Dear Reviewer #1,

Thanks again for your review. Please find below our response to each of the issues that you have raised together with links on how we considered your suggestions.

Reviewer: I think some conclusions need more data to support and more discussions needed. For example, for the wildfire reconstruction using the sugar proxies there should be some discussions about the potential of the movement of these indicators in soluble condition since these indicators themselves are soluble. So, peoples would doubt whether these proxies have experienced movement after their deposition. If so, these proxies could not reflect the wildfires in the corresponding layers. **Response: You are right. We have extended the discussion on the proxy behaviour from source to sink (see below and Chap. 4.1.2)**

Reviewer: For the wildfire reconstruction from Lake El'gygytgyn sediments, the authors selected three glacial-interglacial cases. Due to the uncertainty of chronology reconstruction, the authors integrated the wildfire proxies into two periods, that is, glacial and interglacial. From my side, I think that such integration indeed could give some information for the glacial-interglacial variations of wildfire. But meanwhile it could mix some useful information and sometimes might result in some wrong conclusions. For example, previous studies also suggested that some wildfire occurs mainly during the transition of glacial-interglacial variations. Under such condition, the integration of wildfire into two parts, the glacial periods and the interglacial periods, would not give the real picture of wildfire pattern.

Response: Thanks for your comment. In this study, we don't aim to provide the full picture of a complete, continuous record of glacial-interglacial fire history and also don't consider a full glacial cycle but only the late glacial part.

We state that better now from the beginning on, e.g. in the title: "Relationships between lowtemperature fires, climate and vegetation *during three late glacials and interglacials* of the last 430 kyrs in northeastern Siberia reconstructed from monosaccharide anhydrides in Lake El'gygytgyn sediments" and introduction: "Here, we *assess (1) how far MAs are useful proxies to study* glacial to interglacial fire histories of the last 430 kyrs using sedimentary MA from Lake El'gygytgyn and *(2) discuss* long-term relationships between low-temperature fires and regional vegetation in the Russian Far East." We also replaced "glacial" by "late glacial" at the places in the text where is was misleading.

The split of samples in two groups was done because we are very confident about the exact position of the shift from late glacial to interglacial conditions, because the pollen data from the same samples suggest a very clear shift from cold, dry tundra-steppe to warmer shrub tundra or interglacial forest, independent of the absolute age uncertainties. We are aware that during glacial-interglacial transitions the fire history can be quite variable as suggested, e.g., by Han et al. (2016), which used a record of much higher sedimentation rates compared to Lake El'gygytgyn, or by Bird and Cali (2002) and Daniau et al. (2013) based on African sediments. As below you have commented also on the relationship between insolation and MA trends in time, we reconsidered this part and discuss now also how far fire has occurred during transitions (see below). However, given the age uncertainties for several hundreds of years, the very low sedimentation rates at Lake El'gygytgyn and that our samples are integrating over several hundreds of years, we prefer here to discuss the general trends evidenced by the late glacial versus interglacial using the boxplots, as a starting point for more detailed reconstructions that require a higher resolution.

We mention that now in the conclusion: "Further research will continue exploring lake-sedimentary MAs *in higher resolution together with further sedimentary fire proxies such as charcoal to study* low-intensity fire–climate–vegetation feedbacks in space and time and potential ways of post-depositional degradation in even older interglacials, with CO2 levels similar to those expected in the future.".

Reviewer: In fact, the figure 3 did not give exact information on wildfire pattern at glacial-interglacial scale. There is no clear difference in wildfire between glacial and interglacial periods, except one case of MIS 11c and 12.

Response: In the text, we acknowledge that MA influxes "are consistently higher during interglacials compared to the latter part of their preceding glacials", which doesn't mean that all glacials are equally low. We suspect that the difference didn't become clear visually because we have plotted the biomarker (GAL) with highest influxes on top of those with lower influxes and with rather bold lines. We hope it becomes clearer in a modified Fig 3a that MA influxes are higher during MIS 5e compared to MIS 6 and also in MIS 7e compared to MIS 8 (where just one sample has similarly high influxes as the MIS 7e samples). Please see the slightly modified Fig 3 here:



Reviewer: Specific comments: 1. Lines 68-69. That's good to have an interpretation for fire intensity. But I would like to know the exact factor that controls fire intensity in this study, rather than the factors suggested here including three ones: fire temperatures, combustion efficiencies and fire radiative power. In fact, these factors have no clear relationship between each other. Is temperature more related to fire intensity? Or combustion efficiency is more related to fire intensity? In my view, I think that combustion efficiency is more related with the fire intensity, which has been improved by many previous studies.

Response: According to Keeley (2009), "fire intensity represents the energy released during various phases of a fire" and in a physical sense it is given in W m⁻². Depending on the discipline, fire intensities are reported as fire radiative power (e.g. in remote sensing, Rogers et al., 2015) or when studying emissions, combustion efficiencies are reported (van Leeuwen and van der Werf, 2011). All the parameter mentioned (temperature, combustion efficiency, radiative power) are positively related to each other, as higher burning temperatures combust organic matter more efficiently and also release more energy (see e.g. carbon combustion continuum in several studies, for example, Conedera et al. (2009).

We have now rephrased this part and also added that during each fire several phases of fire intensity can happen, with an average low or high fire intensity characterizing a fire regime that integrates over longer time and larger spatial scales. "While a single fire has several phases of varying fire intensities, fire regimes define larger temporal and spatial scale properties of several fire events with regimes of low fire intensity generally referring to low fire temperatures, low combustion efficiencies and low fire radiative power as typical for smoldering and in contrast to flaming fires (Keeley, 2009;Conedera et al., 2009;van Leeuwen and van der Werf, 2011)."

Reviewer: 2. Line 127, the authors selected three cases of the glacial-interglacial periods for wildfire reconstruction. I would like to know the reason for such selection.

Response: To assess the suitability of MAs as fire proxies in high-latitude lake sediments on long time scales, we have selected three glacial-interglacial periods that differ in terms of biome configuration and climate, including two of them (MIS 5e and 11c) that are generally referred to as potential analogues for future climate change.

We have fully rephrased this part and now write: "We selected three late glacial-to-interglacial periods, i.e. marine isotope stages (MIS) 12–11c, 8–7e, and 6–5e, which reflect varying interglacial biome types and climate conditions, as reconstructed using the pollen records from El'gygytgyn sediments (Melles et al., 2012;Tarasov et al., 2013)."

Reviewer: In fact, a continuous record of three glacial-interglacial intervals would give more robust evidences for the wildfire-climate relationship, I think. Please give some explanations.

Response: In this study, we aim to test how far MAs are useful proxies on long time scales in Arctic lake sediments and to see if we can deduce some first fire-vegetation-climate relationships. Given the amount of time to prepare and analyse a sample in the lab, a continuous record over several glacial-interglacial intervals was beyond the scope of this study. Here, we focus on comparing interglacials and their preceding late glacial period that have previously been identified as being different concerning their climate and vegetation configurations.

Yet, we added in the conclusion: "Further research will continue exploring lake-sedimentary *MAs in higher resolution together with further sedimentary fire proxies such as charcoal to study* low-intensity fire–climate–vegetation feedbacks in space and time and potential ways of post-depositional degradation in even older interglacials, with CO2 levels similar to those expected in the future."

Reviewer: 3. Lines 139-140, please add a parentheses.

Response: As the part in the parentheses did not become clear, we have reordered it to: "(for interglacial astronomical and GHG characteristics see Yin and Berger (2012) and Table 1)".

Reviewer: 4. Line 154, "that cover the time period", which time period? Response: Thanks. We added that we refer to the period "125 to 3600 kyrs".

Reviewer: 5. Lines 203-204, the subhead "2.2 Analyses of source areas". I don't think that we could fully ignore the river input and only consider the atmospheric deposition for source analyses. So, the discussion about the source areas should include the local river inputs.

Response: You are right. We are discussing also fluvial transport in Chap. 4.1.2 and here we write now: "MAs can be transported attached to aerosols in the atmosphere (Sang et al., 2016;Schreuder et al., 2018) and/or via fluvial transport from the catchment (Suciu et al., 2019). To discuss potential source areas of *aeolian-derived* MAs, we calculated exemplary backward trajectory ensembles..." and we start the discussion now by stating: "*Here, we discuss the potential local and regional to extra-regional MA source areas, transport pathways and post-depositional degradation in El'gygytgyn lake sediments.*" And mention catchment sources also in the discussion (chap. 4.2), e.g. "...MA influxes tend to increase with higher amounts of tree and shrub versus tundra-steppe pollen (Figs. 3c, 4b) – *potentially allowing also fires to occur in the El'gygytgyn catchment during warmer than present interglacials (MIS 5e, 11c)*".

Reviewer: 6. Lines 236-239, This could in general work on the condition that you have known that there were clear wildfire changes between glacial and interglacial periods. But, in fact, in your records of three glacial-interglacial periods, I merely find that the last one (MIS 11c-12) could present a clear increasing trend in MA from MIS12 to 11 (although it is not a full glacial-interglacial cycle), however, for the other two cases, there are no clear wildfire variation pattern at the glacial-interglacial intervals. So, I am not convinced of your conclusion that more low-temperature fires occurred during interglacial periods.

Response: To test how far the proposed trends are different, we have now added a non-parametric Wilcoxon rank sum test suitable for small and non-normally distributed sample sets and now mark the respective p > 0.1, 0.05 and 0.001 in fig.2 and provide the test statistics in the supplementary data. We find strong evidence that also based on this distribution test, late glacial samples had lower mean MA influxes compared to interglacial samples, although the p-values partly suggested that more scrutiny is needed in future studies using more samples. In addition, we are choosing a more careful phrasing to not overinterpret the data, as we are aware that our sample size is not very high causing some uncertainties to the statistics.

In the figure 2 caption we have added: "Lines between interglacial (IG) and preceding late glacial (LG) boxplots indicate different mean values for the two periods according to a non-parametric Wilcoxon test, with stars and + indicating respective p-values. IG vs. LG in a subplot marks different means when all interglacial samples are compared to all late glacial samples (see supplement for detailed p-values)." Fig. 2 modified:



Reviewer: 7. Lines 265-275, For those with p > 0.05, I don't think that there are relationships between these two proxies. So, in these cases you couldn't get some relationships for these proxies, especially when there are only very small numbers of samples.

Response: It is common practice to not interpret data with p > 0.05 as being related, but the p-value strongly depends on sample sizes. Hence, we are providing a very careful evaluation of these data sets *sensu* Wasserstein et al. (2019).

In the discussion, we now discuss the relationships more as tendencies (same as for the differences between interglacials and glacials, above).

Reviewer: 8. These proxies for wildfire reconstruction used in this study are soluble sugar materials. So, people would like to know if there is possibility that MA would move downward and thus influence

their indications for wildfire reconstruction? I think such discussions could not be missed in Section 4.1.

Response: We are discussing that now in Chap. 4.1.2 and write: "Assuming that dissolved MAs degrade within days or weeks in oxic, turbulent water (Norwood et al., 2013), the MAs recorded from previous warm periods may rather derive from MAs in particulate phase, which probably did not migrate post-depositionally."

Reviewer: 9. Lines 342-343, Please rethink about the sources and pathway of Mas since river input may be another source.

Response: Yes, we have added more discussion on potential local sources and pathways and depositional processes. Please see chap. 4.1.2 and our response above.

Reviewer: 10. Lines 354-355, I am not sure about the explanation for MA to indicate a background wildfire. I would like to suggest this might be associated (mainly) with the local biofuel availability as well.

Response: We assume that local fires might have contributed MAs only during warmer interglacials (as already today vegetation is sparse to allow fires to occur, see discussion at the beginning of chap 4.1.2). Yet, in this sentence we refer to the presence of MAs in glacial sediments – when it is even more unlikely that fires happened in an even colder and drier climate with sparse tundra steppe vegetation. To not be misleading, we have rephrased the sentences and now write "Accordingly, our rank correlation analysis shows that MA influxes tend to increase with higher amounts of tree and shrub versus tundra-steppe pollen (Figs. 3c, 4b) – *potentially allowing also fires to occur in the El'gygytgyn catchment during warmer than present interglacials (MIS 5e, 11c). In contrast, during glacials,* low temperatures and CO₂ levels limit biomass (fuel availability) and fire spread, which has be shown in previous mid- to high-latitude reconstructions and model simulations (Thonicke et al., 2005;Krawchuk and Moritz, 2011;Daniau et al., 2012;Martin Calvo et al., 2014;Kappenberg et al., 2019). Thus, El'gygytgyn's glacial MA influxes represent a background signal from remote source areas, when fires associated with high-productivity biomes have shifted southwards."

Reviewer: 11. Lines 357-360, In fact, I do not find there is a clear relationship of low-temperature wildfire with maximum summer insolation. There is only one case in MIS 8, while for others more evidences suggest that this is not the case. I think that you should be more precise.

Response: You are right, there is actually no relationship between insolation and MA influxes. We have correlated (Kendall's τ) now the MA concentrations and influxes with the summer insolation (67.5°) at the same time (basically assuming the age uncertainties are low) and found τ around 0 (-0.17 to 0.05) – but more data and a more sophisticated statistical approach would be needed to test that considering potential age and source area uncertainties, which is beyond the scope of this study.

We have rephrased this part now: "We *did not find evidence* for more biomass burning close to the lake in times of high summer insolation, *as suggested by previous* mid- to high latitude studies (Daniau et al., 2012;Remy et al., 2017;Dietze et al., 2018;Kappenberg et al., 2019). *MA influxes rather peaked during transitions from high to low summer insolation and during MIS 8 maximum insolation*, despite pollen data suggesting rather low tree and shrub presence *during MIS 8* (Fig. 3a, c). In addition, lower mean MA influxes during MIS 11c compared to MIS 5e (Fig. 2b) *might be linked* to the rather moderate summer insolation during MIS 11 compared to *a more pronounced insolation cycle* during MIS 5e (Yin and Berger, 2012). Indirectly, high latitude summer insolation could have driven biomass productivity and fires *in the source areas*, for example, by *altering* the length of the growing season,...".

Reviewer: 12. Line 396, a small error "is not adapted to is but able to survive fires" **Response: We have corrected it.**

Reviewer: 13. Line 495. A error "CO2". Response: We have corrected it to "CO₂".

Reviewer: 14. I am not expert to pollen. So, I also would like to know the meaning of the pollen and spores (such as drought or wet?). So, could you please add a brief explain the indications of the pollens and spores in Figure 3. This would facilitate people to know the climate condition. Response: We have added climate conditions for the tundra-steppe taxa that clearly represent the dry cold glacial climate but Sphagnum peatland extent does not only reflect more precipitation but can also be a response to warming and thawing of permafrost, so we would not like to link it one-to-one just to climate: "Tun.steppe (i.e. tundra and steppe taxa reflecting cold and dry glacial conditions): sum of Poaceae, *Artemisia*, Chenopodiaceae, Caryophyllaceae, Cichoriaceae, and *Thalictrum* pollen; summergreen boreal forest taxa (SGB): sum of *Larix, Populus,* and *Alnus* pollen; *Pinus* s/g. *Haploxylon*-type pollen (with SGB and *Pinus* spreading during interglacials); and the *Sphagnum* spore abundance (representative for peatlands)."

References used in the response letter

- Bird, M. I., and Cali, J. A.: A revised high-resolution oxygen-isotope chronology for ODP-668B: implications for Quaternary biomass burning in Africa, Global and Planetary Change, 33, 73-76, <u>https://doi.org/10.1016/S0921-8181(02)00062-0</u>, 2002.
- Conedera, M., Tinner, W., Neff, C., Meurer, M., Dickens, A. F., and Krebs, P.: Reconstructing past fire regimes: methods, applications, and relevance to fire management and conservation, Quaternary Science Reviews, 28, 555-576, http://dx.doi.org/10.1016/j.quascirev.2008.11.005, 2009.
- Daniau, A.-L., Sánchez Goñi, M. F., Martinez, P., Urrego, D. H., Bout-Roumazeilles, V., Desprat, S., and Marlon, J.
 R.: Orbital-scale climate forcing of grassland burning in southern Africa, Proceedings of the National Academy of Sciences, 110, 5069-5073, 10.1073/pnas.1214292110, 2013.
- Daniau, A. L., Bartlein, P. J., Harrison, S. P., Prentice, I. C., Brewer, S., Friedlingstein, P., Harrison-Prentice, T. I., Inoue, J., Izumi, K., Marlon, J. R., Mooney, S., Power, M. J., Stevenson, J., Tinner, W., Andrič, M., Atanassova, J., Behling, H., Black, M., Blarquez, O., Brown, K. J., Carcaillet, C., Colhoun, E. A., Colombaroli, D., Davis, B. A. S., D'Costa, D., Dodson, J., Dupont, L., Eshetu, Z., Gavin, D. G., Genries, A., Haberle, S., Hallett, D. J., Hope, G., Horn, S. P., Kassa, T. G., Katamura, F., Kennedy, L. M., Kershaw, P., Krivonogov, S., Long, C., Magri, D., Marinova, E., McKenzie, G. M., Moreno, P. I., Moss, P., Neumann, F. H., Norström, E., Paitre, C., Rius, D., Roberts, N., Robinson, G. S., Sasaki, N., Scott, L., Takahara, H., Terwilliger, V., Thevenon, F., Turner, R., Valsecchi, V. G., Vannière, B., Walsh, M., Williams, N., and Zhang, Y.: Predictability of biomass burning in response to climate changes, Global Biogeochemical Cycles, 26, GB4007, 10.1029/2011GB004249, 2012.
- Dietze, E., Theuerkauf, M., Bloom, K., Brauer, A., Dörfler, W., Feeser, I., Feurdean, A., Gedminienė, L., Giesecke, T., Jahns, S., Karpińska-Kołaczek, M., Kołaczek, P., Lamentowicz, M., Latałowa, M., Marcisz, K., Obremska, M., Pędziszewska, A., Poska, A., Rehfeld, K., Stančikaitė, M., Stivrins, N., Święta-Musznicka, J., Szal, M., Vassiljev, J., Veski, S., Wacnik, A., Weisbrodt, D., Wiethold, J., Vannière, B., and Słowiński, M.: Holocene fire activity during low-natural flammability periods reveals scale-dependent cultural human-fire relationships in Europe, Quaternary Science Reviews, 201, 44-56, https://doi.org/10.1016/j.quascirev.2018.10.005, 2018.
- Han, Y. M., Peteet, D. M., Arimoto, R., Cao, J. J., An, Z. S., Sritrairat, S., and Yan, B. Z.: Climate and Fuel Controls on North American Paleofires: Smoldering to Flaming in the Late-glacial-Holocene Transition, Scientific Reports, 6, 20719, 10.1038/srep20719, 2016.
- Kappenberg, A., Lehndorff, E., Pickarski, N., Litt, T., and Amelung, W.: Solar controls of fire events during the past 600,000 years, Quaternary Science Reviews, 208, 97-104, <u>https://doi.org/10.1016/j.quascirev.2019.02.008</u>, 2019.
- Keeley, J. E.: Fire intensity, fire severity and burn severity: a brief review and suggested usage, International Journal of Wildland Fire, 18, 116-126, <u>https://doi.org/10.1071/WF07049</u>, 2009.
- Krawchuk, M. A., and Moritz, M. A.: Constraints on global fire activity vary across a resource gradient, Ecology, 92, 121-132, 10.1890/09-1843.1, 2011.
- Martin Calvo, M., Prentice, I. C., and Harrison, S. P.: Climate versus carbon dioxide controls on biomass burning: a model analysis of the glacial–interglacial contrast, Biogeosciences, 11, 6017-6027, 10.5194/bg-11-6017-2014, 2014.
- Melles, M., Brigham-Grette, J., Minyuk, P. S., Nowaczyk, N. R., Wennrich, V., DeConto, R. M., Anderson, P. M., Andreev, A. A., Coletti, A., Cook, T. L., Haltia-Hovi, E., Kukkonen, M., Lozhkin, A. V., Rosén, P., Tarasov, P., Vogel, H., and Wagner, B.: 2.8 Million Years of Arctic Climate Change from Lake El'gygytgyn, NE Russia, Science, 337, 315-320, 10.1126/science.1222135, 2012.

