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# Modern isotope hydrology and controls on $\delta D$ of plant leaf waxes at Lake El'gygytgyn, NE Russia

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## Abstract

Stable isotope data from lipid biomarkers and diatom silica recovered from lake sediment cores hold great promise for paleoclimate and paleohydrological reconstructions. However, these records rely on accurate calibration with modern precipitation and hydrologic processes. Here we investigate the stable isotopic composition of modern precipitation, streams, lake water and ice cover, and use these data to constrain isotope systematics of the Lake El'gygytgyn basin hydrology. Compound specific hydrogen isotope ratios determined from modern vegetation are compared with modern precipitation and lake core top sediments. Multi-species net (apparent) fractionation values between source water (precipitation) and leaf wax lipids (mean value is  $-105 \pm 13\%$ ) agree with previous results in arid environments and provide a basis for application of this proxy downcore. We conclude that although there may be some bias towards winter precipitation signal, overall leaf wax lipids record annual average precipitation within the El'gygytgyn Basin.

## 1 Introduction

Hydrogen isotope ratios ( $\delta D$ ) of lipid biomarkers are emerging as a new paleoclimatic and paleohydrological proxy (e.g. Sauer et al., 2001; Huang et al., 2002; Jacob et al., 2007; Feakins and Sessions, 2010; Castaneda and Schouten, 2011, see references therein). The hydrogen in organic matter derives from meteoric waters, which have  $\delta D$  values that are strongly controlled by climatic variables such as temperature, elevation and water vapour source (Craig, 1961; Dansgaard, 1964; Gonfiantini, 1986; Gat, 1996). The isotopic composition of meteoric waters is also controlled by continental and "latitudinal effects" (Dansgaard, 1964; Craig, 1961; Gonfiantini, 1986; Gat, 1996; Bowen and Revenaugh, 2003; Kurita et al., 2004) which are reflected in the  $\delta D$  of plant leaf waxes ( $\delta D_{wax}$ ) (Sachse et al., 2006; Smith and Freeman, 2006; Hou et al., 2008; Liu and Yang, 2008; Polissar and Freeman, 2010). Changes in  $\delta D_{wax}$  in the geological record have

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been interpreted as shifts in precipitation  $\delta D$  ( $\delta D_p$ ) (e.g. Liu and Yang, 2008; Tierney et al., 2008) or in evapotranspiration (Schefuß et al., 2005; Sachse et al., 2006; Jacob et al., 2007). Qualitatively, a positive  $\delta D_{wax}$  shift interpreted as an increase in  $\delta D_p$  or an increase in evapotranspiration would indicate drier conditions in either case. However, while  $\delta D_{wax}$  is dependent on the  $\delta D$  of the source water, interpretations can be complicated by biosynthetic fractionations, plant life form (e.g. tree, shrub, grass) and water use, evapotranspiration from soil and leaf water and relative humidity (Craig and Gordon, 1965; Sessions et al., 1999; Liu and Yang, 2008; Smith and Freeman, 2006).

In general, abundance and composition of biomarkers can serve to identify past ecosystems and environmental parameters during their formation (Meyers, 2003). Fatty acids such as long-chain even carbon-numbered *n*-alkanoic acids (e.g. > 24 carbon chain length) typically originate from leaf waxes of terrestrial plants (Eglinton and Hamilton, 1967; Cranwell, 1974; Cranwell et al., 1987; Sachse et al., 2006). In contrast, short-chain *n*-fatty acids, (i.e. < 22 carbon atoms), are abundant as major constituents of cell membranes and waxes in many organisms, and are generally used as aquatic biomarkers (Cranwell, 1974; Cardoso et al., 1983; Cranwell et al., 1987; Meyers and Ishiwatari, 1993). Aquatic photosynthetic organisms in lakes obtain hydrogen from lake water to produce their organic compounds whereas terrestrial plants utilize hydrogen within the leaf water, which is subject to evapotranspiration (Leaney et al., 1985; Huang et al., 2004; Sachse et al., 2004; Zhang and Sachs, 2007). Thus, two isotopically distinct source waters are used for the biosynthesis of aquatic versus terrestrial biomarkers (Sachse et al., 2006). Lake water hydrogen isotopic variations are driven by  $\delta D_p$ , lake hydrology, and the residence time of the lake water (i.e. Leng and Marshall, 2004; Jacob et al., 2007). Large, well-mixed lakes with long lake water residence times, such as modern Lake El'gygytgyn ( $\sim 120$  yr; Fedorov et al., 2009, 2012), will “average out” short-term seasonal variations in precipitation and stream input isotopic compositions, and will reflect mean annual precipitation (Leng and Marshall, 2004; Nolan and Brigham-Grette, 2007). Leaf wax lipids derived from terrestrial plants instead record the isotopic ratio of soil water, which is ultimately recharged by precipitation. Snowmelt

is typically the dominant annual hydrological event in Arctic watersheds (Kane et al., 1992) and within the El'gygytgyn Basin soil water is annually recharged by snowmelt (Nolan and Brigham-Grette, 2007; Nolan, 2012).

Proxy use of  $\delta D_{\text{wax}}$  for paleoclimatic reconstructions depends on establishing a “net (or apparent) fractionation” between meteoric water and lipid  $\delta D$  values ( $\epsilon_{\text{wax/w}}$ ). Prior studies have shown a net fractionation for terrestrial plants in more humid climates up to  $-160\text{‰}$  (Chikaraishi and Naraoka, 2003; Sachse et al., 2006; Sessions, 2006; Smith and Freeman, 2006; Hou et al., 2007b), and lower average net fractionation in more arid climates (approx.  $-95\text{‰}$ ) (Hou et al., 2008; Feakins and Sessions, 2010). These values may be offset due to transpiration (i.e. enrichment of leaf water) in semi-arid to arid environments that act to reduce  $\epsilon_{\text{wax/w}}$ , possibly after passing some threshold beyond which  $\epsilon_{\text{wax/w}}$  remains insensitive to increasing aridity (Feakins and Sessions, 2010).

In addition to variable isotopic fractionations, large interspecies variability has been found at the individual plant scale (e.g. Chikaraishi and Naraoka, 2003; Liu and Huang, 2005), particularly in highly arid sites (Feakins and Sessions, 2010). Several studies have linked this variability to life form (e.g. tree, shrub, grass) (Liu et al., 2006; Liu and Yang, 2008), leading to concern for the potential to bias paleoclimatic reconstructions given inevitable vegetation shifts in response to climatic change. However, Feakins and Sessions (2010) find a strong correlation between  $\delta D$  of source water and the average  $\delta D_{\text{wax}}$  value at a site suggesting catchment-scale average  $\delta D_{\text{wax}}$  values reliably record regional climate despite a large spread in values between individuals. This is in good agreement with calibration studies using core-top sediments (e.g. Huang et al., 2004; Sachse et al., 2004; Hou et al., 2008) that show much better correlations to climate than studies of individual plants.

Here we establish local relationships between the  $\delta D$  of modern vegetation, sediment traps and the  $\delta D$  in modern El'gygytgyn lake sediments as the context for reconstruction of El'gygytgyn Basin paleohydrology. These studies are supplemented by isotopic analysis of samples of modern meteoric water, including winter and summer

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precipitation, streams, lake water and ice cover which provide insight into the modern isotope hydrology. This approach enables us to: (i) determine the net fractionation ( $\epsilon_{\text{wax/w}}$ ) between modern vegetation and source water within the El'gygytgyn Basin, (ii) assess the delivery of organic matter and target compounds for  $\delta\text{D}$  analysis, (iii) establish a framework for separating variations in the isotopic composition of lake water from changes in the isotopic composition of precipitation in the  $\delta\text{D}_{\text{wax}}$  paleohydrological proxy.

## 2 Study area: location and climate

Lake El'gygytgyn is situated within a  $3.58 \pm 0.04$  Ma old meteorite crater (Layer, 2000) located in a previously unglaciated region of the Far East Russian Arctic ( $67^{\circ}30' \text{N}$ ,  $172^{\circ}05' \text{E}$ ; Glushkova and Smirnov, 2007; Fig. 1). The crater rim elevations vary between 600 and 930 m, while the lake surface lies at 492 m a.s.l. The crater is approximately 18 km in diameter with a total watershed area of  $293 \text{ km}^2$  ( $110 \text{ km}^2$  is lake surface) and the lake is 12 km wide and 175 m deep with a volume of  $14.1 \text{ km}^3$  (Nolan and Brigham-Grette, 2007). Water temperatures in this cold monomictic, ultra oligotrophic lake are  $< 4^{\circ}\text{C}$  and the lake is fed by 50 streams draining from within the crater rim (Cremmer et al., 2005; Nolan and Brigham-Grette, 2007). Shallow lagoons with seasonal water temperatures  $> 6^{\circ}\text{C}$  are dammed by gravel berms surrounding the lake edge (Nolan and Brigham-Grette, 2007). Mean annual air temperature measured from 2002–2008 was  $-10.4^{\circ}\text{C}$  and total precipitation rose from  $350 \text{ mm a}^{-1}$  to about  $550 \text{ mm a}^{-1}$  over this period with roughly equal rates between winter and summer precipitation (Nolan and Brigham-Grette, 2007; Nolan, 2012).

The El'gygytgyn Basin is located in a zone of hypoarctic tundra (Yurtsev, 1973) and modern vegetation within the basin is dominated by lichen and discontinuous herbaceous taxa (Kohzevnikov, 1993; Minyuk, 2005; Lozhkin et al., 2007). *Salix* and Poaceae species comprise the dominant woody and non-woody taxa, respectively and modern tree line is located ca. 150 km to the south and west of the lake (Lozhkin et al., 2007).

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### 3 Methods

#### 3.1 Sample collection

##### 3.1.1 Water samples

Lake water and precipitation samples were collected during the 2003 El'gygytgyn Field Expedition in 30 ml Nalgene bottles and stabilized with one drop mercuric chloride (Juschus et al., 2005a). Water from incoming streams and the Enmyvaam outflow were likewise collected and all water samples were stored at 4 °C at the Geoscience Dept., UMass Amherst until analysis. Precipitation samples were also collected over the winter 2008–2009 (Table 1).

