We would like to thank Alberto Reyes for his comments provided to help us improve our manuscript. Please find below our responses to these comments and the manuscript changes.

1-Comment from the Editor

Your revised manuscript has now been reviewed by one of the original referees. I agree with the referee that your revisions have resulted in tighter manuscript with improved clarity.

As you can see, the referee provided in-depth and constructive comments regarding interpretation of the TAR data and has recommended major revision. I would like you to consider these comments carefully and submitted another revised manuscript that addresses these concerns. Referee 2 will review your revised submission one more time.

Reviewer 1 (Anders Carlson) asked that you incorporate acknowledgement of the terrestrial record of Cordilleran ice sheet glaciation. It's a good start to include a citation to the 2013 Hidy paper, but this insight (ie. extensive CIS far inland – in fact, the most extensive CIS advances in interior Yukon are the earliest ones) could be better incorporated in the penultimate paragraph of the introduction (lines 15-24, page 2) and likely also in section 4.3.

2-Authors' response

Thank you for your comments. We agree that our manuscript could benefit from a tighter connection between the signs of the Cordilleran glaciation in terrestrial records and our marine record at Site U1417.

3-Authors' changes in the revised manuscript

We have briefly mentioned the timing of the expansion of the Cordilleran glaciation according to the terrestrial records in Hidy et al., 2013 and Duk-Rodkin et al., 2004 (page 2, lines 15 to 17) together with other previous studies of the timing of the Cordilleran glaciation in marine records in Gulick et al., 2014 (page 2, line 18) in our introduction. We have further mentioned these terrestrial records in Section 4.3 (page 12, lines 4 to 7).

1-Comment from the Editor

Please also consider making some reference to the well established stratigraphic, geomorphic, and paleomag record for extensive inland CIS glaciation during the late Pliocene/earliest Pleistocene – you could refer to Duk-Rodkin et al 2004 (Dev. Quat. Sci 2B: 313-345) as a starting point. I think this would strengthen the connection between your relatively distal marine record and the less-well-dated direct terrestrial evidence for ice sheet extent.

2-Authors' response

Thank you for your comments. We agree that previous studies of terrestrial records might strengthen our interpretations of the behaviour of the Cordilleran Ice Sheet during the early and mid-Pliocene and late Pliocene.

3-Authors' changes in the revised manuscript

We have included the valuable insights about the Cordilleran Ice Sheet expansion in Duk-Rodkin et al. (2004) study into our early and mid-Pliocene TAR interpretations (page 10, lines 30 to 32). We have also included Duk-Rodkin et al. (2004)'s interpretations of the development of fan deltas due to coastal uplift to support our TAR interpretations during the late Pliocene (page 11, lines 12 to 14). We have also compared the dates of the oNHG and iNHG in our late Pliocene sections with the Duk-Rodkin et al. (2004) and related papers' timing of the major glaciation in Yukon (page 11, lines 10 to 12; page 12, lines 5 to7).

1-Comment from the Editor

Thank you again for submitting this very interesting and important manuscript to Climate of the Past. I look forward to seeing your revised submission.

2-Authors' response

Thank you.

We would like to thank Anonymous Referee 2 for the constructive comments provided to help us improve our manuscript. Please find below our responses to these comments and the manuscript changes.

1-Comment from Referee

In my first review I stated to major points of criticism, i.e. the presentation of the TAR and CPI indices and the confusing presentation of section 4.4. The authors addressed both points so that the clarity of the entire discussion greatly improved overall. The new section 4.4 is much clearer than before and the authors' inferences are easy to follow, now. So, I don't have further complaints here.

2-Authors' response

Thank you.

3-Authors' changes in the revised manuscript

Please note that have further altered Figure 3 to show the records after applying a 1kyr linear interpolation and 100kyr smoothing (rather than the previous approximate 100kyr smoothing directly applied) to the raw data. Due to the continuity of the data analyses, we have represented the data as continuous lines. We have further included the atmospheric CO2 record from marine δ 11B of Martínez-Botí et al., (2015) to the atmospheric CO2 record from alkenone δ 13C of Seki et al., (2010) (revised in Foster et al., 2017) to represent the agreement/disparity between different atmospheric CO2 reconstructions across the NHG.

1-Comment from Referee

However, I still have some concerns regarding parts of their interpretation of the TAR index which partly arise from having the CPI plotted next to the TAR. At the present stage, the inferences made from the TAR during the oNHG (3-2.8 Ma) are questionable and require further clarification by discussing the different potential factors controlling the TAR in more detail (see below). This is crucial to strengthen the key conclusions that the onset of the Cordilleran Ice Sheet glaciation occurred during this time interval. I am convinced, a more comprehensive discussion around the different factors controlling the TAR will unequivocally strengthen the inferences made from this multi-proxy approach. As already mentioned in my first review, presenting a multi-proxy approach the study is a valuable and important contribution to the ongoing discussion about the glaciation chronology in the Cordillera during the Plio-Pleistocene transition. The paper should be accepted for publication as soon as this last issue regarding the TAR has been addressed.

2-Authors' response

Thank you for your comments. We agree that a further clarification of the TAR would improve the manuscript. Please see the changes we have implemented in our comment below.

1-Comment from Referee

Interpretation of TAR during the oNHG, 3-2.8 Ma (page 10, line 40-page 11, line 5):

As the authors correctly describe in the methods section, the TAR index estimates the relative abundance of long-chain n-alkanes versus the short chain homologues and is commonly used to estimate changes in the relative contributions of plant-wax lipids from higher land plants and aquatic production (page 5, lines 4-7). In the discussion of the onset of the Cordilleran Ice Sheet glaciation (3 to 2.8 Ma) the authors infer an increase in the export of plant-derived organic matter based on a decrease in the TAR-values (page 10, line 40 to page 11, line 5). In fact, this interpretation is contradictory to the general way of interpreting the TAR according to which decreasing values would imply the opposite i.e. an increase in aquatic production and/or a decrease in the contributions from higher land plants. Although I don't doubt the possibility that the export of terrigenous material including the long-chain n-alkanes may increase despite a decrease in the TAR, this interpretation needs further justification and cannot be based on the TAR alone. Concentrations or mass accumulation rates of the shortchain and long-chain n-alkanes are needed to support this statement since they allow to disentangle the individual developments of the input of short chain and long-chain n-alkanes. It may help to compare them to concentrations of other marine biomarkers, e.g. alkenones. Moreover, there are several possibilities to explain the decrease in the TAR-index and not all of them support expanding glaciation in the hinterland. Based on the current presentation of data, the authors don't have a means to rule these options out. Firstly, there could be an increase in aquatic production, which does not necessarily mean that the controls on the export of leaf-wax biomarkers change. That means it would be possible that the vegetation cover

remains as extensive as before (during the early to mid-Pliocene) which would be in conflict with the authors' inference of advancing ice cover in the region. Secondly, assuming that the

source of the long-chain n-alkanes remains the same as during the early to mid-Pliocene, the decreasing TAR may report on a decline in the export of terrigenous organic matter attesting to e.g. a reduction of the vegetation cover. Although this view would be in harmony with advancing ice masses in the hinterland it would be in conflict with the authors' idea of enhanced discharge of leaf-wax lipids from vegetation. Thirdly, the overall low CPI-values (around 1.5) point to a high degree of degradation. So, it is plausible that a large fraction of the long-chain n-alkanes may be petrogenic instead of dominantly vegetation-derived. As the authors point out, there are coal-bearing bedrocks in the region and these are characterized by low TAR (up to 2) and low CPI-values (<1) (page 10, lines 25-28 and page 12, lines 25/26).

As such, the decrease in the TAR may also indicate intensified erosion of bedrock compared to the early and mid-Pliocene. Under these circumstances, decreasing TAR would be compatible with increased export of terrigenous organic matter. This scenario would be in accordance with the idea of reduced vegetation cover and expanding ice masses in the Cordillera.

Since the inferences for the Ice Sheet Glaciation strongly depend on the way of interpreting the TAR, the different potential controls on the index need a more detailed recognition in the discussion in order to strengthen the authors' key conclusions, namely that the onset of the Cordilleran Ice Sheet Glaciation occurred between 3 and 2.8 Ma.

2-Authors' response

Thank you for your comments. We agree that the TAR during the 3 to 2.8 Ma needs clarifying. The confusing sentence (page 10, line 40 to page 11, line 5-now clarified in page 11, lines 1 to 4) arose because we intended to outline a change of TAR pattern from above the TAR average value before 3 Ma to below the TAR average value from 3 Ma onwards (into the Early Pleistocene). Thank you for drawing attention to the confusing statements.

