25.1 ‰ with a mean of –26.1 ‰, (n = 7, Table 3), which is within the range observed for Holocene soils in the delta (e.g. Schirrmeister et al., 2011 and references therein)~~ and references therein~~. Similar to the POC contents, t~~T~~he Lena Delta surface water POM δ^{13}C values varied strongly spatially and annually (Fig. S1 supplement)~~ as described for the organic carbon contents above~~. POM from August 2009 showed more depleted δ^{13}C values compared to the other years, ranging from –34.2 ‰ to –28.8 ‰ (mean value = –30.4 ‰, n = 13). July/August 2010 and June/July 2011 POM isotopic compositions ranged from –30.4 ‰ to –28.3 ‰ (mean value = –29.3 ‰, n = 13) and from –29.3 ‰ to –28.3 ‰ (mean value = –28.7 ‰, n = 3), respectively. The isotopically most enriched POM δ^{13}C value of –26.5 ‰ was determined for the sample from late May 2011 (Table 3). The δ^{13}C of Buor Khaya Bay surface water POM in August 2010 could only be determined for one of the samples, i.e. sample 36 with –30.4 ‰.

3.2.2 Radiocarbon (^{14}C) concentration~~composition~~

The Δ^{14}C values~~concentrations~~ of the two soil profiles decreased with depth from –197 to –377 ‰ for the riverbank profile L09-12 and from –204 to –466 ‰ for profile L09-28 (Table 3, Fig. 3A). The corresponding ^{14}C ages for both profiles together ranged from 1,710 to 4,900 yrs BP. Within profile L09-12 on Samoylov Island an age reversal was observed for the two oldest samples (Table 3), which most likely is due to allochthonous material that was transported to the delta from an upstream location. The same age reversal on Samoylov Island was also observed by Kuptsov and Lisitsin (1996). Overall, our Δ^{14}C values~~concentrations~~

reflect the late Holocene formation of these soils and fit~~s~~ within the range of ages determined for the Lena Delta first terrace (Bolshiyanov et al., 2015~~4~~; Kuptsov and Lisitsin, 1996; Schwamborn et al., 2002).

As with other TSM parameters, the POM Δ^{14}C values~~concentrations~~ showed strong spatial and interannual variability (Fig. 2, panels Q-T and Fig. S1). Lena Delta POM ^{14}C concentrations varied from –262 ‰ to –55 ‰ in August 2009 (mean of –160 ‰, n=13), from –391 ‰ to –143 ‰ in July/August 2010 (mean of –194 ‰, n=13), and from –154 ‰ to –144 ‰ in June/July 2011 (mean of –150 ‰, n=3). The sample from late May 2011 showed a ~~Δ~~^{14}C concentration of –306 ‰ and the Buor Khaya Bay surface samples of –176 ‰ (sample 35) and –143 ‰ (sample 36, Table 3, Fig. 2R-S). Overall these ^{14}C concentrations~~compositions~~ covered a range of ^{14}C ages from 395 to 3920 yrs BP. The samples with the lowest~~most depleted~~ Δ^{14}C values of –262 ‰ (sample 1) and –391 ‰ (sample 31) were taken close to the Pleistocene I~~i~~ce C~~c~~omplex deposits~~ of Kurungnakh Island (Fig. 1B)~~, which likely contributed to the local POM in the Lena River surface water.

4 Discussion

~~4.1 ~~Origin of organic matter in the Lena Delta

~~4.2~~4.1 ~~Particulate carbon to nitrogen ratios (POC:PN)~~

Riverine particulate organic matter consists of a heterogeneous mixture derived from two major sources, i.e. terrestrial (e.g. fresh vegetation and litter, surface and deep soils horizons) and aquatic (phytoplankton/bacterial primary production). The terrestrial OM in the Lena River catchment can further be differentiated into two pools of different age: the late Pleistocene organic-rich I~~i~~ce C~~c~~omplex or Yedoma deposits, particularly in the lowlands (0-400~~-~~m elevation, ~~(~~e.g. Grosse et al., 2013) and the Holocene permafrost soils. POC:PN ratios as well as δ^{13}C and Δ^{14}C values~~signatures~~ of bulk OM can be used to estimate terrestrial and aquatic contributions (e.g. Hedges and Oades, 1997). However, due to overlaps in soil/plant and algal/bacterial signatures it might be difficult to unambiguously differentiate between terrestrial and aquatic sources.

Our POC:PN ratios from the summers 2009 and 2010 as well as spring 2011 vary largely throughout the delta (Table 2 and S4~~, Fig. S2~~) similar to the TSM and lignin phenol concentrations sampled during the same field trips (Winterfeld et al. 2015~~4~~, ~~submitted as~~

companion paper). The ratios range from 3.7 to 19.3 for all samples and seasons with mean atomic POC:PN ratios of 9.6 (2009), 7.6 (2010), and 7.8 (2011). Low POC:PN ratios, i.e. ~6 generally indicate high contributions from phytoplankton and/or bacterial primary production while ratios >20 are indicative of soil and plant contributions (e.g. Hedges et al., 1997; Meyers and Lallier-V~~v~~ergés, 1999). Based on these end-members, our data from the summer and spring seasons would suggest a considerable fraction of primary production OM to be present in our samples. ~~Using a simple two end-member mixing model with POC:PN = 6 for primary production OM and 20.2 for soil-derived OM (mean of OC:TN ratios from the first delta terrace of this study and delta soils as well as tundra and taiga soils south of the delta from Zubrzycki (2013), our soil fractions vary from 0.06 to 0.94 (mean 0.26) in 2009, from 0.10 to 0.30 (mean 0.16) in 2010, and from 0.08 to 0.26 (mean 0.14) in 2011.~~

However, due to possible sorption of inorganic nitrogen to clay minerals (Schubert and Calvert, 2001) our calculated POC:PN ratios might be too low, underestimating the soil-derived OM fraction. This will be discussed in more detail in section 4.3.1. Another possibility for lower, but mainly soil derived POC:PN ratios could be the selective degradation of labile OM compared to total nitrogen (Kuhry and Vitt, 1996).

Analogous to the POC:PN ratios, the $\delta^{13}C_{POM}$ values point to a considerable contribution of primary production OM to our samples from 2009-2011. The $\delta^{13}C_{POM}$ values of our samples taken in 2009-2011 vary over a broad range (–34.2 to –26.5 ‰, Table 3), which is similar to $\delta^{13}C_{POM}$ values previously published by Rachold and Hubberten (1999) for the Lena Delta and Lena River upstream the delta (–31.3 to –25.7 ‰, July/August 1994/95), by A-GRO (www.arcticgreatrivers.org) ~900km upstream the Lena Delta at Zhigansk (–30.3 to –25.2 ‰, May-August 2004-2010), and by Semiletov et al. (2011) for the Lena Delta and Lena River (–30.0 to –25.0 ‰, August-September 1995-2008).

In general, the more enriched $\delta^{13}C_{POM}$ values (around –27 ‰) reflect the dominant contribution from C3 plants and soils (e.g. Hedges et al., 1997) from the river catchment. The more depleted $\delta^{13}C_{POM}$ values (<–29 ‰) point to mixing with riverine plankton utilizing dissolved inorganic carbon (DIC) with depleted $\delta^{13}C$ signatures as suggested for the Lena River (Alling et al., 2012; Rachold and Hubberten, 1999).

The total flux of primary production OM in the Lena River is thought to be negligible due to low light penetration in the turbid waters (Cauwet and Sidorov, 1996; Sorokin and Sorokin, 1996). Yet, the surface water layer from which we took our TSM samples is characterized by an abundance of riverine plankton (Kraberg et al., 2013; Sorokin and Sorokin, 1996), which could explain the small soil-derived OM fraction.

~~However, due to possible sorption of inorganic nitrogen to clay minerals (Schubert and Calvert, 2001) our calculated POC:PN ratios might be too low, underestimating the soil-derived OM fraction. This will be discussed in more detail in section 4.3.1. Another possibility for lower, but mainly soil derived POC:PN ratios could be the selective degradation of labile OM compared to total nitrogen (Kuhry and Vitt, 1996).~~ In chapter 4.3 we present the results of two binary and one three end-member mixing model to estimate the soil OM contribution to our POM samples and to further calculate the ^{14}C concentration of the soil fraction.