- Norwood, M. J., Louchouarn, P., Kuo, L. J., and Harvey, O. R.: Characterization and biodegradation of watersoluble biomarkers and organic carbon extracted from low temperature chars, Organic Geochemistry, 56, 111-119, 10.1016/j.orggeochem.2012.12.008, 2013.
- Remy, C. C., Hély, C., Blarquez, O., Magnan, G., Bergeron, Y., Lavoie, M., and Ali, A. A.: Different regional climatic drivers of Holocene large wildfires in boreal forests of northeastern America, Environmental Research Letters, 12, 035005, 10.1088/1748-9326/aa5aff, 2017.
- Sang, X. F., Gensch, I., Kammer, B., Khan, A., Kleist, E., Laumer, W., Schlag, P., Schmitt, S. H., Wildt, J., Zhao, R., Mungall, E. L., Abbatt, J. P. D., and Kiendler-Scharr, A.: Chemical stability of levoglucosan: An isotopic perspective, Geophysical Research Letters, 43, 5419-5424, 10.1002/2016GL069179, 2016.
- Schreuder, L. T., Hopmans, E. C., Stuut, J.-B. W., Sinninghe Damsté, J. S., and Schouten, S.: Transport and deposition of the fire biomarker levoglucosan across the tropical North Atlantic Ocean, Geochimica et Cosmochimica Acta, <u>https://doi.org/10.1016/j.gca.2018.02.020</u>, 2018.
- Suciu, L. G., Masiello, C. A., and Griffin, R. J.: Anhydrosugars as tracers in the Earth system, Biogeochemistry, 146, 209-256, 10.1007/s10533-019-00622-0, 2019.
- Tarasov, P. E., Andreev, A. A., Anderson, P. M., Lozhkin, A. V., Leipe, C., Haltia, E., Nowaczyk, N. R., Wennrich, V., Brigham-Grette, J., and Melles, M.: A pollen-based biome reconstruction over the last 3.562 million years in the Far East Russian Arctic - new insights into climate-vegetation relationships at the regional scale, Clim. Past, 9, 2759-2775, 10.5194/cp-9-2759-2013, 2013.
- Thonicke, K., Prentice, I. C., and Hewitt, C.: Modeling glacial-interglacial changes in global fire regimes and trace gas emissions, Global Biogeochemical Cycles, 19, 10.1029/2004gb002278, 2005.
- van Leeuwen, T. T., and van der Werf, G. R.: Spatial and temporal variability in the ratio of trace gases emitted from biomass burning, Atmos. Chem. Phys., 11, 3611-3629, 10.5194/acp-11-3611-2011, 2011.
- Wasserstein, R. L., Schirm, A. L., and Lazar, N. A.: Moving to a World Beyond "p<0.05", The American Statistician, 73, 1-19, 10.1080/00031305.2019.1583913, 2019.
- Yin, Q. Z., and Berger, A.: Individual contribution of insolation and CO2 to the interglacial climates of the past 800,000 years, Clim Dyn, 38, 709-724, 10.1007/s00382-011-1013-5, 2012.

Dear John Dodson,

Thanks for your review. Please find below our response to each of the issues that you have raised together with links on how we considered your suggestions.

Reviewer: Actually, it does not cover all of the last 430 kyr and is really a record from MIS stages 5e to 12. Perhaps the title should reflect this.

Response: You are right. We have modified the title that is becomes clearer: "Relationships between low-temperature fires, climate and vegetation during three late glacials and interglacials of the last 430 kyrs in northeastern Siberia reconstructed from monosaccharide anhydrides in Lake El'gygytgyn sediments"

Reviewer: The manuscript and interpretations make a number of assumptions that could be better spelled out. Interpretation of the deeper time record is based on knowledge of contemporary ecosystems in Siberia. Hence assumption number one is that present ecology is a good analogue for the past. Yet we know from the pollen record that the boundary conditions may be different. Response: You are right that past boundary conditions were very different in the past and we are specifically testing how fire-vegetation relationships during several late glacial to interglacial periods (with different boundary conditions represented by the pollen data) have changed with insolation and global ice volume (LR04 record). Thereby, it is one of the major goals of using El'gygytgyn sediments (that comprise proxy records from different interglacials) to test if the modern fire regime – ecosystem association also holds on long time scales, the reason for the MA-pollen correlation analysis. In the discussion and interpretation, we mainly assume that known physico-chemical processes during burning would be transferable also to previous interglacials. These processes are those that determine the MA emission ratios and the physical properties of the modern ecosystems, such as a more open canopy in larch versus spruce forest and certain fire adaptive traits, which are thought to be typical for high latitude biomes.

We have phrased that now more prominently in the abstract: "Combined with pollen and non-pollen palynomorph records from the same samples, we assess *how far the modern* relationships between fire, climate, and vegetation *also persisted during the past*, on centennial to orbital time scales." and in the discussion: "One additional biomass source could be dense moss—lichen mats within the summergreen boreal forest, assuming that past ecosystem properties were similar as today".

Reviewer: I am not aware that the molecular proxies have been tested in the field or lab. So assumption 2 is that the only source of the key compounds is from low-temperature fires.... Response: A comprehensive review paper came out recently that summarizes the state-of-knowledge on anhydrosugars (MAs) in the earth system (Suciu et al., 2019), discussing all relevant processes from source to sink and confirming assumption 2. We acknowledge this review paper throughout the revised manuscript and we added concerning your question:

"A recent review of Suciu et al. (2019) attributes the dominant low temperature production to depolymerization, fragmentation and inter- and intramolecular transglycosylation during pyrolysis, while a minor fraction of MAs can attach to charcoal during higher temperatures, when smoldering overlaps or follows flaming conditions."

Reviewer: The fire record could be readily strengthened if there was some accompanying microcharcoal record from the sediments. Does such data exist?

Response: Thanks for this remark. We are currently analysing charcoal from pollen slides but so far we have microcharcoal data only from the new pollen samples of sediment core PG1351 (MIS 5e-8), but microcharcoal analysis was not included in the previously published pollen data of MIS 11-12 from the ICDP 5011-1 core (Melles et al., 2012). Our main objective is to test the potential of MAs for fire reconstructions on long timescales and we are aware that MAs and charcoal might have partially different sources, transport and degradation pathways (Clark, 1988;Dietze et al., 2019). Hence, we don't

expect identical trends from both proxies and we will discuss a charcoal- versus MA-based fire history in more detail in a next study when we have more data available for both, MAs and charcoal. We mention this now in the concluding sentence: "Further research will continue exploring lake-sedimentary MAs *in higher resolution together with further sedimentary fire proxies such as charcoal to study* low-intensity fire–climate–vegetation feedbacks in space and time and potential ways of post-depositional degradation in even older interglacials, with CO2 levels similar to those expected in the future".

Reviewer: An anonymous review asks the question about how mobile these compounds may be down a sediment profile. This is potentially a serious matter which could make the whole interpretation flawed. It may be ameliorated if the compounds become bound to say clay particles but it needs to be tested. One wonders whether these proxies

may be affected by diagenesis. The authors allude to this in their discussion (lines 280-285, and 340+). Thus, we assume any diagenetic effect is small compared to the magnitude of the changes in abundance of the key compounds. It would be useful if this was made more explicit...

Response: We have now strongly extended the discussion of potential sources, transport and degradation pathways based on the current knowledge of potential source areas, catchment and lake configuration and previously studied processes (e.g. after Suciu et al. (2019) and further studies of the El'gygytgyn expedition team members). For example, we now discuss that the MAs we detect in El'gygytgyn lake sediments might be those that have attached to particles during the production, transport or deposition (and mention this now also in the abstract already) given that all degradation processes are quick (hours to weeks) and are mainly affecting the dissolved MAs. An in-situ production of MAs by diagenesis has been described as "theoretically impossible" (Suciu et al., 2019).

We have added in the abstract: "We find that MAs *attached to particulates* were well-preserved..." and please see the extended chapter 4.1.2. In chapter 4.3.2, we also added: "El'gygytgyn MA ratios are also about two to ten times lower than those previously reported for other lake systems (Kirchgeorg et al., 2014;Schüpbach et al., 2015;Callegaro et al., 2018;Dietze et al., 2019), *suggesting that long-term post-depositional degradation could have altered MA ratios, but the direction of diagenetic decomposition was opposite to what we find, i.e. no MAN and GAL in Miocene and older deposits (Fabbri et al., 2009;Marynowski et al., 2018;Suciu et al., 2019)".*

Reviewer: In lines 200-210 mention is made that the pollen records have been harmonized. What does this mean and how was it done?

Response: As we use pollen data from two different pollen experts, the original pollen names were slightly different and partly describing pollen types in different taxonomic detail. We harmonized the pollen type names by aggregating on a higher taxonomic level (e.g. Betula alba-type and Betula nana-type pollen summed up to "Betula".

In the text, we now have specified: "Existing pollen data (Melles et al., 2012) from the same depths as MA samples of late MIS 12 and 11c were harmonized with the new pollen samples of core PG1351, *i.e. pollen types were aggregated to the highest taxonomic level identified by the two pollen experts,* to compare the same taxa in percentages."

Reviewer: There is an obvious question about how these ancient fire regimes relate to fire in the present day. There is potentially a lot to say here about any differences in natural and anthropogenic fires regimes.

Response: You are right that modern fire regimes might be strongly affected by human fire management, which we know very well from the boreal forests in general (Molinari et al., 2018;Bowman et al., 2011) and that is also the reason to analyse natural processes and fire-vegetation feedbacks in periods when humans have clearly not affected them. However, while it is certainly of strong interest to discuss the human alteration of natural fire-vegetation feedbacks in eastern Siberia, we still have a poor understanding on how humans have affected, for example, the Siberian summergreen boreal forest, as existing data is limited or site-specific (Mollicone et al., 2006;Kukavskaya et al., 2013). Yet, our data is

not suited to answer this question as we are lacking, for example, modern and Holocene lake sediment samples for comparison.

However, based on your comment, we have specified the last sentence in the abstract "extend our knowledge on long-term *natura*/fire–climate–vegetation feedbacks in the high northern latitudes" and added a motivation/explanation in the conclusion on why it is valuable to reconstruct explicitly the natural processes from previous warm periods: "Although limited in samples, we can deduce first fire–climate–vegetation relationships in north-eastern Siberia on long timescales, *which can guide towards a natural, process-based land management of the future.*"

References used in this response letter

- Bowman, D. M., Balch, J., Artaxo, P., Bond, W. J., Cochrane, M. A., D'Antonio, C. M., Defries, R., Johnston, F. H., Keeley, J. E., Krawchuk, M. A., Kull, C. A., Mack, M., Moritz, M. A., Pyne, S., Roos, C. I., Scott, A. C., Sodhi, N. S., Swetnam, T. W., and Whittaker, R.: The human dimension of fire regimes on Earth, J Biogeogr, 38, 2223-2236, 10.1111/j.1365-2699.2011.02595.x, 2011.
- Callegaro, A., Matsubara Pereira, F., Battistel, D., Kehrwald, N. M., Bird, B. W., Kirchgeorg, T., and Barbante, C.: Fire, vegetation and Holocene climate in the south-eastern Tibetan Plateau: a multi-biomarker reconstruction from Paru Co, Clim. Past Discuss., 2018, 1-33, 10.5194/cp-2018-19, 2018.
- Clark, J. S.: Particle motion and the theory of charcoal analysis: Source area, transport, deposition, and sampling, Quaternary Research, 30, 67-80, http://dx.doi.org/10.1016/0033-5894(88)90088-9, 1988.
- Dietze, E., Brykała, D., Schreuder, L. T., Jażdżewski, K., Blarquez, O., Brauer, A., Dietze, M., Obremska, M., Ott, F., Pieńczewska, A., Schouten, S., Hopmans, E. C., and Słowiński, M.: Human-induced fire regime shifts during 19th century industrialization: A robust fire regime reconstruction using northern Polish lake sediments, PLOS ONE, 14, e0222011, 10.1371/journal.pone.0222011, 2019.
- Fabbri, D., Torri, C., Simoneit, B. R. T., Marynowski, L., Rushdi, A. I., and Fabiańska, M. J.: Levoglucosan and other cellulose and lignin markers in emissions from burning of Miocene lignites, Atmospheric Environment, 43, 2286-2295, 10.1016/j.atmosenv.2009.01.030, 2009.
- Kirchgeorg, T., Schüpbach, S., Kehrwald, N., McWethy, D. B., and Barbante, C.: Method for the determination of specific molecular markers of biomass burning in lake sediments, Organic Geochemistry, 71, 1-6, http://dx.doi.org/10.1016/j.orggeochem.2014.02.014, 2014.
- Kukavskaya, E. A., Buryak, L. V., Ivanova, G. A., Conard, S. G., Kalenskaya, O. P., Zhila, S. V., and McRae, D. J.: Influence of logging on the effects of wildfire in Siberia, Environmental Research Letters, 8, 045034, 10.1088/1748-9326/8/4/045034, 2013.
- Marynowski, L., Bucha, M., Smolarek, J., Wendorff, M., and Simoneit, B. R. T.: Occurrence and significance of mono-, di- and anhydrosaccharide biomolecules in Mesozoic and Cenozoic lignites and fossil wood, Organic Geochemistry, 116, 13-22, https://doi.org/10.1016/j.orggeochem.2017.11.008, 2018.
- Melles, M., Brigham-Grette, J., Minyuk, P. S., Nowaczyk, N. R., Wennrich, V., DeConto, R. M., Anderson, P. M., Andreev, A. A., Coletti, A., Cook, T. L., Haltia-Hovi, E., Kukkonen, M., Lozhkin, A. V., Rosén, P., Tarasov, P., Vogel, H., and Wagner, B.: 2.8 Million Years of Arctic Climate Change from Lake El'gygytgyn, NE Russia, Science, 337, 315-320, 10.1126/science.1222135, 2012.
- Molinari, C., Lehsten, V., Blarquez, O., Carcaillet, C., Davis, B. A. S., Kaplan, J. O., Clear, J., and Bradshaw, R. H. W.: The climate, the fuel and the land use: Long-term regional variability of biomass burning in boreal forests, Global Change Biology, 24, 4929-4945, 10.1111/gcb.14380, 2018.
- Mollicone, D., Eva, H. D., and Achard, F.: Human role in Russian wild fires, Nature, 440, 436-437, 10.1038/440436a, 2006.
- Schüpbach, S., Kirchgeorg, T., Colombaroli, D., Beffa, G., Radaelli, M., Kehrwald, N. M., and Barbante, C.: Combining charcoal sediment and molecular markers to infer a Holocene fire history in the Maya Lowlands of Petén, Guatemala, Quaternary Science Reviews, 115, 123-131, http://dx.doi.org/10.1016/j.quascirev.2015.03.004, 2015.
- Suciu, L. G., Masiello, C. A., and Griffin, R. J.: Anhydrosugars as tracers in the Earth system, Biogeochemistry, 146, 209-256, 10.1007/s10533-019-00622-0, 2019.

Relationships between low-temperature fires, climate and vegetation during <u>three late glacials and interglacials of</u> the last 430 kyrs in northeastern Siberia reconstructed from monosaccharide anhydrides in Lake El'gygytgyn sediments

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Keywords: molecular proxies, palaeofire, larch taiga

25 Technical summary (500 characters)

Long-term climate change impacts on fire, vegetation and permafrost in the Arctic are uncertain. Here, we show the high potential of organic compounds from low-temperature biomass burning to serve as proxies for surface fires in lake deposits. During warm periods of the last 430,000 years, surface fires are closely linked to the larch taiga forest with its moss-lichen ground vegetation that isolates the permafrost. They have reduced in warm-wet,

30 spruce-dominated and cool-dry steppe environments.