3-Authors' changes in the revised manuscript

We have addressed this issue by explaining first the TAR during the 3-2.8 Ma (page 10, line 41 and page 11, lines 1 to 10) and then during the 2.7-1.7 Ma time period in Section 4.3 (page 12, lines 22 to 28). To help visualising the discussion behind the TAR, we have included Table 2. Table 2 includes the above or below 4-1.7 average TAR at Site U1417 and terrigenous and aquatic n-alkane mass accumulation rates during the time periods discussed in the text. TAR lowers during the 3-2.8 Ma due to a slight decrease in terrigenous organic matter and a slight increase in aquatic organic matter (page 11, lines 1 to 4). We interpret this as an increase in ice-cover and lower plant wax contribution to the mix of sources of terrigenous OM eroded and transported to Site U1417, despite the overall increase in sedimentation rates (page 11, lines 8 to 10). The increase in sedimentation rates could be related to a change in erosional pathways (i.e., to a softer bedrock) but because our CPI remain similar than during the 4-3 Ma, we interpret rather an increase in erosion due to an advancing ice sheet (page 11, lines 5 to 8). This interpretation also fits well with previously published literature (page 11, lines 12 to 14).

Detailed comments

1-Comment from Referee

Page 5, line 9: remove the sentence in line 9. This seems to be out of context here in the method section. The following paragraph encompassing lines 10-16 stands well by itself.

2-Authors' response and changes in the revised manuscript

This sentence has been modified and moved to Section 4.3 (page 12, lines 22 to 24).

1-Comment from Referee

Page 10, line 20: n-alkane => n-alkane

2-Authors' response and changes in the revised manuscript

This has been modified (page 10, line 20).

1-Comment from Referee

Page 12, line 12-13: Actually, the CPI cannot attest to the maturity of the short-chain alkanes as the formula used here does not consider the homologues shorter than C24.

2-Authors' response and changes in the revised manuscript

This has been now amended and CPI only refers to the maturity of terrigenous OM (page 12, lines 26 to 28).

1-Comment from Referee

The interpretation of the TAR should be justified a little more in detail considering the different interpretation of a similar signal for the period from 3-2.8 Ma. It is not clear why the low TAR values are interpreted as stemming from increased productivity from 2.7 Ma onwards while between 3-2.8 Ma the progressive decrease towards these low values is attributed to an increased export of leaf-wax lipids from land plants. Please, explain.

2-Authors' response and changes in the revised manuscript

This has now been amended (page 10, line 41 and page 12, lines 1 to 14). Table 2 has been included (Page 25).

1-Comment from Referee

Page 2, lines 17 and 20: In line 17 it says St. Elias Mountains while in line 20 it is St Elias Mountains. Please, unify the spelling throughout the entire manuscript.

2-Authors' response and changes in the revised manuscript

This has now been unified to read "St. Elias mountains" throughout the manuscript.

Late Pliocene Cordilleran Ice Sheet development with warm Northeast Pacific sea surface temperatures

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Abstract. The initiation and evolution of the Cordilleran Ice Sheet is relatively poorly constrained. International Ocean Discovery Program (IODP) Expedition 341 recovered marine sediments at Site U1417 in the Gulf of Alaska (GOA). Here we present alkenone-derived sea surface temperature (SST) analyses alongside ice rafted debris (IRD), terrigenous and marine organic matter inputs to the GOA through the late Pliocene and early Pleistocene. The first IRD contribution from tidewater glaciers in southwest Alaska is recorded at 2.9 Ma, indicating that the Cordilleran ice sheet extent increased in the late Pliocene.

- A higher occurrence of IRD and higher sedimentation rates in the GOA during the early Pleistocene, at 2.5 Ma, occur in synchrony with SSTs warming on the order of 1°C relative to the Pliocene. All records show a high degree of variability in the early Pleistocene, indicating highly efficient ocean-climate-ice interactions through warm SST-ocean evaporation-
- 20 orographic precipitation-ice growth mechanisms. A climatic shift towards ocean circulation in the subarctic Pacific similar to the pattern observed during negative Pacific Decadal Oscillation (PDO) conditions today <u>occurs with development of more extensive Cordilleran glaciation, and may have played a role through increased moisture supply to the subarctic Pacificappears to be a necessary pre-requisite to develop the Cordilleran glaciation and increase moisture supply to the subarctic Pacific. The drop in atmospheric CO₂ concentrations since 2.8 Ma is suggested as one of the main forcing mechanisms driving the 25 Cordilleran glaciation.</u>
 - 1 Introduction

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During the Neogene, the global climate transitioned from relatively warm to cooler conditions that enabled the development of ice masses in both hemispheres (Zachos *et al.*, 2001a). The Mid-Piacenzian Warm Period (MPWP, 3.3-3.0 Ma) interrupts this cooling trend, with global temperatures around 2-3 °C above pre-industrial levels (Jansen *et al.*, 2007; Haywood *et al.*, 2004), and more intense warming at higher latitudes (Haywood *et al.*, 2013; Dolan *et al.*, 2015). The MPWP has been suggested

as a potential analogue for the 21^{st} century climate due to the atmospheric CO₂ concentrations (400 ppmv) and largely equivalent continental configurations relative to the present (Salzmann *et al.*, 2011; Raymo *et al.*, 1996, Jansen *et al.*, 2007). Overall, the mid-Pliocene ice masses were smaller than today (Dolan *et al.*, 2011). However, the marine isotope stage (MIS) M2 (~3.3-3.26 Ma) event is characterised by a dramatic cooling in the Atlantic Ocean and is considered to be an unsuccessful

- 5 attempt at a <u>Pleistocene-style glaciation</u> (De Schepper *et al.*, 2013). The later onset (oNHG) or intensification (iNHG) of the Northern Hemisphere Glaciation is marked by the expansion of the Laurentide, Greenland and Scandinavian ice sheets around 2.5 Ma, as indicated by ice rafted debris (IRD) records from the North Atlantic Ocean (i.e. Shackleton *et al.*, 1984) and the advance of the Cordilleran Ice Sheet at 2.7 Ma inferred from a terrestrial record (Hidy *et al.*, 2013). It is still debated whether climatic or tectonic forcing was the main driver of the North Hemisphere Glaciation (NHG) (Haug *et al.*, 2005), as it cannot
- 10 be explained solely by changes in isolation (Lunt *et al.*, 2008). The decrease in atmospheric CO₂ concentrations and radiative forcing at 2.8 Ma has been identified as a potential mechanism for climate cooling of at the oNHG (Seki *et al.*, 2010; Martínez-Botí *et al.*, 2015). However, the timing of the oNHG varies between locations based on IRD delivery, and at some locations the oNHG has been set as far back as 3.5 Ma (Nordic Seas, Mudelsee and Raymo, 2005). Alternative proposals for the oNHG suggest that orogenic changes could have led to an increase in heat transport to the North Atlantic region during the Pliocene.
- 15 potentially increasing precipitation in higher latitudes and promoting glacial development during the Plio-Pleistocene transition (Sarnthein *et al*, 2013; Haug *et al.*, 2005; Bringham-Grette *et al.*, 2013; Fedorov *et al.*, 2013; Lawrence *et al.*, 2010). It remains unclear whether the There is evidence that the Cordilleran Ice Sheet of North America expanded across the period of the oNHG₅ although despite previous studies of as This is indicated by glacial deposits in terrestrial records which date the Cordilleran expansion between 2.9 to 2.6 Ma (i.e. Hidy *et al.*, 2013; Duk-Rodkin *et al*, 2004), and the enhanced delivery of
- 20 terrigenous sediments (IRD) to the Gulf of Alaska (GOA; Northeast Pacific Ocean) (Site 887) since 2.7-6 Ma, has been interpreted as evidence for ice sheet growth (Gulick *et al.*, 2014). The sediments of the Gulf of Alaska (GOA) record Cordilleran glaciation in the St. Elias mMountains, at present the highest coastal mountain range in the world (Enkelmann *et al.*, 2015). It has been proposed that the uplift of the St. Elias mountainsRange from the early Pliocene to early Pleistocene led to an increase in orographic precipitation and a subsequent increase in sedimentation rates in the GOA (Enkelmann *et al.*, 2015).
- 25 2015). Mountain glaciation may have developed in the St Elias <u>mmountains</u> as early as 5.5 Ma (Reece *et al.*, 2011), ultimately developing tidewater glaciers, with the high erosion pathway shifting to the southern St Elias <u>mountains_Range_at 2.6 Ma</u> (Enkelmann *et al.*, 2015). Rather than a tectonic control on Cordilleran glaciation, an alternative explanation could be the reduced radiative forcing and climate cooling associated with the decline in <u>atmospheric</u> CO₂ at 2.8 Ma. However, it is difficult to resolve these hypotheses in the absence of high resolution data for both ice sheet extent and climate from the GOA. Despite
- 30 the global drop in atmospheric CO₂ at 2.8 Ma, it remains unclear whether the Cordilleran Ice Sheet also expanded. Here, we present a new multiproxy data set obtained from IODP core site U1417 (56° 57.58' N, 147° 6.58' W, water depth 4218 m; Fig. 1) in the GOA. The core site allows examination of the land-ocean interactions associated with advance and retreat phases of the Cordilleran Ice Sheet across the Pliocene-Pleistocene transition₇ in the context of mountain uplift. The sediments were collected during IODP Expedition 341 (Jaeger *et al.*, 2014) and were analysed for the time interval from 4 to

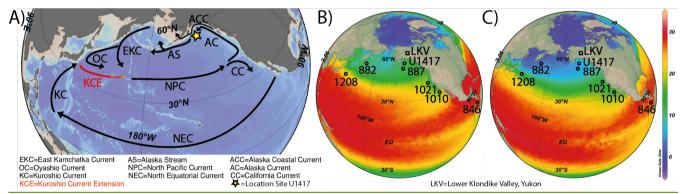
<u>1.7 Ma years ago</u> to reconstruct sea surface conditions <u>using alkenone proxies</u>, by means of alkenone <u>and inputs and properties</u> of IRD-data, and <u>covering the time interval from 4 to 1.7 Ma years ago</u>. T<u>t</u>errestrial organic matter input <u>using to Site U1417</u> is assessed through the abundance of long-chain *n*-alkanes and palynological analysis.