~~4.2.1 Stable carbon isotopes (δ^{13}C)~~

~~The δ^{13}C$_{POM}$ composition of our samples taken in 2009-2011 varies over a broad range (34.2 to 26.5 ‰, Table 3), which is similar to δ^{13}C$_{POM}$ values previously published by Rachold and Hubberten (1999) for the Lena Delta and Lena River upstream the delta (31.3 to 25.7 ‰, July/August 1994/95), by ArcticGRO (www.arcticgreatrivers.org) ~900km upstream the Lena Delta at Zhigansk (30.3 to 25.2 ‰, May-August 2004-2010), and by Semiletov et al. (2011) for the Lena Delta and Lena River (30.0 to 25.0 ‰, August-September 1995-2008).~~

~~In general, the more enriched δ^{13}C$_{POM}$ values (around 27 ‰) reflect the dominant contribution from C3 plants and soils (e.g. Hedges et al., 1997) from the river catchment. The more depleted δ^{13}C$_{POM}$ values (< 29 ‰) point to mixing with riverine plankton utilizing dissolved inorganic carbon (DIC) with depleted δ^{13}C signatures as suggested for the Lena River (Alling et al., 2012; Rachold and Hubberten, 1999). Analogous to the POC:PN ratios, the δ^{13}C$_{POM}$ compositions point to a considerable contribution of primary production OM to our samples from 2009-2011. Possible end-member calculation and implications for the ^{14}C concentration of the POM samples will be further discussed below in Sect. 4.3.2.~~

~~4.3~~4.2 ^{14}C and age heterogeneity ~~Radiocarbon composition~~ of POM (^{14}C$_{POM}$) in the Lena Delta

Terrestrial POM enters a river predominantly by physical weathering of adjacent soils (Raymond and Bauer, 2001 and references therein). Compared to vegetation utilizing modern atmospheric $^{14}CO_2$, the bulk POM of soil is pre-aged. The specific residence time of POM in the soil before entering the river depends on various environmental factors like humidity, temperature, topography, soil type, size and topography of the catchment area (e.g. Kusch et al., 2010; Oades, 1988; Raymond and Bauer, 2001; Trumbore, 2009; 1993). Furthermore, the fluvial transport of POM (and TSM) is governed by hydrological characteristics like runoff, discharge and flow velocity, sedimentation along riverbanks and floodplains as well as resuspension of deposited material. Therefore, the age of terrestrial POM is a combination of its residence time within the soil plus its residence time within the river basin, which can differ substantially for different terrestrial POM fractions (lipids versus lignin) from arctic watersheds (e.g. Feng et al., 2013; Gustafsson et al., 2011). ~~In permafrost affected watersheds with soils storing up to Pleistocene aged OM, the residence time within the watershed after entering the river might have only a minor effect.~~

The Lena Delta and Buor Khaya Bay surface water $^{14}C_{POM}$ concentrations presented here are depleted with respect to the current atmospheric $^{14}CO_2$ (Table 3). The $\Delta^{14}C$ values~~concentrations~~ range~~d~~ from –391 ‰ to –5~~11~~5 ‰ for 2009 to 2011 translating into ^{14}C ages >4~~12~~00 yrs and up to 3,920 yrs BP for samples taken close to the Pleistocene I~~i~~ce C~~c~~omplex deposits of Kurungnakh Island (Fig. ~~s. 1B and~~ 2, panels Q-T, Table 3). These results are within the range of reported $\Delta^{14}C$ values~~concentrations~~ for surface water POM offshore the Lena Delta and influenced by Lena River outflow (Karlsson et al., 2011). Based on these results it seems reasonable to assume that a large fraction of Lena Delta POM originates from physical weathering of relatively young Holocene soils (active layer) of the first delta terrace and south of the delta.

However, soils outcropping along the Lena River south of the delta can be substantially older than late Holocene ages (Kuptsov and Lisitsin, 1996) including some areas of Pleistocene I~~i~~ce C~~c~~omplex deposits (Grosse et al., 2013). Also, OM within the active layer and shallow permafrost table can be as old as 3,000 yrs BP in the Lena Delta (30 cm below surface, ~~(~~Höfle et al., 2013) or more than >10,000 yrs BP south of the delta (Kuptsov and Lisitsin, 1996). As shown by the lignin phenol composition of Lena Delta POM (Winterfeld et al., 2015~~4~~, ~~submitted as~~ companion paper) approximately half of the surface water POM~~particulate organic matter~~ is derived from the catchment south of the delta with considerable contributions from delta soils, particularly in the summer season when riverbank erosion is strongest. Considering that, we would expect generally older POM ^{14}C ages. Additionally, riverbank erosion contributes POM covering the ^{14}C age range of the whole soil~~bluff~~ profile, not only from the active layer.

One possible explanation for our relatively young POM ^{14}C ages could be the contribution of plankton~~algal~~-derived OM with a rather modern ^{14}C concentration~~signature~~ concealing the "true" age of soil-derived OM. Phytoplankton~~Algal~~-derived OM contribution was also suggested to be the reason for relatively young POM from the Ob' River inferred from a core of a floodplain lake (Dickens et al., 2011). Although the overall contribution and flux of autochthonous phytoplankton OM in the Lena River is rather small or even negligible due to the high turbidity and low light penetration (Cauwet and Sidorov, 1996; Sorokin and Sorokin, 1996), phytoplankton can be quite abundant in the surface water (Kraberg et al., 2013; Sorokin and Sorokin, 1996), which we sampled for our POM analyses. In the following section 4.3 we present results from two different approaches to quantify the soil OM fraction in our samples and further estimate its ^{14}C concentration based on the assumption of plankton-derived OM with modern Δ^{14}C values. ~~However, without directly determining the phytoplankton OM by microscopic counts or determining the chlorophyll-*a* content for each radiocarbon-dated sample, indirect evaluations have to be considered estimates. In the following chapterwe describe three different scenarios for possible ^{14}C corrections of POM in the Lena Delta to assess the potential range of Δ^{14}C concentrations and ^{14}C ages of soil-derived particles. In these scenarios we assume that the algal-derived OM is of modern ^{14}C age (Δ^{14}C ~49‰ and) although this might not be true. DIC Δ^{14}C concentrations are not available for the Lena River. The DIC utilized by the phytoplankton is predominantly derived from the carbonate weathering within the Lena watershed and from soils, both, providing a depleted ^{14}C carbon signature and only slowly exchanging with the atmosphere. Therefore, the following calculated Δ^{14}C concentrations of surface water POM from the Lena River have to be considered minimum, i.e. maximum ^{14}C ages.~~

~~4.4~~4.3 ~~^{14}C estimates of soil-derived POM for algal OM contribution~~Quantitative POM source determination and soil ^{14}C concentrations

~~4.4.1~~4.3.1 Binary mixing model s~~S~~cenario 1~~: Soil-derived OM ^{14}C estimates based on POC:PN ratios~~

POC:PN ratios around 6 are usually associated with OM derived from algal/bacterial primary production and higher ratios of >20 with OM derived from soils (Hedges et al., 1997; Meyers and Lallier-vergés, 1999 and references therein). However, as noted above (section 4.1) adsorption of inorganic nitrogen derived from OM decomposition (e.g. ammonium) to clay minerals (Schubert and Calvert, 2001), which is not accounted for when determining the total PN content, may additionally affect the POC:PN ratio in TSM leading to lower values than would be expected from mixing of the two end-members alone. Likewise, selective degradation of labile organic carbon (Kuhry and Vitt, 1996) would also ~~have the same effect~~result in lower POC:PN ratios. Both processes would lead to low estimates of the soil-derived OM contribution and suggest a higher contribution from plankton. Both explanations were also suggested by Sanchéz-García et al. (2011) for POM in the Laptev Sea offshore the Lena Delta with unusually low POC:PN ratios.