Abstract. Landscapes in high northern latitudes are assumed to be highly sensitive to future global change, but the rates and long-term trajectories of changes are rather uncertain. In the boreal zone, fires are an important factor in climate–vegetation–interactions and biogeochemical cycles. Fire regimes are characterized by small, frequent, low-intensity fires within summergreen boreal forests dominated by larch, whereas evergreen boreal forests

dominated by spruce and pine burn large areas less frequently, but at higher intensities. Here, we explore the potential of the monosaccharide anhydrides (MA) levoglucosan, mannosan, and galactosan to serve as proxies of

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low-intensity biomass burning in glacial-to-interglacial lake sediments from the high northern latitudes. We use sediments from Lake El'gygytgyn (cores PG 1351 and ICDP 5011-1), located in the far north-east of Russia, and study glacial and interglacial samples of the last 430 kyrs (marine isotope stages 5e, 6, 7e, 8, 11c, 12) that had 40 different climate and biome configurations. Combined with pollen and non-pollen palynomorph records from the same samples, we assess how far the modern past relationships between fire, climate, and vegetation persisted during the past, on orbital to centennial time scales. We find that MAs attached to particulates were well-preserved in up to 430 kyrs old sediments with higher influxes from low-intensity biomass burning in interglacials compared to glacials. MA influxes significantly increase when summergreen boreal forest spreads closer to the lake, whereas 45 they decrease when tundra-steppe environments and, especially, Sphagnum peatlands spread. This suggests that low-temperature fires are a typical property characteristic of Siberian larch forests also on long timescales. The results also suggest that low-intensity fires would be reduced by vegetation shifts towards very dry environments due to reduced biomass availability, as well as by shifts towards peatlands, which limits fuel dryness. In addition, we observed very low MA ratios, which we interpret as high contributions of galactosan and mannosan from other 50 than currently monitored biomass sources, such as the moss-lichen mats in the understorey of the summergreen boreal forest. Overall, sedimentary MAs can provide a powerful proxy for fire regime reconstructions and extend our knowledge on long-term_natural fire-climate-vegetation feedbacks in the high northern latitudes.

1. Introduction

55 1.1 Background consideration

In recent decades, high northern latitudes have been experienced a warming at more than twice the rate of other regions on Earth (Serreze and Barry, 2011;IPCC, 2014). This Arctic amplification has widespread impacts on the Earth system, such as the hydrological cycle and carbon budgets (Schuur et al., 2015;Linderholm et al., 2018). The likelihood for wildfire is increasing due to warmer temperatures, more lightening-induced ignition and potential
shifts towards more flammable vegetation (Soja et al., 2007;Hu et al., 2010;Tautenhahn et al., 2016;Veraverbeke et al., 2017;Nitze et al., 2018). However, the rates and directions of fire regime and vegetation change are poorly constrained (Abbott et al., 2016).

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Climate and vegetation types shape regional fire regimes (characterized by fire frequency, intensity, severity, seasonality, area, type and amount of biomass burned; Harris et al. (2016)). Climate influences biomass availability and flammability via the length and conditions of the growing season, the type of biomass available to burn, and weather conditions that affect local soil conditions and fuel dryness affecting fire spread (Westerling et al., 2006). Accordingly, biomass burning is related to temperature variability on centennial to orbital time scales (Daniau et al., 2012;Marlon et al., 2013).

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Vegetation drives fires by developing individual plant traits, which determine flammability and regeneration strategies (Rogers et al., 2015;Feurdean et al., 2019) and lead to tight internal fire-vegetation feedbacks (Krawchuk and Moritz, 2011;Pausas et al., 2017). Modern fire regimes differ strongly in the high-northern biomes (Fig. 1), such as between tundra and Siberian summergreen and evergreen boreal forests (Wirth, 2005;Sofronov and Volokitina, 2010;Rogers et al., 2015), with less than 1 % of Arctic and Subarctic tundra being affected by fires compared to c. 8 % of eastern Siberian boreal forest (Nitze et al., 2018).

- 75 In the tundra, grasses and shrubs burn rarely and at low fire intensities, due to soil and organic matter drying-up during the short summer seasons and limited fuel availability (Krawchuk and Moritz, 2011). While a single fire has several phases of varying fire intensities, fire regimes define the larger temporal and spatial scale properties of several fire events. Regimes of low Ffire intensity refers generally refer here to low fire temperatures, low combustion efficiencies and low fire radiative power as typical for smoldering and in contrast to flaming fires
 80 (Keeley, 2009;Conedera et al., 2009;van Leeuwen and van der Werf, 2011). In Siberian summergreen boreal forests, dominated by larch (mainly *Larix gmelenii* and *L. cajanderi*) and shrub alder (*Alnus fruticosa*) (Isaev et al., 2010), frequent low-intensity surface fires burn the understorey during dry summers (Kharuk et al., 2011). These fires characterize a "pyrome" of rare, cool and small events (Archibald et al., 2013) with fires that are mainly non-stand-replacing and of incomplete combustion (Rogers et al., 2015;van der Werf et al., 2017;Chen and
- 85 Loboda, 2018). Fire is suspected to support the existence and regeneration of larch by, for example, selectively reducing regrowth and within-species competition (Kharuk et al., 2011;Zhang et al., 2011;Tautenhahn et al., 2016). In addition, larch is a fire resister with its fire-protecting bark and shedding of the deciduous foliage that limits fire spreading to the crowns (Wirth, 2005). In contrast, Siberian evergreen boreal forests are dominated by Siberian pine (*Pinus sibirica*), spruce (*Picea obovata*), and fir (*Abies sibirica*), which are fire avoiders that rarely burn and
- 90 hardly regenerate after fire, and that suppress fires due to a rather wet understorey, whereas *Pinus sylvestris* stands are highly flammable and can resist infrequent fires (Furyaev et al., 2001;Wirth, 2005;Isaev et al., 2010;Rogers et al., 2015;Tautenhahn et al., 2016). Hence, wildfires are rare, but once ignited they burn large areas at high intensities (Archibald et al., 2013) because fires can spread quickly to the crowns via the resin-rich conifer needles (Rogers et al., 2015;van der Werf et al., 2017).
- 95 Fig.1

While it is well-recognized that alternative stable states of biome configuration can be driven by fire (Lasslop et al., 2016) and can characterize the boreal forest biomes (Scheffer et al., 2012;Rogers et al., 2015), the causes, feedbacks and thresholds that lead to shifts between stable states are still debated (Tchebakova et al., 2009;Gonzalez et al., 2010;Loranty et al., 2014;Abbott et al., 2016). Recently, Herzschuh et al. (2016) proposed that fire plays an important role in long-term climate–vegetation interactions and internal system feedbacks that determine alternative stable states in high northern biomes. However, knowledge of past fire regimes and associated natural feedbacks in the high latitudes on long, centennial to orbital time scales is scarce. Previous interglacials provide analogues for a warming world (Yin and Berger, 2015) beyond human influence in contrast to the Holocene, when lightning was not the only source of ignition (Buchholz et al., 2003;Marlon et al., 2013;Veraverbeke et al., 2017;Dietze et al., 2018).

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Lake El'gygytgyn Saediment cores of Lake El'gygytgyn provide a continuous Pliocene-Pleistocene environmental record in the Arctic that suggestreflecting strong climate and vegetation shifts during the past 3.6 Myrs (Melles et al., 2012;Brigham-Grette et al., 2013;Tarasov et al., 2013;Andreev et al., 2014;Andreev et al., 2016;Wennrich et al., 2016). Lake El'gygytgyn is a meteorite impact crater lake of 110 km², a diameter of ~12 km, and maximum water depth of 170 m formed about 3.6 Myrs ago (Wennrich et al. (2016) and reference therein). The 293-183 km² large catchment and wider region of NE Siberia is dominated by volcanic and metamorphic rocks and permanently frozen Quaternary deposits below an active layer of up to 80 cm depth (Schwamborn et al., 2006). Climate conditions are cold, dry and windy, with a mean annual air temperature of c. -10°C and an annual precipitation of

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c. 180 mm (in 2002), mainly falling as snow (Nolan and Brigham-Grette, 2007). Current treeless herb tundra vegetation is composed of patches of lichens, and herbs, and grasses (mostly Poaceae, Cyperaceae) next to barren ground and a few dispersed dwarf shrubs of Salix and Betula near the lake and of Pinus pumila and Alnus in the surrounding Chukchi uplands (Lozhkin et al., 2007). The treeline towards the summergreen boreal forest is located c. 100-150 km to the south-west (Fig. 1a). In the period 2000 to 2018, no fires have occurred in the El'gygytgyn catchment and only a few in the estimated pollen source area covering several hundreds of kilometres (based on remote sensing data, after Nitze et al. (2018)).

To reconstruct long-term fire regime shifts, sedimentary charcoal can be used as a classical proxy of for fire of various combustion efficiencies (Whitlock and Larsen, 2001;Conedera et al., 2009). Yet, fire intensity reconstructions and the differentiation between surface and crown fires is difficult to specify, especially on long timescales, as well as when considering other fire proxies such as fire scars and fungal spores (Stivrins et al., 125 2019). Molecular burning proxies are currently being explored to infer source- and temperature-specific fire histories (Han et al., 2016;Kappenberg et al., 2019;Dietze et al., 2019). Unique proxies of biomass burning from low-intensity fires are the monosaccharide anhydrides (MAs) levoglucosan (1,6-anhydro-β-D-glucopyranose, LVG) and its isomers mannosan (1,6-anhydro-β-D-mannopyranose, MAN) and galactosan (1,6-anhydro-β-Dgalactopyranose, GAL). While Simoneit et al. (1999) and references therein suggest that MAs form at burn 130 temperatures > 300°C, several studies that have analysed the influence of various combustion conditions in natural samples indicated that MAs are thermal dehydration products at burning temperatures < 350°C, mainly under smouldering as opposed to flaming conditions (Pastorova et al., 1993;Gao et al., 2003;Engling et al., 2006;Kuo et al., 2008;Kuo et al., 2011). A recent review of Suciu et al. (2019) attributes the dominant low temperature production to depolymerization, fragmentation and inter- and intramolecular transglycosylation during pyrolysis, 135 while a minor fraction of MAs can attach to charcoal during higher temperatures, when smoldering overlaps with or follows flaming conditions. In Holocene lake sediments, MAs provide complementary fire proxies to sedimentary charcoal (Elias et al., 2001;Schüpbach et al., 2015;Battistel et al., 2017;Schreuder et al., 2019;Dietze et al., 2019). While LVG is preserved in marine sediment for at least the last 130 kyrs (Lopes dos Santos et al., 2013), to our knowledge, sedimentary MAs not been analysed in either high-latitude or in interglacial lake sediments.

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Here, we assess (1) how far MAs are useful proxies to study late-glacial to interglacial fire histories from during the last 430 kyrs using sedimentary MA from Lake El'gygytgyn and assess (2) discuss long-term relationships between low-temperature fires and regional vegetation in the Russian Far East (Fig. 1). We focus-selectedon three late glacial-to-interglacial periods, which include i.e. marine isotope stages (MIS) 12-11c, 8-7e, and 6-5e,- which 145 reflect varying interglacial biome types and climate conditions, as reconstructed using the pollen records from El'gygytgyn sediments have been used to reconstruct biome types and climate conditions over time (Melles et al., 2012;Tarasov et al., 2013). The so-called "superinterglacial" MIS 11c (c. 420-380 kyrs ago) has been described as warmer and wetter compared to today, which was supported by biomarker based temperature reconstructions (D'Anjou et al., 2013). The presence of spruce (Picea) pollen in El'gygytgyn sediments suggests that the evergreen boreal forest has been much closer to the lake than today (Melles et al., 2012; Tarasov et al., 2013; Lozhkin et al.,

150 2017). In contrast, MIS 7e (c. 240-220 kyrs ago) was cooler than today and only a few coniferous pollen grains were found. Birch and alder pollen suggest that shrub tundra prevailed during this interglacial (Lozhkin et al., 2007;Zhao et al., 2019). MIS 5e interglacial (c. 130-110 kyrs ago) was slightly warmer than today (Tarasov et al., 2013). Larch and alder pollen were found in El'gygytgyn sediments suggesting that a summergreen boreal forest existed close to the lake, whereas spruce pollen was absent (Lozhkin et al., 2007). These differences in regional vegetation and climate conditions have been explained by changes in global ice volume, insolation changes (for interglacial astronomical and GHG characteristics see Yin and Berger (2012) and, Table 1, for interglacial astronomical and GHG characteristics), and interhemispheric ice sheet-ocean-atmosphere feedback mechanisms (Melles et al., 2012;Lozhkin et al., 2017).

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Here, we aim to answer 1) whether sedimentary MAs are suitable proxies to reconstruct low-temperature fires even in interglacial Arctic lake sediments; and 2) whether more biomass burning occurs during interglacials compared to late glacials due to climate-driven changes in biomass availability, since wildfires are expected to be fuel-limited in cold environments (Krawchuk and Moritz, 2011;Daniau et al., 2012). Furthermore, and 3) as different vegetation composition has been reconstructed from past interglacials (Melles et al., 2012;Brigham-165 Grette et al., 2013;Andreev et al., 2014), we consider 3) whether more low-temperature biomass burning occurs in summergreen boreal forest compared to tundra or evergreen boreal forest during interglacials on centennial to millennial timescales.

2 Methods

2.1 Sample selection and preparation

170 We sampled sediment from two cores (Fig. 1b). A 12.7 m long sediment core - PG1351 - was recovered in 1998 and covers the last 270 kyrs, according to 14C- and luminescence dates, as well as magnetostratigraphy (Nowaczyk et al., 2007; Melles et al., 2007; Nowaczyk et al., 2013). A 318 m long composite core from ICDP site 5011-1 comprises three parallel sediment cores that cover the time period <u>125 to 3600 kyrs</u> (Melles et al., 2011;Wennrich et al., 2016), dated based on by magnetostratigraphy, paleoclimatic and orbital tuning with more than 600 175 chronologic age-tie points (Nowaczyk et al., 2013).

For MA analyses, we freeze-dried and homogenized 44 samples of c. 0.7-1.8 g dry sediment from core PG1351 covering late glacials and interglacials of MIS 8 to MIS 5e, integrating sediment of 1 cm core depth. Temporal resolution of these samples ranges from 140 to 960 years per sample. For the period between 430 and 405 kyrs ago (end of MIS 12 to MIS 11c), 13 samples of 0.5-1.3 g of dry sediment from ICDP core 5011-1 were taken for 180 MA analyses, integrating sediment of 2 cm core depth. Eight of these 13 samples are from the same core depths as were previously analysed for pollen (Melles et al., 2012). Temporal resolution of these samples varies between 200 and 970 years per sample comparable to core PG1351. Across all samples, temporal resolution is 333 ± 273 years per sample, giving centennial-to millennial scale averages.

We extracted the polar lipids of all MA samples using a Dionex Accelerated Solvent Extraction system (ASE 350, 185 ThermoFisher Scientific) at 100°C, 103 bar pressure and two extraction cycles (20 min static time) with 100 % methanol, after an ASE cycle with 100 % dichloromethane. For every sample sequence (n=13-18), we extracted a blank ASE cell and included it in all further steps. We added 60 ng of deuterated levoglucosan (C₆H₃D₇O₅; dLVG; Th. Geyer GmbH & Co. KG) as internal standard, and filtered the extract over a PTFE filter using acetonitrile and 5 % HPLC-grade water. We analysed the extracts with an Ultimate 3000 RS ultra-high

- 190 performance liquid chromatograph (U-HPLC) with thermostated autosampler and column oven coupled to a Q Exactive Plus Orbitrap mass spectrometer (Quadrupole-Orbitrap MS; ThermoFisher Scientific) with heated electrospray injection (HESI) probe at GFZ Potsdam, using measurement conditions adapted from earlier studies (Hopmans et al., 2013;Schreuder et al., 2018;Dietze et al., 2019). Briefly, separation was achieved on two Xbridge BEH amide columns in series (2.1 x 150 mm, 3.5 µm particle size) fitted with a 50 mm pre-column of the same
- 195 material (Waters). The compounds were eluted (flow rate 0.2 mL min⁻¹) with 100 % A for 15 minutes, followed by column cleaning with 100 % B for 15 min, and re-equilibration to starting conditions for 25 min. Eluent A was acetonitrile:water:triethylamine (92.5:7.5:0.01) and eluent B acetonitrile:water:triethylamine (70:30:0.01). HESI settings were as follows: sheath gas (N₂) pressure 20 (arbitrary units), auxiliary gas (N₂) temperature of 50 °C, spray voltage -2.9 kV (negative ion mode), capillary temperature
- 300 °C, S-Lens 50 V. Detection was achieved by monitoring *m/z* 150-200 with a resolution of 280,000 ppm. Targeted data dependent MS² (normalized collision energy 13 V) was performed on any signal within 10 ppm of *m/z* 161.0445 (calculated exact mass of deprotonated levoglucosan and its isomers) or *m/z* 168.0884 (calculated exact mass of deprotonated dLVG) with an isolation window of 0.4 *m/z*. The detection limit was 2.5 pg on column, based on injections of 0.5 to 5000 pg on column of authentic standards of LVG, MAN, and GAL (Santa Cruz Biotechnology) and dLVG.
- Integrations were performed on mass chromatograms within 3 ppm mass accuracy and corrected for relative response factors to dLVG (1.08 ± 0.10, 0.76 ± 0.10 and 0.24 ± 0.05 for LVG, MAN, and GAL, respectively), according to known authentic standard mixes injected before and after every measurement sequence and supported by characteristic isomer-specific MS² data. All samples were corrected by subtracting the maximum MA
 concentrations in the blank duplicates of each ASE sequence. To account for biases due to sediment properties and sedimentation rates, MA influxes (mass accumulation rates in ng cm⁻² yr⁻¹) were calculated by multiplying the concentrations (ng g⁻¹) with the sample-specific dry bulk densities (Melles et al., 2007;Wennrich et al., 2016), and
- concentrations (ng g⁻¹) with the sample-specific dry bulk densities (Melles et al., 2007;Wennrich et al., 2016), and the sample's sedimentation rates (cm yr⁻¹) using the age-depth models presented by Nowaczyk et al. (2007) for the PG1351 core and Nowaczyk et al. (2013) for the the PG1351 and the ICDP-5011-1 cores.
 Pollen from sediment core PG1351 was analysed by Lozhkin et al. (2007), but there was no sediment left to sample
- 213 Folder non-sediment core (G155) was analysed by Eozhen et al. (2007), but nere was no sediment et it of sample the same core depths for MA analyses. Therefore, we sampled 19 of the 44 MA sample's core depths for parallel pollen analyses to enable a direct comparison of MA and pollen records without age bias. These new pollen samples were prepared using standard pollen preparation procedures as have been used previously for the El'gygytgyn sediments (Andreev et al., 2012). In addition to pollen and spores, non-pollen-palynomorphs (NPPs)
 220 such as algae remains and coprophilous fungi spores were counted. Existing pollen data (Melles et al., 2012) from the same depths as MA samples of <u>late MIS 12</u> and 11c were harmonized with the new pollen samples of core PG1351, i.e. pollen types were aggregated to the highest taxonomic level identified by the two palynologists, to compare the same taxa in percentages.