##### 3.1.2 Modern vegetation samples

Herbarium samples of typical modern plants growing close to the lake were sampled in summer 2003. All plants were collected during the growing season. This procedure was accomplished in order to establish a standard pollen collection of the local vegetation at Lake El'gygytgyn (Minyuk, 2005). Seven representative herbarium samples (courtesy P. Minyuk, NESRI, RAS Magadan) were subsampled, lyophilized, ground with a mortar and pestle and analysed at the Geoscience Dept., UMass Amherst for lipids and isotopic analysis.

##### 3.1.3 Sediment traps and sediment core tops

Sediment traps were deployed during the spring Field Expedition, 2003 (Fig. 2). Each sediment trap consisted of a base plate made of plastic with a steel rod to fix the trap in vertical position at the rope. Four plastic tubes with a diameter of 46 mm were screwed to the bottom of this plate. The rope was anchored at the lake bottom with a bag filled with pebbles and kept up in the water column by buoys fixed beneath the base of the

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lake ice (Juschus et al., 2005b). Unfortunately, sediment traps were only recovered at one location during the 2003 Expedition yielding only 6 samples.

Samples of the sediment water interface were also collected during the spring 2003 field expedition. Coring was carried out from a floating platform with a gravity corer; attention was paid to horizontal preservation of the sediment water interface. Twelve of these samples were selected for biomarker analysis of which four samples were selected for further preparation for compound specific isotopic analysis. Additionally, a sample from the upper 0–2 cm of a 16 m long sediment core (LZ1024) was also isolated and prepared for analysis. These samples represent modern sedimentation in the lake.

## 3.2 Analytical methods

### 3.2.1 Water isotopic analysis

The oxygen and hydrogen isotopic compositions of the water samples were analyzed at the Alfred Wegener Institute for Polar and Marine Research (AWI) Potsdam, Germany with a Finnigan MAT Delta-S mass spectrometer equipped with two equilibration units for online determination following the methods of Meyer et al. (2000). The stable isotope ratios  $r_{\text{O}} = {}^{18}\text{O}/{}^{16}\text{O}$  and  $r_{\text{H}} = \text{D}/{}^1\text{H}$  are measured for both sample and reference and the final  ${}^{18}\text{O}$  and  $\delta\text{D}$  values are calculated relative to Vienna Standard Mean Ocean Water (V-SMOW) in permil (Gonfiantini, 1978):

$$\delta = \frac{r_{\text{sample}} - r_{\text{VSMOW}}}{r_{\text{VSMOW}}} \times 10^3 \quad (1)$$

The water samples show an analytical precision  $< 0.10\%$  ( $1\sigma$ ) for  $\delta^{18}\text{O}$  and  $< 0.8\%$  for  $\delta\text{D}$ , respectively (Meyer et al., 2000).

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### 3.2.2 Compound-specific isotopic analysis (CSIA)

The method for vegetation, sediment traps and all sediment samples is identical. Samples were freeze-dried, crushed and stored in combusted glassware. Soluble organic matter was extracted using an accelerated solvent extractor (ASE200, Dionex Corp., Sunnyvale, USA) with a dichloromethane/methanol mixture (9 : 1) at 100 °C and 1200 psi for three 15-min cycles. Given the relatively low amount of organic C in the samples (Melles et al., unpublished), the maximum amount of sediment available was used (between ~7–12 g dry sediment). The total lipid extract (TLE) was separated into neutral and carboxylic acid fractions on a solid phase extraction (SPE) column containing 0.5 g of DSC–NH<sub>2</sub> stationary phase (Supelco). A neutral fraction was eluted with 4 ml hexane:DCM (1 : 1) and archived for future analysis. Fatty acids were eluted with 8 ml of 2 % formic acid in DCM and were subsequently derivatized as methyl esters (FAME) with BF<sub>3</sub>MeOH. Compounds (i.e. FAMES) were identified and quantified using a Hewlett Packard 6890 series gas chromatograph – mass selective detector (GC-MSD) and a Hewlett Packard 6890 series gas chromatograph – flame ionization detector (GC-FID), respectively, and with comparisons to external standard mixtures. Both the GC-MSD and GCFID were equipped with 5 % phenyl methyl siloxane columns (30 m × 0.25 mm i.d., film thickness 0.25 μm). The GC oven temperature programs were also identical for both instruments: 60 °C (2 min), 15 °C min<sup>-1</sup> to 315 °C (15 min) with helium as the carrier gas and the inlet temperature set at 320 °C.

The δD of individual lipids were determined at the ESCSIS facility at Yale University using a Thermo Finnigan MAT 253 mass spectrometer with a gas chromatograph combustion interface (GCC) with a high-temperature pyrolysis furnace operated at 1440 °C (Hilkert et al., 1999) coupled with a Thermo TraceGC GC-MS equipped with a DB-5ms column (30 m, ID: 0.32 mm, film thickness: 0.5 μm, Agilent). The gas chromatography (GC) temperature program was: 80 °C (2 min) to 120 °C at 12 °C min<sup>-1</sup> and then to 320 °C at 6 °C min<sup>-1</sup> (held 16 min). For δD analysis, each sample was analysed in triplicate and the H<sub>3</sub><sup>+</sup> factor was determined daily prior to standard calibration and sample

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analysis. The results are reported using standard delta notation (i.e.  $\delta D$  values) in permil (‰) units. Isotope ratios were normalized relative to a co-injected standard mixture containing  $C_{14}$  *n*-alkane to condition the furnace and  $C_{15}$  and  $C_{38}$  *n*-alkanes as compounds of known isotopic values (obtained from A. Schimmelmann, Indiana University).

Instrument performance was also determined daily by measuring a suite of 5 FAMES with a co-injected standard mixture ( $nC_{14}$ ,  $nC_{15}$ ,  $nC_{38}$ ) of known isotopic values. The mean precision for replicate analyses of the standard compounds was typically better than 5‰ ( $1\sigma$ ).

### 3.3 Data handling

#### 3.3.1 Isotope mass balance correction

Fatty acid  $\delta D$  values were measured on methyl ester derivatives. A *n*- $C_{14}$  acid standard was derivatized with  $BF_3MeOH$  (same lot number as used with samples) to calculate the  $\delta D$  contribution from the three H atoms added during the methylation reaction with  $BF_3$  in MeOH. The bulk  $\delta D$  of this *n*- $C_{14}$  fatty acid standard ( $-240.4‰ \pm 1.5‰$ ;  $n = 5$ ) was determined prior to derivatization offline by Thermal Conversion Elemental Analysis (TCEA, ESCSIS Yale University). Derivatized standard was measured in triplicate to yield an average  $\delta D$  value for the methyl contribution from the derivitization agent ( $-172.2‰ \pm 2.5‰$ ). This value was used to correct the measured  $\delta D$  values of fatty acid methyl esters for the added hydrogen atoms by mass balance.

#### 3.3.2 Calculation of net “apparent” fractionations

Isotopic fractionations between two measured substrates,  $\delta D_a$  and  $\delta D_b$  are reported as enrichment factors. Enrichment factors are reported in permil notation, implying a factor of 1000 (Cohen et al., 2007). Here we present the “net (or apparent) fractionation” ( $\epsilon_{wax/w}$ ) between the  $\delta D$  of leaf wax ( $\delta D_{wax}$ ) and the  $\delta D$  of source water ( $\delta D_w$ ),

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commonly used in the plant leaf wax literature, where:

$$\varepsilon_{\text{wax/w}} = (\alpha)_{\text{wax/w}} - 1 = \frac{\delta D_{\text{wax}} + 1}{\delta D_{\text{w}} + 1} - 1 \quad (2)$$

Although  $\varepsilon_{\text{wax/w}}$  is typically calculated relative to  $\delta D$  of mean annual precipitation, this inherently assumes that the isotopic composition of the source water ( $\delta D_{\text{w}}$ ) is identical to local  $\delta D$  values of precipitation ( $\delta D_{\text{precip}}$ ). In this study, the water available for plant uptake is largely spring snowmelt carried in streams and draining through the shallow active layer (see Sect. 4.1; Nolan, 2012), therefore we used the empirically determined average  $\delta D$  for stream inflow. We also calculate the net fractionation between plant leaf wax and annual average precipitation in the basin ( $\varepsilon_{\text{wax/precip}}$ ) for comparison.

### 3.3.3 Model parameters

In order to assess the sensitivity of the lake basin to changes in the lake water residence time and changing  $\delta D_{\text{p}}$  and  $\delta^{18}\text{O}_{\text{p}}$  over time, we constructed an isotope mass balance model of the lake (Fig. 3)

$$\delta_{\text{lake}(i+t)} = \delta_{\text{lake}(i)} + \left( \frac{\delta_{\text{in}(i)} - \delta_{\text{lake}(i)}}{\tau} \right) \cdot \Delta t \quad (3)$$

where  $\delta_{\text{lake}(i)}$  represents the initial  $\delta$  value of the lake water,  $\delta_{\text{in}}$  represents the isotopic composition of input or precipitation,  $\tau$  and  $t$  represent the lake residence time and timestep (here defined as 1 due to the annual resolution of the tree-ring timeseries), respectively. Initial lake conditions were assigned based on the present day mean  $\delta D_{\text{lake}}$  ( $-155\text{‰}$ ) and is similar to reconstructed input approximations. As long term records of past  $\delta D_{\text{p}}$  are lacking in this region, we used  $\delta^{18}\text{O}$  data from tree ring cellulose in Northern Siberia (Sidorova et al., 2008) to reconstruct the isotopic ratios of past precipitation for input to our model. In order to derive isotope ratios of precipitation from  $\delta^{18}\text{O}_{\text{cellulose}}$ , we used the relationship between source water (here assumed to be precipitation) and

$\delta^{18}\text{O}_{\text{cellulose}}$  described in Anderson et al. (2002). We assigned a mid-range value of 0.6 for the fraction of leaf water not subject to evaporation ( $f$ ) (e.g. Saurer et al., 1997) and a relative humidity ( $h$ ) of 78%. Average relative humidity was determined from the automated weather station data collected at Lake El'gygytyn 2002–2008 (Nolan, 2012).

5 Conversion of  $\delta^{18}\text{O}_{\text{reconstructed precip}}$  to our proxy model input,  $\delta\text{D}_{\text{in}}$  where  $\delta\text{D}_{\text{p}} \approx \delta\text{D}_{\text{in}}$ , was accomplished by applying our LMWL equation established below (see Sect. 4.1;  $\delta\text{D} = 7.35\delta^{18}\text{O} - 9.6$ ).

