High resolution leaf wax carbon and hydrogen isotopic record of late Holocene paleoclimate in arid Central Asia

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Abstract

Central Asia is located at the confluence of large scale atmospheric circulation systems. It is thus likely to be highly susceptible to changes in the dynamics of those systems, however little is still known about the regions paleoclimate history. Here we present carbon and hydrogen isotopic compositions of *n*-alkanoic acids from a late Holocene sediment core from Lake Karakuli (eastern Pamir, Xinjiang Province, China). Instrumental evidence and isotope-enabled climate model experiments with the Laboratoire de Météorologie Dynamique Zoom model version 4 (LMDZ4) demonstrate that δD values of precipitation in the region are influenced by both temperature and precipitation amount. We find that those parameters are inversely correlated on an annual scale; i.e. climate varies between cool/wet and dry/warm over the last 50 years. Since the isotopic signals of these changes are in the same direction and therefore additive, isotopes in precipitation are sensitive recorders of climatic changes in the region. Additionally, we infer that plants are using year round precipitation (including snow-melt) and thus leaf wax δD values must also respond to shifts in the proportion of moisture derived from westerly storms during late winter/early spring. Downcore results give evidence for a gradual shift to cooler and wetter climates between 3.5 and 2.5 cal kyr BP, interrupted by a warm/dry episode between 3.0–2.7 kyr BP. Further cool and wet episodes occur between 1.9–1.5 kyr BP and between 0.6–0.1 kyr BP, the latter coeval with the Little Ice Age. Warm and dry episodes between 2.5–1.9 kyr BP and 1.5–0.6 kyr BP coincide with the Roman Warm Period and Medieval Climate Anomaly, respectively. Finally, we find a drying tend in recent decades. Regional comparisons lead us to infer that the strength and position of the Westerlies, and wider Northern Hemispheric climate dynamics control climatic

shifts in arid Central Asia, leading to complex local responses. Our new archive from Lake Karakuli provides a detailed record of the local signatures of these climate transitions in the eastern Pamir.

Keywords: Pamir, Tibetan Plateau; Muztagh Ata, paleolimnology; biomarker; climate model; LMDZ4

1 Introduction

Future climate change associated with anthropogenic disturbance of the Earth system is expected to go in hand with changes in atmospheric circulation dynamics (Seth et al., 2011). In this scenario, certain regions of the Earth are thought to be susceptible to severe and likely abrupt changes in moisture delivery and temperature. One such region is Central Asia, located at the boundaries of influences from the mid-latitude Westerlies, the Siberian High, and the limits of the Indian Monsoon (Aizen et al., 2001; Chen et al., 2008). However, the nature and magnitude of changes in these climatic systems, as well as their Central Asian regional effects are still poorly known. Detailed knowledge about past, naturally-driven climatic variability in this region can contribute to a better understanding of the complex atmospheric circulation system, which can in turn help to better predict possible impacts of future anthropogenically-driven climate changes.

While a large number of studies have analysed climate dynamics in monsoonal eastern Asia and the north- and southeastern Tibetan Plateau (e.g. reviewed in Morill et al., 2003, An et al., 2006 and Herzschuh, 2006), the density of paleoclimate records in continental Central Asia remains comparably low. Central Asian records include studies of glacial extent in the Pamir (e.g. Narama, 2002a and b) and tree-ring width reconstructions (e.g. Esper et al., 2002; Treydte et al, 2006). Lacustrine sedimentary archives exist from Kyrgyzstan (Ricketts et al., 2001; Lauterbach et al., 2014, Mathis et al., 2014), the Aral Sea (Sorrell et al., 2007a and b; Boomer et al., 2009; Huang et al., 2011), the Western and Southern Tarim Basin (Zhao et al., 2012; Zhong et al., 2007), and the Pamirs/Tajikistan (Mischke et al. 2010c; Lei et al., 2014) (Fig 1b). Only one of those studies has included compound specific hydrogen isotopic analyses (Lauterbach et al., 2014), which have elsewhere in Asia shown potential to provide information about moisture sources, precipitation amount and temperature (Mügler et al.; 2010, Aichner et al., 2010c; Liu et al., 2008).

Climatic patterns in Central Asia are complex due to the location on the boundary between various large-scale atmospheric circulation systems, as well as the varied topography of the area (Fig 1). While the easternmost parts are generally arid and receive most of their precipitation during the summer, western regions receive higher proportional input from Westerly-derived winter precipitation (Miehe et al., 2001; Machalett et al., 2008; Lauterbach et al., 2014). Thus a dense network of paleoclimatic records is required to fully understand spatial patterns of climate dynamics over time.