2.2 Analyses of source areas

225 MAs are can be transported attached to aerosols in the atmosphere (Sang et al., 2016;Schreuder et al., 2018) and/or via fluvial transport from the catchment (Suciu et al., 2019). To discuss potential source areas of <u>aeolian-derived</u> MAs, we calculated exemplary backward trajectory ensembles of two days towards Lake El'gygytgyn using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015;Rolph et al., 2017) during the main boreal and tundra fire season of the summers 2017 and 2018 (May to September; <u>http://ready.arl.noaa.gov/hypub-bin/trajtype.pl?runtype=archive</u>). Trajectories considered a mean particle injection height during a wildfire of between 100 and 1000 m after Peters and Higuera (2007).

2.3 Evaluation of relationships

MA influxes and ratios were correlated with the six most indicative pollen and NPP types excluding samples with < 0.5 % of a certain taxon to stabilize the signal-to-noise ratio (Prentice et al., 1996). Evergreen boreal forest is
represented by *Picea* pollen and summergreen boreal forest (SGB) by the sum of *Larix, Alnus*, and *Populus* pollen percentages. The sum of Poaceae, *Artemisia*, Chenopodiaceae, Caryophyllaceae, Cichoriaceae, and *Thalictrum* pollen represent typical taxa of cold tundra-steppe environments. *Pinus* 6-g:S/g. *Haploxylon*-type pollen can be produced by a shrub stone pine (*Pinus pumila*), which does not survive fires, or by *P. sibirica*, a Siberian pine that can survive but is not adapted to fires (Ma et al., 2008;Keeley, 2012). *Sphagnum* spores reflect wet habitats
(peatlands), whereas *Selaginella rupestris* spores from a ledge or rock spike-moss are indicative of extremely dry and cold habitats.

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To test whether mean MA influxes of interglacial and their preceding late glacial sediments were equal (null hypothesis) or significantly higher or lower, we performed the non-parametric Wilcoxon rank sum test with determination of an exact permutation null distribution, suited for few numbers of samples. The test statistics and associated *p*-values were calculated using the function wilcox.test of the stats-package in R (Bauer, 1972). To quantify the relationships between low-temperature fires and vegetation, we log-transformed the MA ratios and pollen percentages to correct for their skewed distributions. Then, we correlated MA records with the major land cover types using the Kendall's τ rank correlation coefficient that is robust against outliers, small sample sizes, and against spurious correlations, in contrast to Pearson's correlation coefficient (Aitchison, 1986;Jackson and Somers, 1991;Arndt et al., 1999). Kendall's τ and associated *p*-values were calculated using the function corr.test of the R package psych (Revelle, 2018) using pairwise complete observations. Kendall's τ is only provided for n > 5. Please note that the reported categorisation of *p*-values in Fig. 2 and 4 does not mean that some distribution differences and relationships are more valuable or "correct" than others (Wasserstein et al., 2019), as uncertainties in the data are not restricted to statistics alone (see Chap. 4.1). Full *p*-values and test statistics are reported in the supplementary data.

3. Results

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MAs are detected well in all samples with LVG, MAN, and GAL concentrations of 77 ± 35 , 109 ± 58 , and 204 ± 129 ng g⁻¹ (mean \pm standard deviation, Fig. 2a), respectively. The standard instrumental errors from duplicate measurements are 4.9 ± 2.9 , 4.8 ± 4.1 , and 7.1 ± 5.2 % for LVG, MAN, and GAL, respectively. Blanks contained 7.6 ± 4.7 , 2.1 ± 0.7 , and 2.1 ± 0.6 % of the respective mean LVG, MAN, and GAL concentrations in the samples, derived from carryover within the ASE preparation step, and are subtracted from the concentrations of the respective sample batch.

Fig. 2

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265 Concentrations vary during interglacials and late glacial stages, with lowest values during MIS 12, although concentrations are strongly affected by the sediment bulk density and sedimentation rate time per sample (Fig. 2a vs. b). Hence, we focus here on relative changes in MA influxes, which are consistently higher during interglacials compared to the latter part of their preceding glacials, according to the boxplots and Wilcoxon rank sum test statistics-(Fig. 2b, supplementary data). Among interglacials, influxes are highest during MIS 5e (e.g., LVGmedian: 270 2.2 ng cm⁻² yr⁻¹, GAL_{median}: 4.2 ng cm⁻² yr⁻¹) and lowest during MIS 7e (LVG_{median}: 0.8 ng cm⁻² yr⁻¹, GAL_{median}: 1.5 ng cm⁻² yr⁻¹; Figs. 2b, 3). Highest late glacial MA influxes are found in MIS 6 samples (LVG_{median}: 1.2 ng cm⁻² yr⁻ ¹, GAL_{median}: 3.8 ng cm⁻² yr⁻¹), whereas MIS 12 samples have the lowest MA influxes (LVG_{median}: 0.2 ng cm⁻² yr⁻¹) ¹, GAL_{median}: 0.2 ng cm⁻² yr⁻¹, Fig. 2b). MA records reach their highest values during the peak of interglacials, with secondary maxima during MIS 8 and MIS 6 at times of high summer insolation (Fig. 3). MA influx records are 275 strongly positively correlated across all intervals (LVG vs. MAN or GAL: Kendall's $\tau = 0.76 - 0.82$, p = 0.00 - 0.020.05; MAN vs. GAL: $\tau = 0.91 - 0.97$, p < 0.001; Fig. 4a) with slightly closer relationships during MIS 8–5e compared to MIS 12-11c.

Fig. 3

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MA influx records are not correlated with MA ratios (Fig. 4a) except for MIS 12-11c samples, which show a significant inverse relationship for MAN and GAL influxes with LVG (MAN+GAL)⁻¹ (Kendall's $\tau = -0.74$ and -0.77, p = 0.05 and 0.02). Fires that produced more MAN and GAL in MIS 11c have a low isomer ratio, whereas MIS 12 samples have very low amounts of all three isomers, yet with relatively high MA ratios. MA ratios do not show elear consistent differences between interglacials and late glacial stages, with highest average ratios during MIS 12 and 7e (LVG MAN-1_{median}: 0.82 and 0.83, LVG (MAN+GAL)-1_{median}: 0.31 and 0.30) and lowest ratios in MIS 11c samples (LVG MAN⁻¹median: 0.54, LVG (MAN+GAL)⁻¹median: 0.17). Only in a single late-MIS 12 sample do LVG MAN-1 and LVG (MAN+GAL)-1 exceed 3 and 1, respectively.

Vegetation compositions as reflected by the pollen and NPP records vary between the interglacials and their preceding late glacials, but also among interglacials (Figs. 3, 5), as stated in previous studies (Lozhkin et al., 2007;Melles et al., 2012). The high proportions of Sphagnum spores during MIS 12-11c and MIS 7e suggest more 290 widespread peatlands in contrast to MIS 8 and 6-5e. The presence of spruce during MIS 11c also indicates much warmer conditions compared to MIS 7e and 5e (Lozhkin et al., 2007; Melles et al., 2012), whereas MIS 7e is the coolest interglacial considered here, as indicated by the low amount of typical summergreen boreal and high amount of tundra steppe taxa pollen (Figs. 3, 5).

Fig. 4, 5

295 Despite few parallel samples with n > 4 and pollen and spore amounts higher than 0.5 %, some linkages between MA and pollen/NPP records using Kendall's t are robust, especially when considering the periods MIS 12-11c and MIS 8-5e separately (Fig. 4b). MA influxes are positively related to the summergreen boreal taxa during MIS 5e to 8 (e.g., LVG vs. log of SGB: $\tau = 0.7$, p = 0.02), and showed a tendency towards positive association between GAL and pine ($\tau = 0.6$, p = 0.14). During late MIS 12 and MIS 11c, we find a weak negative relationship between 300 GAL and Sphagnum ($\tau = -0.65$, p = 0.09) and a tendency towards lower GAL influxes during periods of

widespread tundra steppe (e.g., GAL vs. tun.steppe: $\tau = -0.6$, p = 0.14).

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As both MA ratios show similar temporal trends, we used only LVG (MAN+GAL)⁻¹ in the correlation analysis. LVG (MAN+GAL)⁻¹ is negatively related to tundra-steppe taxa on a log-log scale ($\tau = -0.4$, p = 0.07, Fig. 5) during MIS 8–5e, whereas LVG (MAN+GAL)⁻¹ is negatively related to SGB ($\tau = -0.7$, p = 0.07) during MIS 12– 11c and has higher values during periods of high Sphagnum spore proportions ($\tau = 0.7, p = 0.047$), especially during MIS 12-11c, but also when considering all samples (Fig. 4b).

4. Discussion

4.1 Preservation of monosaccharide anhydrides in high-latitude lake sediments

The abundance of LVG and its isomers in smoke in absolute (influxes) and relative (ratios) terms depends on the 310 amount and type of biomass being burnt and on burning conditions (Engling et al., 2006;Kuo et al., 2008;Fabbri et al., 2009;Kuo et al., 2011). Yet it is unknown, whether sedimentary MAs are suitable biomass burning proxies in Arctic lake sediments on centennial to orbital timescales. Issues such as analytical uncertainties, the source of MAs (i.e. geographic source area, type of biomass burnt, burning conditions) and degradation during transport, deposition and after deposition need to be addressed in order to use the proxies further back in time. We discuss 315 the future potential of MA records in long-term high latitude fire reconstructions and present some first ideas on past fire-vegetation-climate feedbacks on orbital and centennial-to-millennial timescales, as correlation was limited by the few samples that had both MA and > 0.5 % of certain pollen/NPP taxa. Although interglacial climate cannot be discussed independently here from vegetation composition, the main climate trends inferred from pollenbased vegetation reconstructions (Melles et al., 2012; Tarasov et al., 2013; Zhao et al., 2019) as described below 320 have been supported by independent climate reconstructions based on sedimentary diatoms and biomarkers (Chapligin et al., 2012;D'Anjou et al., 2013).

4.1.1 Analytical uncertainties

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MAs are present in up to 430 kyrs old El'gygytgyn lake sediments, and could be analysed well above the detection limit using UHPLC-HRMS and-with average instrumental standard errors of 5-7 % (based on duplicate measurements). LVG influxes are in the same order of magnitude as found in other younger lake sediments from temperate and tropical regions (Schüpbach et al., 2015;Battistel et al., 2017;Callegaro et al., 2018;Dietze et al., 2019). This suggests that emission and preservation conditions of LVG in high latitude sediments are comparable with temperate and tropical regions and that influx calculations based on the age-depth models of lake sediment cores PG1351 and 5011-1 are reasonable, despite absolute age uncertainties that are several hundreds of years

330 (Nowaczyk et al., 2007;Nowaczyk et al., 2013).

> We find a trend towards lower average MA influxes in older sediments (Fig. 2b), which is not as prominent when comparing the trajectories of MA influxes in time (Fig. 3a). This could indicate either a true signal of past fire activity or a certain degree of post-depositional degradation, or a combination of both aspects. As there is no temporal-long-term trend visible in MA ratios (Fig. 2c), the strong-positive relationship between MA isomers (Fig. 4a) suggests that either they derive from the same source and/or have been degraded in a similar way during transport and after deposition.

4.1.2 Potential source areas and degradation pathways from source to sink

While some formation and transport mechanisms are still uncertain (Suciu et al., 2019), MAs are thought be produced as gases during pyrolysis under low burning temperatures, but quickly condensate on co-emitted particulate matter in the smoke plume. A small fraction of MAs can remain at the site and potentially adsorb to char during higher burning temperatures (c. 600° C), becoming available for subsequent transport by overland flows (Suciu et al., 2019). MAs oxidize via several degradation pathways during atmospheric transport and/or get lost by wet or dry deposition within hours to few days (see Suciu et al. (2019) for a review). Hence, Wwe expect higher influxes when fires happen close to the lake because under atmospheric conditions, several degradation pathways limit chemical stability of MAs during aeolian transport to a few hours to days (Sang et al., 2016;Suciu et al., 2019). In central European lake sediments, large fire episodes c. 20-100 km away from the deposit could be traced by robust MA peaks (Dietze et al., 2019). Here, we discuss the potential local and regional to extra-regional MA source areas, transport pathways and post-depositional degradation in El'gygytgyn lake sediments.

Local sources would require biomass burning within the lake catchment, which is rather small (183 km²) compared
 to the 110 km² large lake (Nolan and Brigham-Grette, 2007). In the period 2000 to 2018, no fires have occurred in the El'gygytgyn catchment and only a few in the estimated pollen source area covering several hundreds of kilometres (based on remote sensing data, after Nitze et al. (2018)). The sparse tundra vegetation is currently limited to slopes below 5° (Nolan and Brigham-Grette, 2007), which is c. 55 % of the catchment. Hence, we suggest that during interglacials of similar or cooler conditions (e.g. during MIS 7e) catchment fires were highly unlikely. During warmer interglacials with a potential spread of boreal tree taxa towards the lake (Melles et al., 2012), we cannot exclude a certain contribution from local fires, but given the size of the lake we assume that the majority of MAs in El'gygytgyn sediments derives from extra-local aeolian transport.

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Our HYPSPLIT backward trajectories (Fig. 6) show that MAs attached to aerosols would derive from modern day tundra and larch taiga in the Chukotka region, several hundreds of km away from the lake, transported by north-to south-westerly winds during the main fire season in July and August, in agreement with modern climatology (Mock et al., 1998). Yet, El'gygytgyn MA influx records represent centennial-scale averages integrating over multiple fire events under multiple synoptic conditions that varied in the past, such as changing jet stream position and orientation (Herzschuh et al., 2019). Hence, the potential source area could have been even larger during past integralicials, but still be located in the vegetated realm of eastern Siberia. We assume that shifts in geographic source areas associated with shifts in atmospheric circulation would affect the variability of MA influxes within rather than between integlacials.

Fig. 6

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Beyond dry deposition, rain and snowfall can deposit MAs directly on the (frozen or unfrozen) lake surface and/or within the lake catchment. Snow and lake ice were found to be covered with partially black particulate matter (Melles et al., 2005), with snowmelt and ice-break up currently happening from mid-May to early July (Nolan et al., 2002), i.e. overlapping with the onset of the boreal fire season. Snowmelt causes high-energetic fluvial transport across the alluvial fans that drain the catchment for few days and provide most of the detrital sediments to lake El'gygytgyn (Nolan and Brigham-Grette, 2007). The amount of local aeolian reworking of sediment from barren to sparsely vegetated surfaces during the snow-free season is unknown, but wind intensities are high throughout the year (Nolan and Brigham-Grette, 2007;Fedorov et al., 2013). Hence, a certain amount of MAs from

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within the catchment could have reached the lake in either dissolved or particulate form, e.g. adsorbed to clays (Suciu et al., 2019).

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During interglacials, rather short residence times on snow and quick transport of MAs as part of the turbulent discharge of suspended matter in the main channels (Wennrich et al., 2013) could have a) prevented an important contribution (and potential loss) of dissolved MAs to the local groundwater and b) limited degradation during short distance fluvial transport (Hunsinger et al., 2008;Suciu et al., 2019). During warmer interglacials (MIS 5e and MIS 11c), a denser local vegetation coverage would have prolonged the snowmelt period and reduced the fluvial transport energies, also during rainstorms in summer. These effects would reduce the absolute fluvial and aeolian influxes of MAs from the catchment in warmer compared to cooler interglacials, which might be counterbalanced 385 by an increased likelihood for more local fires during warmer interglacials. During glacials, climate and vegetation reconstructions suggest a dry and windy climate with a multi-year to perennially frozen lake surface and limited local runoff (Nolan et al., 2002; Melles et al., 2007; Melles et al., 2012), limiting also the influxes of MAs. Aeolian material, including extra-regional MAs, would experience much slower deposition times via moats and cracks as currently observed in Antarctica (Rivera-Hernandez et al., 2019), with enough time for certain, not well-390 constrained cryogenic processes to degrade MAs (Suciu et al., 2019).

MA degradation could stillcan also happen in the lake water column or after deposition. Recently, Schreuder et al. (2018) found that LVG was transported and settled attached to organic matter, which might have prevented its degradation within the marine water column, despite its water-solubility., whereas In contrast, Norwood et al. (2013) suggested there is a substantial MA degradation in well-oxygenated river water, which, however, could 395 have been limited when MAs deposit quickly (Suciu et al., 2019). Yet, degradation and desorption of MAs at the sediment-water interface could still be substantial (Schreuder et al., 2018), especially under aerobic conditions (Knicker et al., 2013), as MAs are anhydrous sugars and, thus, potentially more labile and mobile than other organic compounds.