2 Study area

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5 2.1 The Gulf of Alaska (GOA)

The GOA extends from the Alaska Peninsula in the west to the Alexander Archipelago in the east (Hogan, 2013), delimited by the Bering Sea on the west and the Alaska coast in the north and east, which is, in turn, bounded to the north by the Pacific Mountain System (Molnia, 2008). The south of the GOA connects with the North Pacific Ocean (Fig. 1). Glaciers cover 20% of the Gulf of Alaska watershed (Spies, 2007), and the major rivers draining the St. Elias and Chugach mountains towards the GOA (the Alsek River and the Copper River), are fed by meltwater discharge which peaks in August (Weingartner, 2007). The GOA mean annual freshwater discharge derives from high precipitation, runoff and snow melt from watersheds along the SE Alaskan coast (Spies, 2007). High precipitation is due in part to the proximity of the North Pacific Ocean, as a source of moisture, and the high topography of the Pacific Mountain System driving orographic precipitation.



15 Figure 1: Map of modern ocean circulation and SSTs. a) Modern North Pacific Ocean circulation, b) September c) and December 1955-2013 SST average centred in the North Pacific Ocean (NOAA WOA13, Locarnini *et al.*, 2013) and core and sample sites discussed in this study. Map made using Ocean Data View (Schlitzer, 2018).

The Alaskan Coastal Current (ACC) flows anti-clockwise along the GOA coastline and westward to the Bering Sea (Fig. 1a), and its properties are dominated by nutrient and meltwater supply from the coastal Alaskan glaciers (Spies, 2007). Further offshore, the Alaska Current (AC) also flows anti-clockwise, controlled in strength by the Alaska Gyre (Kato *et al.*, 2016) (Fig. 1a). The location of Site U1417 rests under the modern influence of the AC (Fig. 1). The Alaskan Gyre is, in turn, influenced by atmospheric circulation via the Aleutian Low (AL) and the Pacific High Pressure Systems, which are coupled in an annual cycle. High pressures dominate during the summer season and low pressures dominate during autumn to spring (Hogan, 2013), when the AL also migrates eastward across the North Pacific Ocean, becoming most intense when located in

25 the GOA during winter (Pickart et al., 2009). The coast of Alaska receives high winter precipitation because of the AL winter

position and strength (Rodinov *et al.*, 2007) and Alaska's high topography which drives orographic precipitation. The GOA locally receives annual precipitation of ~800 cm (Powell and Molnia, 1989). During summer, the AL is less intense and almost disappears when it is located in the Bering Sea. A weaker AL is translated into reduced precipitation over the GOA. A strong winter AL also creates a strong zonal SST gradient in the North Pacific Ocean (Fig. 1b). During winter, the ocean

5 responds to a more intense AL through southward movement of the cold Arctic waters, and northward flow of mid-latitude warm waters into the Gulf of Alaska through the AC. During the summer migration of the AL northwards, the GOA registers higher SSTs due to higher insolation on the North Pacific Ocean, and as the zonal SST gradient is reduced, the storms diminish (Pickart *et al.*, 2009) (Fig. 1c).

3 Material and Methods

10 **3.1 Age model and sedimentation rates**

The shipboard age model was calculated using magnetostratigraphy (Jaeger *et al.*, 2014, Fig. S1-3). The recovery of the Pliocene-early Pleistocene sediments averaged 70 % (Expedition 341 Scientists, 2014), with a number of core breaks in the record. Poor carbonate preservation across the Pliocene and early Pleistocene prevents the production of a higher resolution stable isotope stratigraphy. The shipboard depth models place all discrete core biscuits to the upper depth range of each core, and a continuous core break below; it is possible that the biscuits were originally distributed through the core barrel before recovery on the ship. We have converted the depth scale of our data sets to assume an even distribution of core biscuits and

(Fig. 2 and Figs. S1-S3). The magnetostratigraphy ages were similar between the shipboard and new age model; The Gauss/Matuyama magnetic reversal (2.581 \pm 0.02 Ma and 330.76 \pm 1 m CCSF-A) was well constrained in multiple holes to

core breaks (Fig. S1), converted these depths to age and interpolated the ages of the samples between core top and bottom

20 provide an important age control point for this study (Fig. S1). The shipboard age model sedimentation rates show a marked but temporary increase between 2.5-2.0 Ma, which has been attributed to the first major erosion of the landscape by expansion of the Cordilleran Ice Sheet (Gulick *et al.*, 2014). Our new sedimentation rates detail a two-step increase from 2.5-2.4 <u>Ma</u> and from 2.4-2.0 Ma (Fig. S3).

3.2 Biomarkers

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- A total of 119-biomarker samples between 4 and 1.7 Ma were analysed for biomarkers, which corresponds to an average sampling resolution of 19 kyr. Microwave lipid biomarker extraction of freeze-dried and homogenized sediment was carried out following the method of Kornilova and Rosell-Melé (2003). The total lipid extract was separated into 4 fractions by silica column chromatography, through sequential elution with <u>h</u>Hexane (3 ml), <u>h</u>Hexane: <u>d</u>Dichloromethane (9:1)-(;_1.5 ml), <u>Dichloromethane-dichloromethane (5.5 ml) and Ethylacetateethylacetate</u>:Hexane-hexane (20:80)-(;_4 columns) to generate
- 30 <u>obtain</u>: *n* -alkanes, aromatics, ketone and polar fractions.

The *n*-alkane fraction was analysed by different sets of gas chromatography (GC) configurations for compound quantification and identification. A Thermo Scientific Trace 1310 gas chromatograph was fitted with flame ionization detector (GC-FID) and a split-splitless injector. Compressed air is set as the air flow, helium (He) is set as the carrier flow, nitrogen (N) as a makeup flow and hydrogen (H) helps with ignition. The oven temperature was set at 70 °C for 2 min, then increased to 170 °C at

- 5 12 °C min⁻¹, then increased to 310 °C at 6.0 °C min⁻¹, then held at 310 °C for 35 min. *N*-alkanes were separated using a 60 m x 0.25 mm i.d., Restek RXi-5ms column. (0.25 μm 5% diphenyl-95% dimethyl polysiloxane coating). Compound identification was confirmed using a Thermo Scientific Trace 1310 gas chromatography mass spectrometer (GC-MS), equipped with a programmable temperature vaporizer (PTV) injector. He-Helium was used as a carrier flow. The oven temperature program was set at 60 °C during-for 2 min and then raised at 12 °C min⁻¹ until reaching 150 °C and then raised
- 10 again to 310 °C at 6 °C min⁻¹ and held for 25 min. Compounds were quantified with reference to internal standards (5α-cholestane) and normalised to the original extracted dry weight of sediment, and to sedimentation rate changes by calculating the mass accumulation rates (MAR). The ratio of higher land-plant derived long-chain *n*-alkanes against aquatic sourced short-chain *n*-alkanes (TAR) (Eq. (1); Cranwell, 1973) and the carbon preference index (CPI) (Eq. (2); Bray and Evans, 1961) were calculated using GC-FID peak areas of the respective compounds:

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$$TAR = \frac{[C27]+[C29]+[C31]}{[C15]+[C17]+[C19]}$$
 (1)

(2)

$$CPI = \frac{\frac{[C23 - 32(even)]}{[C24 - 32(even)]} + \frac{[C25 - 34(even)]}{[C26 - 34(even)]}}{2}$$

Terrigenous and aquatic organic matter sources increase during the early Pleistocene in comparison with the late Pliocene. High TAR values can be indicative of relative increases in terrigenous organic matter transported to the ocean and/or to relative decreases in aquatic microorganism production. The opposite could explain low TAR values.- To disentangle the presence of petrogenic old organic matter contamination from the fresh *n*-alkane signal, we include the CPI index (Bray and Evans, 1961). High CPI values indicate a fresher or relatively newly produced organic matter transported to the ocean. CPI close to 1 indicates mature / petrogenic or old organic matter sources, such as coal or oil deposits, eroded to the ocean. Higher CPI values indicate a fresher or relatively newly produced to the ocean. This distinction may be important in the GOA, where the onshore bedrock includes units with high contents of terrigenous organic matter (e.g. the Yakutat Terrain, Childress, 2016; Walinsky *et al.*, 2009).