## 4 Results

### 4.1 $\delta\text{D}$ and $\delta^{18}\text{O}$ values of water samples

10 Precipitation collected in summer 2003 (rain), winter 2008/2009 (snow), streams, lake ice and lake water profile samples were analysed for their oxygen and hydrogen isotopic compositions (Fig. 4). Mean  $\delta^{18}\text{O}$  and  $\delta\text{D}$  values for the different hydrological components are overall lower than previously reported values for the El'gygytyn basin (Table 1; Schwamborn et al., 2006). Snow samples  $\delta^{18}\text{O}$  values ranged from  $-33.90\text{‰}$  to  $-14.10\text{‰}$  (mean:  $-23.16\text{‰}$ ) and  $\delta\text{D}$  values from  $-271.0\text{‰}$  to  $-100.5\text{‰}$  (mean:  $-179.9\text{‰}$ ) (Table 1). Rain  $\delta^{18}\text{O}$  values range from  $-15.91\text{‰}$  to  $-12.37\text{‰}$  (mean  $-14.29\text{‰}$ ) and  $\delta\text{D}$  values range from  $-127.5\text{‰}$  to  $-98.1\text{‰}$  (mean  $-114.8\text{‰}$ ) (Table 1). Summer precipitation shows much lower deuterium excess (d-excess) values (mean d-excess =  $-0.5\text{‰}$ ) than winter precipitation and is offset from the GMWL consistent with observations by Schwamborn et al. (2006). The mean d-excess values for both snow and stream samples ( $5.4\text{‰}$  and  $6.7\text{‰}$  respectively) are similar while lake water d-excess values are lower. Annual average precipitation is calculated partitioning precipitation into a 60% winter contribution and 40% summer input (annual average  $\delta^{18}\text{O} = -19.3\text{‰}$ ,  $\delta\text{D} = -152.9\text{‰}$ ) based on Nolan and Brigham-Grette (2007). These proportions are derived from measurements of water equivalency made in a single year

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but are thought to be representative for modern climate (Nolan and Brigham-Grette, 2007).

The  $\delta D$ - $\delta^{18}O$  relationship in winter precipitation samples from the El'gygytyn Basin closely follows the global meteoric water line (GMWL; Craig, 1961) and defines a linear relationship of:

$$\delta D = 7.35(\pm 0.24)\delta^{18}O - 9.62(\pm 5.64) \quad (4)$$

$n = 35; R^2 = 0.97$

where  $n$  is the number of data points and  $R^2$  is the coefficient of determination (Fig. 4). Streams, lake ice and lake water profile samples also display similar linear relationships, close to the GMWL and consistent with a circum-polar regression compiled from the International Atomic Energy Agency/World Meteorological Organization (IAES/WMO) Global Network of Isotopes in Precipitation (GNIP) database ( $\delta D = 7.26\delta^{18}O - 5.98; R^2 = 0.99$ ; Lacelle, 2011; IAEA/WMO, 2006). Summer precipitation defines a Local MWL (LMWL) different from the GMWL ( $\delta D = 6.80\delta^{18}O - 22.3; R^2 = 0.95$ ) indicating kinetic fractionation (Fig. 4). These results are consistent with previous observations within the El'gygytyn basin (Schwamborn et al., 2006) and in Northern Siberia (Meyer et al., 2002a,b; Sugimoto et al., 2003; Kurita et al., 2004; Opel et al., 2011).

The isotopic composition of precipitation and temperature are strongly correlated at mid- and high latitudes (Dansgaard, 1964), occurring because the degree of rain-out from the atmosphere through condensation is highly correlated with the condensation temperature (see Jouzel et al., 1997 for review). Figure 5 shows the  $\delta D$ - $T$  and  $\delta^{18}O$ - $T$  relationships for the El'gygytyn basin for both winter and summer precipitation ( $n = 26$ ). Positive spatial relationships ( $\delta^{18}O = 0.43 T - 18.5, R^2 = 0.68$ ;  $\delta D = 3.1 T - 143.7, R^2 = 0.64$ ) are observed between the isotopic compositions of precipitation and air temperature. Temperatures used are "precipitation temperature" (i.e. surface air temperature during a precipitation event) as isotopic values are discrete events, not weighted means. The slope coefficients of the  $\delta$ - $T$  relationships presented

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here are similar to the spatial  $\delta^{18}\text{O}$ -mean air temperature (MAT) relation for Taymyr and Lena data ( $\delta^{18}\text{O} = 0.59 \text{ MAT} - 11.47$ ,  $R^2 = 0.91$ ; Boike et al., 1997) and data from 47 European IAEA/WMO stations to the west ( $\delta^{18}\text{O} = 0.59 \text{ MAT} - 14.35$ ; Rozanski et al., 1992). However, the slope coefficient for  $\delta D$ - $T$  relationship at El'gygytgyn is lower than previously reported by Rozanski et al. (1992) ( $4.5\% \text{ } ^\circ\text{C}^{-1}$ ) and from the Siberian Network of Isotopes in Precipitation ( $\delta D_{\text{winter}} = 5.6 T_{\text{DJF}} - 77.9$ ;  $R^2 = 0.93$ ;  $\delta D_{\text{summer}} = 5.1 T_{\text{JJA}} - 157.3$ ;  $R^2 = 0.55$ ; Kurita et al., 2004).

Spatial variations in the isotopic composition of stream water samples were observed. Stream  $\delta^{18}\text{O}$  values ranged from 24.23‰ to -16.68‰ (mean -18.91‰) and  $\delta D$  values ranged from -179.5‰ to -127.8‰ (mean -144.9‰; Table 1). Relatively isotopically enriched values cluster predominantly along the south facing and western slopes (Fig. 2.). Most negative  $\delta^{18}\text{O}$  values and  $\delta D$  values were found along the eastern side of the crater, in streams with relatively high topographic gradients (Fig. 2). Decreasing  $\delta^{18}\text{O}$  and  $\delta D$  values broadly correlate with higher relief ( $R^2 = 0.44$ ), with the exception of the shortest stream (Stream 37). There is a general trend toward increasing  $\delta^{18}\text{O}$  and  $\delta D$  values over the course of the summer sampling interval ( $R^2 = 0.33$ ).

Sampled profiles of the lake water show small ( $\delta^{18}\text{O}$ :  $\sim 1.5\%$ , Chapligin et al., 2012;  $\delta D$ :  $\sim 11\%$ , Fig. 6) seasonal variations in the upper 2 m surface waters. These variations are likely due to spring (May) input from streams carrying isotopically depleted snowmelt and evaporative enrichment during summer (August) (Chapligin et al., 2012). Below 2 m water depth, the lake is well mixed with an average  $\delta D$  value of  $-155.1 \pm 0.7\%$  ( $\delta^{18}\text{O}$ :  $-19.82\%$ ; Chapligin et al., 2012).

### 4.2 Modern vegetation samples

We report abundances and hydrogen isotope ratios for  $C_{20}$ ,  $C_{24}$ ,  $C_{26}$ ,  $C_{28}$ , and  $C_{30}$   $n$ -alkanoic acids from representative samples of modern vegetation from within the Lake El'gygytgyn basin (Fig. 7). Despite the small sample set ( $n = 7$ ), we observe a large interspecies variation in the distribution of compounds with some species

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producing relatively high concentrations of short-chain compounds typically associated with aquatic organisms (i.e.  $nC_{20}$  and  $nC_{22}$  acids; Cranwell et al., 1987; Ficken et al., 2000). The average chain length (ACL =  $\sum(C_n \cdot n) / \sum C_n$ , where  $n = 20, 22, 24, 26, 28, 30$ ) for modern vegetation varied between species, ranging from 24 to 27 (Table 2).

Concentrations are normalized per g of total lipid extract (TLE). The highest concentrations of  $C_{30}$ - $n$ -alkanoic acid were found in the *Douglasia ochotensis* and *Rumex sp.* samples whereas lowest concentrations were observed in *Artemisia* and *Bryophyta sp.* High concentrations of  $nC_{28}$  acid were found in the *Bryophyta sp.* but concentrations of all other  $n$ -alkanoic acids were very low or below detection limit.

For all plants yielding measurable  $n$ -alkanoic acids,  $\delta D$  values are reported in Table 2. We observe differences in  $n$ -alkanoic acid chain length abundances for different species as well as isotopic offsets between different chain lengths in some species. Thus, a mass-weighted mean  $\delta D$  value ( $\delta D_{wax}$ ) was calculated to provide a common basis for comparison of the  $\delta D$  of different species regardless of chain length abundances (Table 2). Within lake sediments, only  $nC_{30}$  acid can be attributed solely to a terrestrial source (Holland et al., 2012) therefore, we also compared  $\delta D_{wax}$  values with the  $\delta D$  values of  $nC_{30}$  acid (i.e.  $\delta D_{30}$ ). The hydrogen isotope compositions of  $C_{30}$   $n$ -alkanoic acid ( $\delta D_{30}$ ) strongly correlate with the concentration-weighted  $\delta D_{wax}$  values ( $r^2 = 0.97$ ) confirming  $\delta D_{30}$  values are representative of integrated  $\delta D$  values for terrestrial leaf waxes from plants within the Lake El'gygytgyn watershed (ranging between  $-227.5\%$  and  $-260.5\%$ ). Net fractionation values calculated between plant leaf wax (i.e.  $nC_{30}$  acid) and two different source waters (stream water,  $\varepsilon_{wax/streams}$  and annual average precipitation,  $\varepsilon_{wax/precip}$ ) are similar but show slightly higher values with annual average precipitation (mean  $\varepsilon_{wax/precip} = -107.4 \pm 12\%$ ; Table 2).

### 4.3 Sediment traps and lake sediments

Concentrations of even chain  $n$ -alkanoic acids ( $nC_{20}$ ,  $C_{22}$ ,  $C_{24}$ ,  $C_{26}$ ,  $C_{28}$ , and  $C_{30}$ ) varied widely between sediment traps and samples of the upper 0–2 cm of lake sediments (Fig. 8). Sediment traps show variable distributions of compounds and concentrations

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with water depth with highest concentrations of all compounds at 90 m water depth (Fig. 8). Individual compounds were normalized by total yield of *n*-alkanoic acids to show changes in the relative proportions of compounds (normalized to total yield of *n*-alkanoic acids) with water depth (Fig. 9). The upper surface waters show high percentages of  $C_{20}$ ,  $C_{22}$ ,  $C_{24}$ , and  $C_{26}$  acids with a marked decline of  $C_{22}$  acid by 50 m depth. The shortest chain *n*-alkanoic acid,  $nC_{20}$ , decreases with depth. Below 50 m,  $C_{30}$  and  $C_{22}$  acids appear to co-vary, whereas  $C_{24}$ ,  $C_{26}$ , and  $C_{28}$  follow a similar general trend down to 130 m water depth. Concentrations of  $C_{24}$  and  $C_{26}$  increase at the deepest sediment trap (146 m water depth), yielding higher percentages of these compounds near the lake bottom than in surface waters. Abundances of all compounds in all sediment traps were too low for  $\delta D$  analysis.