To further decipher past climatic processes in our study we generated a high-resolution, mid to late Holocene paleoclimatic record from Lake Karakuli (western China), located in the eastern Pamir mountain range, at the very westernmost edge of the Tibetan Plateau. Building upon the work of Liu et al., (2014) who inferred glacial fluctuations from grain-size parameters and elemental composition at the same lake, we use compound-specific carbon ($\delta^{13}C$) and hydrogen (δD) isotopic compositions of long-chain ($>C_{24}$) *n*-alkanoic acids originating from plant leaf waxes to deduce past climatic changes in our study area. To evaluate the hydrogen isotopic data it is essential to understand what drives the variability of the isotopic signal which is recorded by the biomarker in a specific study area. Therefore we draw comparisons to isotope-enabled model experiments using the Laboratoire de Météorologie Dynamique Zoom model version 4 (LMDZ4) simulations (Hourdin et al., 2006; Risi et al., 2010; Risi et al., 2012a and b; Lee et al., 2012). On basis of this data we characterize the processes controlling isotopic composition of precipitation over Central Asia and discuss the implications for the interpretation of the biomarker isotopic evidence.

2 Study site

Lake Karakuli (also: Lake Kala Kule) is a small lake (ca. 1 x 1.5km) located at the westernmost edge of Xinjiang Province (PR China) at an altitude of 3650 m, between the massifs of Kongur Shan and Muztagh Ata, both exceeding 7500 m (Fig. 1a). Those mountains which form the eastern edge of the Pamir plateau and the very westernmost edge of the Tibetan Plateau are directly adjacent to the mountain ranges of Karakorum and Tien Shan. The climate in this high altitude region is cold and dry. At Taxkorgan climate station, 80 km south of Lake Karakuli (3090 m), average annual temperatures and precipitation amounts are 3.2°C and 69 mm, respectively (1957-1990; Miehe et al., 2001) with June and July being the wettest months. Climatic data from Bulun Kul (3310 m), 30 km north of our study area, are in a similar range (0.6 °C and 127 mm) with a precipitation maximum during spring and summer

(1956-1968; Miehe et al., 2001). At higher altitudes, precipitation amounts increase by orographic forcing. At the Muztagh Ata, annual rain- and snowfall was estimated to account for about 300 mm at the glacier accumulation zone (at 5919 m; Seong et al., 2009a) while other studies estimated a water equivalent depth of 605 mm for snow accumulation at 7010 m (Wu et al., 2008).

Lake Karakuli is an open freshwater lake with a maximum depth of 20 m. The relatively small catchment comprises meltwater mainly derived from glaciers on the western flank of Mt. Muztagh Ata. Those form an alluvial fan with several creeks which discharge into the lake from the south while the single outflow drains towards the north (see Fig. 1 and Fig. S1). Most of the glacial runoff derived from the surrounding massifs incl. the main glacier of Muztagh Ata and Mt Kongur Shan does currently not discharge into the lake.

The sparse vegetation consists of alpine grasslands, partly used for pasture (see Fig S1), with alpine desert at higher altitudes. Above 5500 m the landscape is fully glaciated (with valley glaciers descending to 4300 m; Tian et al., 2006). Compared to other shallow lakes on the Tibetan Plateau where macrophytes are numerous (Aichner et al., 2010b), there are only a few emergent and submerged macrophytes on or close to the shores, and few indications for submerged plants in the deeper parts of the lake.

3 Material and methods

3.1 Coring and chronology

A sediment core with a composite length of ca. 820 cm was taken in September 2008 at 38.43968 °N and 75.05725 °E from a water depth of 16 m, using an UWITEC coring system and a floating platform (the coring position is shown in supplement S7). The chronology was based-on seventeen radiocarbon ages derived from ^{14}C AMS dating conducted on total organic carbon (TOC) (Liu et al., 2014*).* The 0 cal. yr BP (1950 A.D.) was derived from ^{210}PB/^{137}Cs dating and appeared at ca. 10.5 cm depth. A reservoir-effect of 1880 years was extrapolated from dating of core-top samples and assumed to be constant throughout the core. The ^{14}C-ages indicate a nearly constant sedimentation rate across 4.3 kyr. For calibration of the ages and construction of the age-depth model the IntCal09 dataset was used (Reimer et al., 2009) applying a Bayesian method (Blaauw and Christen, 2011); for details see Liu et al. (2014).