Alkenones (ketone fractions) were quantified by a GC coupled with chemical ionisation mass spectrometry (GC-CIMS), adapted from the method of (Rosell-Melé *et al.*, 1995). Analyses were performed using a Trace Ultra gas chromatograph directly coupled to a Thermo DSQ single quadrupole mass spectrometer, fitted with a programmed temperature vaporising

30 (PTV) injector. 1.2 ml of sample is injected. Alkenones were separated using a 30 m x 0.25 mm i.d., Restek RXi-5ms column (0.25 μm 5% diphenyl-95% dimethyl polysiloxane coating). Helium was employed as the carrier gas (2 ml min⁻¹). The injector was held at 120 °C and splitless mode (1.2 min) during injection, and then immediately temperature programmed from 120 °C

to 310 °C at 10 °C s⁻¹, then held for 0.6 min. The oven was programmed to hold at 175 °C for 1.7 min, then increased to 310 °C at 11 min⁻¹, and held at from 310 °C for 12 min. The mass spectrometer was operated in positive chemical ionisation mode (PICI), using high-purity anhydrous ammonia (N6.0, BOC) introduced to the ion source through the CI gas inlet. Selected ion monitoring was performed, targeting the 8 ions corresponding to the $[M + NH4]^+$ adducts of the target C₃₇ and C₃₈ alkenones

5 and the internal standard (2-nonadecanone), each with a selected ion monitoring (SIM) width of 1 m z⁻¹ and a dwell time of 30 min. The target m z⁻¹ were: 300 (2-nonadecanone), 544 (C_{37:4}), 546 (C_{37:3}), 548 (C_{37:2}), as detailed by (Rosell-Melé *et al.*, 1995).

-The alkenone $U^{K_{37}}$ index has been converted into SST according to the core-top to annual mean SST correlation constructed with samples spanning 60° S to 60° N (including from the Pacific Ocean), which withand an which accuracy is beignis

- 10 constrained by an standard error of ±1.5 °C (Eq. (3); Müller *et al.*, 1998). The more recently developed BAYSPLINE SST calibration (Tierney and Tingley, 2018) provides similar SST estimates in the northern latitudes to previous calibrations. <u>SThe seasonality</u> in the alkenone production has been evidenced in the North Pacific (Tierney and Tingley, 2018). The SST calibration of Prahl *et al.* (1988) (Eq. (4)), which includes the C_{37:4} alkenone, is also displayed here for comparison, as some concerns have arisen with the use of the U^K₃₇' index in samples with high C_{37:4} in the Nordic Seas (Bendle *et al.*, 2005). The
- 15 standard error of Prahl *et al.* (1988) (Eq. (4)) is ±1.0 °C. We identify samples with high C_{37:4} by presenting the percentage of C_{37:4} relative to the other C₃₇ alkenones, as %C_{37:4} (Bendle and Rosell-Melé, 2004) (Eq. (5)). The %C_{37:4} represents fresher and cooler surface water characteristics (Bendle *et al.*, 2005). In the Nordic Seas this has been linked to subpolar and polar water masses (Bendle *et al.*, 2005), whereas elsewhere in the North Atlantic it has been linked to freshwater inputs (e.g. during Heinrich events, Martrat *et al.*, 2007). In the subarctic Pacific, the %C_{37:4} proxy has been less well studied (McClymont *et al.*, 2008), but high %C_{37:4} is also proposed to reflect cooler and fresher water masses (Harada *et al.*, 20086).

$$U_{37}^{K'} = \frac{[C_{37:2}]}{[C_{37:2}] + [C_{37:3}]} = 0.033SST - 0.044$$
(3)

$$U_{37}^{K} = \frac{[C_{37:2}] - [C_{37:4}]}{[C_{37:2}] + [C_{37:4}]} = 0.040SST - 0.104$$
(4)

$$\%C_{37:4} = \frac{[C_{37:4}]}{[C_{37:2}] + [C_{37:3}] + [C_{37:4}]} *100$$
(5)

3.3 IRD

- IRD were-was quantified by weighing the coarse sand fraction (2 mm 250 µm⁻²-mm) following the method of Krissek (1995). Coarse sand was separated from 10 cm³ samples by wet sieving after air drying and rinsing with distilled water to remove salts. Each sand sample was examined with a binocular microscope to estimate the volume of terrigenous ice-rafted sediment (in volume percent) in order to exclude biogenic components and burrow fills of manganese and pyrite, which do not have an ice-rafted origin. The volume of terrigenous ice-rafted sediment was estimated in volume percent. The mass accumulation rate
- 30 of IRD (in grams per cm² kyr⁻¹) was calculated as in Eq. (6): IRD MAR = CS% * IRD% * DBD * LSR

(6)

where CS% is the coarse sand abundance (multiplied as a decimal), IRD% is the IRD abundance in the coarse-sand fraction (as a volume ratio), DBD is the dry bulk density of the whole sediment sample (in grams per cm³) determined from discrete shipboard measurements and LSR is the interval average linear sedimentation rates (in cm kyr⁻¹).

- Closed-form Fourier analysis was used to describe the shape of quartz grains in the IRD fraction imaged on a Quanta FEI 200
 Scanning Electron Microscope (in the high vacuum mode at 20 kV) following methods that have been used to describe sedimentary particles for more than 40 years (Ehrlich and Weinberg, 1970; Ehrlich *et al.*, 1980; Dowdeswell, 1982; Livsey *et al.*, 2013). Two-dimensional SEM images (from 200 to 500 X magnification) were input into ImageJ to produce a line trace of the boundary for each grain. The output was inspected to verify that the trace was representative of the grain. 120 xy coordinate points were output from the boundary to represent the grain and these were input into the software program PAST
- 10 (Hammer *et al.*, 2001). Harmonic amplitudes 1-20 were calculated, lower orders (1-10) represent grain shape, a function of provenance and higher order harmonics (11-20) represent grain roundness (Dowdeswell, 1986; Haines and Mazzullo, 1988; Livsey *et al.*, 2013). An average dimensionless roughness coefficient (Rca-b) was calculated for each sample using the harmonics 16-20 for each grain in the population. Higher Rc16-20 values indicate increasing roughness and lower coefficients indicate smoother grains (Dowdeswell, 1982; Livsey *et al.*, 2013). The roughness coefficient is calculated as in Eq. (7):

15
$$Rc_{a-b} = \sqrt{0.5} \sum Rn^2$$
 (7)

Where Rn is the nth harmonic coefficient and a-b is the harmonic range used, in our case 16-20 (Ehrlich and Weinberg, 1970). This value represents the average roundness for the grains in each sample, numbering at least 25.

3.4 Pollen Analysis

Palynological treatments were performed on 13 samples according to the procedure routinely used at GEOTOP (de Vernal *et al.*, 1996). Before sieving and chemical treatments, one *Lycopodium clavatum* spore tablet was added in each sample to estimate palynomorph concentrations (Matthews, 1969; Mertens *et al.*, 2009). Wet sample volumes were measured by water displacement and weighed after being dried. The fraction between 10 and 120 μm was treated chemically to dissolve carbonate and silicate particles with repeated cold HCl (10 %) and HF (48 %). A small drop of the final residue was mounted on a microscope slide with glycerine jelly. Counting and identification of pollen grains and spores were carried out with a LEICA DM 5000B microscope.

4 Results and Discussion

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4.1 Early and mid-Pliocene (4 to 3 Ma): early Cordilleran Ice Sheet and first glaciation attempts

Early to late-Pliocene (4.0 to 2.76 Ma) SSTs at Site U1417 are highly variable (max and min SST difference of 10 °C) with an average value of 8.2 °C (Fig. 2a; Table 1). We compare our palaeo-SST with the modern SST (here "modern" refers to the averaged decadal statistical mean SST of 6.5 °C (standard deviation of 3.4 °C) during the 1955 to 2012 time period, NOAA

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WOD13; Boyer *et al.*, 2013) at the location of Site U1417 to <u>observe-identify</u> changes in the behaviour of the Alaskan Current. Early to late-Pliocene average SSTs at Site U1417 were approximately 1.7 °C warmer than modern<u>, while</u>. The Pliocene and Pleistocene SSTs at <u>the GOASite U1417</u> have a similar is similar to the modern range observations <u>SST range to modern</u> (e.g. NOAA WOD13; Boyer *et al.*, 2013; Fig. 2<u>a</u>). <u>Within that context, t</u>The MPWP (3.2 to 3.0 Ma) contains the highest SST peak