High concentrations of all compounds were observed in the upper 0–2 cm of lake sediments, increasing in concentrations with increasing lake water depth (note log scale, Fig. 8). The distribution of all compounds is consistent across all samples as indicated by calculated values of average chain length ( $ACL = \sum(C_n \cdot n) / \sum C_n$ , where  $n = 20, 22, 24, 26, 28, 30$ ) (Table 2). The hydrogen isotope compositions of sedimentary *n*-alkanoic acids are also reasonably consistent across samples (i.e. sedimentary  $\delta D_{30}$  values varied by  $\sim 10\text{‰}$ ; within  $2\sigma$  analytical error). Net fractionation values calculated between terrestrially sourced  $nC_{30}$  acid and the different source water signals (stream water,  $\epsilon_{30/\text{streams}}$  or annual average precipitation,  $\epsilon_{30/\text{precip}}$ ) are similar but show slightly higher values with annual average precipitation (mean  $\epsilon_{30/\text{streams}} = -114.0 \pm 13\text{‰}$ ; mean  $\epsilon_{30/\text{precip}} = -105.4 \pm 13\text{‰}$ ; Table 2).

## 5 Discussion

### 5.1 Modern isotope hydrology

The isotopic composition of modern precipitation and various components of the hydrological system provide the basis for applying paleoclimatic interpretations to stable



isotope compositions of sediment archives (Schwamborn et al., 2006). Here we outline the modern isotope hydrology from precipitation to stream runoff to lake water and discuss the major controls on each component. Each of these components affects the hydrogen isotope composition ultimately recorded in the  $\delta D$  values from leaf wax lipids (Fig. 10).

Precipitation regimes are influenced by air temperature, proximity to moisture sources and air mass trajectories over landmasses. Consequently, the  $\delta^{18}O$  and  $\delta D$  values in local precipitation (i.e.  $\delta D_p$ ) are highly variable and the slope values of local meteoric precipitation often deviate from the GMWL. Spatially, lower slope values are located in areas with local orographic effects and continental climatic regimes, while higher slope values are from sites with maritime climates (Lacelle, 2011). East Siberian Arctic winter precipitation exhibits a wide range of stable isotopic compositions (e.g. Kurita et al., 2004) and although the values presented here are not weighted means, the range of variation (e.g.  $\Delta\delta^{18}O$  values = 17‰ and  $\Delta\delta D$  values = 19‰) is similar to that previously reported within the El'gygytgyn Basin (Schwamborn et al., 2006). Modern winter precipitation at Lake El'gygytgyn closely follows the  $\delta D$  and  $\delta^{18}O$  values of precipitation from other Arctic sites in the GNIP database. The values of the slope (7.35) and the y-intercept (-9.62) are lower than the GMWL but very close to the GNIP-based circum-polar regression, indicating a nearly unaltered precipitation signal from the original moisture source during winter months. In contrast, summer precipitation shows a clear kinetic fractionation of isotopes with lower slope and d-excess values. This may be related to enhanced evaporation occurring during summer months in typical tundra settings due to the water surplus on the land surface and continuous 24 h net radiation (Ohmura, 1982; Boike, 1997). Reprecipitated moisture derived from recycled water masses from the land surface may also be included (Sugimoto et al., 2003; Kurita et al., 2004; Schwamborn et al., 2006).

The seasonal contrast in modern precipitation may also be explained by the strong seasonality in the dominant weather patterns. Modern synoptic weather patterns identified from National Centers for Environmental Prediction (NCEP) global reanalysis data

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show strong low pressure systems over the Aleutians during winter and broad high pressure systems to the south and east with weaker lows to the north during the summer (Nolan et al., 2012). Strong Aleutian Lows during winter tend to bring cold Arctic air from the east and north to the lake, whereas summer synoptic weather patterns bring warm continental air from the south and west (Nolan et al., 2012). Thus, these dominant weather patterns affect both the seasonal temperature change and source of moisture delivery to Lake El'gygytgyn. Seasonal air temperature changes affect the saturation vapour pressure of air masses and subsequently the degree of rainout, yielding decreasing precipitation isotope ratios with decreasing temperatures and along trajectories of atmospheric vapour transport. These trajectories deliver winter precipitation from a proximal north and eastern moisture source (i.e. E. Siberian, Chukchi and Bering seas and possibly North Pacific) and recycled moisture from the Siberian land surface from the south and west during the summer months. High d-excess values of winter precipitation in the El'gygytgyn Basin support a proximal moisture source with minimal alteration of the original isotopic composition while low d-excess values during summer months is consistent with recycled continental moisture from the south and west. Furthermore, Nolan et al. (2012) suggest that these weather patterns have been relatively stable with time and are likely representative of this and other interglacial periods.

Input to the lake via streams draining from the crater rim is predominantly driven by winter snowmelt and shows spatial variations in isotopic composition according to stream gradient and slope aspect (Fig. 2). Stream samples are more  $^{18}\text{O}$  and D depleted than overall summer precipitation isotope ratios with values closer to mean annual precipitation ( $\delta D_p$ ). This suggests that stream waters carry a large winter precipitation signal offset by some input of isotopically enriched summer precipitation and/or evaporative enrichment of the soil water. Water tracks, characteristic of permafrost terrain and immature drainage systems, are common on El'gygytgyn slopes with rapid drainage of moisture through the active layer in early spring/summer (Nolan and Brigham-Grette, 2007; Nolan, 2012; Federov et al., 2012). The residence time

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of the water in the soil typically produces a dampening of the seasonal variations observed in precipitation (Buhay and Edwards, 1995; Anderson et al., 2002), however, this residence time is exceedingly short within the El'gygytgyn Basin. Therefore, stream water effectively represents the soil moisture in the basin, draining rapidly and recharged annually by snowmelt. Soil moisture data collected from different depths within the El'gygytgyn Basin (to base of active layer; 2002–2008) shows peak soil moisture in early spring prior to peak soil temperatures and with individual summer precipitation events having only a minor effect on soil moisture content (Federov et al., 2012). Deep permafrost in this region ( $\sim 500$  m; Yershov, 1998; Schwamborn et al., 2006) precludes deep groundwater input and streams are therefore representative of soil moisture recharged by winter precipitation draining through the shallow active layer during the short summer season.

Streams with south facing and western slopes display relatively enriched isotope compositions. Lower gradients for these slopes, longer transit times and seasonally greater net incoming solar radiation likely resulted in  $^{18}\text{O}$  and D enrichment of these stream waters due to evaporation. These streams were also sampled late in the summer season, well after peak discharge and when flow in many of the smaller streams was reduced ( $< 1 \text{ m}^3 \text{ s}^{-1}$ ) and is often just a trickle (Nolan and Brigham-Grette, 2007). Thus, despite summer sampling during maximum expected evaporative enrichment, the isotopic ratios of stream samples appear biased towards more negative winter precipitation isotope ratios though offset by some minor evaporative enrichment.

Small seasonal changes are also observable in the isotopic composition of the lake water. Profiles of the lake water column show seasonal enrichment of the surface waters in summer and depleted surface waters in early spring. Although thermally stratified in winter due to ice cover, the lake is rapidly mixed following snowmelt and ice breakup in late May (Nolan and Brigham-Grette, 2007) with little variation below the uppermost surface waters. Isotopically depleted lake surface waters (2 m depth, Fig. 6) collected in late May are likely the combined result of melting lake surface snow and ice and input from streams carrying isotopically-depleted snowmelt. Evaporative

enrichment of the lake surface waters in summer (August) is small and likely very brief given vertical mixing due to strong winds (Nolan and Brigham-Grette, 2007) and has little to no influence on the overall isotopic composition of the lake (Chapligin et al., 2012).

5 The mean hydrogen isotope composition of the lake water ( $\delta D_{\text{lake}}$ ) is largely controlled by the hydrogen isotope composition of precipitation ( $\delta D_p$ ), duration of ice cover and the residence time of the lake. Input to the lake is a combination of direct precipitation on the lake surface and stream discharge. Streams account for 85 % of water input to the lake (Fedorov et al., 2012) and prolonged or perennial ice cover prevents evaporative enrichment of surface waters. Accordingly, the average isotopic ratio of modern lake water is slightly more depleted in  $^{18}\text{O}$  and D when compared to mean annual precipitation values, reflecting either greater input from isotopically depleted snowmelt and/or a lag in response based on lake residence time to past changes in  $\delta D_p$ .

10 To assess the impact of this potential lag, lake water isotopic response was modelled by isotope mass balance (Fig. 3). Variations in  $\delta D_p$  are reflected in the lake water isotopic response within decades ( $\geq 10$  yr) but the magnitude of the response is strongly attenuated by the residence time. Modern residence times for Lake El'gygytgyn are  $\sim 120$  yr (Fedorov et al., 2009, 2012) but could have been greater during ice-covered, glacial conditions. Although the modelled  $\delta D_{\text{lake}}$  does not record the full magnitude of minor fluctuations of  $\delta D_p$ , these results suggest  $\delta D_{\text{lake}}$  does still respond to large magnitude changes in input on long glacial-interglacial timescales (see also Chapligin et al., 2012).

## 5.2 Climate signal preserved in plant leaf waxes

### 5.2.1 Modern vegetation

25 Modern vegetation within the El'gygytgyn Basin produced a range of *n*-alkanoic acids with variable concentrations and  $\delta D$  values. Reconstructions of the  $\delta D$  of meteoric water rely on a constant "net fractionation" between lipid and meteoric water.