In order to estimate the inorganic nitrogen content for our sample sets from each year, we used the intercept of the POC versus PN content regression line at ~~zero~~ POC = 0 (Fig. S2~~2 supplement~~). By subtracting these amounts from the analyzed total PN content of the respective samples, we could calculate new soil POC:PN ratios (POC:PN$_{\mathrm{NEW}}$) for our samples (Table S4~~2~~ supplement). Based on these POC:PN$_{\mathrm{NEW}}$ ratios (and their uncertainty, derived from the standard error of the POC versus PN regressions), we calculated the soil-derived fraction within our POM samples using a simple two end-member mixing model as following:

POC:PN$_{\mathrm{NEW}}$~~$_{\mathrm{POM}}$~~ = $f_{\mathrm{soil}} \times$ POC:PN$_{\mathrm{soil}}$ + $f_{\mathrm{plankton}} \times$ POC:PN$_{\mathrm{plankton}}$ (1)

and 1 = $f_{\mathrm{soil}} + f_{\mathrm{plankton}}$ (2)

where POC:PN$_{\mathrm{NEW}}$~~$_{\mathrm{POM}}$~~ is the corrected~~measured~~ value of~~in~~ the POM sample (Table S4~~1~~). As the end-member for POC:PN$_{\mathrm{soil}}$ we chose~~assumed~~ ~~a value of 20.2~~23.7 ±~~±-~~ 11 (~~1sd~~Table 4 and S1 supplement), which is an average calculated from Lena Delta first terrace soils presented in Winterfeld et al. (2015~~4~~, ~~submitted as~~ companion paper) and delta soils as well as soil from the Lena River watershed covering the taiga to tundra transition from Höfle et al. (2013), Zubrzycki (2013), and Zubrzycki e al. (2012, individual values in Table S1 supplement). The POC:PN$_{\mathrm{plankton}}$ end-member value is 6 ± ~~+-~~1 (T~~1sd[1]~~able 4, ~~) (~~e.g. Meyers, 1994~~ and references therein~~).

The calculated soil-derived OM fractions of Lena Delta POM varied from 0.26~~8~~ to 0.70~~55~~ (mean of 0.35~~41~~, n=2~~1~~1, Table 5) for summer 2009, from 0.26~~26~~ to 0.44~~51~~ (mean of 0.34~~43~~, n=11~~10~~) in summer 2010, and from 0.39~~48~~ to 0.50~~69~~ (mean of 0.43~~54~~, n=9~~3~~) for late June/early July 2011 (Table 5~~3~~). We used the POC versus PN (wt%) regression line from the summer 2010 samples within the delta to correct the two samples taken outside of the delta in the Buor Khaya Bay. They had calculated soil fractions of 0.39~~43~~ (sample 35) and 0.12~~03~~ (sample 36). The same was done for the single sample from late May 2011. We used the regression line from the samples taken 4 weeks later in June/July and the soil fraction of sample 37 was 0.13~~25~~ (Table 5~~3~~). Note that these soil- and plankton-derived OM fractions can only serve as rough estimates. Without determining the particulate organic nitrogen directly for every sample our POC:PN$_{\mathrm{NEW}}$ ratios (Table S4) might be highly over- and underestimating OM fractions in individual samples.

The~~ so~~ calculated soil and plankton OM fractions (f_{soil} and $f_{plankton}$, Table 5~~3 supplement~~) were further used in an isotopic mass balance to determine the ~~Δ~~^{14}C concentration of the soil fraction assuming the plankton-derived OM is modern ($\Delta^{14}C_{plankton}$ = 41.9 ± 4.2~~49~~‰, Table 4):

$$f_{POM} \times \Delta^{14}C_{POM} = f_{soil} \times \Delta^{14}C_{soil} + f_{plankton} \times \Delta^{14}C_{plankton} \quad (3)$$

and $f_{POM} = f_{soil} + f_{plankton}$ (4)

where f_{POM}, f_{soil}, and $f_{plankton}$ are the fractions of POM, soil- and plankton-derived OM and $\Delta^{14}C_{POM}$, $\Delta^{14}C_{soil}$, and $\Delta^{14}C_{plankton}$ are the Δ^{14}C values~~concentrations~~ of these ~~s~~ sources. As not~~mention~~ed above, 41.9 ± 4.2~~49~~‰ is assumed as a maximum estimate for $\Delta^{14}C_{plankton}$~~.~~ based on atmospheric CO_2 Δ^{14}C values for May to August 2009-2011 from the Schauinsland observatory, Germany.

The results of the newly calculated soil Δ^{14}C~~$_{soil}$~~ values~~concentrations~~ using POC:PN$_{NEW}$~~$_{corr}$~~ ratios ($\Delta^{14}C_{POC:PN}$) to partition between soil- and plankton-derived OM are shown in T~~t~~able 5~~4~~. The soil $\Delta^{14}C_{POC:PN}$ values~~concentrations~~ range from –884~~786~~ ‰ to –322~~258~~ ‰ for the sampling period of 2009-2011 translating into soil ^{14}C~~$_{soil}$~~ ages >~~2~~3,000 yrs BP with an average of 6,~~62~~70~~0~~0 yrs BP (Table 5~~4~~). Analogous to the two comparatively old bulk $^{14}C_{POM}$ ages off Kurungnakh Island taken in 2009 and 2010 (Table 3), the calculated $^{14}C_{POC:PN}$ age~~s~~ for ~~these~~ sample 1~~s~~ was~~ere with~~ ~~∼~~17~~1~~,~~600~~ 250 years BP and yielded a fossil age (>50,000 years BP) for sample 31~~ the oldest~~. Considering that the riverbank outcrops of Kurungnakh Island cover an age range of approximately 100 kyrs with organic-rich ice complex deposits of about 50 kyrs (Wetterich et al., 2008) these ^{14}C ages seem to be realistic.

Again, the uncertainties associated with the contribution of inorganic nitrogen to our total PN contents are rather high resulting in a relatively rough estimation of soil and plankton OM fractions. These uncertainties are further affecting the calculation of the soil $\Delta^{14}C_{POC:PN}$ concentrations. That means the calculated ^{14}C ages are estimates. Yet, they demonstrate that a possible underestimation of soil-derived OM ages can be considerable.

~~4.4.2~~4.3.2 Binary mixing model s~~S~~cenario 2

Similar to the~~ correction~~ approach discussed in section 4.3.1~~above~~, we used a second scenario based on δ^{13}C values~~compositions~~ to distinguish between soil- and plankton-derived OM in our Lena Delta POM samples. The vegetation in the Lena River catchment (taiga and tundra) is dominated by C3 plants with a δ^{13}C of around –25 ‰ to –27 ‰ (Rachold and Hubberten, 1999). This is also reflected in our δ^{13}C data from the first delta terrace soils with an average of –26.2 ‰ (n=7, Table 3). Bird et al. (2002) determined the δ^{13}C composition of taiga and tundra soils (excluding peatlands) along Yenisey River on a latitudinal transect. ~~Their data suggests an average δ^{13}C concentration of –26.69 +-1 (1sd) ‰ for tundra and taiga soils combined~~For the binary mixing model we used a soil OM end-member value –26.6 ± 1‰ (Table 4), which is a combination of δ^{13}C data from this study and the literature (Table S2, Bird et al., 2002; Pitkänen et al., 2002; Xu et al., 2009) covering tundra and taiga soils in Siberia and Alaska~~, which we use as the soil OM end-member in our two end-member model~~.

The δ^{13}C composition of riverine plankton POM depends on the fractionation between phytoplankton and dissolved inorganic carbon (DIC). The distribution of the different DIC fractions (dissolved CO_2, bicarbonate (HCO_3^-), and carbonate ion (CO_3^{2-}) varies depending on temperature and pH. In the lower reaches of the Lena River and the Lena Delta >90 % of the DIC are made up of bicarbonate (Alling et al., 2012), i.e. the δ^{13}C of bicarbonate represents the δ^{13}C of DIC. Sources for DIC are generally the CO_2 derived from soil OM degradation, ~~and~~ CO_2 released during the dissolution of carbonates, and CO_2 from the atmosphere. The Lena River geochemistry is mainly influenced by carbonate weathering and groundwater in the summer season (Gordeev and Sidorov, 1993). Assuming a fractionation of –22.5 ± 2.5~~5~~ ‰ between phytoplankton and DIC (Mook and Tan, 1991) ~~these high latitude waters~~ and a $\delta^{13}C_{DIC}$ of –8 ‰ for the Lena Delta (Alling et al., 2012) a plankton ~~δ~~ δ^{13}C~~^{13}C~~ end-member value of –30.5 ± 2.5~~3~~ ‰ would be expected. Similar or more depleted δ^{13}C values of bicarbonate and phytoplankton POM were also determined in the Yenisey and Ob' Rivers (Galimov et al., 2006) and in temperate estuaries (e.g. Ahad et al., 2008; Chanton and Lewis, 2002). Our most depleted $\delta^{13}C_{POM}$ of –34.2 ‰ for a sample from the summer 2009 (Table 3) is even lower than the plankton end-member used here ($\delta^{13}C_{plankton}$ = –30.5 ‰). The sample location is offshore Muostakh Island (Fig. 1B) and influenced by mixing with marine waters, which complicates the DIC composition and processes affecting δ^{13}C of DIC (Alling et al., 2012).