- 5 of the Pliocene with a, SST of =12.4 °C, 5.9 °C warmer than modern SST in the GOA (Fig. 2a). The average MPWP SST of 8.9 °C is around 2.4 °C warmer than modern. Similar to the MPWP, the MG1-Gi1 warm period (3.6 to 3.4 Ma) contains the second highest peak in SST during the Pliocene, with a SST reaching =11.7 °C, 5.2 °C warmer SST than modern conditions (Fig. 2a)GOA. Also oOther SST peaks during the MG1-Gi1 are up to 2-3 °C warmer than modern (Fig. 2a). The average SST during the MG1-Gi1 period is 9.5 °C, around 3 °C warmer than modern. C_{37:4} concentrations during the Pliocene remain below
- 10 the threshold of subpolar/subarctic water masses identified in the Nordic Seas (Bendle and Rosell-Melé, 2004;)Fig. 2b) and are consistent with a warm surface ocean and/or minimal meltwater inputs to the GOA. The wide range of "warmer than modern" SSTs occurring during the MPWP together with higher than modern atmospheric CO₂ levels (Fig. 3a) and similar continental configuration, further supports the proposal to use this time period as an analogue for future climate predictions (Hansen, 2006). The MG1-Gi1period represents the opportunity for studies to focus on a prolonged period of sustained warm
- During the early to mid-Pliocene, IRD is absent (Fig. 2c) and sedimentation rates are the lowest of the 4-1.7 Ma record (Fig. 2g). Small glaciers in Alaska since or before 4 Ma have been indicated from neodymium and lead isotope records from the Bering Sea (Horikawa *et al.*, 2015). However, our data show that during the early and mid-Pliocene, the Cordilleran Ice Sheet was not yet extensive enough to erode or transport large volumes of sediment and runoff to the GOA. In contrast, IRD at ODP

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SST but with similar SST peaks than the MPWP.

- 20 Site 887 (located 200 km southwest of U1417) suggests glacial influence in the GOA since 5.5 Ma (Reece *et al.*, 2011). Early Pliocene and even Miocene evidence of tidewater glaciation (δ^{18} O, IRD) has been found at other locations in the North Atlantic (Mudelsee and Raymo, 2005; Bachem *et al.*, 2016). Reece *et al.* (2011) attributed the initiation of glaciation in the GOA to the uplift of the Yakutat formation. However, IRD mass accumulation rates at ODP 887 prior to 2.6 Ma are very small, being close to 0 and < 0.2 g cm⁻² Ky⁻¹ (Krissek, 1995). The low sedimentation rate, high TAR, low %C_{37:4} and absence of IRD during this
- 25 period at Site U1417 suggest that although the GOA experienced intervals of relatively cool SSTs (Fig. 2a), any glaciation was not sufficiently extensive to generate icebergs capable of reaching Site U1417 (Fig. 2). limited mountain glaciation but not full scale continental glaciation resulting in tidewater glaciers marked the early and mid Pliocene presented here. There are two intervals of significant cooling recorded during the Pliocene at Site U1417: the MIS M2 (3.3 Ma) and KM2 (3.2

Ma) (Fig. 2a). Neither of these cold intervals record IRD delivery to Site U1417. Both intervals are punctuated by core breaks,

30 suggesting a change in the sediment lithology which made core recovery difficult (Fig. S1). The M2 has been proposed as a significant Pliocene glaciation, <u>but though</u>-smaller than early Pleistocene glaciations, possibly due to the prevalent high atmospheric CO₂ levels (De Schepper *et al.*, 2013) (Fig. 3a). However, if this event, and the climatic conditions we record in the GOA, triggered the appearance of glaciation in Alaska at all (De Schepper *et al.*, 2013), our data suggests the glaciation was not intense enough to support an ice sheet with a tidewater margin that delivered icebergs to Site U1417. Our record

provides evidence for relatively cold SST conditions during M2, as cold as conditions during major glacial cycles of the Pleistocene, but with no evidence for the development of a major Cordilleran Ice Sheet.

Between 4 and 3 Ma ago, we observe maximum TAR values (up to 16; Fig. 2<u>e</u>), pointing to a higher export of terrigenous (i.e. land-plant leaf waxes) relative to aquatic organic matter to the GOA. We assume that the warm and wet climate of the early

- 5 Pliocene during high atmospheric CO₂ levels potentially sustained a highly vegetated landscape in Alaska and west Canada which delivered high amounts of plant wax lipids and pollen grains into the GOA. The absence of IRD and higher pollen counts may refer to infer an airborne transport of the leaf wax lipids rather than an export via icebergs (Fig. 2). The colder SST during the Pliocene (relative to the early Pleistocene) could have promoted a deeper AL and dust driven transport of terrigenous organic matter may have developed. Strong winds could have transported plant waxes to Site U1417 during the Pliocene, as
- 10 is also observed in the North Atlantic during the NHG (Naafs *et al.*, 2012). Müller *et al.* (2018) also proposed an export of long-chain *n*-alkanes to the GOA via dust storms. We suggest that, in addition to wind transport, also-coastal river discharge of terrigenous organic matter may also have contributed to higher TAR values recorded at Site U1417.

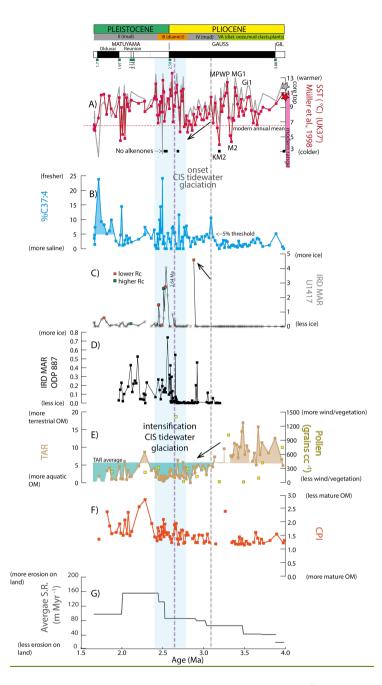


Figure 2: Site U1417 across the Pliocene-Pleistocene transition. a) red line: SST from U^K₃₇' index according to Müller *et al.* (1998) calibration; grey line: SST from U^K₃₇ index according to Prahl *et al.* (1988) calibration. Black squares are samples where alkenones were not detected. Dashed red line: Modern averaged decades (1955-2012) annual statistical mean SST=6.4 °C at 0 m water depth (NOAA WOD13, Boyer *et al.*, 2013) at Site U1417, similar to the modern annual average SST=7 °C at GAK1 station during the 1970-2018 time intervals for the 0-100 m water column depth (Weingartner *et al.* 2016) in the Gulf of Alaska; Red star on y-axis: value of our youngest sample analysed at Site U1417 (U1417D 1H-1W 44-48; 0.016 Ma; SST=10.6 °C with Müller *et al.* (1998) calibration; SST=11.8 °C with Prahl *et al.*, 1988 calibration); Pink rectangle on y axis: modern averaged decades (1955-2012) statistical mean SST during winter and summer at Site U1417 and 0 m water depth (NOAA WOD13, Boyer *et al.*, 2013) SST=0-11,3 °C; b) abundance of the cold and/or freshwater

alkenone $C_{37:4}$ (%). Horizontal line shows the threshold of Bendle *et al.* (2005) above which subarctic/subpolar water masses were determined for the Nordic Seas; c) IRD MAR (g cm⁻² ka⁻¹). Orange and green squares reflect lower and higher average roughness coefficient (Rc) of the IRD quartz grains, respectively; d) IRD MAR (g cm⁻² ka⁻¹) at ODP 887 (Prueher and Rea, 2001); e) terrestrial/aquatic *n*-alkane index (TAR, defined in the main text), horizontal line shows the average TAR value for the study interval, yellow squares represent pollen grains

- 5 concentrations in grains cc⁻¹-: and f) CPI (defined in the main text): and g) average sedimentation rates (see Fig. S3b) in m Myr⁻¹ at Site U1417. Upper panel: Plicene-Pleistocene boundary, magnetostratigraphy events and interpretations (see Fig. S2 and S3) and Lithostratigraphic units of Site U1417 with simplified lithology (orange: colouring represents ice rafted diamict interbedded with mud, brown: colouring represents marine mud_and green: colouring represents diatom ooze interbedded with debris flow deposits containing mud clasts and plant fragments) (Jaeger et al., 2014). Grey vertical line represents the onset of the Cordilleran Ice Sheet (CIS) glaciation (or
- 10 oNHG) climate transition at 3 Ma, blue shading represents the 2.5-2.0 Ma climate transition with the intensification of the Cordilleran Ice Sheet (CIS) tidewater glaciation (or iNHG) as in Table 1. Purple vertical line represents the onset of the Cordilleran Ice Sheet at the Lower Klondike Valley, Yukon interior (Hidy *et al.*, 2013; Fig. 1). Missing <u>TAR or CPI</u> data points are either a result of samples analysed for SSTs at the early stages of the project which were not subsequently analysed for *n*-alkane distributions, <u>and-or</u> the result of samples where chromatograms reflected poorly resolved *n*-alkane peaks.
- 15 We further note that rivers and ocean currents could have transported bedrock material from the Yakutat Terrain (Childress, 2016) to Site U1417, 700 km offshore from the Alaskan coast. This would imprint the sediments delivered to the ocean with a <u>petrogenic n ancient</u> signal of terrigenous organic matter, rather than reflecting erosion of contemporary 'fresh' organic matter from vegetation and soils. The CPI is often used to estimate the maturity of the organic matter and determine its source (Fig. 2f). Previous studies suggest that elevated TAR values and CPI values close to 1 reflect coal particles found in sediments
- 20 in the GOA (Rea *et al.*, 1995; Gulick *et al.*, 2015). However, the coal-bearing Kulthieth rocks (McCalpin *et al.*, 2011), have a TAR signature of a maximum value of 2 and CPI values of less than 1 (Childress, 2016). Since Site U1417 TAR (up to 16) and CPI values (> 1) do not overlap with these TAR (up to 2) and CPI (< 1) values found onshore values, we (Childress, 2016), the TAR and CPI values at Site U1417 thus suggest a mix of sources of organic matter during this time dominated by contemporaneous vegetation, although we cannot exclude the possibility of some coal erosion. The mix of terrigenous sources</p>
- 25 delivered to GOA agrees well with previous studies that suggest the mergingmeeting of ice bodies during the Pliocene westward expansion of the east-central Alaska Cordilleran Ice Sheet at XX Ma-during the Pliocene (Duk-Rodin et al., 2004),, , although we cannot exclude the possibility of some coal erosion.