Monitoring and sediment properties of Lake El'gygytgyn suggest rapid depositional processes in a turbulent, wind-400 mixed water column (Wennrich et al., 2013)_and-with well-oxygenated bottom waters during summers and past warm periods_(Wennrich et al., 2013; Melles et al., 2012).7 whereas dDuring glacial periods long-term lake stratification led to rather anoxic bottom water conditions that improved the preservation of total organic carbon (Melles et al., 2007; Melles et al., 2012). Assuming that dissolved MAs degrade within days or weeks in oxic, turbulent water (Norwood et al., 2013), the MAs recorded from previous warm periods may rather derive from 405 MAs in particulate phase, which probably did not experience post-depositional migration and which also cannot be produced by diagenesis (Suciu et al., 2019). If we assume a constant influx of particulate phase MAs and high organic matter degradation at the lake bottom, we would expect higher preservation during glacials than interglacials - but we find higher MA influxes in interglacial sediments (Fig. 2b), similar to total organic carbon percentages (Melles et al., 2012). Hence, we assume that MA degradation was limited even-over longer time, when 410 occluded within or adsorbed to a mineral matrix or iron oxides (Lalonde et al., 2012;Hemingway et al., 2019). MAs are known to adsorb well to minerals and organic particles such as co-emitted soot and are chemically prone to form organo-metal complexes via chelation <u>(Tobo et al., 2012;Suciu et al., 2019)</u>, Adsorption (protecting) and desorption (destabilizing) processes can happen already during emission, transport and at the sediment-water interface, most likely in rather short time of days to weeks (Suciu et al., 2019) and may vary with climate

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conditions, as described above. These short-term processes would affect all isomers in a similar way, <u>MAs are known to adsorb well to particles (Tobo et al., 2012;Suciu et al., 2019)</u>, and all isomers show <u>indeed</u> the same trends <u>withover centennial and orbital</u> time <u>scales</u> (Fig. 2b), which is indicative for bound and protected compounds (Hemingway et al., 2019). Hence, we <u>assume-propose</u> that <u>MA influxes from</u> El'gygytgyn sediments<u>-based MA influxes represent particle-bound MAs that</u> are only marginally affected by degradation on centennial to orbital timescales, and, instead, represent relative changes in biomass burnt from a <u>similar regional</u> source during low-intensity fires.

4.2 Long-term relationships between low-intensity fires, climate, and vegetation

4.2.1 Orbital-scale fire occurrence and the role of biomass and fuel availability, insolation and climate

425 More low-intensity biomass burning occurs during interglacials compared to glacials, as we find a tendency towards higher MA influxes during interglacials compared to the preceding late glacials. Considering source areas and transport pathways (Sect. 4.1.2), we suggest that during interglacials more biomass has burnt closer to Lake El'gygytgyn (Fig. 2b). Absolute biomass availability and land cover change cannot be assessed quantitatively from pollen percentage data due to (partly) unknown taxa-specific pollen and NPP dispersal, productivity, and 430 preservation (Sugita, 2007) and our low sample resolution prevents the use of new quantitative land cover reconstructions (Theuerkauf and Couwenberg, 2018). YetHowever, more biomass availability is suggested by significantly higher abundance of tree and shrub pollen in interglacial sediments compared to glacial cold tundrasteppe assemblages dominated by herbs and grasses (Fig. 5) (Melles et al., 2012; Tarasov et al., 2013). Accordingly, our rank correlation analysis shows that MA influxes tend to increase with higher amounts of tree and shrub versus 435 tundra-steppe pollen (Figs. 3c, 4b) - potentially allowing also fires to occur in the El'gygytgyn catchment during warmer-than-present interglacials (MIS 5e and MIS 11c). Reasons could be that during During glacials, in contrast, low temperatures and CO2 levels limit biomass (fuel availability) and fire spread, which has be shown in previous mid- to high-latitude reconstructions and model simulations (Thonicke et al., 2005;Krawchuk and Moritz, 2011;Daniau et al., 2012;Martin Calvo et al., 2014;Kappenberg et al., 2019). Thus, glacial Lake El'gygytgyn's 440 glacial MA influxes represent a background signal from remote source areas, when fires associated with highproductivity biomes have shifted southwards.

We find did not find also evidence for more biomass burning close to the lake in times of high summer insolation, in agreement withas suggested by previous mid- to high latitude studies (Daniau et al., 2012;Remy et al., 2017;Dietze et al., 2018;Kappenberg et al., 2019). This-Yet, is visible during the period of MIS 8–5e, when MA
influxes not only rather peaked during transitions from high to low summer insolation and during the MIS 8 insolation maximum during MIS 7e and 5e, but also during maximum summer insolation of the preceding MIS 8 and 6 late glacial stages, despite pollen data suggesting rather low tree and shrub presence during these intervals MIS 8 (Fig. 3a, c). In addition, lower mean MA influxes during MIS 11c compared to MIS 5e (Fig. 2b) might be linked to the rather moderate summer insolation during MIS 11 compared to a more pronounced insolation cycle during MIS 5e (Yin and Berger, 2012). OverallIndirectly, increased high latitude summer insolation could haveseems to driven more biomass productivity and fires in the source areas, for example, by altering the length

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of the growing season, but also by affecting land-atmosphere feedbacks and potential interhemispheric ice-oceanland feedbacks that alter regional precipitation-evaporation patterns (Yin and Berger, 2012;Melles et al., 2012;Martin Calvo et al., 2014).

455 While it is difficult to fully disentangle biomass availability and climate conditions, we suggest that climate was the main driver of boreal forest fires on orbital timescales. This confirms previous studies from Holocene reconstructions, wheren, depending on regional biome configurations, wildfires have increased with increasing temperature and reduced precipitation patterns (Anderson et al., 2006; Remy et al., 2017; Molinari et al., 2018).

4.2.2 Centennial- to millennial scale fire occurrence and the role of climate and vegetation composition

- 460 The differences in MA influxes among interglacials seem to reflect centennial to millennial-scale temperature and moisture changes, as we find the lowest interglacial MA influxes during MIS 7e, known to be the coolest of the three interglacials considered here, when pollen of a birch and willow shrub tundra prevailed with little abundance of summergreen boreal and pine pollen (Lozhkin et al., 2007;Zhao et al., 2019) (Figs. 2b, 3a, 5). Yet, insolation and temperature control alone cannot explain the lower MA influxes during the "super-interglacial" MIS 11c 465 compared to MIS 5e.

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Regional moisture availability and biome configuration, hence fuel composition, seem to affect low-temperature fire occurrence on centennial-to millennial scales for two reasons. First, we find a negative relationship of MA influxes with Sphagnum spores across all samples and especially during late MIS 12-and early MIS 11c (Fig. 4b), when Sphagnum spore abundance was highest (Fig. 5), indicating widespread peatlands. The presence of spruce pollen also confirmed that MIS 11c was wetter than MIS 5e (Melles et al., 2012), suggesting that fuel wetness

limits fires on centennial time scales.

Second, most biomass was burnt (i.e. highest MA influxes) during the warm and dry MIS 5e, which was dominated by summergreen boreal (larch) and pine forest (Lozhkin et al., 2007; Melles et al., 2012). Our rank correlation analysis suggests a significant positive correlation of MA influxes with summergreen boreal tree pollen during MIS 8-5e (Fig. 4b). In addition, during the late MIS 12 and early MIS 11c, MA influxes peaked at the time of

- 475 high summergreen boreal pollen, before spruce pollen reached their maximum and after the maximum in Sphagnum spore abundance (Figs. 3a, c). As there was also no relationship between spruce pollen and MA influxes (Fig. 4b), we suggest that evergreen spruce forest was not an important source of MAs on long timescales, in contrast to summergreen boreal forest.
- 480 There is also a tendency towards more higher MA influxes with increasing pine pollen abundance during MIS 8-5e (Fig 3 a, c), whereas during MIS 12-11c Pinus s-g-s/g. Haploxylon-type pollen was not or weakly negatively related to MA influxes (t not significant, Fig. 4b), maybe because different pine species with different fire-related traits could have produced the P. s.e.s./g. Haploxylon-type pollen during the two periods. The shrubby P. pumila that dominates in modern treeline ecotones does not survive frequent low-intensity surface fires, whereas P. 485 sibirica, occurring in evergreen and high-elevation boreal forest, is not adapted to is but able to survive fires (Ma et al., 2008;Keeley, 2012). Pinus s.g.s/g. Diploxylon-type pollen, derived from P. sylvestris, are almost not presentabsent in Lake El'gygytgyn sediments. However, as independent high-resolution climate proxy data is

lacking for our samples, we cannot fully disentangle the role of vegetation composition and centennial- to millennial-scale climate conditions.

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4.3.2 Centennial- to millennial scale burning conditions: -Importance of the understorey

The relationships between MA influxes, summergreen- and evergreen boreal taxa and Sphagnum are independently confirmed by MA ratios that depend on the type of biomass burnt and on burning conditions such as duration (Engling et al., 2006;Kuo et al., 2008;Fabbri et al., 2009;Kuo et al., 2011). Before relating the MA influx-based with the MA ratio-based evidence, we have to state that, surprisingly, El'gygytgyn MAN and GAL 495 influxes were up to ten times higher than LVG influxes during all periods, and LVG MAN-1 and LVG (MAN+GAL)⁻¹ ratios were much lower than 3 and 1, respectively (Fig. 2c). This is in contrast to previous observations and experimental burnings (Fig. 2d) that report MA ratios of about an order of magnitude higher than those found here (Oros and Simoneit, 2001b, a;Oros et al., 2006;Iinuma et al., 2007;Fabbri et al., 2009). El'gygytgyn MA ratios are also about two to ten times lower than those previously reported for other lake systems 500 (Kirchgeorg et al., 2014;Schüpbach et al., 2015;Callegaro et al., 2018;Dietze et al., 2019). suggesting that longterm post-depositional degradation could have altered MA ratios. However, the direction of diagenetic decomposition was opposite to what we find, i.e. no MAN and GAL in Miocene and older deposits (Fabbri et al., 2009;Marynowski et al., 2018;Suciu et al., 2019)- As biomass differs in its relative proportions of cellulose (LVG precursor), hemicellulose (MAN and GAL precursor), lignin and others components (Simoneit et al., 505 1999;Simoneit, 2002), the emission rates of MA isomers differ, with a tendency towards highest MA ratios (i.e. high LVG production) in grasses, followed by deciduous hardwood trees, whereas coniferous softwood trees are characterized by and relatively low MA ratios in coniferous softwood trees (Oros and Simoneit, 2001b, a;Engling et al., 2006;Oros et al., 2006;Schmidl et al., 2008;Jung et al., 2014).

Only Oros and Simoneit (2001b) and Otto et al. (2006) report similarly low MA ratios from burning of a mixed
 bark, needle, cone and wood sample of a temperate pine (*Pinus monticola*) and from a charred pine cone (*P. banksiana*), respectively. Yet, these softwood tree species are not present in Siberia and there was-is no relation between MA ratios and pine pollen during MIS 8–5e, and-but a tendency towards higher ratios LVG (MAN+GAL)⁻¹ when with pine-increased pine during MIS 12–11c – which cannot explain the low El'gygytgyn MA ratios. There was also no relationship between MA ratios and spruce pollen in our samples, supporting the suggestion from MA
 influx-based evidence that evergreen conifers do not significantly influence El'gygytgyn MA records.

In the studied MIS 12–11c samples, *Sphagnum* is significantly positively related to LVG (MAN+GAL)⁻¹, with highest ratios in times of highest *Sphagnum* abundance, but also in times of very low, background MA influxes (Fig. 3b, c; 4b). LVG MAN⁻¹ ratios in modern-day aerosols of peatland fires in Russia were found to be > 7 (Fujii et al., 2014), that is, higher than softwood-derived MA ratios, but lower than those from hardwoods and grasses (Fig. 2d) – which cannot explain the low El'gygytgyn MA ratios.

Instead, we find a significant negative correlation between El'gygytgyn MA ratios and the summergreen boreal pollen sum during MIS 12–11c, confirming reports that burning of larch wood produce relatively higher levels of GAL (= lower LVG (MAN+GAL)⁻¹ ratios) than other softwoods (Schmidl et al., 2008). The reported larch LVG MAN⁻¹ ratios, however, are still about 1.8 to 5 times higher than <u>what we have measuredours</u>. In addition, we find low ratios across all samples, mainly independent of high or low influxes. Hence, we hypothesise that an additional 14

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biomass source and/or specific burning conditions have contributed to the low sedimentary MA ratios found in Lake El'gygytgyn sediments.

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One additional biomass source could be dense moss–lichen mats within the summergreen boreal forest., <u>assuming</u> that past ecosystem properties were similar as today. There, <u>L</u>ight can penetrate more easily through <u>its the</u> open canopy <u>of summergreen</u> compared to evergreen boreal forest, enabling the understorey to dry up quickly during summer droughts. The more open the canopy is, the denser the moss–lichen mats in the understorey become, providing several percent of the total biomass and carbon stored in NE Siberian larch forests and <u>helping</u> <u>supporting theto</u> insulatione the <u>underlain</u> permafrost (Isaev et al., 2010;Loranty et al., 2018). Together with larch needles and deadwood, moss–lichen layers are highly flammable, promoting fast surface fires, because winds can penetrate easily to the forest floor under an open canopy (Sofronov et al., 2004). Although we could not find any literature on MA emissions after burning of mosses (other than *Sphagnum*) and lichens, their cell walls are also composed of cellulose and hemicellulose (Honegger and Bartnicki-Garcia, 1991;Roberts et al., 2012) – suggesting them as a likely source of MAs. As cellulose and hemicellulose have slightly different thermal stabilities (Yang et al., 2007), different proportions in moss–lichen mats might have favoured the release of hemicellulose-derived MAN and GAL compared to LVG favoured in woody biomass burning.

Specific burning conditions can favour MAN and GAL over LVG release. In experiments, MAN and GAL reach higher yields at slightly lower temperatures (c. 200°C) compared to LVG (c. 250°C), with a strong decline in overall MA yields at temperatures higher than 350°C (Engling et al., 2006;Kuo et al., 2008;Kuo et al., 2011;Knicker et al., 2013). Although average burn temperatures of low-intensity surface fires in larch forests are unknown from low-intensity surface fires in larch forests, the suggested high speed of surface fires under open canopies (Sofronov et al., 2004) would decrease burn durations, with absolute MA yields increasing during shorter burn durations (Engling et al., 2006;Kuo et al., 2011). After burning of the moss–lichen layer, rejuvenation of larch and tree growth is promoted by active–layer thickening and nutrient release for several decades post fire, until moss–lichen mats recover (Loranty et al., 2018).

- 550 Overall, all samples and sedimentary MA influxes and ratios reported here are integrated over centennial to millennial timescales. Together with the MA influx-based evidence (i.e. no significant differences and very low MA ratios across glacial-interglacial periods, Fig. 2c), we propose that during all times there was a high contribution of MAs from low-temperature surface fires in summergreen larch forest presumably including the burning of moss–lichen mats of currently unknown MA emissions ratios. In periods of low MA influxes (background influxes), higher MA ratios probably included more burning residues from remote grass, peatland and forest fires, whereas higher influxes suggest that summergreen boreal fires happened closer to Lake El'gygytgyn with the northward spread of larch forest being well-documented in MIS 11c and 5e pollen records (Lozhkin et al., 2007;Melles et al., 2012;Lozhkin et al., 2017).
- Considering a future warming and wetting of the high northern latitudes (Hoegh-Guldberg et al., 2018), we would
 expect an increase in the availability of flammable biomass on long timescales, but low-intensity fire might decrease when fuel moisture exceeds a certain threshold either by transition from a stable forest state to peatlands or by shifts from summergreen to evergreen boreal forest despite potentially increasing fire ignitions (Veraverbeke et al., 2017).

5. Conclusion

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- 565 Molecular proxies are increasingly being used in palaeoenvironmental studies, providing insights into past biogeochemical cycles during periods that provide natural analogues of the expected future regional change. Here, we have shown the potential of MA influxes and ratios in high northern lake sediments as proxies for the amount and type of low-intensity biomass burning. Although limited in samples, we can deduce first fire-climate-vegetation relationships in north-eastern Siberia on long timescales, which can guide towards a natural, process-based land management of the future.
 - MAs can be measured well above the detection limit using UHPLC-HRMS on sediment samples of Lake El'gygytgyn for three previous glacial-to-interglacial periods and seem little affected by degradation. A declining trend in MA influxes with time is thought to represent the past amount of biomass burnt during lowtemperature fires that can be related to climate, regional biomass availability and biomass composition on orbital and centennial- to millennial-timescales.
 - Low-temperature fires are an important component of the fire regime and biogeochemical cycles of modern Siberian larch forests (Kharuk et al., 2011;Rogers et al., 2015;Chen and Loboda, 2018): a relationship that seems to hold on centennial to millennial timescales during past interglacials.
- Relatively higher MA influxes during interglacials and times of high summer insolation suggest that low-temperature fires are closely linked to biomass availability and climate conditions that favour fuel dryness on orbital timescales. Differences between interglacials are revealed by higher MA influxes when summergreen boreal forest has spread closer towards Lake El'gygytgyn, although there is no clear relationship to evergreen coniferous taxa.
- Surprisingly high influxes of MAN and GAL compared to LVG (i.e. low MA ratios across all periods) cannot be explained solely by woody biomass burning. We hypothesize that MA can serve as a proxy for fuels that derive primarily from understorey and moss–lichen mats, typical of open-canopy larch forests, that have shifted their geographic distribution southwards during glacial times.