We used the following two end-member model to estimate the soil- and plankton-derived OM fractions ($f_{soil}, f_{plankton}$) to our POM samples:

$$\delta^{13}C_{POM} = f_{soil} \times \delta^{13}C_{soil} + f_{plankton} \times \delta^{13}C_{plankton} \quad (5)$$

and $$1 = f_{soil} + f_{plankton} \quad (6)$$

where $\delta^{13}C_{POM}$ is the analyzed δ^{13}C value~~composition~~ of our POM samples (Table 3), $\delta^{13}C_{soil}$ is the soil end-member value of –26.6 ± 1‰, and $\delta^{13}C_{plankton}$ is plankton end-member value of –30.5 ± 2.5 ‰ (Tables 4 and S3 supplement).

The calculated soil OM fractions varied from 0.12~~07~~ to 0.51~~68~~ in summer 2009 (mean of 0.32~~44~~, n=12, Table 5~~1~~), from 0.34~~47~~ to 0.59~~77~~ in summer 2010 (mean of 0.45~~62~~, n=12~~0~~), and from 0.44~~60~~ to 0.59~~76~~ in spring 2011 (mean of 0.53~~70~~, n=3). The Buor Khaya Bay POM sample 21 showed a soil fraction of 0.75 and the spring freshet value (sample 37) a soil fraction of 0.80 (Table 5)~~45~~.~~ Two samples are not considered in this end-member model, because their δ^{13}C values are outside the range of the end-member values chosen here (sample 37 and 19, see Table 3). This illustrates the limitations of our approach. Constraining a model with appropriate end-members strongly influences the quality of the model output and here we were not able to account for the whole range of natural variability observed in the Lena River.~~ As discussed above in Sect. 4.3.1, these soil OM fractions are rough estimates in the absence of direct plankton determination.

Furthermore, ~~The~~ Δ^{14}C values~~concentrations~~ of the soil-derived OM fraction were~~are~~ ~~further corrected~~ calculated for the contribution of modern plankton OM as described in Sect. 4.3.1 using the Eq. (3) and (4). Because the soil contributions calculated in this scenario are slightly higher than in the POC:PN-based scenario, the Δ^{14}C calculations resulted in less radiocarbon depleted estimates compared to the POC:PN scenario (table 5~~4~~). Nonetheless, the Δ^{14}C values~~concentrations~~ based on the δ^{13}C end-member model are considerably ^{14}C-depleted compared to bulk POM ~~Δ~~^{14}C concentrations. The estimated Δ^{14}C~~$_{corr}$~~ values~~concentrations~~ range from –944~~704~~ ‰ to –495~~191~~ ‰ for all samples 2009-2011 representing ^{14}C ages from 23,100~~ 9720~~ to 2,370~~1770~~ yrs BP (Table 5~~4~~). The oldest samples are, again, the ones taken close to~~ Kurungnakh Island with its~~ Pleistocene I~~i~~ce C~~c~~omplex deposits (samples 1 and 31). ~~In contrast to scenario 1, it is more obvious in this scenario that the POM samples taken in late June/early July 2011 are more enriched in ^{14}C (younger) than the POM samples taken later in the summer (Aug 2009 and July/Aug 2010). Similar observation were made for DOM Δ^{14}C concentrations of the Lena River at Zhigansk ~900km south of the delta (Raymond et al., 2007) and the Kolyma River in East Siberia (Neff et al., 2006). Their explanation was that due to the deepening of the active layer in summer older soil layers are accessible for melt water and groundwater contributing an older DOM signature to the river than in spring when most soil is still frozen. A comparable scenario could explain estimated Δ^{14}C concentration decreasing from spring freshet to summer. Additional to active layer deepening riverbank erosion is strongest during the summer and might contribute a considerable amount of soil-derived OM to the Lena Delta surface water.~~

4.3.3 Comparison of scenario 1 and 2

Of all samples from 2009-2011 presented here, 28 samples have a soil fraction estimates from both scenarios. For these samples the POC:PN scenario gives a lower mean soil fraction of 0.33 compared to 0.43 from scenario 2 (based $\delta^{13}C$, Fig. S3). The comparison demonstrates that the error estimates determined in the end-member calculation are realistic, as only 6 (2) of 28 samples are outside of one (two) standard deviations of the estimated error. As the uncertainty of the estimates is high, both estimates are only weakly correlated (removing the clear outlier sample 37 we get R=0.3,p=0.1).

Because we assumed a modern $\Delta^{14}C$ value for the phytoplankton OM, the estimated soil ^{14}C concentrations are related to the amount of soil OM calculated in both scenarios. The lower mean fraction in scenario 1 therefore resulted in a lower mean soil $\Delta^{14}C$ value of –569‰ (^{14}C age = 6700 year BP, n= 26) compared to –495‰ (^{14}C age = 5430 years BP, n= 28) for scenario 2. Besides the spring freshet sample from 2011, the two samples with the lowest bulk $\Delta^{14}C$ values (sample 1 & 31, Table 3) also showed the one of the lowest estimated soil $\Delta^{14}C$ values, namely –884‰ and radiocarbon-free (fossil) for scenario 1 as well as –944‰ and –754‰ for scenario 2. The two samples were taken close to Ice Complex deposits within the delta, which illustrates the locally pronounced influence of these deposits. Another sample standing out is sample 19 taken outside the delta close to Muostakh Island, which is also composed of Ice Complex deposits (Fig. 1B). Its bulk POM $\Delta^{14}C$ value was highest ($\Delta^{14}C$ = –55‰, Table 3) pointing to comparatively young OM and its $\delta^{13}C$ value was lowest (–34.2‰) pointing to a rather high contribution of phytoplankton. This is in our line of argumentation assuming a contribution of plankton-derived OM with a modern $\Delta^{14}C$ value. Because of our chosen end-members, the soil fraction in sample 19 was rather small in the ^{13}C-based scenario and the resulting $\Delta^{14}C$ value of the soil fraction was with –774‰ (^{14}C age of 11,890 years BP) the second lowest.

In contrast to scenario 1, it is more obvious in scenario 2 that the POM samples taken in late June/early July 2011 are more enriched in ^{14}C (younger) than the POM samples taken later in the summer (Aug 2009 and July/Aug 2010). Similar observation were made for DOM $\Delta^{14}C$ concentrations of the Lena River at Zhigansk ~900km south of the delta (Raymond et al., 2007) and the Kolyma River in East Siberia (Neff et al., 2006). Their explanation was that due to the deepening of the active layer in summer older soil layers are accessible for melt water and groundwater contributing an older DOM signature to the river than in spring when most soil is still frozen. A comparable scenario could explain estimated $\Delta^{14}C$ concentration decreasing from spring freshet to summer. In addition to active layer deepening riverbank erosion is strongest during the summer and might contribute a considerable amount of soil-derived OM with a large ^{14}C age range to the Lena Delta surface water.

4.3.4 Holocene soil versus Pleistocene Ice Complex deposits

As an extension of the two end-member model, we applied a dual-carbon isotope (δ^{13}C, Δ^{14}C) three end-member model (after Karlsson et al., 2011; Vonk et al., 2010, 2012) to separate the source contributions of phytoplankton, Holocene soil, and Pleistocene Ice Complex deposits (ICD) using the following equations:

$\delta^{13}C_{POM} = f_{\text{soil Holocene}}\ \text{X}\ \delta^{13}C_{\text{soil}} + f_{\text{soil ICD}} \times \delta^{13}C_{\text{soil}} + f_{\text{plankton}} \times \delta^{13}C_{\text{plankton}}$ (7)

$^{14}C_{POM} = f_{\text{soil Holocene}}\ \text{X}\ {}^{14}C_{\text{soil}} + f_{\text{soil ICD}} \times {}^{14}C_{\text{soil}} + f_{\text{plankton}} \times {}^{14}C_{\text{plankton}}$ (8)

and $1 = f_{\text{soil Holocene}} + f_{\text{soil ICD}\cancel{\text{pleistocene}}} + f_{\text{plankton}}$ (9),

where $f_{\text{soil Holocene}}$, $f_{\text{soil ICD}}$, and f_{plankton} are the fractions of Holocene soil, Ice Complex deposits, and riverine plankton contributing to each POM sample. The end-member values chosen for Holocene soil OM were δ^{13}C –26.6 ± 1‰ and Δ^{14}C of –282 ± 133‰. As the two soil end-members have very similar δ^{13}C values (see Table 4, Holocene soil δ^{13}C = –26.6 ± 1.0 ‰, Ice Complex δ^{13}C = –26.3 ± 0.67 ‰), the δ^{13}C mainly determines the phytoplankton vs. total (Holocene + Pleistocene) soil fraction, whereas Δ^{14}C mainly determines the fraction of Holocene to Pleistocene soil. Therefore, the results concerning the total soil fraction are very similar to those derived from the two-end-member model (R^2=0.92, mean two end-member soil fraction is 0.43, mean three end-member is 0.46, Fig. S4) than derived with the two end-member model.

