



Relationships between low-temperature fires, climate and vegetation during the last 430 kyrs in northeastern Siberia reconstructed from monosaccharide anhydrides in Lake El'gygytyn sediments

Elisabeth Dietze¹, Kai Mangelsdorf², Andrei Andreev^{1,3}, Cornelia Karger², Laura T.

5 Schreuder⁴, Ellen C. Hopmans⁴, Oliver Rach⁵, Dirk Sachse⁵, Volker Wennrich⁶, Ulrike
Herzschuh^{1,7,8}

¹Polar Terrestrial Environmental Systems, Alfred-Wegener-Institute for Polar and Marine Research, Research Unit Potsdam, Telegrafenberg, 14473 Potsdam, Germany

²GFZ German Research Centre for Geosciences, Helmholtz Centre Potsdam, Organic Geochemistry,
10 Telegrafenberg, 14473 Potsdam, Germany

³Institute of Geology and Petroleum Technologies, Kazan Federal University, Kremlyovskaya str. 4/5, 420008, Kazan, Russia

⁴Department of Marine Microbiology and Biogeochemistry, Royal Netherlands Institute for Sea Research (NIOZ) and Utrecht University, Texel, The Netherlands

15 ⁵GFZ, German Research Centre for Geosciences, Helmholtz Centre Potsdam, Geomorphology, Surface Organic Geochemistry lab, Telegrafenberg, 14473 Potsdam, Germany

⁶University of Cologne, Institute of Geology and Mineralogy, Zùlpicher str. 49a, 50674 Cologne, Germany

⁷ Institute of Geosciences, University of Potsdam, Karl-Liebknecht-Str. 24-25, 14476 Potsdam-Golm

⁸ Institute of Biochemistry and Biology, University of Potsdam, Karl-Liebknecht-Str. 24-25, 14476 Potsdam-
20 Golm

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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
25 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
30 low-intensity biomass burning in glacial-to-interglacial lake sediments from the high northern latitudes. We use sediments from Lake El'gygytyn (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 different climate and biome configurations. Combined with pollen and non-pollen palynomorph records from the same samples, we assess past relationships between fire, climate, and vegetation on orbital to centennial time scales. We find that MAs were well-preserved in up to 430 kyrs old sediments with higher influxes from low-
35 intensity biomass burning in interglacials compared to glacials. MA influxes significantly increase when summergreen boreal forest spreads closer to the lake, whereas they decrease when tundra-steppe environments and, especially, *Sphagnum* peatlands spread. This suggests that low-temperature fires are a typical property of Siberian larch forest on long timescales. The results also suggest that low-intensity fires would be reduced by



40 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 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 fire–climate–vegetation feedbacks
in the high northern latitudes.

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1. Introduction

1.1 Background consideration

50 In recent decades, high northern latitudes have been 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).

55 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
60 al., 2012;Marlon et al., 2013).

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;Rogers et al., 2015),
65 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).

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). Fire intensity refers
here to fire temperatures, combustion efficiencies and fire radiative power (Keeley, 2009). In Siberian
70 summergreen boreal forests, dominated by larch (mainly *Larix gmelinii* 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 Loboda, 2018). Fire is suspected to support the existence and regeneration of larch by, for
75 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 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).

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;Lorantny 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).

Sediment cores of Lake El'gygytgyn provide a continuous Pliocene-Pleistocene environmental record in the Arctic that suggest 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 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 c. 180 mm (in 2002), mainly falling as snow (Nolan and Brigham-Grette, 2007). Current treeless herb tundra vegetation is composed of lichens, herbs, and grasses (mostly Poaceae, Cyperaceae) 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 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., 2019). Molecular burning proxies are currently being explored to infer source- and temperature-specific fire histories (Kehrwald et al., 2016;Kapenberg et al., 2019;Dietze et al., in review). 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 -D-galactopyranose, GAL). While Simoneit et al. (1999) and references therein suggest that MAs form at burn temperatures $> 300^{\circ}\text{C}$, several studies that have analysed the influence of various combustion conditions in natural samples indicated that MAs are thermal dehydration products at burning temperatures $< 350^{\circ}\text{C}$, 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). 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., in review). 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.