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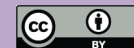
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Differences between plant species and vegetation type may affect the net fractionation (e.g. Chikaraishi and Naraoka, 2003; Smith and Freeman 2006; Hou et al., 2007), however within watersheds these differences are integrated and a representative multi-species net fractionation factor may be appropriate for paleohydrologic reconstructions (Feakins and Sessions, 2010). Modern vegetation within the El'gygytgyn Basin includes 249 identified plant species and approximately an additional 100 rare species (Kohzenikov, 1993; Minyuk, 2005), yet, despite the small sample set, the seven species analysed here appear to largely represent the modern vegetation contribution to modern lake sediments.

In order to evaluate how representative these 7 species are of all vegetation within the basin, we calculated an average distribution of compounds from modern vegetation samples assuming an equal weighting of all species to produce a representative assemblage of *n*-alkanoic acids within the El'gygytgyn Basin (Fig. 11). This modern vegetation distribution pattern was then compared with an average distribution of compounds from modern sediments calculated from all four sediment core tops. The two patterns are similar although as expected there are discrepancies given the relatively small sample set and the modern floristic diversity within the basin. The modern vegetation distribution pattern appears particularly sensitive to input from Bryophyta, as it produces *n*C<sub>28</sub> acid almost exclusively in high concentrations. Reducing the contribution from Bryophyta from the overall modern vegetation pattern results in a distribution similar to that observed in modern lake sediments. This may be because Bryophyta may not be a significant portion of overall plant biomass in the basin or alternatively may not be major contributor of terrestrially sourced *n*-alkanoic acids to the lake. Additionally, *Salix arctica* and *Artemisia spp.* are abundant on El'gygytgyn slopes and modification of their proportional contributions produces a nearly identical distribution of compounds to modern lake sediments (Fig. 11). Minor differences may certainly be attributed to and accounted for with the further inclusion and proportional representation of the remaining vegetation types and species. This preliminary survey suggests that although the proportional contributions of each species to the overall pattern may

change, these six species (excluding Bryophyta) are generally representative of the overall production and input of even-chain  $n$ -alkanoic acids from modern vegetation to the lake basin.

The hydrogen isotope composition observed in sampled vegetation may also be representative basin-wide. The range of interspecies  $\delta D$  variability observed around Lake El'gygytgyn is within other published ranges (eg. Chikaraishi and Naraoka, 2003; Liu and Huang, 2005; Sachse et al., 2006, Hou et al., 2007; Feakins and Sessions, 2010) although, these authors note that there is significant scatter at the level of individual plants across a wide range of climatic conditions. Some studies have linked  $\delta D$  values to life form (e.g. tree, shrub, and grass; Smith and Freeman, 2006; Liu et al., 2006; Liu and Yang, 2008) however, no clear linkage between  $\delta D$  values and life form was found in our study. Yet, despite a large spread between individuals, catchment scale average  $\delta D_{\text{wax}}$  values generally do seem to reliably record the climate conditions of the region (e.g. Huang et al., 2004; Sachse et al., 2004; Feakins and Sessions, 2010) thereby allowing for a constant offset to be used for paleoenvironmental reconstructions.

Our calculated  $\varepsilon_{\text{wax/w}}$  values (Table 2) are also within published ranges (Liu and Yang, 2008 and references therein) with an overall average net fractionation between modern vegetation and stream water ( $\varepsilon_{\text{wax/streams}}$ ) of  $-116.0 \pm 12\text{‰}$ . However, this assumes that the source water available for plant uptake is directly represented by streams in the El'gygytgyn Basin (i.e.  $\delta_w = \delta_{\text{streams}}$ ). Average net fractionation between modern vegetation and annual average precipitation ( $\varepsilon_{\text{wax/precip}}$ ) is slightly more positive ( $-107.0 \pm 12\text{‰}$ ). These values are similar to average net fractionations of  $-99 \pm 8\text{‰}$  reported from a North American transect of  $C_{28}$   $n$ -alkanoic acids (Hou et al., 2008) and  $-94 \pm 21\text{‰}$  from long chain ( $C_{27}$ ,  $C_{29}$  and  $C_{31}$ )  $n$ -alkanes in arid to semi-arid ecosystems (Feakins and Sessions, 2010). In a global compilation, Liu and Yang (2008) report a high latitude effect with smaller  $\varepsilon_{\text{wax/w}}$  values for woody plants versus grasses at high latitudes. Within the El'gygytgyn Basin, *Salix* and *Poaceae* comprise the dominant woody and non-woody taxa, respectively (Minyuk, 2005; Lozhkin et al., 2007). We find

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no clear evidence of D/H fractionations related to life form and although limited, our data instead agrees with the average multi-species net fractionation ( $-94\text{‰} \pm 21\text{‰}$ ) found by Feakins and Sessions (2010), despite the lack of grasses in their dataset. Both net fractionations ( $\epsilon_{\text{wax}/\text{streams}}$  and  $\epsilon_{\text{wax}/\text{precip}}$ ) determined here are very similar.

5 However, potential changes in the seasonality of precipitation and Arctic summer productivity (i.e. timing of biosynthesis with respect to peak snowmelt and runoff) suggests caution in application of  $\epsilon_{\text{wax}/\text{streams}}$  in paleohydrological reconstructions. These values could also be affected by inclusion of more plant samples and species given the large range of known  $\delta D$  diversity between individuals and the small sample set used here.

10 Nevertheless, these results contribute towards establishing a constant net fractionation factor for arid environments and provide the basis for paleoclimatic reconstructions from sedimentary records of  $\delta D_{\text{wax}}$ .

### 5.2.2 Sediment traps

15 The abundances and distribution of compounds produced by vegetation directly affects the integrated signal captured in lake sediments and, thus, any sedimentary isotopic signature could be complicated by the inclusion of compounds from multiple sources with differing isotopic composition. Parallel compound-specific  $\delta^{13}\text{C}$  analysis shows that within El'gygytgyn sediments,  $n\text{C}_{30}$  acid derives from a strictly terrestrial source while other even long chain  $n$ -alkanoic acids derive from a mixed terrestrial and aquatic source (Wilkie et al., 2012; Holland et al., 2012). This same study further shows that

20  $n\text{C}_{20}$  and  $n\text{C}_{22}$  acids likely derive from an aquatic source, however production of these compounds in modern vegetation (this study, see above) initially suggests some contribution of a terrestrial source of these compounds to lake sediments. This explicitly requires intact delivery of these terrestrial sourced compounds to the lake bottom. Our

25 results show the abundances of these compounds vary with depth in the water column, suggesting multiple processes in the water column influencing the ultimately integrated signal captured in lake sediments.



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Even short chain *n*-alkanoic acids (e.g.  $nC_{14} - nC_{22}$ ) typically produced by phytoplankton and/or bacteria (Cranwell et al., 1987; Gong and Hollander, 1997), are more labile and subject to rapid turnover in the water column (Lee et al., 2004; Jones et al., 2008). Thus, the observed trend in  $nC_{20}$  acid is consistent with aquatic production in the upper surface waters and degradation with depth. Combined terrestrial inputs and aquatic production of even chain  $nC_{20} - C_{26}$  acids likely contributes a higher proportion of these compounds to surface waters, while the sharp decline by 50 m depth could reflect early degradation of the more labile shorter chain  $nC_{22}$  acid. Although  $nC_{24}$  and  $nC_{26}$  acids are typically attributed to terrestrial sources, some aquatic production (i.e. subaquatic/floating macrophytes and filamentous algae similar to those observed in El'gygytgyn streams and lagoons) of these mid-chain fatty acids has been reported (Cranwell et al., 1987; Ficken et al., 2000).

Relative abundances of  $nC_{24}$  and  $nC_{26}$  acids increase in the deepest water column sample, exceeding values observed in upper surface waters. This suggests either additional input at depth (e.g. delivery of terrestrial material from dense hypopycnal flows from warming moat and lagoon waters), autochthonous production at intermediate to lower depths in the water column (> 90 m water depth), and/or advantageous capture of a sinking organic debris entrained in and subsequently released by melting snow and lake ice. However, delivery of terrestrial sourced  $C_{24}$  and  $C_{26}$  acids by hypopycnal flows is unlikely as this would also result in a concomitant increase in concentrations and relative proportions of  $nC_{28}$  and  $nC_{30}$  acids. While we cannot rule out autochthonous preferential production of  $nC_{24}$  and  $nC_{26}$  acids below 90 m water depth, capture of previously ice entrained sinking organic debris seems likely during the period of sediment trap deployment (late May to mid-July). Substantial lake ice persisted for most of the trap deployment, with extensive ice melting, development of leads and moat formation occurring throughout sample collection (Fig. 9). Ice-rafted fatty acids (IRFAs), possibly entrained during the previous fall freeze and accumulated over winter through aeolian deposition on lake snow and ice, would be released to the water column sporadically during lake ice melt and breakup. These settling plumes of IRFAs

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could explain the increase in total concentrations of all fatty acids at 90 m water depth, while shifting proportions with depth could reflect variable melt and release times of the ice over the site. The isotopic ratios of these IRFAs would still reflect their original biological sources regardless of transport and ice residence times though the signal from compounds of both aquatic and terrestrial sources would be obscured. Further deployment and study of time-series sediment traps is required to fully assess modern water column processes and contributions to lake floor sediments.

### 5.2.3 Lake sediments

Lake sediments represent an integrated signal from the watershed and the carboxylic acids within them typically originate from multiple sources (Meyers, 2003). Concentrations of fatty acids in core top sediments increase exponentially towards the deepest part of the basin consistent with greater accommodation space and lower sediment supply resulting in less dilution of the organic signal. An age model developed on the deepest core from the central part of the lake (LZ1024, 170 m water depth) yields an age of 200 yr for the upper 0–1 cm. This sampling resolution for the lake surface sediments implies that seasonal to interannual variations are lost and the “modern” sediments are an integrated average of climate and deposition for the last 200 yr. The consistent distribution of compounds across surface sediment samples regardless of water depth confirms a catchment-integrated organic signal reaching the lake bottom. Thus, the climatic signal from leaf waxes in lake surface sediments is a combined average for all vegetation changes in the catchment in the last 200 yr (i.e. molecular abundances). Additionally, the sedimentary  $\delta D_{wax}$  also represents an integrated signal of precipitation for the last 200 yr. Long-term  $\delta D_p$  records are lacking at this site, however the reconstructed estimates of  $\delta D_p$  from the Northern Yakutia  $\delta^{18}O_{cellulose}$  record (Sidorova et al., 2008) yields a 97-yr average reconstructed  $\delta D_p$  of about  $-147\text{‰}$ . Net fractionations between core top sediment and either streams ( $-100.8 \pm 5.0\text{‰}$ ) or precipitation ( $-94.5 \pm 5.0\text{‰}$ ) closely agree with  $\epsilon_{wax/w}$  values from modern vegetation (streams:

$\epsilon_{\text{wax}/\text{streams}} = -116.0 \pm 12\%$ ; precipitation:  $\epsilon_{\text{wax}/\text{precip}} = -107.4 \pm 12\%$ ). Calculation of these values inherently applies present day input to a time-integrated signal yet recalculation with the derived time-averaged  $\delta D_p$  still yields near identical net fractionation values ( $\sim -100\%$ ).