As shown in Table S5 and Figure 4, the POM source determinations resulted in relative fractions of 0.19-0.83, 0.11-0.65, and 0.07-0.34 for plankton, Holocene soil, and Ice Complex deposits, respectively. Again, sample 31 taken off Kurungnakh Island (Ice Complex) stands out with the highest contribution from ICD (fraction of 0.34) and sample 19 taken outside the delta with highest contribution of phytoplankton (fraction of 0.83). Overall, with the end-members chosen here and despite the high spatial variability within the delta, the riverine phytoplankton fraction contributes most (mean of 0.55) to the surface water POM compared to the Holocene soil OM (0.32) and Ice Complex deposits (0.14). The rather low OM contribution from Ice Complex deposits reflects the distribution of these deposits in the Lena watershed, where they are only locally concentrated within elevations up 400m (Grosse et al., 2013).

~~4.5~~4.4 Implications of estimated soil-derived POM Δ^{14}C

The two binary mixing models (scenario 1 & 2)~~s~~ discussed above allow an estimate ~~of the contribution of plankton-derived OM to our bulk POM samples and~~ of the soil Δ^{14}C values~~concentration estimates~~ based on a~~this~~ contribution of modern phytoplankton-derived OM~~-contribution~~. Both scenarios show considerably ^{14}C-depleted soil-derived OM compared to the bulk ~~Δ~~^{14}C$_{POM}$ concentrations. This implies that the bulk POM ^{14}C age of samples taken in surface water during the summer, when the riverine primary production is high,~~ have~~ likely underestimate~~d~~ the age of the soil-derived OM transported by the Lena River. The estimated soil Δ^{14}C values~~concentrations~~ and ^{14}C ages in both scenarios give a more plausible picture for soil-derived POM in the Lena River watershed. In contrast to DOM that is restricted in its flow path to the unfrozen soil layers, POM is not exclusively derived from surface soils. It also originates from resuspension of accumulated pre-aged material along the river channels and from riverbank erosion. The latter contributes POM with ~~Δ~~^{14}C concentrations representing the whole range covered by the respective riverbank bluffs. In the Lena Delta this is predominantly OM of late Holocene age with local inputs from ice complex deposits of Pleistocene age (e.g. Bolshiyanov et al., 2015~~4~~; Schirrmeister et al., 2011; Schwamborn et al., 2002). About half of the POM in the Lena Delta originates from the boreal forest hinterland south of the delta (Winterfeld et al. 2015~~4~~, ~~submitted as~~ companion paper). In the hinterland~~Here~~ the soils along the Lena River and its tributaries can be older than the delta soils, i.e. covering the whole age range from Holocene soils to Pleistocene I~~i~~ce C~~c~~omplex deposits (e.g. Grosse et al., 2013; Kuptsov and Lisitsin, 1996). Our estimated soil ^{14}C ages of about 2,37~~0~~0 to 23~~11~~,16~~6~~00 years (Table 5~~4~~) for both scenarios therefore better reflect these hinterland deposits contributing a heterogeneous ^{14}C age mix to riverine POM than the bulk POM ^{14}C ages.

The soil POM ^{14}C estimates as well as the Lena Delta first terrace soil data (δ^{13}C and Δ^{14}C) presented here improve our knowledge of the stable and radiocarbon isotopic range characteristic for soil-derived OM exported by the Lena River to the Laptev Sea. This information is critical for modeling the OM contribution from different terrestrial (fluvial vs. coastal erosion) and marine sources to Laptev Sea sediments and thus help characterizing and quantifying the OM pools released from permafrost thawing. Recent studies suggest that OM exported by arctic rivers and OM derived from erosion of ice complex coasts differ in their mineral and OM composition and thus show different potential for remineralization by microorganisms after thawing as well as different modes of transport and burial (e.g. Feng et al., 2013; Gustafsson et al., 2011; Knoblauch et al., 2013; Vonk et al., 2012). This has a direct impact on how to assess the possibility of a positive carbon-climate feedback from permafrost degradation, which has the potential to enhance global greenhouse warming by releasing huge amounts of previously frozen OM to the atmosphere.

A promising approach to distinguish OM sequestered in arctic sediments is the dual-carbon-isotope end-member simulation applied by Karlsson et al. (2011) and Vonk et al. (2012; 2010) to Laptev and East Siberian Sea sediments. The authors use the $\delta^{13}C$ and $\Delta^{14}C$ values~~concentrations~~ of surface water suspended matter and surface sediments to quantify OM derived from Pleistocene ice complex deposits, soil/top-soil OM exported by Siberian rivers, and marine phytoplankton OM. Their end-member definitions for ice complex deposits and marine primary production are rather well constrained. In contrast, the soil/topsoil end-member is more difficult to define, particularly when using indirect parameters such as riverine DOM and POM. Here the~~our~~ $\delta^{13}C$ end-member chosen for surface soil including our first terrace soil data (Table S2) and our estimates for soil-derived POM (Table 5~~4~~) provide new $\delta^{13}C$ and $\Delta^{14}C$ value~~concentration~~ ranges (Fig. 3B) for fluvially exported soil POM. Together with published $\delta^{13}C$ values~~concentrations~~ from tundra and taiga soils in Siberia and Alaska (Bird et al., 2002; Pitkänen et al., 2002; Xu et al., 2009) we ~~can~~ defined a Lena River soil OM end-member with a $\delta^{13}C$ value of –26.6 ± 1~~.1~~ ‰. Based on the $\delta^{13}C$ binary mixing model (scenario 2) t~~T~~he corresponding $\Delta^{14}C$~~^{14}C~~ value~~ concentration~~ is –495~~362~~ ± ~~123~~ 153 ‰ ~~is taken from the second scenario discussed above~~ (Table 5~~4~~). ~~The $\delta^{13}C$-based scenario is favored~~We favor scenario 2 ~~here~~ over the POC:PN-based scenario, because it allows to calculate the soil fraction of each POM sample based on its bulk $\delta^{13}C$ value~~composition~~, which is a mixture of soil and phytoplankton OM. In contrast, in the POC:PN-based scenario a constant value for particulate inorganic nitrogen is subtracted, which does not necessarily represent the actual inorganic nitrogen content of the particular sample. Moreover, it does not account for selective degradation of labile carbon, which could result in overestimating the phytoplankton contribution. ~~The uncertainties associated with the $\delta^{13}C$-based estimates are smaller, as the $\delta^{13}C$ end-member for soil-derived and phytoplankton-derived OM are fairly well constrained (see also Sect. 4.3.2).~~

T~~However, ~~he above proposed $\delta^{13}C$ and $\Delta^{14}C$ values for the~~our~~ soil OM end-member of~~for~~ the Lena River catchment make~~s~~ it more complicated to distinguish between soil/top-soil derived OM from the river and ice complex deposits from coastal erosion (see Fig. 3B). The $\delta^{13}C$ values~~signatures~~ of both end-members are almost indistinguishable. The $\Delta^{14}C$ range of our soil-derived POM estimates is lower~~more depleted~~ than the end-members used by Karlsson et al. (2011) and Vonk et al. (2012; 2010). Furthermore, Höfle et al. (2013) have shown that OM within the first 30cm of a polygon rim of the Lena Delta first terrace can be 3,000 ^{14}C years old, which make the end-members chosen for fluvial exported soil/top-soil OM by Karlsson et al. (2011) and Vonk et al. (2012; 2010) appear too young. Using the bulk surface water POM and DOM δ^{13}C and Δ^{14}C values~~compositions~~ as end-member could therefore highly over- or underestimate the soil OM contribution from permafrost watersheds and in turn highly over- or underestimate OM contribution from ice complex to marine sediments.