4.2 The late Pliocene onset of the Cordilleran Ice Sheet glaciation (3.0 to 2.8 Ma)

- The interval from 3.<u>0</u> to 2.8 Ma is characterised by a shift of climate conditions from those observed during the early and mid-30 Pliocene (Fig. 2) to more glacial conditions. At 3.<u>0</u> Ma, average SSTs at Site U1417 remain relatively warm (around 8 °C), yet, there is the first evidence of cooling evidence at Site U1417 deduced from C_{37:4} crossing the threshold of 5 % (Bendle *et al.*; 2005). %C_{37:4} increases can be related to colder sea surface conditions, but due to Site U1417's location and climatic context, it may also be possible that we suggest that increases in %C_{37:4} relate to meltwater discharge from the expanding icesheet. From 3.<u>0+</u> to 2.8 Ma, SST decreases gradually from 8 to 5.5 °C (Fig. 2a) recording again-colder SSTs than the modern
- 35 GOA. We attribute this 0.3 Ma~2300 kyr progressive cooling to the oNHG in as a response to the overall decrease in the atmospheric CO₂ (Seki *et al.*, 2010; Martínez-Botí *et al.*, 2015). From 3.0 Mato 2.8 Ma, TAR values decrease to below the average of the entire TAR record suggesting a decrease in leaf-wax lipid transport to Site U1471 and/or higher input of aquatic derived organic matter. Compared to the 4.40-3.0 Ma time interval, accumulation rates of long-chain *n*-alkanes, reflecting

terrigenous organic matter input, slightly decreased from average values of 6.19 μ g cm⁻² kyr⁻¹ to This pattern can be, indicating <u>attributed to a decrease in that transport of leaf-wax lipid transports to Site U1417 increased (5.94 (+3.76/-1.59) μ g cm⁻² kKyr⁻¹ while accumulation rates of short-chain *n*-alkanes, reflecting <u>-combined with and increase in</u> aquatic organic matter export, <u>productivity</u> increased from 0.66 μ g cm⁻² kyr⁻¹ to (1.7 (+1.54/-0.73) μ g cm⁻² Kkyr⁻¹ (Table 2).-compared to in comparison with</u>

- 5 the Pliocene_(6.19 (+33.29/-4.15)k and 0.66 (+3.46/-0.34)k, respectively)., which may be related to an increase in erosion on land due to the advancing ice sheet. This increase in the abundance of short-chain *n*-alkanes during a period of SST cooling may point to a higher algal productivity stimulated by a successive supply of nutrients through the erosion of bedrock. In fact, tThe coincident increase in average sedimentation rates (from 65 to 79 m Myr⁻¹) indicates a more efficient erosive agent onshore than before 3 Ma. and/or a change in the source of terrestrial matter. However, As CPI values at Site U1417 (Fig. 2fe)
- 10 remain similar to early and mid-Pliocene values, which suggests a similar (immatuire) source of the terrigenous organic matter is suggested. We propose that The most reasonable explanation is that instead, as the land was becoming increasingly ice covered during the expansion of the Cordilleran Ice Sheet, limiting vegetation growth became limited and there was less and, so that the erosion of vegetation higher plant materials during the oNHG became limited. decreased despite the increase in and terrigenous organic matter eroded and transported to the ocean increased. The timing of ourthe oNHG fits well with the first
- 15 majoroldest Cordilleran glaciation recorded in the Yukon between 2.9 and 2.6 Ma (Duk-Rodkin and Barendregt, 1997; Duk-Rodkin et al., 2001; Froese et al., 2000). as well as oOur interpretation of the TAR data is also consistent interpretation agrees well-with the deposition of terrigenous fan deltas due to coastal uplift (Duk-Rodkin et al., 2004), where the higher topography during the onset of the Cordilleran glaciationa colder climate would translate in lower vegetation growth and higher sedimentation rates. (Duk-Rodkin et al., 2004).
- 20 The peak in %C_{37:4} at 3 Ma is followed by lower %C_{37:4} values (close to 5 %) and the first significant pulse of IRD <u>as</u> identified by a single sample with the highest IRD MAR. This IRD MAR peak (4.5 g cm⁻² ka⁻¹) and an increase in sedimentation rates (from 79 to 85 m Myr⁻¹) at 2.9 Ma constitute the first evidence that tidewater glaciers were present in southwest Alaska delivering icebergs to Site U1417. IRD quartz grains do not appear crushed or abraded by glacial activity, indicating small tidewater valley glaciers producing icebergs which could contain grains that were introduced by rockfall or fluvial sediment.
- 25 The abrupt peak in IRD delivery to U1417 at 2.9 Ma could be due to ice growth on land and cold enough SSTs to permit distal iceberg-drift and release of debris to Site U1417. <u>A smaller number of icebergs also drifted to ODP 887 (St John and Krissek, 1999) during this time (Fig. 2d).</u> The increase in sedimentation rates has been suggested to mark the maximum Cordilleran Ice Sheet extension during the Pliocene (Gulick *et al.*, 2015). Following this first peak, IRD MAR decreases to values between 0 and 1 g cm⁻² ka⁻¹ until 2.6 Ma. This abrupt decrease in IRD indicates<u>d</u> lower iceberg delivery to Site U1417. A synchronous
- 30 increase of C_{37:4} above 5 % suggests that the melting of tidewater glaciers was responsible for the decrease in iceberg delivery despite the cold SSTclimate. A colder and drier climate associated with lower SSTs and atmospheric CO₂ concentrations (Fig. 3a) during the last stages of the oNHG could have limited moisture transport to land and iceberg delivery to the GOA (Fig. 2c and d). Atmospheric CO₂ concentration peaks during this time (Fig. 3a), raising the possibility that enhanced radiative forcing may have contributed to a reduced ice sheet due to radiative forcing.

4.3 The intensification of the Cordilleran Ice Sheet glaciation (2.7-2.4 Ma) and its evolution during the early Pleistocene (2.4-1.7 Ma)

At Site U1417, the iNHG during the Plio-Pleistocene transition (PPT) is characterised by a rise in SST, followed by highly variable values (between 5.6 to 13.6 °C) with an average of 9.7 °C, which are 3.2 °C warmer than modern. The iNHG is defined here as the period containing sustained signs of glaciation (i.e. Maslin *et al.*,1996; Bartoli *et al.*, 2005), which at Site U1417 are confirmed by glacial meltwater and IRD delivery. The relatively high %C_{37:4} (up to 24 %) in the early Pleistocene correlates well with the period of high IRD delivery (up to 4 g cm⁻² kKa⁻¹) between 2.7 to 2.4 Ma (Fig. 2cb and de). This suggests this period marks an expansion/intensification of the Cordilleran glaciation following a gradual SST cooling during the oNHG. The timing of the increase in IRD at Site U1417 coincides with the increase in IRD at Site 887 (St John and Krissek,

- 10 1999) and, the maximum extent of the CIS as recorded onshore in the eastern Cordillera by the extensive Klondike gravels at 2.64 Ma (+0.20/-0.18 Ma) (Hidy et al., 2013) and the oldest glacial records in stratigraphical sections in south-western Yukon and south-eastern Alaska, 2.9 to 2.6 Ma (Barendregt et al., 1996, Duk-Rodkin et al, 19967, Duk-Rodkin and Barendregt, 1997; Froese et al., 2000; Duk-Rodkin et al., 2001; Duk-Rodkin et al., 2004). The lithology at Site U1417 includes diamict layers that alternate with bioturbated mud from 2.7 Ma, indicating that the Cordilleran Ice Sheet remained very variable after the
- 15 oNHG and maintained glacial tidewater margins discharging icebergs into the sea. Yet the intensification of the Alaskan tidewater glaciation occurred with a GOA that was overall either warmer than, or at least as warm as, the mid to late Pliocene (considering Müller *et al.* 1998 SST calibration error).