3.2 Lab chemistry

Sediments were extracted with Accelerated Solvent Extraction system (ASE 350; Dionex), under high pressure (1500psi) and temperature (100°C) and using DCM/MeOH (9:1) as solvent. Alkanoic acids were separated from the total lipid extract using column chromatography (5 cm x 40 mm Pasteur pipette, NH_2 sepra bulk packing, 60 Å), eluting with 2:1 DCM/isopropanol, followed by 4% formic acid in diethylether, yielding neutral and acid fractions respectively. The acid fraction was esterified with 5% HCl and 95% methanol (of known isotopic composition) at 70°C for 12 h to yield corresponding fatty acid methyl esters (FAMEs). Lipids were obtained by liquid-liquid extraction using hexane as the non-polar solvent, and dried by passing through a column of anhydrous Na_2SO_4. They were further purified using column chromatography (5 cm x 40 mm Pasteur pipette, 5% water-deactivated silica gel, 100–200 mesh), eluting with hexane, followed by FAMEs eluted with DCM.

3.3 Biomarker isotopic analysis

Compound specific isotopic values were obtained using gas chromatography isotope ratio mass spectrometry (GC-IRMS). We used a Thermo Scientific® Trace gas chromatograph equipped with a Rxi-5ms column (30 m x 0.25 mm, film thickness 1µm) and a programmable temperature vaporizing (PTV) injector operated in solvent split mode with an evaporation temperature of 60°C. The GC was connected via a GC Isolink with pyrolysis/combustion furnace (at 1400/1000 °C) and a Conflo IV interface to a DeltaV$_{Plus}$ isotope ratio mass spectrometer. The H_3^+-factor (Sessions et al., 2001) was determined daily to test measurement-linearity of the system and accounted for 5.8 ppm mv^{-1} on average. Reference peaks of H_2/CO_2 bracket *n*-alkanoic acid peaks during the course of a GC-IRMS run; two of these peaks were used for standardization of the isotopic analysis, while the remainders were treated as unknowns to assess precision. Except for the case of co-elution, precision of these replicates was better than 0.6‰.

Data were normalized to the Vienna Standard Mean Ocean Water (VSMOW)-Standard Light Antarctic Precipitation (SLAP) hydrogen isotopic scale and to Vienna Pee Dee Belemnite (VPBD) carbon isotopic scale by comparing with an external standard containing 15 *n*-alkane compounds (C_{16} to C_{30}) of known isotopic composition (obtained from A. Schimmelmann, Indiana University, Bloomington). The RMS error of replicate measurements of the standard across the course of analyses was below 5‰ (hydrogen) and 0.7% (carbon) For hydrogen isotopes we further monitored for instrument drift by measuring the δD values of a C_{34} *n*-

alkane internal standard co-injected with the sample (-240.6±3.0‰; *n*=105). The isotopic composition of H and C added during methylation of alkanoic acids was estimated by methylating and analyzing phthalic acid as a dimethyl ester (isotopic standard from A. Schimmelmann, University of Indiana) yielding $\delta D_{methanol}$ = -198.3±3.9‰, $\delta^{13}C_{methanol}$ = -25.45±0.42‰ (n=7) . Correction for H and C added by methylation was then made by way of mass balance.

3.4 LMDZ4 simulations

To understand the control of spatial and seasonal isotopic variations, we use the climate model LMDZ4 (Hourdin et al., 2006) to characterize the processes controlling isotopes of precipitation over our study area. Details about the model and methodology are described in Risi et al., (2010; 2012a and b) and Lee et al., (2012). Briefly, the applied model version incorporates the entire cycle of stable water isotopes and includes fractionation when phase changes occur. The resolution of the model is 2.5° x 3.75° with 19 vertical levels in the atmosphere. To obtain more realistic simulations of the hydrology and isotope values compared with free-running simulations and to better reproduce the observed circulation pattern, simulated winds from LMDZ4 are relaxed toward the pseudo-observed horizontal wind field from the ERA-40 reanalysis results (Uppala et al., 2005) with a time constant of 1 hour. Boundary conditions used observed sea surface temperatures and sea ice fractions from the HadISST data set (Rayner et al., 2003) from 1958 to 2009.

4 Results

4.1 Lipid concentrations

Due to the sparse vegetation in and around the lake, concentrations of leaf wax biomarkers in the sediments were relatively low. For compound-specific isotopic analysis we chose fatty acids (FAs) which showed higher concentrations than alkanes in a set of test samples. Here, C_{24}, C_{26} and C_{28} *n*-alkanoic acids were the most abundant compounds, which average concentrations of ca 1050, 1000 and 750 ng/g dw (nanograms per grams dry weight; Fig. S2). We found fatty acid concentrations were relatively constant with depth, suggesting no major change in productivity, dilution or preservation during the late Holocene.