However, we are aware of the limitations and uncertainties associated with the soil Δ^{14}C estimates discussed above (and in sections 4.3.1, 4.3.2). Without determining the phytoplankton biomass of each sample by microscopic counting or from chlorophyll-*a* analysis, the plankton OM fraction calculated based on POC:PN ratios (section 4.3.1) and based δ^{13}C values~~composition~~ (section 4.3.2) can only be regarded as rough estimates. These estimates give orders of magnitude of OM contribution from the individual sources rather than exact values, which in turn provide a possible range of Δ^{14}C values~~concentrations~~ for soil-derived POM. An additional source of uncertainty is our assumption of a modern ~~Δ~~^{14}C concentration of plankton OM. Without determining the ~~Δ~~^{14}C concentration of the Lena River DIC, which is utilized by phytoplankton, we cannot be sure of a modern ^{14}C concentration~~signature~~. Lena River DIC is derived from several sources providing carbon with different ^{14}C concentrations. DIC derived from carbonate and silicate weathering is ^{14}C-depleted, DIC derived from soil respiration has a broad range of Δ^{14}C values due decomposition of soil OM pools of varying age, and DIC derived from exchange with the atmosphere has modern Δ^{14}C values. The resulting DIC Δ^{14}C value is mixture of these sources depending on the varying contribution from each source. Furthermore, Tank et al. (2012) found that DIC yields are negatively correlated with continuous permafrost extent in the watersheds of the six large Arctic rivers including the Lena. This would imply that carbonate weathering is to some extent hampered by continuous permafrost making up 77% of the Lena catchment area (Tank et al., 2012). In summary, because there is no DIC Δ^{14}C value available for the Lena River and there are too many factors influencing the DIC Δ^{14}C value, we made the simplified assumption of modern Δ^{14}C of 41.9 ± 4.2 (average value of atmospheric CO$_2$ Δ^{14}C May-Aug 2009-2011 from Levin et al., 2013). ~~geochemistry and DIC are dominated by dissolution of carbonates in the watershed contributing fossil ^{14}C to the DIC pool, leading likely to ^{14}C-depletion in river phytoplankton. The true contribution of modern phytoplankton OM is thus likely smaller than estimated above.~~ Consequently, if true Δ^{14}C values of DIC in the Lena were depleted relative to the modern atmosphere, the soil-derived

POM would be less ^{14}C depleted than estimated~~calculated~~ here. Thus, Δ^{14}C values for soils have to be considered minimum estimates or in other words, the ~~. Therefore, our~~ estimated ^{14}C soil ages have to be considered maximum ages.

The best way to determine the Δ^{14}C value~~concentration~~ of riverine soil-derived OM would be a biomarker-specific radiocarbon analysis using source-specific compounds, e.g., short- and long-chain alkanoic~~fatty~~ acids for plankton- and terrestrial-derived OM, respectively. However, for these analyses large samples of POM are needed. The samples analyzed during our study were too small to allow for compound-specific dating.

5 Conclusions

There are~~is~~ only few~~scarce~~ data available on ^{14}C concentrations~~tents~~ of POM from Lena River surface water, but regarding~~within~~ the likely positive carbon-climate feedback to greenhouse warming the quality and fate of this permafrost OM pool in the coastal waters of the Laptev Sea is currently under debate (e.g. Feng et al., 2013; Gustafsson et al., 2011; Karlsson et al., 2011; Vonk et al., 2012). With this study we provide ~~the~~ one of the first data sets on surface water POM ~~Δ~~^{14}C concentrations from the Lena Delta sampled during the summers 2009 and 2010 and during spring 2011 (n=30 samples). The contribution of modern phytoplankton POM to these samples was on the one hand estimated using binary mixing models based on POC:PN ratios and δ^{13}C values, which ~~to~~ allowed us ~~for~~ to calculate~~ion~~ ~~of~~ the Δ^{14}C values~~concentrations~~ of the soil-derived POM fraction. These soil Δ^{14}C estimates were low~~are depleted~~ compared to the bulk POM Δ^{14}C values~~concentrations~~ and therefore seem to represent the heterogeneous ^{14}C mix of soil OM ranging from Holocene to Pleistocene age (e.g. ice complex deposits) in the Lena River watershed more accurately. Moreover, we applied a dual carbon-isotope three end-member model to further distinguish between OM contributions from Holocene soils and Pleistocene Ice Complex deposits to the soil fraction. Here, we could show that the overall contribution of Ice Complex deposits to surface water POM in the Lena Delta was relatively low, which reflects the restricted spatial distribution of these deposits within the Lena watershed. Only samples taken close to Ice Complex deposits exhibited higher contributions of this source in the model implying a small, locally pronounced influence on surface water POM before it becomes mixed with other soil-derived POM during fluvial transport.

Because of the limitations of our approach, particularly the assumption of modern phytoplankton OM without determining the ~~Δ~~^{14}C concentration of the Lena River DIC utilized by phytoplankton, our ^{14}C estimates for the soil-derived fraction have to be considered minimum $\Delta^{14}C$ concentrations and maximum ^{14}C ages, respectively. Nonetheless, we propose average values for the soil POM isotopic composition based on our data and published values of ~~(~~$\delta^{13}C$ = –26.6 ± 1~~.1~~ ‰ and ~~;~~ $\Delta^{14}C$ = –495~~362~~ ± 15~~2~~3 ‰ (Tables 5 and S2), which will be useful for dual-carbon-isotope simulations focusing on unraveling the OM contributed by different terrigenous (fluvial vs. coastal erosion) and marine sources to arctic sediments.

The complete data set presented here can also be found in PANGAEA (www.pangaea.de).

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Table 1. Soil s~~S~~amples from riverbank bluffs and total suspended matter (TSM) samples presented in this study analyzed for particulate organic carbon, stable, and radiocarbon isotope composition. ~~Information on additional samples analyzed for organic carbon content can be found in Winterfeld et al. (2014, submitted as companion paper) in table 1 and S1.~~ Latitude and longitude are given in decimal degrees (dec). Bluff height is given in meters above river level [m a.r.l.] measured in August 2009 and only applicable for the soil samples. The TSM samples were taken from the surface water layer at a sampling depth of ca. 0.5m.~~Not applicable denoted by n.a.~~

Sample code	Sample & site description	Date of sampling	Lat.N~~titude~~ [dec]	Long.E~~itude~~ [dec]	Bluff height [m a.r.l.]
Lena Delta first terrace soil profiles					
L09-12	Samoylov Island, 5 depths sampled	18-Aug-2009	72.3775	126.4954	7.5
L09-28-2	Bykovskaya Channel, 2 depths sampled	21-Aug-2009	72.0586	128.6309	1.7
Lena Delta TSM					
1	Olenyokskaya Channel	14-Aug-2009	72.4772	125.2856	~~n.a.~~
2	Olenyokskaya Channel	14-Aug-2009	72.3598	125.6728	~~n.a.~~
3	Lena River main channel	16-Aug-2009	72.1526	126.9160	~~n.a.~~
4	Lena River main channel south of Tit Ari Island	16-Aug-2009	71.9040	127.2544	~~n.a.~~
5	Sardakhskaya/Trofimovskaya Channel	17-Aug-2009	72.5825	127.1891	~~n.a.~~
6	Sardakhskaya Channel	17-Aug-2009	72.7002	127.4930	~~n.a.~~
7	Sardakhskaya/Trofimovskaya Channel	17-Aug-2009	72.6268	127.3860	
8	near Kurungnakh Island	18-Aug-2009	72.2904	126.0909	
9	Lena River main channel	19-Aug-2009	72.2987	126.7080	
10	Lena River main channel	19-Aug-2009	72.2760	126.9041	~~n.a.~~
11	Lena River main channel	19-Aug-2009	72.5159	126.7142	~~n.a.~~
12	Bykovskaya Channel	20-Aug-2009	72.4140	126.9124	~~n.a.~~
13	Lena River Bykovskaya Channel	20-Aug-2009	72.2352	127.9619	~~n.a.~~
14	Lena River Bykovskaya Channel	20-Aug-2009	72.0341	128.5232	~~n.a.~~
15	Lena River Bykovskaya Channel	21-Aug-2009	72.0354	128.0974	
16	Lena River Bykovskaya Channel	21-Aug-2009	72.0586	128.6309	
17	offshore Bykovsky Peninsula	22-Aug-2010	71.7889	129.4189	
18	NE of Muostakh Island	22-Aug-2010	71.6761	130.1728	
19	NE of Muostakh Island	22-Aug-2010	71.7062	130.2900	~~n.a.~~
20	W of Muostakh Island	23-Aug-2010	71.6088	129.9393	
21	close to Muostakh Island shoreline	23-Aug-2010	71.5750	129.8200	~~n.a.~~
22	close to Samoylov Island	30-Jul-2010	72.3650	126.4628	~~n.a.~~
24	Sardakhskaya/Trofimovskaya Channel	31-Jul-2010	72.5343	126.8794	~~n.a.~~
25	Lena River Trofimoskaya Channel	31-Jul-2010	72.4764	126.6250	~~n.a.~~
26	Lena River Trofimoskaya Channel	31-Jul-2010	72.4764	126.8588	~~n.a.~~