Further research will continue exploring lake-sedimentary MAs in higher resolution together with further sedimentary fire proxies such as charcoal and to study low-intensity fire-climate-vegetation feedbacks in space
 and time and potential ways of post-depositional degradation in even older interglacials, with CO₂ levels similar to those expected in the future.

Author contributions. The study was conceptualized by ED, UH, and KM. Method development and lab analysis were performed by ED, CK, and AA, with support by ECH, LTS, OR, DS. ED analysed the data, with further data support by VW. ED wrote the manuscript with contributions from all authors.

Data availability.- Data will be available in the PANGAEA data repository.

Competing interests. The authors declare that they have no conflict of interest.

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605 Figure Captions

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Fig. 1: (a) Distribution of high-northern summer- and evergreen boreal forest (light and dark green, respectively) and location of Lake El'gygytgyn (red star). Black lines roughly mark approximate modern continuous permafrost extent (after Williams and Ferrigno (2012)), land cover classification based on data from ESA Climate Change Initiative Land cover project, land cover CCI, provided via the Centre for Environmental Data Archival (CEDA).
(b) Lake El'gygytgyn and location of analysed sediment cores (map based on Landsat-7 image, courtesy of the U.S. Geological Survey).

Fig. 2: Ranges of monosaccharide anhydride (MA) composition during selected marine isotope stages (<u>MIS</u>) at an orbital timescale: (a) levoglucosan (LVG), mannosan (MAN), and galactosan (GAL) concentrations; (b) influxes and (c) ratios of the three isomers. Boxplots show median, interquartile ranges (IQR: box), 1.5 x the IQR (whisker) and extreme outliers (ticks outside of whiskers) of samples that cover different time spans; MIS 12: 430–424 kyrs

and extreme outliers (ticks outside of whiskers) of samples that cover different time spans; MIS 12: 430–424 kyrs (n = 5), 11c: 422–406 kyrs (n = 8), 8: 256–246 kyrs (n = 5), 7e: 242–232 kyrs (n = 16), 6: 145–134 kyrs (n = 5), 5e: 132–117 kyrs (n = 18), with blue (orange) boxes marking late glacial (interglacial) periods. (d) Modern MA ratios from observations in aerosol and experimental burning after Fabbri et al. (2009), for comparison with (c). Lines between interglacial and preceding late glacial boxplots indicate different mean values for the two periods according to a non-parametric Wilcoxon test, with stars and + indicating respective *p*-values (see supplement for more details).

Fig. 3: Records of low-intensity fires and vegetation at a centennial timescale, (a) MA influxes against summer insolation (after Laskar et al. (2004)); (b) MA ratios against a proxy for past ice-sheet extent (after Lisiecki and Raymo (2005)); and (c) selected pollen records (MIS 6–5e, 8–7e: this study; MIS 12–11c (after Melles et al. (2012)). Tun.steppe (i.e. tundra and steppe taxa reflecting cold and dry glacial conditions): sum of Poaceae, *Artemisia*, Chenopodiaceae, Caryophyllaceae, Cichoriaceae, and *Thalictrum* pollen; summergreen boreal forest taxa (SGB): sum of *Larix, Populus,* and *Alnus* pollen; *Pinus* s-g-s/g. *Haploxylon*-type pollen (with SGB and *Pinus* spreading during interglacials); and the *Sphagnum* spore abundance (representative for peatlands).

Fig. 4: Kendall's t rank correlation coefficients between (a) MA influxes (LVG.yr, MAN.yr, GAL.yr) and ratios
 (L.M = LVG MAN⁻¹; L.MG = LVG (MAN+GAL)⁻¹) and (b) selected influx and ratio record and selected pollen records. SGB: pollen sum of summergreen boreal forest taxa; Tun.steppe: sum of indicative taxa for (typical glacial) tundra-steppe environment, for further taxa see text and Fig. 3.

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Fig. 5: Boxplots of vegetation composition for the marine isotope stages of interest based on pollen samples. MIS 6–5e, 8–7e: this study; MIS 12–11c: after Melles et al. (2012), SGB: sum of *Larix*, *Populus*, and *Alnus* pollen; *Pinus* = *P*. <u>s-g-s/g</u>. *Haploxylon*-type pollen; trees & and shrubs: sum of *Betula*, SGB, *Salix*, *Pinus*, and *Picea*; MIS 12: 430–424 kyrs (n = 4), <u>MIS</u> 11c: 423–406 kyrs (n = 10), <u>MIS</u> 8: 272–243 kyrs (n = 11), <u>MIS</u> 7e: 242–193 kyrs (n = 21), <u>MIS</u> 6: 190–134 kyrs (n = 37), <u>MIS</u> 5e: 132–110 kyrs (n = 25), with blue (orange) boxes marking late glacial (interglacial) periods.

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Fig. 6: Aerosol backward trajectories of summer 2018 as examples of potential modern analogues for source areas of burning residues being deposited at-<u>in</u> Lake El'gygytgyn (star) during interglacial summers. The HYSPLIT transport and dispersion model was kindly provided by the NOAA Air Resources Laboratory and the READY website (http://www.ready.noaa.gov).

References

- 645 Abbott, B., W., Jones, J., B., Schuur, E., A. G., Chapin, F. S., Bowden, W. B., Bret-Harte, M. S., Epstein, H. E., Flannigan, M., D., Harms, T. K., Hollingsworth, T. N., Mack, M. C., McGuire, A. D., Natali, S. M., Rocha, A. V., Tank, S. E., Turetsky, M. R., Vonk, J. E., Wickland, K. P., Aiken, G. R., Alexander, H. D., Amon, R. M. W., Benscoter, B. W., Bergeron, Y., Bishop, K., Blarquez, O., Bond-Lamberty, B., Breen, A. L., Buffam, I., Cai, Y., Carcaillet, C., Carey, S. K., Jing, M. C., Han, Y. H. C.,
- 650 Christensen, T. R., Cooper, L. W., Cornelissen, J. H. C., de Groot, W. J., DeLuca, T. H., Dorrepaal, E., Ned, F., Jacques, C. F., Bruce, C. F., Nancy, H. F. F., Sylvie, G., Martin, P. G., Scott, J. G., Johann, G. G., Laura, G., Paul, G., Laodong, G., Higuera, P. E., Larry, H., Feng Sheng, H., Gustaf, H., Elchin, E. J., Randi, J., Jill, F. J., Jan, K., Eric, S. K., Gerhard, K., Ryan, K., Frida, K., George, W. K., Pirkko, K., Jari, K., Peter, K., Hjalmar, L., Isabelle, L., Robie, W. M., Paul, J. M., Pertti, J. M., James, W. M., Ulf, M., Steven, F. O.,
- David, O., David, P., Marc-André, P., Payette, S., Peng, C., Pokrovsky, O. S., Rastetter, E. B., Raymond, P. A., Raynolds, M. K., Rein, G., Reynolds, J. F., Robards, M., Rogers, B. M., Schädel, C., Schaefer, K., Schmidt, I. K., Shvidenko, A., Sky, J., Spencer, R. G. M., Starr, G., Striegl, R. G., Teisserenc, R., Tranvik, L. J., Virtanen, T., Welker, J. M., and Zimov, S.: Biomass offsets little or none of permafrost carbon release from soils, streams, and wildfire: an expert assessment, Environmental Research Letters, 11, 034014, 2016.
 - Aitchison, J.: The statistical analysis of compositional data, Chapham and Hall, London, New York, 1986.

Anderson, R. S., Hallett, D. J., Berg, E., Jass, R. B., Toney, J. L., de Fontaine, C. S., and DeVolder, A.: Holocene development of Boreal forests and fire regimes on the Kenai Lowlands of Alaska, The Holocene, 16, 791-803, 10.1191/0959683606hol966rp, 2006.

- Andreev, A. A., Morozova, E., Fedorov, G., Schirrmeister, L., Bobrov, A. A., Kienast, F., and Schwamborn, G.: Vegetation history of central Chukotka deduced from permafrost paleoenvironmental records of the El'gygytgyn Impact Crater, Clim. Past, 8, 1287-1300, 10.5194/cp-8-1287-2012, 2012.
- 670 Andreev, A. A., Tarasov, P. E., Wennrich, V., Raschke, E., Herzschuh, U., Nowaczyk, N. R., Brigham-Grette, J., and Melles, M.: Late Pliocene and Early Pleistocene vegetation history of northeastern Russian Arctic inferred from the Lake El'gygytgyn pollen record, Clim. Past, 10, 1017-1039, 10.5194/cp-10-1017-2014, 2014.
- Andreev, A. A., Tarasov, P. E., Wennrich, V., and Melles, M.: Millennial-scale vegetation changes in
 the north-eastern Russian Arctic during the Pliocene/Pleistocene transition (2.7–2.5 Ma) inferred
 from the pollen record of Lake El'gygytgyn, Quaternary Science Reviews, 147, 245-258,
 https://doi.org/10.1016/j.quascirev.2016.03.030, 2016.

Archibald, S., Lehmann, C. E. R., Gómez-Dans, J. L., and Bradstock, R. A.: Defining pyromes and global syndromes of fire regimes, Proceedings of the National Academy of Sciences, 110, 6442-6447, 10.1073/pnas.1211466110, 2013.

680 10.1073/pnas.1211466110, 2013. Arndt, S., Turvey, C., and Andreasen, N. C.: Correlating and predicting psychiatric symptom ratings: Spearmans r versus Kendalls tau correlation, Journal of Psychiatric Research, 33, 97-104, <u>https://doi.org/10.1016/S0022-3956(98)90046-2</u>, 1999.

Battistel, D., Argiriadis, E., Kehrwald, N., Spigariol, M., Russell, J. M., and Barbante, C.: Fire and
human record at Lake Victoria, East Africa, during the Early Iron Age: Did humans or climate cause
massive ecosystem changes?, The Holocene, 27, 997-1007, 10.1177/0959683616678466, 2017.
Bauer, D. F.: Constructing Confidence Sets Using Rank Statistics, Journal of the American Statistical
Association, 67, 687-690, 10.1080/01621459.1972.10481279, 1972.

Brigham-Grette, J., Melles, M., Minyuk, P., Andreev, A., Tarasov, P., DeConto, R., Koenig, S.,
Nowaczyk, N., Wennrich, V., Rosén, P., Haltia, E., Cook, T., Gebhardt, C., Meyer-Jacob, C., Snyder, J.,
and Herzschuh, U.: Pliocene Warmth, Polar Amplification, and Stepped Pleistocene Cooling Recorded
in NE Arctic Russia, Science, 340, 1421-1427, 10.1126/science.1233137, 2013.
Buchholz, G., Goldammer, J. G., and Martell, D. L.: Application of Logistic Models to Predict Human-Caused Forest Fires in Siberia, in: Early Warning Systems for Natural Disaster Reduction, edited by:

Zschau, J., and Küppers, A., Springer Berlin Heidelberg, Berlin, Heidelberg, 593-599, 2003.
 Callegaro, A., Matsubara Pereira, F., Battistel, D., Kehrwald, N. M., Bird, B. W., Kirchgeorg, T., and Barbante, C.: Fire, vegetation and Holocene climate in the south-eastern Tibetan Plateau: a multi-biomarker reconstruction from Paru Co, Clim. Past Discuss., 2018, 1-33, 10.5194/cp-2018-19, 2018.
 Chapligin, B., Meyer, H., Swann, G. E. A., Meyer-Jacob, C., and Hubberten, H. W.: A 250 ka oxygen

isotope record from diatoms at Lake El'gygytgyn, far east Russian Arctic, Clim. Past, 8, 1621-1636, 10.5194/cp-8-1621-2012, 2012.
 Chen, D., and Loboda, T. V.: Surface forcing of non-stand-replacing fires in Siberian larch forests, Environmental Research Letters, 13, 045008, 2018.

- Conedera, M., Tinner, W., Neff, C., Meurer, M., Dickens, A. F., and Krebs, P.: Reconstructing past fire
 regimes: methods, applications, and relevance to fire management and conservation, Quaternary
 Science Reviews, 28, 555-576, <u>http://dx.doi.org/10.1016/j.quascirev.2008.11.005</u>, 2009.
 D'Anjou, R. M., Wei, J. H., Castañeda, I. S., Brigham-Grette, J., Petsch, S. T., and Finkelstein, D. B.:
 High-latitude environmental change during MIS 9 and 11: biogeochemical evidence from Lake
 El'gygytgyn, Far East Russia, Clim. Past, 9, 567-581, 10.5194/cp-9-567-2013, 2013.
- 710 Daniau, A. L., Bartlein, P. J., Harrison, S. P., Prentice, I. C., Brewer, S., Friedlingstein, P., Harrison-Prentice, T. I., Inoue, J., Izumi, K., Marlon, J. R., Mooney, S., Power, M. J., Stevenson, J., Tinner, W., Andrič, M., Atanassova, J., Behling, H., Black, M., Blarquez, O., Brown, K. J., Carcaillet, C., Colhoun, E. A., Colombaroli, D., Davis, B. A. S., D'Costa, D., Dodson, J., Dupont, L., Eshetu, Z., Gavin, D. G., Genries, A., Haberle, S., Hallett, D. J., Hope, G., Horn, S. P., Kassa, T. G., Katamura, F., Kennedy, L. M.,
- 715 Kershaw, P., Krivonogov, S., Long, C., Magri, D., Marinova, E., McKenzie, G. M., Moreno, P. I., Moss, P., Neumann, F. H., Norström, E., Paitre, C., Rius, D., Roberts, N., Robinson, G. S., Sasaki, N., Scott, L., Takahara, H., Terwilliger, V., Thevenon, F., Turner, R., Valsecchi, V. G., Vannière, B., Walsh, M., Williams, N., and Zhang, Y.: Predictability of biomass burning in response to climate changes, Global Biogeochemical Cycles, 26, GB4007, 10.1029/2011GB004249, 2012.
- 720 Dietze, E., Theuerkauf, M., Bloom, K., Brauer, A., Dörfler, W., Feeser, I., Feurdean, A., Gedminienė, L., Giesecke, T., Jahns, S., Karpińska-Kołaczek, M., Kołaczek, P., Lamentowicz, M., Latałowa, M., Marcisz, K., Obremska, M., Pędziszewska, A., Poska, A., Rehfeld, K., Stančikaitė, M., Stivrins, N., Święta-Musznicka, J., Szal, M., Vassiljev, J., Veski, S., Wacnik, A., Weisbrodt, D., Wiethold, J., Vannière, B., and Słowiński, M.: Holocene fire activity during low-natural flammability periods reveals scale-
- dependent cultural human-fire relationships in Europe, Quaternary Science Reviews, 201, 44-56, https://doi.org/10.1016/j.quascirev.2018.10.005, 2018.
 Dietze, E., Brykała, D., Schreuder, L. T., Jażdżewski, K., Blarquez, O., Brauer, A., Dietze, M., Obremska, M., Ott, F., Pieńczewska, A., Schouten, S., Hopmans, E. C., and Słowiński, M.: Human-induced fire

regime shifts during 19th century industrialization: A robust fire regime reconstruction using
 northern Polish lake sediments, PLOS ONE, 14, e0222011, 10.1371/journal.pone.0222011, 2019.
 Elias, V. O., Simoneit, B. R. T., Cordeiro, R. C., and Turcq, B.: Evaluating levoglucosan as an indicator of

- biomass burning in Carajás, amazônia: a comparison to the charcoal record2, Geochimica et Cosmochimica Acta, 65, 267-272, <u>http://dx.doi.org/10.1016/S0016-7037(00)00522-6</u>, 2001. Engling, G., Carrico, C. M., Kreidenweis, S. M., Collett Jr, J. L., Day, D. E., Malm, W. C., Lincoln, E., Min
- Hao, W., Iinuma, Y., and Herrmann, H.: Determination of levoglucosan in biomass combustion aerosol by high-performance anion-exchange chromatography with pulsed amperometric detection, Atmospheric Environment, 40, 299-311, 10.1016/j.atmosenv.2005.12.069, 2006.
 Fabbri, D., Torri, C., Simoneit, B. R. T., Marynowski, L., Rushdi, A. I., and Fabiańska, M. J.: Levoglucosan and other cellulose and lignin markers in emissions from burning of Miocene lignites,
- Atmospheric Environment, 43, 2286-2295, 10.1016/j.atmosenv.2009.01.030, 2009.
 Fedorov, G., Nolan, M., Brigham-Grette, J., Bolshiyanov, D., Schwamborn, G., and Juschus, O.:
 Preliminary estimation of Lake El'gygytgyn water balance and sediment income, Clim. Past, 9, 1455-1465, 10.5194/cp-9-1455-2013, 2013.
- Feurdean, A., Vannière, B., Finsinger, W., Warren, D., Connor, S. C., Forrest, M., Liakka, J., Panait, A.,
 Werner, C., Andrič, M., Bobek, P., Carter, V. A., Davis, B., Diaconu, A. C., Dietze, E., Feeser, I.,
 Florescu, G., Gałka, M., Giesecke, T., Jahns, S., Jamrichová, E., Kajukało, K., Kaplan, J., Karpińska-Kołaczek, M., Kołaczek, P., Kuneš, P., Kupriyanov, D., Lamentowicz, M., Lemmen, C., Magyari, E. K.,
 Marcisz, K., Marinova, E., Niamir, A., Novenko, E., Obremska, M., Pędziszewska, A., Pfeiffer, M.,
 Poska, A., Rösch, M., Słowiński, M., Stančikaitė, M., Szal, M., Święta-Musznicka, J., Tanţău, I.,
- 750 Theuerkauf, M., Tonkov, S., Valkó, O., Vassiljev, J., Veski, S., Vincze, I., Wacnik, A., Wiethold, J., and Hickler, T.: Fire risk modulation by long-term dynamics in land cover and dominant forest type in Eastern and Central Europe, Biogeosciences Discuss., 2019, 1-36, 10.5194/bg-2019-260, 2019. Fujii, Y., Iriana, W., Oda, M., Puriwigati, A., Tohno, S., Lestari, P., Mizohata, A., and Huboyo, H. S.: Characteristics of carbonaceous aerosols emitted from peatland fire in Riau, Sumatra, Indonesia,
- 755 Atmospheric Environment, 87, 164-169, <u>https://doi.org/10.1016/j.atmosenv.2014.01.037</u>, 2014. Furyaev, V. V., Vaganov, E. A., Tchebakova, N. M., and Valendik, E. N.: Effects of Fire and Climate on Successions and Structural Changes in The Siberian Boreal Forest, Eurasian Journal of Forest Research, 2, 1-15, 2001.