Here, we study late glacial to interglacial fire histories from the last 430 kyrs using sedimentary MA from Lake El'gygytgyn and assess long-term relationships between low-temperature fires and regional vegetation in the Russian Far East (Fig. 1). We focus on three late glacial-to-interglacial periods, which include marine isotope stages (MIS) 12–11c, 8–7e, and 6–5e. 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., 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 global ice volume, insolation changes (see Yin and Berger (2012), Table 1, for interglacial astronomical and GHG characteristics), and interhemispheric ice sheet–ocean–atmosphere feedback mechanisms (Melles et al., 2012; Lozhkin et al., 2017).

Here, we aim to answer 1) whether sedimentary MAs are suitable proxies to reconstruct low-temperature fires even in interglacial Arctic lake sediments; 2) whether more biomass burning occurs during interglacials compared to 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), and 3) as different vegetation composition has been reconstructed from past interglacials (Melles et al., 2012; Brigham-Grette et al., 2013; Andreev et al., 2014), we consider 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

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 ^{14}C - and luminescence dates, as well as magnetostratigraphy (Nowaczyk



et al., 2007;Melles et al., 2007). A 318 m long composite core from ICDP site 5011-1 comprises three parallel
sediment cores that cover the time period (Melles et al., 2011;Wennrich et al., 2016), dated by
155 magnetostratigraphy, paleoclimatic and orbital tuning with more than 600 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 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
160 samples of 0.5–1.3 g of dry sediment from ICDP core 5011-1 were taken for 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-scale averages.

We extracted the polar lipids of all MA samples using a Dionex Accelerated Solvent Extraction system (ASE 350,
165 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 ($C_6H_3D_7O_5$;
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
170 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., in review). Briefly, separation was achieved on two
Xbridge BEH amide columns in series (2.1 x 150 mm, 3.5 μ m particle size) fitted with a 50 mm pre-column of the
175 same 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)
pressure 3 (arbitrary units), auxiliary gas (N₂) temperature of 50 °C, spray voltage -2.9 kV (negative ion mode),
180 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
185 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
190 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 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 ICDP-5011-1 cores.

195 Pollen from sediment core PG1351 was analysed by Lozhkin et al. (2007), but there was no sediment left to 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) 200 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 MIS 12 and 11c were harmonized with the new pollen samples of core PG1351 to compare the same taxa in percentages.

2.2 Analyses of source areas

205 MAs are transported attached to aerosols in the atmosphere (Sang et al., 2016; Schreuder et al., 2018). To discuss potential source areas of MAs, we calculated exemplary backward trajectory ensembles of two days towards Lake El'gygytgyn using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPPLIT) 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; <http://ready.arl.noaa.gov/hypub-bin/trajtype.pl?runtype=archive>). Trajectories considered a mean particle injection height during a wildfire of between 100 and 1000 m after Peters and Higuera (2007).

210 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* 215 pollen represent typical taxa of cold tundra-steppe environments. *Pinus* 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.

220 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 225 `corr.test` of the R package `psych` (Revelle, 2018) using pairwise complete observations. Kendall's τ is only provided for $n > 5$.



3. Results

230 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.

235 *Fig. 2*

Concentrations vary during interglacials and late glacial stages, with low values during MIS 12, although concentrations are strongly affected by the sediment bulk density and 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 (Fig. 2b). Among interglacials, influxes are highest 240 during MIS 5e (e.g., LVG_{median}: 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, 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 245 insolation (Fig. 3). MA influx records are strongly positively correlated across all intervals (LVG vs. MAN or GAL: Kendall's $\tau = 0.76 - 0.82$; MAN vs. GAL: $\tau = 0.91 - 0.97$; Fig. 4a) with slightly closer relationships during MIS 8–5e compared to MIS 12–11c.