## 6 Conclusions

This study provides a detailed assessment of the stable isotopic composition of modern hydrology within the El'gygytyn Basin and the controls on the  $\delta D$  signature from plant leaf waxes within the El'gygytyn Basin. Our results suggest different source regions for winter and summer precipitation, characterized by an unaltered isotopic signature from a northern and eastern moisture source in winter and a south-southwest continental moisture source including recycled moisture from the land surface. LMWL determined within the El'gygytyn Basin is consistent with other circumpolar regressions (Kurita et al., 2004; Lacelle, 2011), though the local spatial  $\delta$ - $T$  relationships are lower than previously reported. However, evaporative enrichment is not a strong control on  $\delta D_{\text{lake}}$  and the lake responds relatively quickly to large magnitude changes in  $\delta D_p$ . Changes in the length of the residence time are reflected in the degree of dampening of the  $\delta D_p$  driven signal, with longer residence times averaging out shorter term (e.g. decadal) changes. Inflow from streams in the basin represent soil water ( $\delta_{\text{soil water}} = \delta_{\text{streams}}$ ) that is annually recharged by snowmelt, drained rapidly and relatively unaffected by summer precipitation events. Enhanced evaporative enrichment on southfacing slopes causes  $\delta_{\text{streams}}$  to more closely reflect annual average  $\delta D_p$ . Plants utilize this water within the active layer so  $\delta D_{\text{wax}}$  should reflect  $\delta_{\text{streams}}$ . However, large interspecies variability and potential changes in seasonality of precipitation suggest caution in applying  $\epsilon_{\text{wax}/\text{streams}}$  in paleohydrological reconstructions. Therefore, it may be more prudent to use  $\epsilon_{\text{wax}/\text{precip}}$  for paleohydrological reconstructions (both values are within  $1\sigma$  and agree with previously published ranges). Differences between samples are more likely due to randomly distributed net fractionations rather than a systematic offset between

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woody vs. grass samples (as predicted for high latitudes). However, larger sample sets including more species and multiple individuals within each genus are needed. In modern lake sediments:  $\varepsilon_{C_{30}/precip} = -94.5 \pm 5\text{‰}$  which is determined only from terrestrial sourced  $nC_{30}$  acid and represents catchment wide integrated signal (200 yr of all basin vegetation and  $\delta D_p$ ). Utilizing reconstructed ( $\sim 100$  yr average)  $\delta D_{p, cellulose}$  to calculate net fractionations for 200 yr-integrated lake sediments yields  $\varepsilon_{30/precip} = -96 \pm 8\text{‰}$ . This time averaged  $\varepsilon_{30/precip}$  value provides a robust net “apparent” fractionation to be used in future paleohydrological reconstructions.

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**Table 1.** Mean values ( $\delta^{18}\text{O}$ ,  $\delta D$ , d-excess) for  $\text{H}_2\text{O}$  sample sets.

$\text{H}_2\text{O}$ sample sets	$\delta^{18}\text{O}$ (‰) vs. VSMOW	$\delta D$ (‰) vs. VSMOW	d-excess	$\delta^{18}\text{O}$ (‰) OIPC <sup>a</sup>	$\delta D$ (‰) OIPC <sup>a</sup>
Snow	−23.16	−179.9	5.4	−21.4 <sup>b</sup>	−163 <sup>b</sup>
Rain	−14.29	−114.8	−0.5	−15.4 <sup>c</sup>	−123 <sup>c</sup>
Streams	−18.91	−144.9	6.7		
Lake Ice	−19.27	−151	3.1		
Lake profiles	−19.79	−154.9	3.4		

<sup>a</sup> 95 % confidence interval of model values determined from the Online Isotopes in Precipitation Calculator (OIPC), version 2.2 (Bowen and Revenaugh, 2003).

<sup>b</sup> Arithmetic mean of OIPC modelled monthly values for Siberian winter months (November–April).

<sup>c</sup> Arithmetic mean of OIPC modelled monthly values for Siberian summer months (June–August).

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**Table 2.** Concentrations and  $\delta D$  values for selected *n*-alkanoic acids from vegetation, sediment traps from various water depths, and lake bottom sediment from various depths with the El'gygytgyn Basin.

Sample	Concentration ( $\mu\text{g g}^{-1}$ TLE)						ACL*	$\delta D_{20}$	$\delta D_{22}$	$\delta D_{24}$	$\delta D_{26}$	$\delta D_{28}$	$\delta D_{30}$	$\delta D_{\text{max}}$	$\epsilon_{\text{streams}}^{\text{C}_{30\text{W}}}$	$\epsilon_{\text{precip}}^{\text{C}_{30\text{W}}}$	$\epsilon_{\text{streams}}^{\text{wax/w}}$	$\epsilon_{\text{precip}}^{\text{wax/w}}$
	C <sub>20</sub>	C <sub>22</sub>	C <sub>24</sub>	C <sub>26</sub>	C <sub>28</sub>	C <sub>30</sub>												
<i>Rumex sp.</i>	6.7	16.6	28.2	28.1	66.4	46.8	27	-247.5	-262.6	-258.5	-263.1	-262.4	-259.3	-260.5	-134.0	-125.6	-135.4	-127.0
<i>Saxifrage</i>	19.6	17.8	45.1	13.6	13.9	29.2	25		-226.1				-229.6	-227.5	-99.3	-90.6	-96.8	-88.1
<i>Douglasia ochotensis</i>	18.4	34.8	91.4	61.6	96.4	111.6	26	-244.7	-234.1	-242.0	-243.3	-248.9	-244.0	-243.8	-116.1	-107.5	-115.8	-107.3
<i>Poacea</i>	18.0	32.1	35.7	72.5	23.6	10.2	25		-236.6	-236.3	-256.1	-238.6		-245.4			-117.8	-109.3
<i>Artemisia</i>	7.5	19.1	21.6	7.3	7.9	2.6	24	-224.8	-245.7	-249.2	-253.3	-256.9	-242.0	-246.5	-113.7	-105.2	-119.0	-110.5
<i>Bryophyta</i>	3.2	6.1			71.2	3.9	27					-258.8		-245.4			-117.7	-109.2
<i>Salix arctica</i>	29.4	63.1	111.2	178.9	131.6	15.0	25	-219.4	-222.8	-227.5	-247.2	-247.4	-236.0	-238.3	-106.8	-98.2	-109.5	-100.8
<b>AVERAGE</b>													<b>-242.2</b>	<b>-243.9</b>	<b>-114.0</b>	<b>-105.4</b>	<b>-116.0</b>	<b>-107.4</b>
Sediment trap (30 m)	14.3	20.6	21.4	15.4	7.9	7.2	24											
Sediment trap (50 m)	8.2	8.7	10.2	8.1	4.3	5.6	24											
Sediment trap (90 m)	23.5	41.9	30.3	25.5	12.7	25.8	24											
Sediment trap (130 m)		6.0	7.7	4.7	3.5	4.2	25											
Sediment trap (146 m)		1.1	1.6	1.2	0.6	0.7	25											
Sediment water interface (159 m)	12.2	39.2	78.3	72.5	68.4	21.1	25	-235.3	-246.2	-243.7	-241.6	-244.6	-238.5	-243.0	-107.2	-101.0	-114.9	-106.4
Sediment water interface (168 m)	136.6	412.0	818.3	776.6	626.8	230.1	25	-253.2	-242.7	-236.1	-235.5	-236.8	-228.2	-237.2	-95.2	-88.9	-108.1	-99.5
Sediment water interface (170 m)	1340.7	4273.0	9016.1	8819.4	7488.4	2660.2	25	-245.4	-249.5	-239.1	-242.3	-243.5	-232.8	-242.0	-100.6	-94.3	-113.8	-105.2
Sediment core top (LZ1024; 170 m)	1879.8	5585.9	11258.1	10564.1	9172.4	3215.6	25	-245.8	-244.1	-234.4	-231.1	-232.4	-232.4	-234.8	-100.2	-93.9	-105.3	-96.7
<b>AVERAGE</b>													<b>-233.0</b>	<b>-239.2</b>	<b>-100.8</b>	<b>-94.5</b>	<b>-110.5</b>	<b>-101.9</b>

\* ACL = Average Chain Length as defined in text.