27	Lena River main channel south of Samoylov	1-Aug-2010	72.3776	126.7478	~~n.a.~~
28	Lena River main channel north of Tit Ari Island	1-Aug-2010	72.2102	126.9423	~~n.a.~~
29	Lena River main channel south of Tit Ari Island	1-Aug-2010	71.9514	127.2582	~~n.a.~~
30	Lena River main channel off Kurungnakh	2-Aug-2010	72.2808	126.2091	~~n.a.~~
31	Lena River main channel	2-Aug-2010	72.3567	126.3521	~~n.a.~~
32	Lena River Bykovskaya Channel	3-Aug-2010	72.3604	127.6761	~~n.a.~~
33	Bykovskaya Channel	3-Aug-2010	72.3604	127.6765	~~n.a.~~
34	off Bykovsky Peninsula	4-Aug-2010	71.7015	129.7523	
35	offshore Lena Delta	6-Aug-2010	72.0379	129.7694	~~n.a.~~
36	offshore Lena Delta	6-Aug-2010	72.2753	129.8248	~~n.a.~~
37	Lena River main channel off Samoylov Island	29-May-2011	72.3651	126.4757	~~n.a.~~
38	Lena River main channel, south of Stolb	26-Jun-2011	72.3705	126.6538	~~n.a.~~
39	off Kurungnakh	26-Jun-2011	72.3334	126.2914	
40	close to Samoylov Island	29-Jun-2011	72.3681	126.4738	
41	close to Samoylov Island	29-Jun-2011	72.3681	126.4738	
42	close to Samoylov Island	30-Jun-2011	72.3681	126.4738	~~n.a.~~
43	close to Samoylov Island	30-Jun-2011	72.3681	126.4738	
44	close to Samoylov Island	1-Jul-2011	72.3681	126.4738	
45	close to Samoylov Island	1-Jul-2011	72.3681	126.4738	
46	close to Samoylov Island	2-Jul-2011	72.3681	126.4738	
47	close to Samoylov Island	2-Jul-2011	72.3681	126.4738	~~n.a.~~

Table 2. Particulate organic carbon (POC) contents in Lena Delta surface water (2009 to 2011) given in milligram per liter (mg/L) and percent based on sediment dry weight (wt%) as well as atomic particulate organic carbon (POC) to particulate total nitrogen (PN) ratios. Data on individual samples can be found in the supplement (Table S4). Note that for Aug 2009 there are only n=20 samples for POC (wt%), because the total suspended matter concentration was not determined for sample 19.

		POC [mg/L]	POC [wt%]	atomic POC:PN
Lena Delta TSM Aug 2009		*n=21*	*n=20*	*n=21*
	mean	1.21	7.2	9.6
	median	0.83	4.7	9.2
	min	0.35	1.9	6.8
	max	7.24	37.7	19.3
Lena Delta TSM Jul/Aug 2010		*n=13*	*n=13*	*n=13*
	mean	0.57	3.05	7.6
	median	0.47	3.05	7.8
	min	0.15	1.42	3.7
	max	1.30	4.74	10.3
Lena Delta TSM late May 2011				
sample code:	37	8.20	1.66	7.5
Lena Delta TSM late Jun/early Jul 2011		*n=9*	*n=9*	*n=9*
	mean	0.74	4.32	7.8
	median	0.69	4.61	7.8
	min	0.29	3.20	5.9
	max	1.51	4.99	9.7

Table 3. Stable carbon isotope (δ^{13}C) and radiocarbon composition (^{14}C) of Lena Delta first terrace soil~~bluff~~ profiles and surface water particulate organic matter (2009-2011). Soil~~Bluff~~ profile samples are given in meters below ~~bluff~~ surface (m b.s.). Not determined is denoted by *n.d.*

Sample code	δ^{13}C [‰ VPDB]	*fMC*[1]	1σ *fMC*	Δ^{14}C [‰]	conv. ^{14}C age[2] [yrs BP]	1σ ^{14}C age [yrs BP]	Lab ID
Lena Delta first terrace soils							
L09-12, 0.45m b.s.	-26.8	0.8084	0.0039	-197	1710	40	OS-84097
L09-12, 1.35m b.s.	-26.3	0.7311	0.0029	-274	2510	30	OS-84073
L09-12, 2.50m b.s.	-25.2	0.7023	0.0025	-303	2840	30	OS-84072
L09-12, 4.70m b.s.	-27.0	0.5708	0.0024	-433	4500	35	OS-84071
L09-12, 5.80m b.s.	- 25.1	0.6275	0.0027	-377	3740	35	OS-84070
L09-28, 030m b.s.	-26.1	0.8015	0.0026	-204	1780	25	OS-84074
L09-28, 070m b.s.	-26.6	0.5430	0.0028	-461	4900	40	OS-84087
Lena Delta Aug 2009							
1	-30.5	0.7436	0.0029	-262	2380	30	OS-84096
2	-32.6	0.8173	0.0034	-189	1620	35	OS-84090
3	-30.9	0.8735	0.0031	-133	1090	30	OS-84093
4	-29.6	0.8259	0.0029	-180	1540	25	OS-84091
5	-31.3	0.8717	0.0031	-134	1100	30	OS-84098
6	-30.5	0.8524	0.0032	-154	1280	30	OS-84127
10	-29.8	0.8419	0.0032	-164	1380	30	OS-84101
11	-28.9	0.8458	0.0031	-160	1340	30	OS-84100
12	-30.6	0.8913	0.0031	-115	925	25	OS-84102
13	-29.9	0.8672	0.0036	-139	1140	30	OS-84133
14	-28.8	0.7971	0.0031	-209	1820	30	OS-84099
19	-34.2	0.9522	0.0042	-55	395	35	OS-84086
21	-27.1	0.8210	0.0028	-185	1580	25	OS-84088
mean	-30.4	0.8462	0.0032	-160	1353	30	
standard deviation	1.7	0.0480	0.0003	48	457	3	
Lena Delta TSM Jul/Aug 2010							
22	-29.7	0.8344	0.0029	-172	1450	30	OS-95088
24	-29.4	0.8201	0.0034	-186	1590	35	OS-95100
25	-28.3	0.8288	0.0030	-177	1510	30	OS-95266
26	-28.9	0.8001	0.0028	-206	1790	30	OS-95382

27	-28.9	0.8386	0.0034	-167	1410	30	OS-95268
28	-28.8	0.8046	0.0035	-201	1750	35	OS-95380
29	-28.9	0.8353	0.0117	-171	1440	110	OS-94853
30	-30.2	0.8389	0.0033	-167	1410	30	OS-95238
31	-28.6	0.6139	0.0021	-391	3920	25	OS-95239
32	-29.6	0.8390	0.0033	-167	1410	30	OS-95267
33	-29.8	0.8125	0.0144	-193	1670	140	OS-94857
35	*n.d.*	0.8299	0.0035	-176	1500	35	OS-95377
36	-30.4	0.8628	0.0150	-143	1180	140	OS-94858
mean	-29.3	0.8122	0.0056	-194	1695	54	
standard deviation	0.6	0.0594	0.0045	59	661	42	
Lena Delta TSM late May 2011							
37	-26.5	0.6988	0.0028	-306	2880	30	OS-94760
Lena Delta TSM late June/early July 2011							
38	-28.3	0.8623	0.0030	-144	1190	30	OS-95378
42	-29.3	0.8558	0.0033	-151	1250	30	OS-95384
47	-28.5	0.8519	0.0117	-154	1290	110	OS-94854
mean	-28.7	0.8567	0.0060	-150	1243	57	
standard deviation	0.4	0.0043	0.0040	4	41	38	

[1] *fMC*=fraction modern carbon, [2] conv. age=conventional radiocarbon age in years before present [yrs BP], i.e. before 1950

Table 4. End-member values used for the binary mixing models based on POC:PN and $\delta^{13}C$, and for dual-carbon isotope three end-member mixing model. If not directly taken from the literature, individual values from the literature and this study used to calculate the end-members can be found in Tables S1-S3 in the supplement.