The overall increase in $C_{37:4}$ in the GOA during the early Pleistocene coincides with an SST warming (ca. 1 °C relative to the Pliocene; Fig. 2a and b), suggesting a stronger link between $C_{37:4}$ and meltwater fluxes rather than an expansion of subarctic

- 20 water masses. Additionally, maxima and minima in %C_{37:4} during the iNHG are unrelated to elevated or lowered SSTs, respectively. There is no-little information available about the origin of C_{37:4} in the North Pacific to explain the high %C_{37:4} values recorded at Site U1417 (i.e. Harada *et al.*, 2008; McClymont *et al.*, 2008), nor their association with intermediate SSTs rather than minima/maxima. It has been suggested that stratification of the water column due to glacier dischargeatmospheric CO₂ changes in the North Pacific could result in warmer sea surface in comparison to deeper water masses and the surrounding
- 25 <u>land</u> due to an increase in surface absorption of solar radiation and seawater heat capacity (Haug *et al.* 2005)Meheust *et al.*, 2013). Haug *et al.* (2005) proposed this could lead to an increase in ocean evaporation and orogenic precipitation, ultimately encouraging North American ice sheet growth.

Terrigenous and aquatic organic matter accumulation rates both sources-increase during the the early Pleistocene 2.4 to 1.7 Ma interval -in comparison with the late Pliocene 4-2.8 Ma time period, resulting in a-variable but overall lower than average TAR

30 values (Table 2), despite enhanced terrigenous organic matter inputs to Site U1417 (Fig. 2ge). Over the iNHG, low TAR values (< 1) and small variations in IRD MAR (the order of 0.1 to 2.8 g cm⁻² kKa⁻¹) coincide with intermediate SSTs (7 to 11 °C) and %C_{37:4} between (2-24 %). This could point to an increase in marine productivity export related to an enhanced nutrient delivery to Site U1417 via glacial runoff. The increase in CPI values discard mature sources of organic matter to the GOA-at this time interval suggesting an enhanced contemporary leafve wax-aquatic organic matter contribution. IRD peaks are observed

typically present during SST minima suggesting the importance of SSTs <u>forin</u> the delivery of icebergs to distal sites such as Site U1417. The average Rc of IRD is low (Fig. 2c) even during IRD MAR peaks, indicating minimal glacial crushing during the iNHG. In comparison, samples from 1.6---1.5 Ma show a higher Rc and appear to have greater evidence of glacial crushing, suggesting development of a larger ice sheet or scouring and evacuation of sediment from the non-glacial, weathered landscape.

5 This could indicate that the first IRD in icebergs delivered to the GOA during the late Pliocene and early Pleistocene originated from smaller marine terminating valley glaciers which removed sediment and weathered rock from the landscape rather than eroding bedrock and allowed IRD generation.

The comprehensive data set obtained from Site U1417 sediments (Fig. 2) supports a climate role in the ice-sheet expansion during the early Pleistocene and the iNHG, with an increase in precipitation from a warmer and/or stratified ocean, and cooler

- 10 periods associated with IRD delivery. An additional-alternative and/or complementary explanation for the changing TAR during the early Pleistocene is that tectonic uplift of the Chugach/St Elias area from 2.7 Ma (Enkelmann *et al.*, 2015) led to enhanced orogenic precipitation and a change in erosional pathways (Enkelmann *et al.*, 2015). The glaciation could have altered the main source of terrestrial input to the Surveyor Channel, to higher metamorphic and plutonic sources with lower or null TAR values (Childress, 2016). An increase in CPI variability to concentrations up to 2 and 3 during the early Pleistocene
- 15 (starting from 2.7 Ma) supports the change of source of organic matter away from the more mature coal bedrock <u>into-to</u> more immature terrestrial organic matter (plant waxes). However, this comes at a time of increasing IRD, which adds a new source of terrigenous sediment to Site U1417. The <u>shift inhigher frequency of highshift in</u>-CPI values <u>fromat</u> 2.7 Ma <u>in comparison</u> with the period before 2.7 Ma is synchronous agrees with the shift towards the erosion of sediments sourced from metamorphic and plutonic sources, described in Enkelmann *et al.* (2015), -delivered to Site U1417. Lower pollen counts suggest a less
 20 vegetated landscape, which <u>also</u> could help explain the overall lower TAR_during the early Pleistocene in comparison with

4.4 The Pliocene and Pleistocene climate across the North Pacific Ocean.

the Pliocene.

The overall cooling trend during the Neogene, briefly interrupted by the MPWP and intense cooling events such as the M2, is believed to be a dominant pattern in records of the global climate. This notion is largely based onreflects the global increase
in ice volume (as reflected in the e.g. LR04 Benthic δ¹⁸O Stack₅₁. (Lisiecki and Raymo, 2005; Fig. 3b) as well as cooling trends reconstructed in and from studies in the North Atlantic SSTs (e.g. i.e. ODP Site 982, Lawrence *et al.*, 2009). In contrast, our understanding of the contribution of the North Pacific into our understanding of to global climate evolution from the Pliocene to the Pleistocene is limited. Our study at Site U1417 adds valuable regional climate information during the evolution of the Cordilleran Ice Sheet. Unlike the LR04 stack, average Pliocene SST values (4.0 to 2.8 Ma) at Site U1417 are 1 °C ± 0.5
(s.d.) colder than the average early Pleistocene values (2.7 to 1.7 Ma) (the Pliocene Pleistocene SST difference of 1°C has an standard deviation of 0.5°C). In the wider North Pacific, aA warming trend from the late Pliocene to early Pleistocene has also been observed at ODP Site 882 in the subarctic Pacific (Martínez-García *et al.*, 2010), at Site 1010 and potentially at Site

1021 (mid-latitude east Pacific) (Fig. 3). Beyond the North Pacific, warmer SSTs during the early Pleistocene compared to the

Pliocene have also been recorded i.e. DSDP Site 593 in the Tasman Sea (McClymont *et al.*, 2016) and Site 1090 (Martínez-García *et al.*, 2010) in the South Atlantic. In contrast, long-term cooling trends mark the early Pleistocene for the mid-latitude west Pacific (Site 1208) and tropical east Pacific (Site 846), more consistent with the development of a cooler and/or more glaciated climate (Fig. 3).

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The North Pacific warming occurs despite an atmospheric CO₂ drop from <u>an average of 280-450-347ppmv</u> (Pagani *et al.*, 2010; Seki *et al.*, 2010) and 357 (Martínez-Botí *et al.*, 2015) ppmto 250-300 ppmv to (similar to pre-industrial levels (268 ppm and 315 ppm) from 3.03.2 to 2.68 Ma (Pagani *et al.*, 2010; Seki *et al.*, 2010) and an associated reduction in global radiative forcing (Foster *et al.*, 2017). The early Pleistocene warming signal in the GOA (and the north Pacific more generally) thus implies an important role for local or regional processes. We have discussed above the potential role played by ocean stratification in the North Pacific, and a possible link to the evolving Cordilleran Ice Sheet in the GOA through evaporation/precipitation feedbacks. The synchrony of these changes with observed tectonic uplift (e.g. Enkelmann *et al.* 2015)

makes it difficult to disentangle the potential climatic and tectonic mechanisms behind ice sheet expansion.

- 15 To understand the evolution of the ocean eurrents governing the North Pacific at the present core sites (Fig. 1) and to find possible explanations of the observed SST distributions during the Pliocene and Pleistocene climate evolution, the modern elimate system is used here as an analogue. Modern monthly mean SSTs at ODP 882 SSTs are colder than Sites U1417 and 1021 all year around. During the late Pliocene and early Pleistocene, ODP 882 SSTs are 3-4 °C warmer than in the east (Fig. 3f and g). Modern seasonal climate analogues cannot be used to explain to Pliocene and Pleistocene subarctic SST distribution.
 20 However, on longer timescales, the strength of the AL is currently linked to the wider Pacific Ocean circulation by the Pacific Decadal Oscillation (PDO) over periods of 20 30 years (Furtado *et al.*, 2011). The Pliocene Pleistocene North Pacific SST gradients show similarities with the negative phase of the PDO (-PDO), which is characterized by positive SST anomalies in the central North Pacific surrounded by negative SST anomalies along the North American coast and in the east equatorial Pacific. If the PDO can be used as an analogue for the late Pliocene, The -PDO the associated route of wind systems might have increased the precipitation in the Gulf of Alaska, and could therefore represent a key factor for the fast building ofgrowth
- of glaciers and/or the CIS_ice in the Alaskan mountains.