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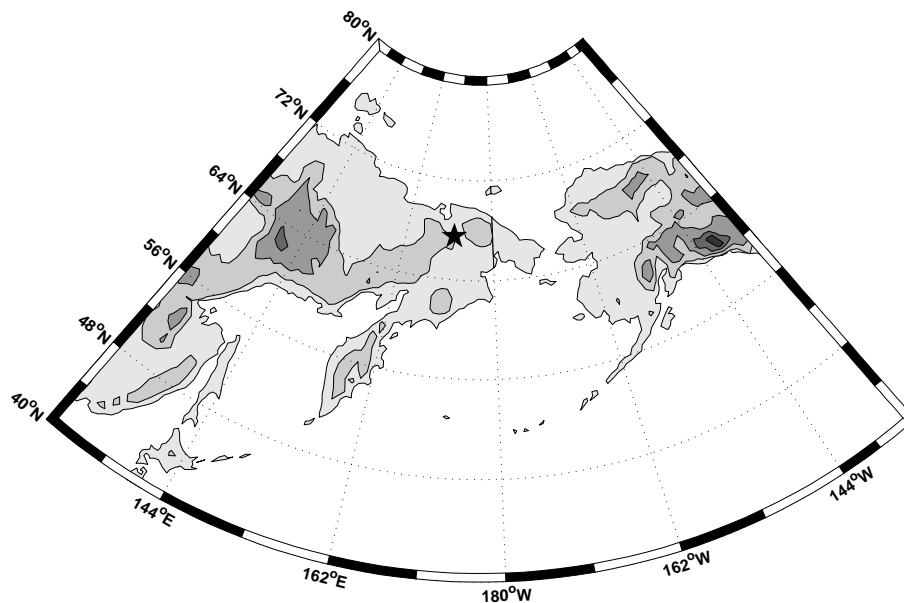
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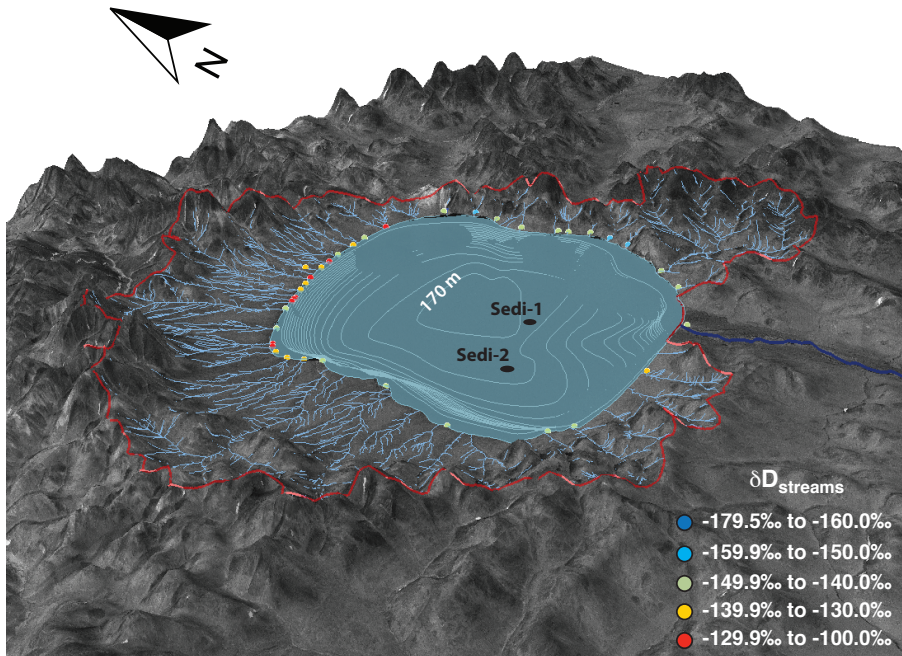
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**Fig. 1.** Location of Lake El'gygytyn (black star) in NE Russia.

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**Fig. 2.** Location of the sediment traps deployed in Lake El'gygytyn in 2003, streams and the  $\delta D$  of sampled stream water. View to the NE with 5 $\times$  vertical exaggeration. Coloured dots indicate measured  $\delta D$  of stream waters collected summer 2003. Red line denotes watershed, streams are indicated in light blue lines. Contours indicate lake bathymetry.

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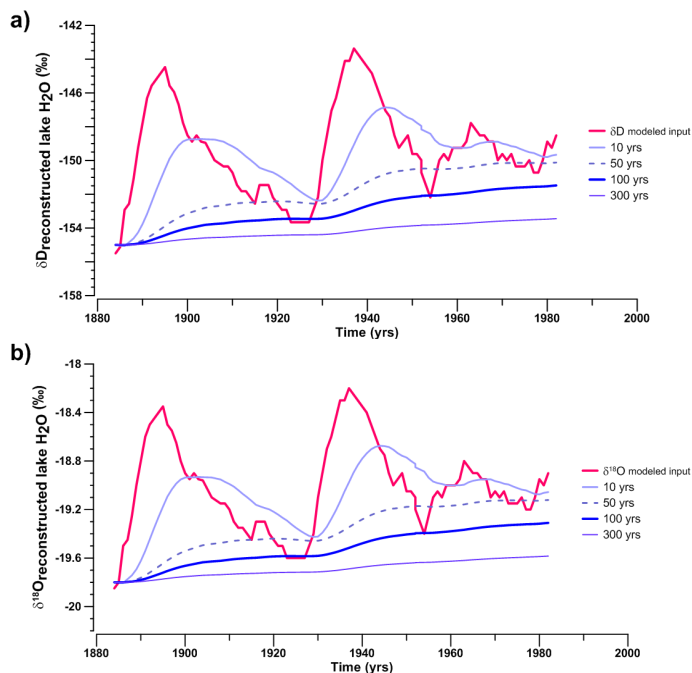
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**Fig. 3.** Isotopic response of the lake water to changes in residence time and variable  $\delta D_p$  of input (precipitation). Pink lines denotes reconstructed (a)  $\delta D$  and (b)  $\delta^{18}\text{O}$  of precipitation from  $\delta^{18}\text{O}_{\text{cellulose}}$  (Sidorova et al., 2008) based on Anderson et al. (2002). Blue curves show modeled response of  $\delta D_{\text{lake}}$  and  $\delta^{18}\text{O}_{\text{lake}}$  with various residence times (10, 50, 100 and 300 yr).

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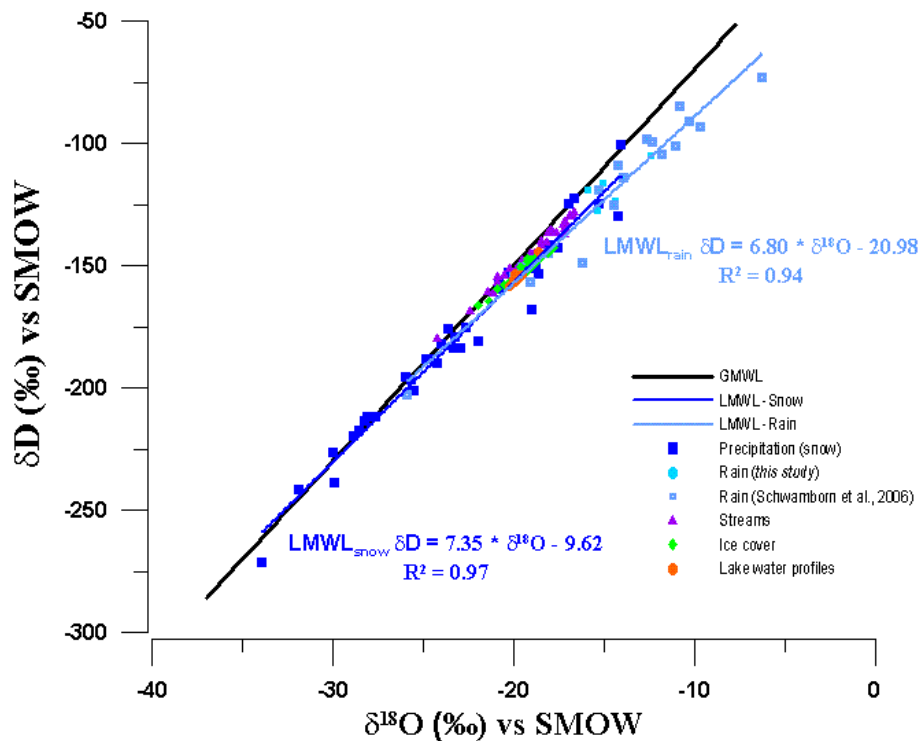
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**Fig. 4.**  $^{18}O/D$  bi-plot of  $H_2O$  samples from modern precipitation (snow, rain), streams, lake water profiles, surface waters, and ice cover.

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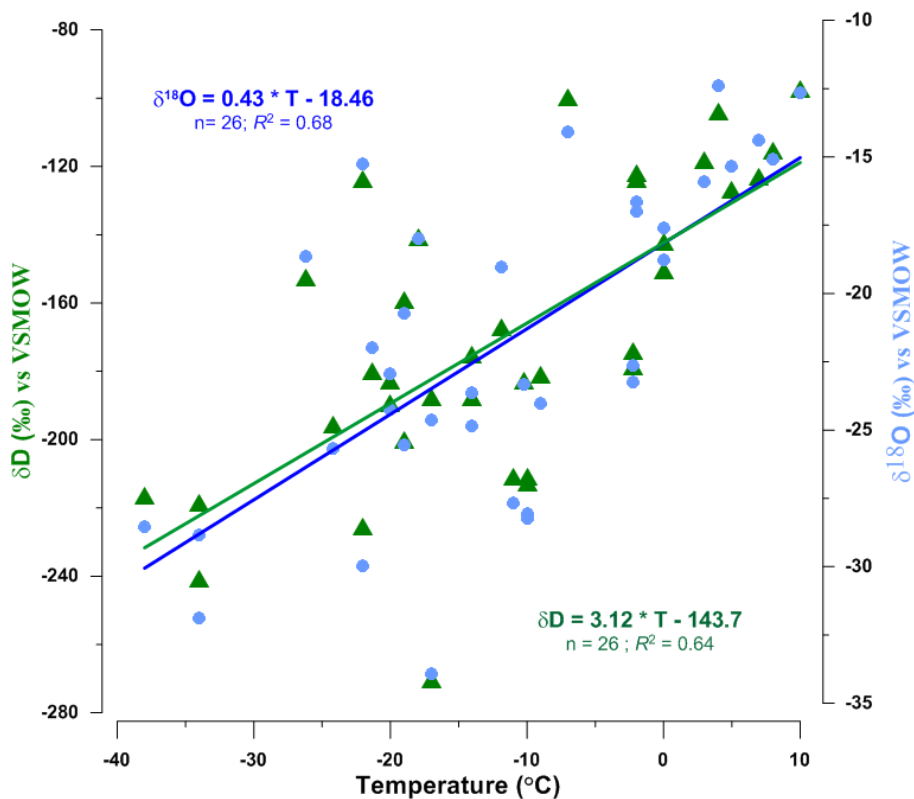
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**Fig. 5.** Relationship between  $\delta^{18}O$  and  $\delta D$  of precipitation with temperature within the El'gygytyn Basin.