Fig. 3

MA influx records are not correlated with MA ratios (Fig. 4a) except for MIS 12–11c samples, which show a 250 significant inverse relationship for MAN and GAL influxes with LVG (MAN+GAL)⁻¹ (Kendall's $\tau = -0.74$ and -0.77). 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 clear 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⁻¹_{median}: 0.54, LVG (MAN+GAL)⁻¹_{median}: 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 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 widespread 260 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

265 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 τ 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
270 GAL and pine ($\tau = 0.6, p = 0.14$). During late MIS 12 and MIS 11c, we find a weak negative relationship between
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: $\tau = -0.6, p = 0.14$).

As both MA ratios show similar temporal trends, we used only LVG $(\text{MAN}+\text{GAL})^{-1}$ in the correlation analysis.
LVG $(\text{MAN}+\text{GAL})^{-1}$ 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 $(\text{MAN}+\text{GAL})^{-1}$ is negatively related to SGB ($\tau = -0.7, p = 0.07$) during MIS 12–
275 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

280 The abundance of LVG and its isomers in smoke in absolute (influxes) and relative (ratios) terms depends on the
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 use the proxies further back in time. We discuss the
285 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 pollen-based
vegetation reconstructions (Melles et al., 2012;Tarasov et al., 2013;Zhao et al., 2019) as described below have
290 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

295 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 lake sediments (Schüpbach et
al., 2015;Battistel et al., 2017;Callegaro et al., 2018;Dietze et al., in review). 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,



300 despite absolute age uncertainties that are several hundreds of years (Nowaczyk et al., 2007; Nowaczyk et al., 2013).

305 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 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

310 We 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; Bhattarai 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., in review). Our HYPSPPLIT 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 315 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 interglacials, 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 interglacials.

320 *Fig. 6*

325 MA degradation could still 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 Norwood et al. (2013) suggest there is substantial MA degradation in well-oxygenated river water. Yet, degradation 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 than other organic compounds.

330 Monitoring and sediment properties of Lake El'gygytgyn suggest mixed and well-oxygenated bottom waters during summers and past warm periods, whereas during glacial periods long-term lake stratification led to rather anoxic bottom water conditions (Melles et al., 2007; Nolan and Brigham-Grette, 2007). If we assume a constant influx of MAs and high degradation at the lake bottom, we would expect higher preservation during glacial periods 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 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 particles (Tobo et al., 2012), and all isomers show the same trend with time (Fig. 335 2b), which is indicative for bound and protected compounds (Hemingway et al., 2019). Hence, we assume that



El'gygytgyn sediment-based MA influxes are only marginally affected by degradation on centennial to orbital timescales, and, instead, represent relative changes in biomass burnt from a similar source during low-intensity fires.

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

340 4.2.1 Orbital-scale fire occurrence and the role of biomass and fuel availability, insolation and climate

More low-intensity biomass burning occurs during interglacials compared to glacial, as we find higher MA influxes during interglacials compared to the preceding late glacial. 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
345 percentage data due to (partly) unknown taxa-specific pollen and NPP dispersal, productivity and preservation (Sugita, 2007) and our low sample resolution prevents the use of new quantitative land cover reconstructions (Theuerkauf and Couwenberg, 2018). Yet, more biomass availability is suggested by significantly higher abundance of tree and shrub pollen in interglacial sediments compared to glacial cold tundra-steppe assemblages dominated by herbs and grasses (Fig. 5) (Melles et al., 2012; Tarasov et al., 2013). Accordingly, our rank
350 correlation analysis shows that MA influxes tend to increase with higher amounts of tree and shrub versus tundra-steppe pollen (Figs. 3c, 4b). Reasons could be that during glacial, low temperatures and CO₂ levels limit biomass (fuel availability) and fire spread, which has been 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
355 remote source areas, when fires associated with high-productivity biomes have shifted southwards.