4.2 δD and δ^{13}C values of leaf wax lipids and water samples

In total, we measured 125 core samples for hydrogen isotopic composition and 66 samples for carbon isotopic composition (Tables S6). Samples contained C_{16}- C_{28} *n*-alkanoic acids with an even:odd chain length preference. We report isotopic results for the C_{24}, C_{26} and C_{28} *n*-alkanoic acids as these are target long chain compounds within the dynamic range of isotopic measurement capabilities (Tables S6, Fig. S3).

$\delta^{13}C$ values are generally more depleted with increasing chain-length, with C_{24} averaging to -27.9±1.4‰, C_{26} to -29.3±1.0‰, and C_{28} *n*-alkanoic acids to -31.0±0.9‰ (Figs. 2 and S3). For the C_{28} we find no significant downcore trend. C_{24} shows the largest variations in $\delta^{13}C$ values with generally more ^{13}C-depleted values in the middle of the core (min: -30.7‰) compared to the core-base and core-top (max: -24.3‰) (Fig. S3). For hydrogen isotopes, compounds are also more D-depleted with increasing chain-length (C_{24}: -173±6‰; C_{26}: -182±7‰; C_{28}: -185±6‰; Fig. 2). We observe downcore variations in δD values for C_{26} and C_{28} ranging from -196 to -167‰.

Six water samples (two from inflows, two from Lake Karakuli and two from ponds nearby) have been analysed for isotopic composition (Table 1). Both inflows show similar isotopic signatures (ca. -83‰). The lake water averages +3.5‰ ($\delta^{18}O$) and +15‰ (δD) enriched relative to the inflow due to evaporation. Closed ponds nearby are also evaporatively enriched relative to inflow.

5 Discussion

5.1 Origin of organic compounds and implications for source water

5.1.1 Molecular abundance distribution

Organic compounds in lake sediments originate from a mixture of terrestrial and aquatic organisms, with molecular abundance distributions and isotopic compositions that may be diagnostic of source. Most plants contain a broad of range of biomarkers (e.g. *n*-alkanes or fatty acids) but the fingerprints of the different compound classes are mainly dominated by compounds of a specific chain-length. Terrestrial and emergent aquatic plants for instance produce higher proportional abundances of long-chain *n*- alkanes (e.g. C_{29} and C_{31}) while submerged macrophytes contain higher amounts of mid-chain *n*-alkanes (e.g. C_{23} and C_{25}) (Ficken et al., 2000; Aichner et al., 2010b). *n*-Alkanoic acids show a less distinct pattern (Ficken et al., 2000), but also here long-chain compounds (e.g. C_{28}-FAs) are mostly interpreted to be originated from terrestrial sources (e.g. Kusch et al., 2010; Feakins et al., 2014).

In the sediments of Lake Karakuli the contribution of aquatic plants to the lipid pool is considered to be relatively low compared to other Tibetan high altitude lakes. A submerged aquatic plant sample collected close to the shore-line (ca. 20 cm water depth) shows a dominance of C_{16} and C_{18}-FAs and minor relative amounts of C_{20} to C_{30} even-chain FAs (see Fig. S4). This fatty acid-pattern is in agreement with published fingerprints of other aquatic plants collected on the Tibetan Plateau (Wang and Liu, 2012). Hence, the low relative abundance of C_{16} and C_{18}-FAs in our sediment samples suggests a relatively low contribution of plant material derived from aquatic macrophytes to the sedimentary organic matter in Lake Karakuli~~.~~

5.1.2 Carbon isotopic signal

Additional indication for the source of compounds comes from their carbon isotopic signature. Lipids of terrestrial C_3-plants usually show values around -30 to -35‰, while compounds derived from terrestrial C_4-plants and from submerged aquatic macrophytes can reach significantly more enriched values in the range -15 to -20‰ (Chikaraishi and Naraoka 2005; Aichner et al, 2010a). The difference between C_3 and C_4-plants can be explained by different isotopic fractionation in carbon assimilation of those two plant types, while the enriched values of submerged aquatic plants are due to the uptake of different carbon sources i.e. isotopically enriched bicarbonate instead of dissolved CO_2 (Allen and Spence, 1981; Prins and Elzenga, 1989).

In our sediment core from Lake Karakuli $\delta^{13}C$-values of the C_{28}-FA are similar to that of terrestrial C_3-plants without a clear trend (Fig. 2; Fig. S3). Thus, we conclude that this compound is predominantly derived from terrestrial C_3 grasses in the lake catchment. $\delta^{13}C$ values of C_{24} and C_{26}-*n*-alkanoic acids are slightly higher than for C_{28}, indicating an increasing contribution of submerged aquatic plant material and/or lipids derived from C_4-plants with decreasing chain lengths.