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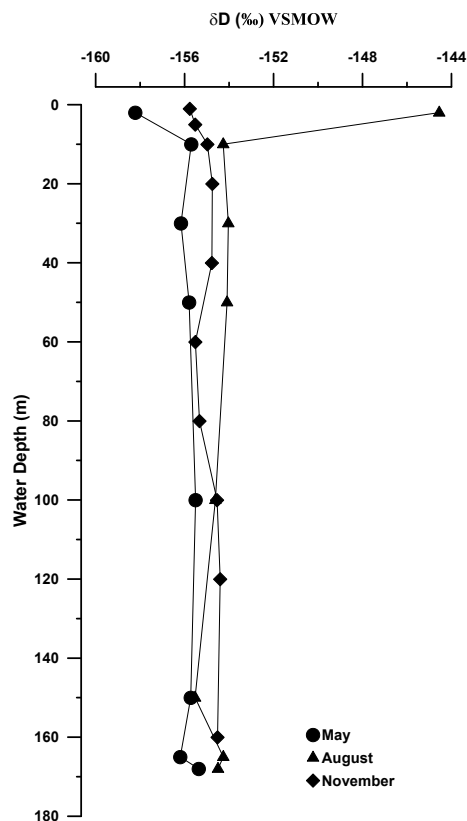
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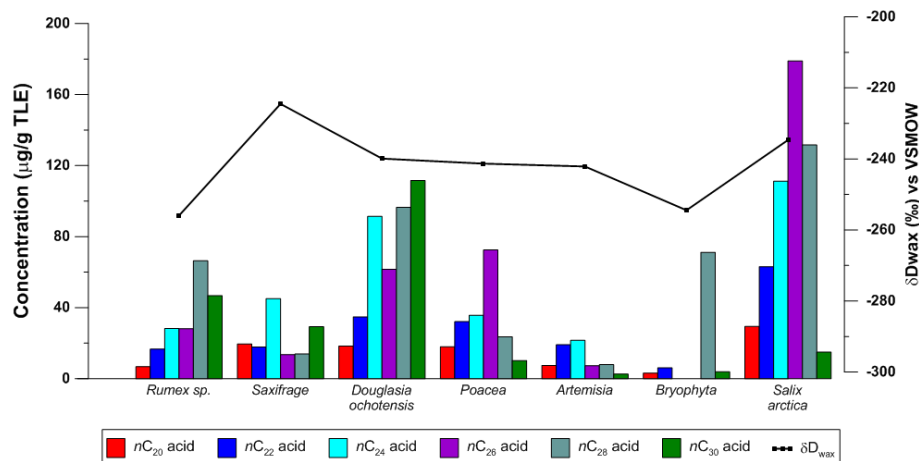


**Fig. 6.** Hydrogen isotope composition of water from Lake El'gygytgyn at various depths, sampled in different months (May and August, 2003; November 2008).

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**Fig. 7.** Concentrations and  $\delta D$  values of  $n$ -alkanoic acids measured in modern vegetation samples from within the El'gygytgyn Basin.

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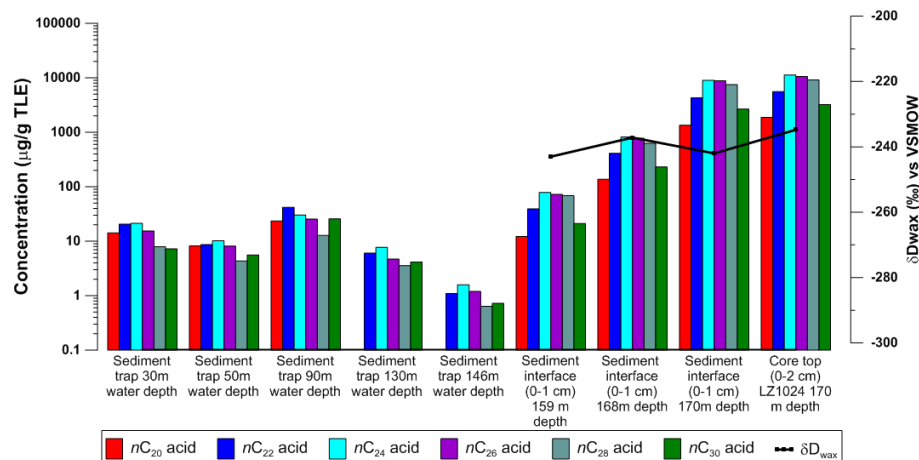
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**Fig. 8.** Concentrations of *n*-alkanoic acids with water column depth in sediment traps and lake surface sediments (note log scale).  $\delta D_{wax}$  values are plotted for lake surface sediments only as concentrations of *n*-alkanoic acids in sediment traps were insufficient for compound-specific  $\delta D$  analysis.

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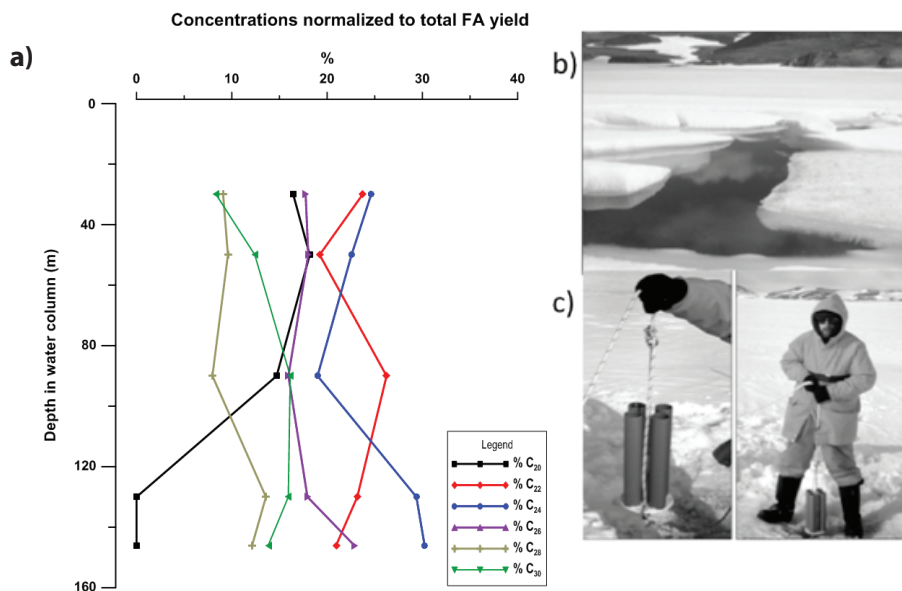
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**Fig. 9.** Concentrations of *n*-alkanoic acids normalized to total yield of *n*-alkanoic acids by water column depth in Lake El'gygytyn. **(b)** Large moat formed by ice melt 500 m to the north of the southern shore on 17 June 2003 – approximately 1/3 way through sediment trap deployment. (from Melles et al., 2005). Note extent of snow and lake ice cover during typical summer month. **(c)** Sediment traps deployed 31 May 2003 and recovered 19 July 2003. (from Melles et al., 2005).

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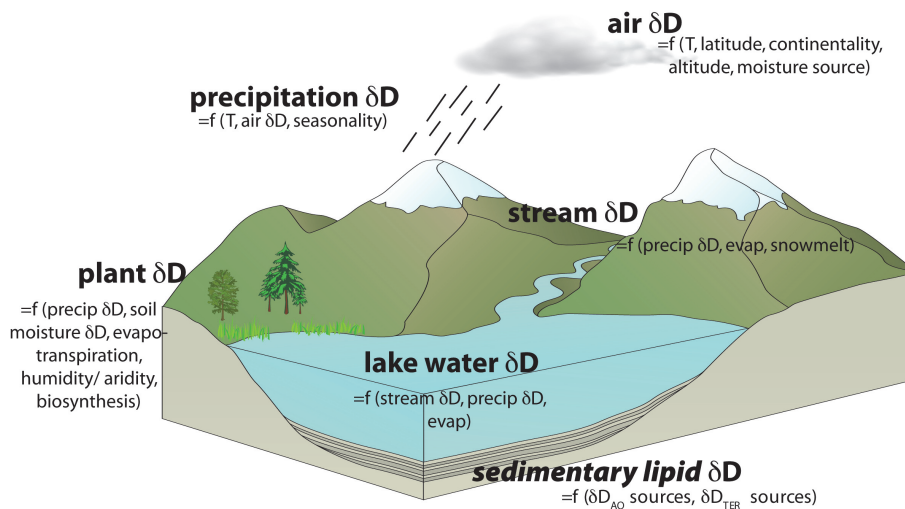
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**Fig. 10.** Conceptual diagram of major factors controlling the modern isotope hydrology at Lake El'gygytgyn.

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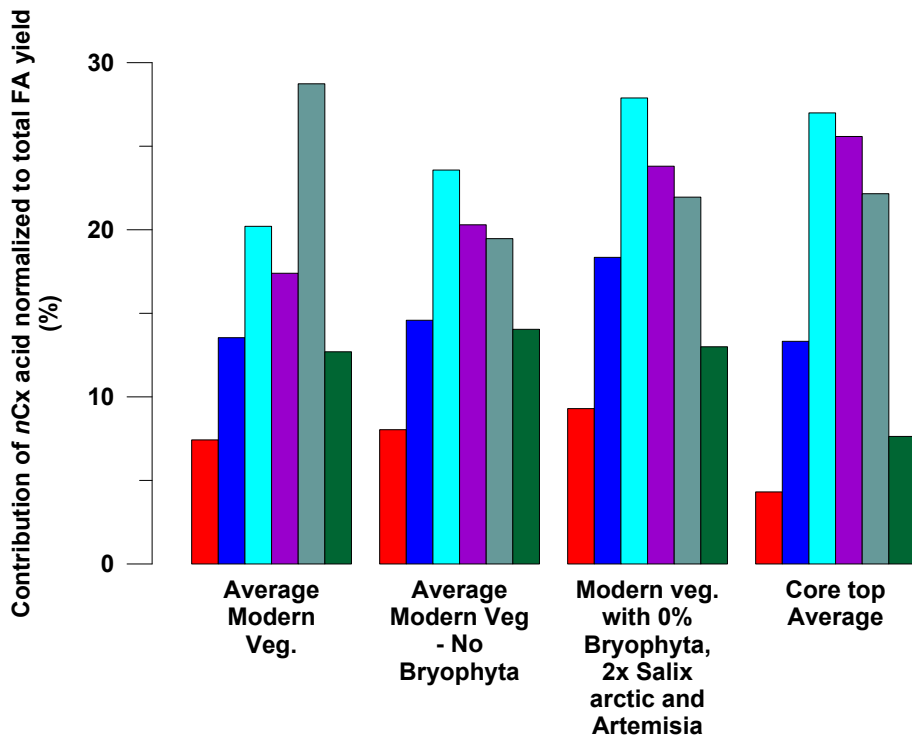
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**Fig. 11.** Average distribution of *n*-alkanoic acids from modern vegetation within the El'gygytgyn Basin (far left) and in core top sediments (far right). Modeled distributions are shown with varying proportions of vegetation species. Compounds are normalized to total yield of *n*-alkanoic acids.

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