Gao, S., Hegg, D. A., Hobbs, P. V., Kirchstetter, T. W., Magi, B. I., and Sadilek, M.: Water-soluble
 organic components in aerosols associated with savanna fires in southern Africa: Identification,
 evolution, and distribution, Journal of Geophysical Research: Atmospheres, 108,
 10.1029/2002jd002324, 2003.

Gonzalez, P., Neilson, R. P., Lenihan, J. M., and Drapek, R. J.: Global patterns in the vulnerability of ecosystems to vegetation shifts due to climate change, Global Ecology and Biogeography, 19, 755-765
 768, doi:10.1111/j.1466-8238.2010.00558.x, 2010.

Han, Y. M., Peteet, D. M., Arimoto, R., Cao, J. J., An, Z. S., Sritrairat, S., and Yan, B. Z.: Climate and Fuel Controls on North American Paleofires: Smoldering to Flaming in the Late-glacial-Holocene Transition, Scientific Reports, 6, 20719, 10.1038/srep20719

http://www.nature.com/articles/srep20719#supplementary-information, 2016.

770 Harris, R. M. B., Remenyi, T. A., Williamson, G. J., Bindoff, N. L., and Bowman, D. M. J. S.: Climate– vegetation–fire interactions and feedbacks: trivial detail or major barrier to projecting the future of the Earth system?, Wiley Interdisciplinary Reviews: Climate Change, 7, 910-931, 10.1002/wcc.428, 2016.

Hemingway, J. D., Rothman, D. H., Grant, K. E., Rosengard, S. Z., Eglinton, T. I., Derry, L. A., and Galy,
V. V.: Mineral protection regulates long-term global preservation of natural organic carbon, Nature,
570, 228-231, 10.1038/s41586-019-1280-6, 2019.
Herzschuh, U., Birks, H. J. B., Laepple, T., Andreev, A., Melles, M., and Brigham-Grette, J.: Glacial
legacies on interglacial vegetation at the Pliocene-Pleistocene transition in NE Asia, Nature
Communications, 7, 11967, 10.1038/ncomms11967

780 <u>https://www.nature.com/articles/ncomms11967#supplementary-information</u>, 2016. Herzschuh, U., Cao, X., Laepple, T., Dallmeyer, A., Telford, R. J., Ni, J., Chen, F., Kong, Z., Liu, G., Liu, K.-B., Liu, X., Stebich, M., Tang, L., Tian, F., Wang, Y., Wischnewski, J., Xu, Q., Yan, S., Yang, Z., Yu, G., Zhang, Y., Zhao, Y., and Zheng, Z.: Position and orientation of the westerly jet determined Holocene rainfall patterns in China, Nature Communications, 10, 2376, 10.1038/s41467-019-09866-8, 2019.

785 Honegger, R., and Bartnicki-Garcia, S.: Cell wall structure and composition of cultured mycobionts from the lichens Cladonia macrophylla, Cladonia caespiticia, and Physcia stellaris (Lecanorales, Ascomycetes), Mycological Research, 95, 905-914, <u>https://doi.org/10.1016/S0953-7562(09)80085-3</u>, 1991.

 Hopmans, E. C., dos Santos, R. A. L., Mets, A., Sinninghe Damsté, J. S., and Schouten, S.: A novel
 method for the rapid analysis of levoglucosan in soils and sediments, Organic Geochemistry, 58, 86-88, <u>http://dx.doi.org/10.1016/j.orggeochem.2013.02.003</u>, 2013.

Hu, F. S., Higuera, P. E., Walsh, J. E., Chapman, W. L., Duffy, P. A., Brubaker, L. B., and Chipman, M. L.: Tundra burning in Alaska: Linkages to climatic change and sea ice retreat, Journal of Geophysical Research: Biogeosciences, 115, doi:10.1029/2009JG001270, 2010.

795 Hunsinger, G. B., Mitra, S., Warrick, J. A., and Alexander, C. R.: Oceanic loading of wildfire-derived organic compounds from a small mountainous river, Journal of Geophysical Research: Biogeosciences, 113, 10.1029/2007jg000476, 2008.

 linuma, Y., Brüggemann, E., Gnauk, T., Müller, K., Andreae, M. O., Helas, G., Parmar, R., and Herrmann, H.: Source characterization of biomass burning particles: The combustion of selected
 European conifers, African hardwood, savanna grass, and German and Indonesian peat, Journal of Geophysical Research: Atmospheres, 112, 10.1029/2006jd007120, 2007.

IPCC: Climate Change 2014: Impacts, Adaptation, and Vulnerability. Part B: Regional Aspects. Contribution of Working Group II to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Barros, V.R., C.B. Field, D.J. Dokken, M.D. Mastrandrea, K.J. Mach, T.E. Bilir, M.

805 Chatterjee, K.L. Ebi, Y.O. Estrada, R.C. Genova, B. Girma, E.S. Kissel, A.N. Levy, S. MacCracken, P.R. Mastrandrea, and L.L. White (eds.)], Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 688 pp., 2014.

Isaev, A. P., Protopopov, A. V., Protopopova, V. V., Egorova, A. A., Timofeyev, P. A., Nikolaev, A. N.,
Shurduk, I. F., Lytkina, L. P., Ermakov, N. B., Nikitina, N. V., Efimova, A. P., Zakharova, V. I., Cherosov,
M. M., Nikolin, E. G., Sosina, N. K., Troeva, E. I., Gogoleva, P. A., Kuznetsova, L. V., Pestryakov, B. N.,

Mironova, S. I., and Sleptsova, N. P.: Vegetation of Yakutia: Elements of Ecology and Plant Sociology, in: The Far North: Plant Biodiversity and Ecology of Yakutia, edited by: Troeva, E. I., Isaev, A. P., Cherosov, M. M., and Karpov, N. S., Springer Netherlands, Dordrecht, 143-260, 2010. Jackson, D. A., and Somers, K. M.: The spectre of 'spurious' correlations, Oecologia, 86, 147-151,

815 1991. Jung, J., Lee, S., Kim, H., Kim, D., Lee, H., and Oh, S.: Quantitative determination of the biomassburning contribution to atmospheric carbonaceous aerosols in Daejeon, Korea, during the riceharvest period, Atmospheric Environment, 89, 642-650, <u>https://doi.org/10.1016/j.atmosenv.2014.03.010</u>, 2014.

Kappenberg, A., Lehndorff, E., Pickarski, N., Litt, T., and Amelung, W.: Solar controls of fire events during the past 600,000 years, Quaternary Science Reviews, 208, 97-104, https://doi.org/10.1016/j.quascirev.2019.02.008, 2019.
 Keeley, J. E.: Fire intensity, fire severity and burn severity: a brief review and suggested usage, International Journal of Wildland Fire, 18, 116-126, https://doi.org/10.1071/WF07049, 2009.

Keeley, J. E.: Ecology and evolution of pine life histories, Annals of Forest Science, 69, 445-453, 10.1007/s13595-012-0201-8, 2012.
 Kharuk, V. I., Ranson, K. J., Dvinskaya, M. L., and Im, S. T.: Wildfires in northern Siberian larch

dominated communities, Environmental Research Letters, 6, 045208, 2011. Kirchgeorg, T., Schüpbach, S., Kehrwald, N., McWethy, D. B., and Barbante, C.: Method for the

determination of specific molecular markers of biomass burning in lake sediments, Organic Geochemistry, 71, 1-6, <u>http://dx.doi.org/10.1016/j.orggeochem.2014.02.014</u>, 2014.

Knicker, H., Hilscher, A., de la Rosa, J. M., González-Pérez, J. A., and González-Vila, F. J.: Modification of biomarkers in pyrogenic organic matter during the initial phase of charcoal biodegradation in soils, Geoderma, 197–198, 43-50, http://dx.doi.org/10.1016/j.geoderma.2012.12.021, 2013.

Krawchuk, M. A., and Moritz, M. A.: Constraints on global fire activity vary across a resource gradient, Ecology, 92, 121-132, 10.1890/09-1843.1, 2011.

Kuo, L.-J., Herbert, B. E., and Louchouarn, P.: Can levoglucosan be used to characterize and quantify char/charcoal black carbon in environmental media?, Organic Geochemistry, 39, 1466-1478, http://doi.org/10.1016/j.orggeochem.2008.04.026, 2008.

- 840 Kuo, L.-J., Louchouarn, P., and Herbert, B. E.: Influence of combustion conditions on yields of solventextractable anhydrosugars and lignin phenols in chars: Implications for characterizations of biomass combustion residues, Chemosphere, 85, 797-805, 10.1016/j.chemosphere.2011.06.074, 2011. Lalonde, K., Mucci, A., Ouellet, A., and Gélinas, Y.: Preservation of organic matter in sediments promoted by iron, Nature, 483, 198, 10.1038/nature10855
- https://www.nature.com/articles/nature10855#supplementary-information, 2012.
 Laskar, J., Robutel, P., Joutel, F., Gastineau, M., Correia, A. C. M., and Levrard, B.: A long-term numerical solution for the insolation quantities of the Earth, A&A, 428, 261-285, 2004.
 Lasslop, G., Brovkin, V., Reick, C. H., Bathiany, S., and Kloster, S.: Multiple stable states of tree cover in a global land surface model due to a fire-vegetation feedback, Geophysical Research Letters, 43, 6324-6331, 10.1002/2016gl069365, 2016.
- Linderholm, H. W., Nicolle, M., Francus, P., Gajewski, K., Helama, S., Korhola, A., Solomina, O., Yu, Z., Zhang, P., D'Andrea, W. J., Debret, M., Divine, D. V., Gunnarson, B. E., Loader, N. J., Massei, N., Seftigen, K., Thomas, E. K., Werner, J., Andersson, S., Berntsson, A., Luoto, T. P., Nevalainen, L., Saarni, S., and Väliranta, M.: Arctic hydroclimate variability during the last 2000 years: current
- understanding and research challenges, Clim. Past, 14, 473-514, 10.5194/cp-14-473-2018, 2018.
 Lisiecki, L. E., and Raymo, M. E.: A Pliocene-Pleistocene stack of 57 globally distributed benthic δ180 records, Paleoceanography, 20, doi:10.1029/2004PA001071, 2005.
 Lopes dos Santos, R. A., De Deckker, P., Hopmans, E. C., Magee, J. W., Mets, A., Sinninghe Damste, J. S., and Schouten, S.: Abrupt vegetation change after the Late Quaternary megafaunal extinction in
- southeastern Australia, Nature Geosci, 6, 627-631, 10.1038/ngeo1856
 http://www.nature.com/ngeo/journal/v6/n8/abs/ngeo1856.html#supplementary-information, 2013. Loranty, M. M., Natali, S. M., Berner, L. T., Goetz, S. J., Holmes, R. M., Davydov, S. P., Zimov, N. S., and
- Zimov, S. A.: Siberian tundra ecosystem vegetation and carbon stocks four decades after wildfire, Journal of Geophysical Research: Biogeosciences, 119, 2144-2154, doi:10.1002/2014JG002730, 2014.
 Loranty, M. M., Berner, L. T., Taber, E. D., Kropp, H., Natali, S. M., Alexander, H. D., Davydov, S. P., and Zimov, N. S.: Understory vegetation mediates permafrost active layer dynamics and carbon dioxide fluxes in open-canopy larch forests of northeastern Siberia, PLOS ONE, 13, e0194014,
- 10.1371/journal.pone.0194014, 2018. Lozhkin, A. V., Anderson, P. M., Matrosova, T. V., and Minyuk, P. S.: The pollen record from
- El'gygytgyn Lake: implications for vegetation and climate histories of northern Chukotka since the late middle Pleistocene, Journal of Paleolimnology, 37, 135-153, 10.1007/s10933-006-9018-5, 2007. Lozhkin, A. V., Minyuk, P. S., Anderson, P. M., Nedorubova, E. Y., and Korzun, J. V.: Variability in landscape and lake system responses to glacial and interglacial climates during the Middle Pleistocene based on palynological and geochemical data from Lake El'gygytgyn, Eastern Arctic,
- Review of Palaeobotany and Palynology, 246, 1-13, <u>https://doi.org/10.1016/j.revpalbo.2017.06.004</u>, 2017.
 No. V. Margar, Y. Margar, W. and Gan, A. A supersoftwarders and superstations.

Ma, Y., Liu, K.-b., Feng, Z., Sang, Y., Wang, W., and Sun, A.: A survey of modern pollen and vegetation along a south–north transect in Mongolia, Journal of Biogeography, 35, 1512-1532, 10.1111/j.1365-2699.2007.01871.x, 2008.

880 Marlon, J. R., Bartlein, P. J., Daniau, A. L., Harrison, S. P., Maezumi, S. Y., Power, M. J., Tinner, W., and Vanniére, B.: Global biomass burning: A synthesis and review of Holocene paleofire records and their controls, Quaternary Science Reviews, 65, 5-25, 10.1016/j.quascirev.2012.11.029, 2013. Martin Calvo, M., Prentice, I. C., and Harrison, S. P.: Climate versus carbon dioxide controls on biomass burning: a model analysis of the glacial–interglacial contrast, Biogeosciences, 11, 6017-6027, 10.5194/bg-11-6017-2014, 2014.

Marynowski, L., Bucha, M., Smolarek, J., Wendorff, M., and Simoneit, B. R. T.: Occurrence and significance of mono-, di- and anhydrosaccharide biomolecules in Mesozoic and Cenozoic lignites and fossil wood, Organic Geochemistry, 116, 13-22, <u>https://doi.org/10.1016/j.orggeochem.2017.11.008</u>, 2018.

885

- Melles, M., Minyuk, P., Brigham-Grette, J., Juschus, O., and participants, w. c. b. t.: The Expedition Lake El'gygytgyn 2003 (Siberian Arctic). In: Ber. Polarforsch. Meeresforsch., 2005.
 Melles, M., Brigham-Grette, J., Glushkova, O. Y., Minyuk, P. S., Nowaczyk, N. R., and Hubberten, H.-W.: Sedimentary geochemistry of core PG1351 from Lake El'gygytgyn—a sensitive record of climate variability in the East Siberian Arctic during the past three glacial–interglacial cycles, Journal of
- Paleolimnology, 37, 89-104, 10.1007/s10933-006-9025-6, 2007.
 Melles, M., Brigham-Grette, J., Minyuk, P., Koeberl, C., Andreev, A., Cook, T., Fedorov, G., Gebhardt, C., Haltia-Hovi, E., Kukkonen, M., Nowaczyk, N., Schwamborn, G., Wennrich, B., and the El'gygytgyn Scientific, P.: The Lake El'gygytgyn Scientific Drilling Project Conquering Arctic Challenges through Continental Drilling, Sci. Dril., 11, 29-40, 10.2204/iodp.sd.11.03.2011, 2011.
- 900 Melles, M., Brigham-Grette, J., Minyuk, P. S., Nowaczyk, N. R., Wennrich, V., DeConto, R. M., Anderson, P. M., Andreev, A. A., Coletti, A., Cook, T. L., Haltia-Hovi, E., Kukkonen, M., Lozhkin, A. V., Rosén, P., Tarasov, P., Vogel, H., and Wagner, B.: 2.8 Million Years of Arctic Climate Change from Lake El'gygytgyn, NE Russia, Science, 337, 315-320, 10.1126/science.1222135, 2012.
- Mock, C. J., Bartlein, P. J., and Anderson, P. M.: Atmospheric circulation patterns and spatial climatic
 variations in Beringia, International Journal of Climatology, 18, 1085-1104, doi:10.1002/(SICI)1097-0088(199808)18:10<1085::AID-JOC305>3.0.CO;2-K, 1998.
 Molinari, C., Lehsten, V., Blarquez, O., Carcaillet, C., Davis, B. A. S., Kaplan, J. O., Clear, J., and
- Bradshaw, R. H. W.: The climate, the fuel and the land use: Long-term regional variability of biomass burning in boreal forests, Global Change Biology, 24, 4929-4945, 10.1111/gcb.14380, 2018.
 Nitze, I., Grosse, G., Jones, B. M., Romanovsky, V. E., and Boike, J.: Remote sensing quantifies
- 910 NIZE, I., Grosse, G., Jones, B. M., Romanovsky, V. E., and Borke, J.: Remote sensing quantities widespread abundance of permafrost region disturbances across the Arctic and Subarctic, Nature Communications, 9, 5423, 10.1038/s41467-018-07663-3, 2018. Nolan, M., Liston, G., Prokein, P., Brigham-Grette, J., Sharpton, V. L., and Huntzinger, R.: Analysis of

 lake ice dynamics and morphology on Lake El'gygytgyn, NE Siberia, using synthetic aperture radar
 (SAR) and Landsat, Journal of Geophysical Research: Atmospheres, 107, ALT 3-1-ALT 3-12, 10.1029/2001jd000934, 2002.