$\delta^{13}C$ values of C_{24} *n*-alkanoic acids are controlled by relative contributions of aquatic macrophytes and/or macrophyte productivity, with higher productivity leading to higher $\delta^{13}C$ values (Aichner et al., 2010b). We hypothesize that a higher proportional input of aquatic material to the sedimentary organic matter is indicative of warmer and possibly also drier conditions. Longer ice-free periods and a lower lake level could be the driving factors behind enhanced macrophyte growth during warmer years.

C_4-plants are widely absent on the central and eastern Tibetan Plateau at present, but they are wide-spread in Central Asian deserts and some *Chenopodiaceae* which use the C_4-pathway have occasionally been observed at high altitude alpine deserts of the Pamir (Sage et al., 2011). Thus we cannot totally exclude the contribution of C_4-derived lipids to the sedimentary organic matter of Lake Karakuli, however, we consider these sources as of secondary importance. Nevertheless, if we have underestimated the input of alkanoic acids derived from C_4-plants this would not bias the overall interpretation, because higher abundances of C_4-plants resulting in higher sedimentary $\delta^{13}C$ would indicate a drier/warmer climate, which is similar to the hypothesis that drying/warming leads to increased macrophyte productivity.

5.1.3 Hydrogen isotopic signal

Hydrogen isotopes provide further evidence for the origins of C_{24} and C_{26} or C_{28} *n*-alkanoic acids. The average δD values of C_{24} are ca. 9-12‰ higher than that of C_{26} and C_{28} (Fig. 2). A different water source i.e. isotopically enriched lake water (see Tab. 1) instead of water derived from precipitation or snow-melt could explain this. We assume that C_{24} is derived from mixed aquatic and terrestrial sources, while C_{28} and also C_{26} can be considered as of mainly terrestrial origin.

The δD-values of these terrestrial biomarkers is representative of the hydrogen isotopic composition of the source water which –for terrestrial plants– could be expected to be spring and summer precipitation during the growing season (Sachse et al., 2012), although a contribution of D-depleted melt-water from snow in the early spring growth period is highly likely (Fan et al., 2013). The fractionation factors between source water and lipids are variable but previous studies found that for terrestrial C_3-grasses they average to -149±28‰ (n=47) for the C_{29} *n*-alkane, while they are ca. -134±28‰ (n=53) for C_4-grasses and in similar range for forbs (Sachse et al., 2012). In arid ecosystems, soil-water evaporation (for grasses; Smith and Freeman, 2006) and transpiration from the leaf, lead to isotopic enrichment of leaf water above the meteoric water (Feakins and Sessions, 2010; Kahmen et al., 2013a and b). Recent results from the central Tibetan Plateau, a similar environmental setting to our study, quantified the apparent isotopic fraction between meteoric water and *n*-alkanes to be ca. -95‰ due to ca. +70‰ evapotranspirational isotopic enrichment above meteoric water (Günther et al., 2013). This is in agreement with the average fractionation from Feakins and Sessions (2010) who suggested ca. -95‰ as net fractionation factor between meteoric water and leaf wax *n*-alkanes in an arid ecosystem (southern California), and found similar values for *n*-alkanoic acids in a later study from that region (Feakins et al., 2014).

While the fractionation was not directly determined on modern plant *n*-alkanoic acids in this catchment, based on core-top δD_{lipid}-values of ca. -190‰ and knowledge of hydrogen isotope values of modern precipitation and waters in the catchment we can infer a reasonable catchment average apparent fractionation (Fig. 3). Summer precipitation in the catchment averages ca. -45‰ at Lake Karakuli, compared to mean annual precipitation average of ca. -90‰ (derived from the Online Isotopes in Precipitation Calculator, OIPC; Bowen and Revenaugh, 2003.; Fig. 4b). If the summer precipitation (-45‰; OIPC) is indicative of source water, and given the measured sedimentary value of C_{28} *n*-alkanoic acids (-190‰) we would compute an apparent fractionation of ca. -150‰ (see supplement S5 for formula to calculate isotopic fractionation factors). Whereas if we use mean annual precipitation (ca. -90‰; OIPC) then the calculated apparent fractionation would be ca. -110‰ which is closer to the reported fractionation factors for arid ecosystems (Feakins and Sessions, 2010; Günther et al., 2013).