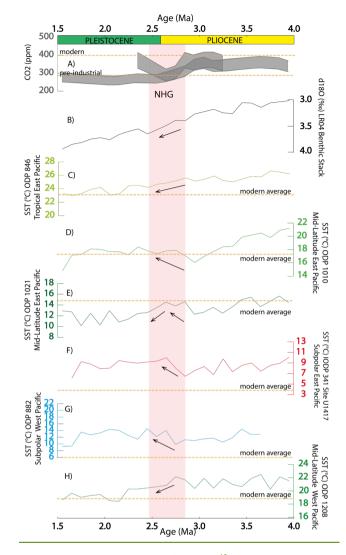


Figure 3: Pliocene-Pleistocene SST across the North Pacific. a) Alkenone δ¹³C atmospheric pCO₂ upper and lower end (ppm+) estimates at Site ODP 999A (Seki *et al.*, 2010, revised in Foster *et al.*, 2017) (4.0-1.5 Ma) and marine δ¹¹B atmospheric CO upper and lower end (ppm) estimates at Site ODP 999 (2.3-3.2 Ma) (Martínez-Botí *et al.*, 2015, revised in Foster *et al.*, 2017); b) δ¹⁸O (‰) LR04 Benthic Stack (Lisiecki and Raymo, 2005); Alkenone SST (°C) from c) ODP Site 846 (Herbert *et al.*, 2017), d) Site 1010 (Herbert *et al.*, 2018), e) Site 1021 (Herbert *et al.*, 2018), f) IODP 341 Exp. Site U1417 (Sánchez-Montes *et al.*, 2019), g) ODP Site 882 (Martínez-García *et al.*, 2010) and h) ODP Site 1208 (Herbert *et al.*, 2018). The SST records are arranged from top to bottom following an anticlockwise direction from tropical-east to subpolar east, subpolar west and tropical-west locations. Orange horizontal lines indicate reference levels of pre-industrial times and/or modern values for each of the records or sites (SSTs from NOAA WOD13, Boyer *et al.*, 2013). Arrows indicate position of M2 and KM2 periods and cooling or warming trends across the oNHG in the Pacific Ocean. The KM2 event is located within the MPWP. The MG1-Gi warming precedes the M2 event. b) to h)All elimate data-records are 1kyr linearly interpolated and –100 kyr smoothed-records₂, and the Ages (Ma)x axis have been determined through the calculation of the mean for each interval.

To understand the evolution of the ocean currents governing the North Pacific at the present core sites (Fig. 1) and to find possible explanations of the observed SST distributions during the Pliocene and Pleistocene climate evolution, the modern

15 climate system is used here as an analogue. Modern monthly mean SSTs at ODP 882 SSTs are colder than Sites U1417 and

1021 all year around. During the late Pliocene and early Pleistocene, ODP 882 SSTs are 3-4 °C warmer than in the east (Fig. 3f and g). Modern seasonal climate analogues cannot be used to explain the Pliocene and Pleistocene subarctic SST distribution. However, on longer timescales, the strength of the AL is currently linked to the wider Pacific Ocean circulation by the Pacific Decadal Oscillation (PDO) over periods of 20-30 years (Furtado *et al.*, 2011). The Pliocene-Pleistocene North

- 5 Pacific SST gradients show similarities with the negative phase of the PDO (-PDO), which is characterized by positive SST anomalies in the central North Pacific surrounded by negative SST anomalies along the North American coast and in the east equatorial Pacific. If the –PDO can be used as an analogue for the late Pliocene, the associated route of wind systems might have increased the precipitation in the Gulf of Alaska and could therefore represent a key factor for the growth of glaciers and/or the CIS in the Alaskan mountains.
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5 Conclusions

The sea surface temperature (SST) evolution from the Pliocene to the early Pleistocene in the subarctic Northeast northeast and east-mid latitude North Pacific is very different from the North Atlantic, with a colder Pliocene than early Pleistocene. The early Pliocene appears to be characterised by a heavily vegetated landscape where there is no obvious noticeable glaciation

- 15 in the St. Elias mountains. A series of cooling events during the Pliocene (including the M2 event) could have initiated glaciation in Alaska but <u>if so, the glaciation was it was</u>-limited to mountain glaciers probably due to <u>relatively</u> high atmospheric CO₂ concentrations and the lower topography in coastal Alaska. The first evidence of glaciation starts at 3 Ma with an increase in glacial meltwater followed by a progressive 2.5 °C SST cooling from 3.04 to 2.8 Ma and the first IRD peak at 2.9 Ma since the late Pliocene. Glacial meltwater, IRD and sedimentation rates increase <u>areis indicative of the intensification</u>, identified as
- 20 the intensification of the Cordilleran glaciation (2.7-2.4 Ma). This occurs with warm SSTs suggesting an efficient warm oceanland precipitation-Cordilleran Ice Sheet growth interactions.

A permanent-warm surface ocean in the west mid-latitude Pacific during the late Pliocene and early Pleistocene compared to modern was potentially a key mechanism for increasing moisture supply to the Gulf of Alaska (GOA) and triggering the growth of the Cordilleran Ice Sheet. A similar to modern negative PDO-like climate could have set a more efficient route for moisture transport from the west subarctic Pacific to the GOA since the MPWP and could have been a key mechanism for glacial growth. Unlike duringIn contrast to the Pliocene, the early Pleistocene drop in atmospheric CO₂-concentrations could have been decisive in developing an expansion continuous glaciation of the Cordilleran Ice Sheet during the variable climate of the late Pliocene / early Pleistocene intensification of the Cordilleran tidewater glaciation. However, the synchronousThe tectonic uplift of the St Elias mountains could also have been a contributing factor for the Cordilleran Ice Sheet expansion, by increasing

30 the potential for precipitation to fall as snow over the ice sheet source regions, despite warm SST in the GOA during the <u>Pliocene</u>. In contrast to the Pliocene, the early Pleistocene drop in atmospheric CO₂ concentrations could have been decisive in an expansion of the Cordilleran Ice Sheet.

Data availability

The data presented in this manuscript has been submitted to Pangaea.de and it is under review. After the publication of this manuscript, the data would be accessible through this link <u>https://doi.org/10.1594/PANGAEA.899064</u> and could be cited as Sánchez-Montes *et al.* (2019). The SST data in this publication will also be published in the PlioVAR database.

5 Author contribution

New data sets presented in this manuscript derive from the PhD project of MLSM supervised by ELM and JML. JM was closely engaged from early stages of this project including aspects of method development. EAC generated the IRD data, and CZ generated pollen data. All authors have contributed to data interpretations. MLSM prepared the manuscript with contributions from all co-authors.

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| Age intervals (Ma) | Average SST (°C) | Peak SST (°C) average | Trough SST (°C) average | Average SST variability (°C) | Average C _{37:4} (%) | Peak C _{37:4} (%) |
|-------------------------|---------------------|--------------------------|-------------------------------|---------------------------------|----------------------------------|----------------------------|
| 4.0-3. <u>0</u> 1 | 10.2/8.7 | 12.5/11.4 | 7.2/4.4 | 5.3/7.0 | 1.9 | 10.5 |
| 3. <mark>0</mark> 1-2.8 | 8.5/ 7.3 | 9.9/ 9.0 | 7.4/5.7 | 2.4/ 3.3 | 3.9 | 4.9 |
| 2.7-2.4 | 10.2/9.8 | 13.2 12.6 | 8.4/ 6.6 | 4.8/5.9 | 4.8 | 24.1 |
| 2.4-1.7 | 9.0/ 8.6 | 10.7/ 10.4 | 6.8/ 4.8 | 3.9/5.6 | 5.2 | 23.8 |
| 4-2.8 -Ma | 9.6/ 8.2 | 11.2/ 10.2 | 7.3/5.1 | 3.9/5.2 | 2.5 | 10.5 |
| 2.7-1.7 <mark>Ma</mark> | 9.6/ 9.1 | 12.0/11.5 | 7.6/5.7 | 4.4/ 5.8 | 5.0 | 24.1 |

Table 2: Overall above (+) and below (-) the 4-1.7 Ma TAR average values and terrigenous and aquatic *n*-alkane mass accumulation rates (μ g cm⁻² kyr⁻¹) at Site U1417 during the time intervals described in the text.

Table 1: Average SST (°C) and %C_{37:4} during key climatic intervals 4.0-3.0 Ma, 3.0-2.8 Ma, 2.7-2.4 Ma, 2.4-1.7 Ma. Average SST (°C) is the average of all the data points of the time interval, peak SST (°C) average is the average of the highest data points of each interval selected (Fig. 2), trough SST (°C) average is the average of the lowest data points of each interval (Fig. 5.2) and the average SST (°C) variability is the difference between average SST peak and the average SST trough. In black: data calculated from U_{37}^{K} (Prahl *et al.*, 1988) and in black bold, data from U_{37}^{K} (Müller *et al.*, 1998).

| Age intervals (Ma) | <u>Above (+) or</u> <u>below (-)</u> average TAR | Average terr <i>n</i> - alkane MAR | <u>Average aq. <i>n</i>-</u> alkane MAR | |
|-----------------------|--|---------------------------------------|--|--|
| <u>4.0-3.0</u> | + | <u>6.19</u> | <u>0.66</u> | |
| 3.0-2.8 | <u>=</u> | <u>5.94</u> | <u>1.70</u> | |
| 2.7-2.4 | = | 12.39 | <u>6.89</u> | |
| 2.4-1.7 | = | 35.85 | 11.37 | |
| 2.7-1.7 | | 24.12 | 9.13 | |