Nolan, M., and Brigham-Grette, J.: Basic hydrology, limnology, and meteorology of modern Lake El'gygytgyn, Siberia, Journal of Paleolimnology, 37, 17-35, 10.1007/s10933-006-9020-y, 2007. Norwood, M. J., Louchouarn, P., Kuo, L. J., and Harvey, O. R.: Characterization and biodegradation of

- 920 water-soluble biomarkers and organic carbon extracted from low temperature chars, Organic Geochemistry, 56, 111-119, 10.1016/j.orggeochem.2012.12.008, 2013.
 Nowaczyk, N. R., Melles, M., and Minyuk, P.: A revised age model for core PG1351 from Lake El'gygytgyn, Chukotka, based on magnetic susceptibility variations tuned to northern hemisphere insolation variations, Journal of Paleolimnology, 37, 65-76, 10.1007/s10933-006-9023-8, 2007.
- 925 Nowaczyk, N. R., Haltia, E. M., Ulbricht, D., Wennrich, V., Sauerbrey, M. A., Rosén, P., Vogel, H., Francke, A., Meyer-Jacob, C., Andreev, A. A., and Lozhkin, A. V.: Chronology of Lake El'gygytgyn sediments - a combined magnetostratigraphic, palaeoclimatic and orbital tuning study based on multi-parameter analyses, Clim. Past, 9, 2413-2432, 10.5194/cp-9-2413-2013, 2013. Oros, D. R., and Simoneit, B. R. T.: Identification and emission factors of molecular tracers in organic
- aerosols from biomass burning Part 2. Deciduous trees, Applied Geochemistry, 16, 1545-1565, https://doi.org/10.1016/S0883-2927(01)00022-1, 2001a.
 Oros, D. R., and Simoneit, B. R. T.: Identification and emission factors of molecular tracers in organic aerosols from biomass burning Part 1. Temperate climate conifers, Applied Geochemistry, 16, 1513-1544, http://dx.doi.org/10.1016/S0883-2927(01)00022-1, 2001a.

- Oros, D. R., Abas, M. R. b., Omar, N. Y. M. J., Rahman, N. A., and Simoneit, B. R. T.: Identification and emission factors of molecular tracers in organic aerosols from biomass burning: Part 3. Grasses, Applied Geochemistry, 21, 919-940, <u>http://dx.doi.org/10.1016/j.apgeochem.2006.01.008</u>, 2006.
 Otto, A., Gondokusumo, R., and Simpson, M. J.: Characterization and quantification of biomarkers from biomass burning at a recent wildfire site in Northern Alberta, Canada, Applied Geochemistry, 21, 166-183, <u>http://dx.doi.org/10.1016/j.apgeochem.2005.09.007</u>, 2006.
- Pastorova, I., Arisz, P. W., and Boon, J. J.: Preservation of d-glucose-oligosaccharides in cellulose chars, Carbohydrate Research, 248, 151-165, <u>https://doi.org/10.1016/0008-6215(93)84123-N</u>, 1993.
 Pausas, J. G., Keeley, J. E., and Schwilk, D. W.: Flammability as an ecological and evolutionary driver, Journal of Ecology, 105, 289-297, doi:10.1111/1365-2745.12691, 2017.
- 945 Peters, M. E., and Higuera, P. E.: Quantifying the source area of macroscopic charcoal with a particle dispersal model, Quaternary Research, 67, 304-310, <u>http://dx.doi.org/10.1016/j.yqres.2006.10.004</u>, 2007.

Prentice, C., Guiot, J., Huntley, B., Jolly, D., and Cheddadi, R.: Reconstructing biomes from palaeoecological data: a general method and its application to European pollen data at 0 and 6 ka,
Clim Dyn, 12, 185-194, 10.1007/bf00211617, 1996.

- Remy, C. C., Hély, C., Blarquez, O., Magnan, G., Bergeron, Y., Lavoie, M., and Ali, A. A.: Different regional climatic drivers of Holocene large wildfires in boreal forests of northeastern America, Environmental Research Letters, 12, 035005, 10.1088/1748-9326/aa5aff, 2017. Revelle, W.: psych: Procedures for Personality and Psychological Research
- 955 <u>https://CRAN.R-project.org/package=psych</u> Northwestern University, Evanston, Illinois, USA, 2018. Rivera-Hernandez, F., Sumner, D. Y., Mackey, T. J., Hawes, I., and Andersen, D. T.: In a PICL: The sedimentary deposits and facies of perennially ice-covered lakes, Sedimentology, 66, 917-939, 10.1111/sed.12522, 2019.

Roberts, A., Roberts, E., and Haigler, C.: Moss cell walls: structure and biosynthesis, Frontiers in Plant
 Science, 3, 10.3389/fpls.2012.00166, 2012.

Rogers, B. M., Soja, A. J., Goulden, M. L., and Randerson, J. T.: Influence of tree species on continental differences in boreal fires and climate feedbacks, Nature Geosci, 8, 228-234, 10.1038/ngeo2352

http://www.nature.com/ngeo/journal/v8/n3/abs/ngeo2352.html#supplementary-information, 2015.

965 Rolph, G., Stein, A., and Stunder, B.: Real-time Environmental Applications and Display sYstem: READY, Environmental Modelling & Software, 95, 210-228, <u>https://doi.org/10.1016/j.envsoft.2017.06.025</u>, 2017. Sang, X. F., Gensch, I., Kammer, B., Khan, A., Kleist, E., Laumer, W., Schlag, P., Schmitt, S. H., Wildt, J.,

Zhao, R., Mungall, E. L., Abbatt, J. P. D., and Kiendler-Scharr, A.: Chemical stability of levoglucosan: An
 isotopic perspective, Geophysical Research Letters, 43, 5419-5424, 10.1002/2016GL069179, 2016.
 Scheffer, M., Hirota, M., Holmgren, M., Van Nes, E. H., and Chapin, F. S.: Thresholds for boreal biome transitions, Proceedings of the National Academy of Sciences, 109, 21384-21389, 10.1073/pnas.1219844110, 2012.

Schmidl, C., Marr, I. L., Caseiro, A., Kotianová, P., Berner, A., Bauer, H., Kasper-Giebl, A., and 975 Puxbaum, H.: Chemical characterisation of fine particle emissions from wood stove combustion of

common woods growing in mid-European Alpine regions, Atmospheric Environment, 42, 126-141, <u>https://doi.org/10.1016/j.atmosenv.2007.09.028</u>, 2008.

Schreuder, L. T., Hopmans, E. C., Stuut, J.-B. W., Sinninghe Damsté, J. S., and Schouten, S.: Transport and deposition of the fire biomarker levoglucosan across the tropical North Atlantic Ocean,
 Geochimica et Cosmochimica Acta, <u>https://doi.org/10.1016/j.gca.2018.02.020</u>, 2018.

Schreuder, L. T., Donders, T. H., Mets, A., Hopmans, E. C., Sinninghe Damsté, J. S., and Schouten, S.: Comparison of organic and palynological proxies for biomass burning and vegetation in a lacustrine sediment record (Lake Allom, Fraser Island, Australia), Organic Geochemistry, 133, 10-19, https://doi.org/10.1016/j.orggeochem.2019.03.002, 2019. 985 Schüpbach, S., Kirchgeorg, T., Colombaroli, D., Beffa, G., Radaelli, M., Kehrwald, N. M., and Barbante, C.: Combining charcoal sediment and molecular markers to infer a Holocene fire history in the Maya Lowlands of Petén, Guatemala, Quaternary Science Reviews, 115, 123-131, <u>http://dx.doi.org/10.1016/j.quascirev.2015.03.004</u>, 2015.

Schuur, E. A. G., McGuire, A. D., Schädel, C., Grosse, G., Harden, J. W., Hayes, D. J., Hugelius, G.,
 Koven, C. D., Kuhry, P., Lawrence, D. M., Natali, S. M., Olefeldt, D., Romanovsky, V. E., Schaefer, K.,
 Turetsky, M. R., Treat, C. C., and Vonk, J. E.: Climate change and the permafrost carbon feedback,

- Nature, 520, 171, 10.1038/nature14338, 2015.
 Schwamborn, G., Meyer, H., Fedorov, G., Schirrmeister, L., and Hubberten, H.-W.: Ground ice and slope sediments archiving late Quaternary paleoenvironment and paleoclimate signals at the margins
 of El'gygytgyn Impact Crater, NE Siberia, Quaternary Research, 66, 259-272,
- https://doi.org/10.1016/j.yqres.2006.06.007, 2006. Serreze, M. C., and Barry, R. G.: Processes and impacts of Arctic amplification: A research synthesis, Global and Planetary Change, 77, 85-96, <u>https://doi.org/10.1016/j.gloplacha.2011.03.004</u>, 2011. Simoneit, B. R. T., Schauer, J. J., Nolte, C. G., Oros, D. R., Elias, V. O., Fraser, M. P., Rogge, W. F., and
- 1000 Cass, G. R.: Levoglucosan, a tracer for cellulose in biomass burning and atmospheric particles, Atmospheric Environment, 33, 173-182, <u>http://dx.doi.org/10.1016/S1352-2310(98)00145-9</u>, 1999.
 Simoneit, B. R. T.: Biomass burning — a review of organic tracers for smoke from incomplete combustion, Applied Geochemistry, 17, 129-162, <u>http://dx.doi.org/10.1016/S0883-2927(01)00061-0</u>, 2002.
- 1005 Sofronov, M. A., Volokitina, A. V., Kajimoto, T., and Uemura, S.: The Ecological Role of Moss-Lichen Cover and Thermal Amelioration of Larch Forest Ecosystems in the Northern Part of Siberia, Eurasian Journal of Forest Research, 7, 11-19, 2004.

Sofronov, M. A., and Volokitina, A. V.: Wildfire Ecology in Continuous Permafrost Zone, in:
 Permafrost Ecosystems: Siberian Larch Forests, edited by: Osawa, A., Zyryanova, O. A., Matsuura, Y.,
 Kajimoto, T., and Wein, R. W., Springer Netherlands, Dordrecht, 59-82, 2010.

- Soja, A. J., Tchebakova, N. M., French, N. H. F., Flannigan, M. D., Shugart, H. H., Stocks, B. J., Sukhinin, A. I., Parfenova, E. I., Chapin, F. S., and Stackhouse, P. W.: Climate-induced boreal forest change: Predictions versus current observations, Global and Planetary Change, 56, 274-296, https://doi.org/10.1016/j.gloplacha.2006.07.028, 2007.
- 1015 Stein, A. F., Draxler, R. R., Rolph, G. D., Stunder, B. J. B., Cohen, M. D., and Ngan, F.: NOAA's HYSPLIT Atmospheric Transport and Dispersion Modeling System, Bulletin of the American Meteorological Society, 96, 2059-2077, 10.1175/bams-d-14-00110.1, 2015. Stivrins, N., Aakala, T., Ilvonen, L., Pasanen, L., Kuuluvainen, T., Vasander, H., Gałka, M., Disbrey, H.
- R., Liepins, J., Holmström, L., and Seppä, H.: Integrating fire-scar, charcoal and fungal spore data to
 study fire events in the boreal forest of northern Europe, The Holocene, 0959683619854524,
 10.1177/0959683619854524, 2019.

Suciu, L. G., Masiello, C. A., and Griffin, R. J.: Anhydrosugars as tracers in the Earth system, Biogeochemistry, 146, 209-256, 10.1007/s10533-019-00622-0, 2019.

- Sugita, S.: Theory of quantitative reconstruction of vegetation I: pollen from large sites REVEALS
 regional vegetation composition, The Holocene, 17, 229-241, 10.1177/0959683607075837, 2007.
 Tarasov, P. E., Andreev, A. A., Anderson, P. M., Lozhkin, A. V., Leipe, C., Haltia, E., Nowaczyk, N. R., Wennrich, V., Brigham-Grette, J., and Melles, M.: A pollen-based biome reconstruction over the last 3.562 million years in the Far East Russian Arctic new insights into climate-vegetation relationships at the regional scale, Clim. Past, 9, 2759-2775, 10.5194/cp-9-2759-2013, 2013.
- 1030 Tautenhahn, S., Lichstein, J. W., Jung, M., Kattge, J., Bohlman, S. A., Heilmeier, H., Prokushkin, A., Kahl, A., and Wirth, C.: Dispersal limitation drives successional pathways in Central Siberian forests under current and intensified fire regimes, Global Change Biology, 22, 2178-2197, doi:10.1111/gcb.13181, 2016.
- Tchebakova, N. M., Parfenova, E., and Soja, A. J.: The effects of climate, permafrost and fire on vegetation change in Siberia in a changing climate, Environmental Research Letters, 4, 045013, 2009.

Theuerkauf, M., and Couwenberg, J.: ROPES Reveals Past Land Cover and PPEs From Single Pollen Records, Frontiers in Earth Science, 6, 10.3389/feart.2018.00014, 2018.

Thonicke, K., Prentice, I. C., and Hewitt, C.: Modeling glacial-interglacial changes in global fire regimes and trace gas emissions, Global Biogeochemical Cycles, 19, 10.1029/2004gb002278, 2005.

- 1040 Tobo, Y., DeMott, P. J., Raddatz, M., Niedermeier, D., Hartmann, S., Kreidenweis, S. M., Stratmann, F., and Wex, H.: Impacts of chemical reactivity on ice nucleation of kaolinite particles: A case study of levoglucosan and sulfuric acid, Geophysical Research Letters, 39, 10.1029/2012gl053007, 2012. van der Werf, G. R., Randerson, J. T., Giglio, L., van Leeuwen, T. T., Chen, Y., Rogers, B. M., Mu, M., van Marle, M. J. E., Morton, D. C., Collatz, G. J., Yokelson, R. J., and Kasibhatla, P. S.: Global fire
- 1045 emissions estimates during 1997–2016, Earth Syst. Sci. Data, 9, 697-720, 10.5194/essd-9-697-2017, 2017.

van Leeuwen, T. T., and van der Werf, G. R.: Spatial and temporal variability in the ratio of trace gases emitted from biomass burning, Atmos. Chem. Phys., 11, 3611-3629, 10.5194/acp-11-3611-2011, 2011.

1050 Veraverbeke, S., Rogers, B. M., Goulden, M. L., Jandt, R. R., Miller, C. E., Wiggins, E. B., and Randerson, J. T.: Lightning as a major driver of recent large fire years in North American boreal forests, Nature Climate Change, 7, 529, 10.1038/nclimate3329

https://www.nature.com/articles/nclimate3329#supplementary-information, 2017.

 Wasserstein, R. L., Schirm, A. L., and Lazar, N. A.: Moving to a World Beyond "p < 0.05", The American</th>

 1055
 Statistician, 73, 1-19, 10.1080/00031305.2019.1583913, 2019.

- Wennrich, V., Francke, A., Dehnert, A., Juschus, O., Leipe, T., Vogt, C., Brigham-Grette, J., Minyuk, P. S., Melles, M., and El'gygytgyn Science, P.: Modern sedimentation patterns in Lake El'gygytgyn, NE Russia, derived from surface sediment and inlet streams samples, Clim. Past, 9, 135-148, 10.5194/cp-9-135-2013, 2013.
- 1060 Wennrich, V., Andreev, A. A., Tarasov, P. E., Fedorov, G., Zhao, W., Gebhardt, C. A., Meyer-Jacob, C., Snyder, J. A., Nowaczyk, N. R., Schwamborn, G., Chapligin, B., Anderson, P. M., Lozhkin, A. V., Minyuk, P. S., Koeberl, C., and Melles, M.: Impact processes, permafrost dynamics, and climate and environmental variability in the terrestrial Arctic as inferred from the unique 3.6 Myr record of Lake El'gygytgyn, Far East Russia – A review, Quaternary Science Reviews, 147, 221-244, https://doi.org/10.1016/j.quascirev.2016.03.019, 2016.

Westerling, A. L., Hidalgo, H. G., Cayan, D. R., and Swetnam, T. W.: Warming and Earlier Spring Increase Western U.S. Forest Wildfire Activity, Science, 313, 940-943, 10.1126/science.1128834, 2006.

- Whitlock, C., and Larsen, C.: Charcoal as a Fire Proxy, in: Tracking Environmental Change Using Lake
 Sediments. Terrestrial, Algal, and Siliceous Indicators., edited by: Smol, J. P., Birks, H. J. B., and Last,
 W. M., KluwerAcademic Publishers, Dordrecht, The Netherlands, 75-97, 2001.
 Williams, R. S. J., and Ferrigno, J. G.: State of the Earth's cryosphere at the beginning of the 21st
- century–Glaciers, global snow cover, floating ice, and permafrost and periglacial environments, U.S.
 Geological Survey Professional Paper, 1386–A, edited by: USGS, United States Government Printing
 Office, Washington, 546 pp., 2012.

Wirth, C.: Fire Regime and Tree Diversity in Boreal Forests: Implications for the Carbon Cycle, Ecological Studies: Forest Diversity and Function: Temperate and Boreal Systems edited by: Scherer-Lorenzen, M., Körner, C., and Schulze, E. D., Springer-Verlag, Berlin Heidelberg, 2005. Yang, H., Yan, R., Chen, H., Lee, D. H., and Zheng, C.: Characteristics of hemicellulose, cellulose and

- lignin pyrolysis, Fuel, 86, 1781-1788, <u>https://doi.org/10.1016/j.fuel.2006.12.013</u>, 2007.
 Yin, Q., and Berger, A.: Interglacial analogues of the Holocene and its natural near future, Quaternary Science Reviews, 120, 28-46, <u>https://doi.org/10.1016/j.quascirev.2015.04.008</u>, 2015.
 Yin, Q. Z., and Berger, A.: Individual contribution of insolation and CO2 to the interglacial climates of the past 800,000 years, Clim Dyn, 38, 709-724, 10.1007/s00382-011-1013-5, 2012.
- 1085 Zhang, N., Yasunari, T., and Ohta, T.: Dynamics of the larch taiga–permafrost coupled system in Siberia under climate change, Environmental Research Letters, 6, 024003, 2011.

Zhao, W., Andreev, A. A., Tarasov, P. E., Wennrich, V., and Melles, M.: Vegetation and climate during the penultimate interglacial of the northeastern Russian Arctic: the Lake El'gygytgyn pollen record, Boreas, 48, 507-515, 10.1111/bor.12373, 2019.