We find also evidence for more biomass burning close to the lake in times of high summer insolation, in agreement with previous mid- to high latitude studies (Daniau et al., 2012; Remy et al., 2017; Dietze et al., 2018; Kappenberg et al., 2019). This is visible during the period of MIS 8–5e, when MA influxes not only peaked during MIS 7e and 5e, but also during maximum summer insolation of the preceding MIS 8 and 6 late glacial stages, despite pollen
360 data suggesting rather low tree and shrub presence during these intervals (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 MIS 5e (Yin and Berger, 2012). Overall, increased high latitude summer insolation seems to drive more biomass productivity and fires, for example, by the length of the growing season, but also by affecting land-atmosphere feedbacks and potential interhemispheric ice–ocean–land feedbacks that alter regional
365 precipitation–evaporation patterns (Yin and Berger, 2012; Melles et al., 2012; Martin Calvo et al., 2014).

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, where, 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).

370 4.2.2 Centennial- to millennial scale fire occurrence and the role of climate and vegetation composition



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 compared to MIS 5e.

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 MIS 12–11c (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 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.

There is also a tendency towards more MA influxes with increasing pine pollen abundance during MIS 8–5e (Fig. 3 a, c), whereas during MIS 12–11c *Pinus* s.g. *Haploxyton*-type pollen was not or weakly negatively related to MA influxes (τ not significant, Fig. 4b), maybe because different pine species with different fire-related traits could have produced the *P.* s.g. *Haploxyton*-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. 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. *Diploxyton*-type pollen, derived from *P. sylvestris*, are almost not present 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.

4.3.2 Centennial- to millennial scale burning conditions - Importance of the understory

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 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).
410 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., in review). As biomass differs
in its relative proportions of cellulose (LVG precursor), hemicellulose (MAN and GAL precursor), lignin and
others components (Simoneit et al., 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
415 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 no relation
420 between MA ratios and pine pollen during MIS 8–5e, and a tendency towards higher ratios LVG (MAN+GAL)⁻¹
when pine increased 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
425 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
430 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 ours. In addition, we find low ratios across all
samples, mainly independent of high or low influxes. Hence, we hypothesise that an additional biomass source
and/or specific burning conditions have contributed to the low sedimentary MA ratios found in Lake El'gygytgyn
435 sediments.

One additional biomass source could be dense moss–lichen mats within the summergreen boreal forest. There,
light can penetrate more easily through its open canopy 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
440 larch forests and helping to insulate the permafrost (Isaev et al., 2010; Lorant 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
445 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 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).

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

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.

- 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 low-temperature 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.
- 485 • 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.
- 490 • 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 and low-intensity fire-climate-vegetation feedbacks in space and time and potential ways of post-depositional degradation in even older interglacials, with
495 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.

500 **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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Figure Captions

510 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 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'gygytyn and location of analysed sediment cores (map based on Landsat-7 image, courtesy of the U.S. Geological Survey).
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Fig. 2: Ranges of monosaccharide anhydride (MA) composition during selected marine isotope stages 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 (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).
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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: sum of Poaceae, *Artemisia*, Chenopodiaceae, Caryophyllaceae, Cichoriaceae, and *Thalictrum* pollen; SGB: sum of *Larix*, *Populus*, and *Alnus* pollen; *Pinus* s.g. *Haploxyton*-type pollen; *Sphagnum* spore abundance.
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Fig. 4: Kendall's τ rank correlation coefficients between (a) MA influxes (LVG.yr, MAN.yr, GAL.yr) and ratios ($L.M = LVG \text{ MAN}^{-1}$; $L.MG = LVG \text{ (MAN+GAL)}^{-1}$) 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.* s.g. *Haploxyton*-type pollen; trees & shrubs: sum of *Betula*, SGB, *Salix*, *Pinus*, and *Picea*; MIS 12: 430–424 kyrs (n = 4), 11c: 423–406 kyrs (n = 10), 8: 272–243 kyrs (n = 11), 7e: 242–193 kyrs (n = 21), 6: 190–134 kyrs (n = 37), 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 Lake El'gygytyn (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>).
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