	Scenario 1 POC:PN		Scenario 2 $\delta^{13}C$		Dual-carbon isotope three end-member		
	riverine phytoplankton	soil	riverine phytoplankton	soil	riverine phytoplankton	Holocene soils	Ice Complex deposits
POC:PN	6 ±1[1]	23.7 ± 11	–	–	–	–	–
$\delta^{13}C$ [‰ vs. VPDB]	–	–	30.5 ± 2.5	–26.6 ± 1.0	30.5 ± 2.5	26.6 ± 1.0	–26.3 ± 0.67[3]
$\Delta^{14}C$ [‰]	–	–	–	–	41.9 ± 4.2[2]	–282 ± 133	–940 ± 84[3]

[1] Meyers (1994) and references therein

[2] average and standard deviation of atmospheric $\Delta^{14}C$ values from May-Aug 2009-2011 from Levin et al. (2013)

[3] from Vonk et al. (2012)

Table ~~4[2].~~5 Soil fractions calculated based on POC:PN ratios and $\delta^{13}C$ values and associated estimated ^{14}C concentrations of the soil-derived POM fraction. Soil fractions are given with standard deviation (1σ). $\Delta^{14}C$ values are given with lower quantile (q16) and upper quantile (q84) representing the 68% confidence interval (1σ for normal distributions). $\Delta^{14}C$ values lower than -1000‰ were marked as 'fossil'. Not determined denoted by *n.d.*

	POC:PN based estimates					plankton $\delta^{13}C$-based estimates				
sample code	fraction soil $POM_{POC:PN}$	$\Delta^{14}C_{POC:PN}$ [‰]	$\Delta^{14}C_{POC:PN}$ q16 [‰]	$\Delta^{14}C_{POC:PN}$ q84 [‰]	conv. $^{14}C_{POC:PN}$ age [yrs BP]	fraction soil $POM_{\delta13C}$	$\Delta^{14}C_{\delta13C}$ [‰]	$\Delta^{14}C_{\delta13C}$ q16 [‰]	$\Delta^{14}C_{\delta13C}$ q84 [‰]	conv. $^{14}C_{\delta13C}$ age [yrs BP]
Lena Delta TSM Aug 2009										
1	0.33 ± 0.18	–884	fossil	–550	17,250	0.31 ± 017	–944	fossil	–585	23,100
2	0.32 ± 0.18	–688	fossil	–416	9,300	0.17 ± 0.12	fossil	–	–	fossil
3	0.30 ± 0.18	–535	fossil	–313	6,100	0.28 ± 0.16	–591	fossil	–354	7,120
4	0.26 ± 0.17	–811	fossil	–465	13,320	0.40 ± 0.20	–511	–967	–327	5,690
5	0.31 ±0.18	–528	fossil	–314	5,970	0.24 ± 0.15	–681	fossil	–399	9,120
6	0.36 ±0.19	–506	–998	–492	5,600	0.31 ± 0.17	–594	fossil	–363	7,180
7	0.27 ± 0.18	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
8	0.29 ± 0.18	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
9	0.38 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
10	0.38 ± 0.19	–506	–985	–322	5,600	0.38 ± 0.19	–500	–953	–317	5,510
11	0.32 ± 0.18	-595	fossil	–359	7,200	0.50 ± 0.21	–364	–655	–245	3,580
12	0.42 ± 0.20	–332	–625	–213	3,190	0.30 ± 0.17	–483	–981	–291	5,240
13	0.43 ± 0.20	–382	–709	–249	3,810	0.37 ± 0.19	–451	–899	–282	4,760
14	0.34 ± 0.19	–704	fossil	–434	9,720	0.51 ± 0.22	–447	–793	–307	4,700
15	0.38 ± 0.20	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
16	0.70 ± 0.16	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
17	0.36 ± 0.18	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
18	0.38 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
19	0.27 ± 0.17	–322	–718	–174	3,060	0.12 ± 0.09	–774	fossil	–407	11,890
20	0.38 ± 0.20	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
21	0.30 ± 0.20	–724	fossil	–413	10,280	0.75 ± 0.20	–261	–377	–214	2,370
Lena Delta TSM Jul/Aug 2010										
22	0.30 ± 0.18	–675	fossil	–402	8,970	0.39 ± 0.20	–505	–976	–322	5,590
24	0.30 ± 0.18	–708	fossil	–424	9810	0.43 ± 0.20	–491	–914	–320	5,370
25	0.37 ± 0.19	–544	fossil	–349	5100	0.59 ± 0.22	–330	–553	–237	3,160
26	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	0.50 ± 0.21	–456	–807	–308	4,830
27	0.44 ± 0.20	–431	–787	–284	3760	0.50 ± 0.21	–456	–679	–256	3,780
28	0.41 ± 0.20	–544	fossil	–355	5360	0.51 ± 0.21	–380	–756	–296	4,490
29	0.35 ± 0.18	–570	fossil	–353	5890	0.50 ± 0.21	–384	–690	–258	3,830
30	0.26 ± 0.17	–764	fossil	–437	11700	0.34 ± 0.18	–581	fossil	–360	6,930
31	0.31 ± 0.18	fossil	–	–	fossil	0.54 ± 0.22	–754	fossil	–536	11,210
32	0.34 ± 0.19	–564	fossil	–349	6190	0.40 ± 0.20	–476	–903	–306	5,130
33	0.32 ± 0.18	–694	fossil	–419	8890	0.38 ± 0.19	–579	fossil	–366	6,890

34	0.36 ± 0.21	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
35	0.39 ± 0.20	515	–997	–329	5190	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
36	0.12 ± 0.13	fossil			*n.d.*	0.34 ± 0.18	–510	fossil	–312	5,670
Lena Delta TSM late May 2011										
37	0.13 ± 0.13	fossil			*n.d.*	0.80 ± 0.19	–393	–539	–341	3,950
Lena Delta TSM late Jun/early Jul 2011										
38	0.43 ± 0.19	–391	–704	–258	2870	0.59 ± 0.22	–274	–460	–196	2,510
39	0.50 ± 0.20	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
41	0.39 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
42	0.41 ± 0.19	–429	–798	–280	3340	0.44 ± 0.21	–394	–738	–256	3,970
43	0.43 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
44	0.41 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
45	0.43 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
46	0.44 ± 0.19	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*	*n.d.*
47	0.40 ± 0.19	–451	–848	–290	3620	0.56 ± 0.22	–310	–534	–215	2,920

Figure captions

Figure 1. A) Lena River catchment area within the northern hemisphere permafrost zone (map by Hugo Ahlenius, UNEP/GRID-Arendal, see www.grida.no/graphicslib/detail/permafrost-extent-in-the-northern-hemisphere_1266; source data from Brown et al. 1998), B) Lena Delta and Buor Khaya Bay sampling sites from 2009 to 2011 analyzed for radiocarbon content (see also table 1).

Figure 2. Spatial distribution of organic carbon concentrations, POC:PN ratios and stable and radiocarbon isotopic composition in surface water total suspended matter (TSM) samples from 2009-2011. TSM concentrations of the years 2009 to late May 2011 (panels A-C) taken from Winterfeld et al. (2015, companion paper~~4~~)~~, submitted as companion paper~~.

Figure 3~~3~~. Stable and radiocarbon isotopic values of Lena Delta surface water POM of A) this study and literature data as well as B) estimated isotopic data of soil-derived POM based on POC:PN and $\delta^{13}C$ ratios. Ranges for different end-members for topsoil, ice complex, and marine are taken from the literature.

Figure 4. Results of dual carbon-isotope ($\delta^{13}C$, $\Delta^{14}C$) three end-member mixing model showing the fractions for riverine phytoplankton, Holocene soils, and Pleistocene Ice Complex deposits contributing to individual surface water POM samples. The end-member used in the model can be found in Table 4. Individual values comprising the Holocene soil $\delta^{13}C$ and $\Delta^{14}C$ end-member are given in Tables S2 and S3 in the supplement.