The δD-values of the two lake inflows sampled in September 2008 (average -83‰; Table 1) provide a reasonable constraint on catchment average water isotopic composition in September, presumably including a mix of contributions from precipitation runoff, groundwater, and snow melt from winter precipitation and higher elevations. A calculated source water δD value based on published fractionation factors mentioned above (ca. -95‰) would be -110‰ (Fig. 3) which is in range of late-winter/early spring precipitation in the study area according to OIPC-data (Fig. 4b). These are helpful constraints on the proxy, however, regardless of knowing the exact season of source water and the appropriate fractionation which are needed for absolute isotopic conversions, we can infer relative variations in δD values of the C_{28} *n*-alkanoic acid down core in terms of variations in the δD of precipitation. We therefore use the δD values of the C_{28} and C_{26} *n*-alkanoic acids to reconstruct past variations in the isotopic composition of precipitation.

5.2 Controls on the isotopic signature of precipitation in the eastern Pamir

5.2.1 Monthly signal

The isotopic composition of precipitation is influenced by multiple isotope effects including those associated with precipitation amount, condensation temperature, or vapour source (Gat, 1996). In subtropical and tropical latitudes, the ‘amount effect’ has usually been identified as most relevant controlling factor with lower δD values reflecting more humid episodes in sedimentary records (Schefuss et al., 2005; Tierney et al., 2008, Lee et al., 2008). At mid- and high latitudes temperature and vapour source mostly have interpreted to be the dominant factors (Dansgaard, 1964; Thompson, 2000; Rach et al., 2014). In addition, large scale circulation changes or a shift in the balance of two or more different moisture sources and transport trajectories can result in isotopic shifts over time (Dansgaard, 1964; Thompson, 2000; Rach et al., 2014).

Evaluating isotopes of precipitation in context with climatic parameters in Asia, Araguas-Araguas et al. (1998) and Yao et al. (2013) came to the conclusion that the amount effect is the dominant factor in monsoonal east Asia while in arid Central Asia temperature mainly controls δD and $\delta^{18}O$ values of precipitation. The closest meteorological stations to Lake Karakuli are the station at Bulun Kul (ca. 30 km northeast) and Taxkorgan (ca. 80 km south). Both stations record low winter precipitation and slightly enhanced amounts during the summer (Fig., 4a and b). Higher isotopic values in the summer compared to the winter (Yao et al., 2013; Bowen and Revenaugh, 2003) suggest that monthly values are indeed driven by temperature. If these seasonal controls are also determining interannual variations in the isotopic composition of precipitation then temperature is likely to be a major factor explaining the reconstructed hydrogen isotopic variability.

We also observe amount effect modulation of the summer season precipitation isotopes associated with increased precipitation totals in June 2004 and more pronounced in June 2005 (Fig. 4a), which lowers the $\delta^{18}O$ values. This amount effect lowers the summer precipitation isotopic composition, dampens the seasonality of mean precipitation of isotopic values, and lowers the integrated annual precipitation isotopic composition. Hence in drier years average δD values will be D-enriched relative to wetter years; and likewise warmer years will be D-enriched relative to colder years (Fig. 4b). Given the low precipitation amounts in this arid region today, the amount effect is likely to remain secondary to the temperature controls on isotopic composition apparent in the seasonal cycle.

5.2.2 Annual/seasonal signal

To further establish the connections between climate anomalies and isotopic signatures of precipitation in Central Asia, we compare instrumental data and climate model simulations. At Taxkorgan meteorological station we find a negative correlation between annual temperature and precipitation amount over a period of 43 yrs (1957-2000; Fig. 5; data provided from Tian at al., 2006). Similar trends can be observed when comparing simulated data over a period of 50 yrs (1958-2009; Fig. 5). We use the LMDZ4 climate model (Hourdin et al., 2006) to characterize the climatic processes in our study area (as described in Lee et al. 2012). We find higher annual precipitation amounts in the LMDZ4 model simulations compared to instrumental observations at Taxkorgan meteorological stations. This is related to the scale of the model resolution of 3.75° x 2.5° (Lee et al., 2012) which includes the relatively high precipitation amounts in higher altitudes during winter (Seong et al., 2009a and b; Wu et al., 2008) within the grid box. Significant negative correlations ($r=0.58$; $p<0.0001$) between temperature and precipitation amount can be inferred for the summer months (April-September), while comparisons over the winter or whole year deliver non-significant correlations ($p>0.01$; Fig. 5).

As a consequence of the negative correlation between temperature and precipitation amount we observe positive/negative correlations between precipitation isotopes and those climatic parameters for our larger study area (Fig. 6). Considering temperature, we found a positive correlation ($0.4 < r < 0.6$) for both winter and summer over large parts of Central Asia indicating the broad regional significance of our record. For the summer, no correlations are seen in India and SE Asia, where distinct monsoonal circulation and precipitation patterns exert independent controls on the isotopic values of precipitation (Morill et al., 2003;Yao et al., 2013). Considering precipitation amount, negative correlations ($-0.6 < r < -0.2$) can be deduced for the summer months for a large region around Lake Karakuli, spanning from SW to NE and covering parts of Iran, Central Asia and NW China. During winter, no correlation can be observed directly at the location of the lake, however, precipitation isotopes seem to negatively correlate with precipitation amounts located westwards to our study area (Fig. 6).

In a recent study Tian et al. (2006) found a positive correlation between $\delta^{18}O$ in the local Muztagh Ata ice core (which covers the period 1957-2003) and annual temperatures from Taxkorgan climate station. In contrast they found no significant relationship between ice-core $\delta^{18}O$ and annual precipitation amount at Taxkorgan (Tian et al., 2006). Different precipitation dynamics between middle and high altitudes, and/or seasonal differences, as supported by our LMDZ4-data, could explain this discrepancy. Elevation differences may play a role in different precipitation patterns and these may be associated with isotope effects. The Muztagh Ata glacier accumulation zone receives higher annual precipitation amounts and also a higher proportional input from winter precipitation compared to lower altitudes (Seong et al., 2009a and b). Whilst instrumental and modelling data inferred a slight increase of precipitation amount throughout the last 50 years in the westernmost part of China (Yao et al., 2012; Zhang and Cong, 2014), a decreasing accumulation rate at the Muztagh Ata ice core since 1976 was measured by Duan et al. (2007). Even if the instrumental data from Taxkorgan do not show a significant trend in precipitation amount between 1957 and 2000, this does not rule out changes of snowfall at higher altitudes. Increasing temperatures could have further contributed to the lower observed accumulation rates.

Since temperature and precipitation amounts are anti-correlated on an interannual timescale (Fig. 5), we interpret low δD values to indicate both relatively cool and wet conditions. In addition to fluctuations in mean annual precipitation isotopes, snow-melt and delivery to plants may vary. We suggest that a high proportional contribution of water derived from snow-melt, after relatively long and wet winters with high amounts of snowfall, can further lead to more negative δD leaf wax values.

5.3 Paleoclimatic interpretation of downcore data

δD and $\delta^{13}C$ values from Lake Karakuli sediment core suggest relatively warm and dry conditions between ca. 4-3.5 kyrs BP (Fig. 7). $\delta^{13}C$ values are highest for C_{24} during this interval and even C_{28} shows slightly enriched values (>-30‰; Fig. S3). Also δD shows maximum values during this episode. Even though an increased input from C_4-plants or enhanced productivity of aquatic macrophytes could slightly have biased δD-values towards a more positive signal, we infer that this period probably was the warmest/driest in our studied time-interval. After 3.5 kyrs a gradual cooling trend started (interrupted by a warmer/drier period between ca. 3.0 and 2.7 kyrs BP), peaking in coolest and wettest conditions around 2.5 kyrs BP. Between ca. 2.5 and 1.9 kyrs BP we observe a reversal to a slightly warmer and drier climate, based on δD evidence. We note that the $\delta^{13}C$ values are rather variable and inconclusive in this core-section, and we observe an offset between δD-C_{26} and δD-C_{28} (these are normally within analytical error of each other). We hypothesize that a warming influenced precipitation isotopes but that the change wasn´t intense and stable enough to trigger a large-scale ecosystem response to be recorded in the $\delta^{13}C$ values. Between ca. 1.9 and 1.4 kyrs BP, cool and wet conditions occurred again before returning to a warm and dry episode from ca. 1.4 to 0.6 kyrs BP (possibly interrupted by a cooling event around 1 kyrs BP). The last 0.6 kyrs have been mainly cool and wet again, except for the last ca. 100 years, where the topmost three samples of the sediment core indicate another reversal to relatively warm and dry conditions.

Enhanced precipitation, rather than lower temperatures, has been argued to be the main driving force behind growth of glaciers in Asian high-altitude regions (Seong et al., 2009b). The cool/wet episodes deduced from our organic geochemical record match relatively well to reconstructed glacial advances at Mts. Muztagh Ata and Kongur Shan. On basis of ^{10}Be-

dating of erratic boulders Seong et al. (2009a) estimated maximal glacial advances at 4.2±0.3 kyrs, 3.3±0.6 kyrs, 1.4±0.1 kyrs, and a few hundred years before present (Fig. 7). Further, the δD-data are in good agreement with silt-contents in the same sediment core (Fig. 7). These have been interpreted to be influenced by glacial input and thus higher contents indicating cooler/wetter conditions (Liu et al., 2014).

Our interpretation of lower δD-values indicating both relatively cool and wet conditions fits well with results from other late Holocene records in arid Central Asia (Fig. 8c, e and g). The Little Ice Age (LIA) corresponds to the cool/humid period between 0.6 and 0.1 cal. ka BP at Lake Karakuli and has been well documented as a widely humid episode in arid Central Asia (paleoclimatic data compiled in Chen et al., 2010; Fig. 8c). For instance, the Guliya ice core, located ca. 630 km SE from Lake Karakuli, shows relatively high accumulation rates during that period (Fig. 8e), indicating that higher precipitation amounts and not just higher effective moisture (induced by decreased evaporation during cooler conditions) was the main driving force behind e.g. higher lake levels. This very much contrasts the situation in eastern/monsoonal Asia where many records show a relatively dry LIA due to a weakened summer monsoon (Chen et al., 2010 and references therein).

Similarly a number of records have shown a pronounced warm/dry period during the Medieval Climate Anomaly (MCA; Fig 8 c,e; Chen et al., 2010; Lauterbach et al., 2014; Esper et al., 2002) also seen in our record from Late Karakuli. At ca. 1 cal. ka BP we observe a ca. 100-year interruption of this event indicated by three samples with lower δD-values. Recently, Lei et al. (2014) observed a similar spike in carbonate $\delta^{18}O$ values from Lake Sasi Kul, which is located ca. 190 km west of our study site (Fig. 8b). Thus we hypothesize that this interruption was not just a local phenomenon. Warm and dry conditions during the MCA have also been observed at Kashgar (western Tarim Basin; just ca. 150 km north of Lake Karakuli; Zhao et al., 2012), and from from Lakes Bangong Co on the western Tibetan Plateau (Gasse et al., 1996) and large Karakul in the Tajik Pamir (Mischke et al., 2010).

Applying these findings to the complete record we see fluctuating climatic conditions throughout the late Holocene with clearly identifiable warmer/drier and cooler/wetter episodes (Fig. 8). During the oldest section of our record (ca. 4.2-3.4 kyrs BP) average conditions appeared having been warmer and drier than during the medieval and today, followed by a general (even though non-continuous) cooling trend until ca. 2.4 kyrs BP. A cool and wet phase of roughly 1000 years starting at ca. 3.5 kyrs BP has been observed in numerous global climate records (Mayewski et al., 2004). At the nearby oasis of Kashgar, conditions prevailed relatively wet from ca. 4.0 until ca. 2.6 kyrs BP (Zhao et al., 2012). At the large Lake Karakul in Tajikistan a rapid drop of TOC-contents occurred at ca. 3.5 cal. ka BP, indicating a drop of lake productivity probably induced by low-temperatures and eventually associated with shorter ice-free periods in the summer (Mischke et al., 2010; Fig. 8h). At Lake Balikun (northeastern Xinjiang) a reversal to wetter conditions occurred after a pronounced dry event lasting from 4.3-3.8 kyrs BP (An et al., 2012). In Lake Manas (northern Xinjiang) a wet episode was reconstructed for 4.5-2.5 kyrs BP, interrupted by a short dry period between 3.8-3.5 kyrs BP (Rhodes et al., 1996). Low $\delta^{18}O$-values in the Guliya ice core between 3.5 and 3.0 kyrs BP also give evidence for low temperatures on the northwestern Tibetan Plateau (Thompson et al., 1997) while in the southern Tarim Basin a rapid shift to wetter conditions at ca. 3.0 kyrs BP have been observed (Zhong et al., 2007).

After a ca. 500 year slight warming (ca. 2.4-1.9 kyrs BP; synchronous with the Roman Warm Period; RWP), another reversal into cool and wet condition occurred, peaking at ca. 1.8-1.6 kyrs BP (often referred to as Dark Ages Cool Period, DACP, or Migration Period). Both of these events have also been observed in the nearby Kashgar (Zhao et al., 2012). Afterwards that the climatic trend gradually transitioned into the above mentioned warm period during the medieval, followed by the LIA and the current warming period (CWP), the latter indicated by increased δD values in the topmost three samples of the sediment core.

5.4 Implications for Central Asian climate dynamics

The sequence of relatively cool/wet and warm/dry episodes displays coherency with other records of Northern Hemisphere climate records. There is a similarity between cyclicity of cooling events at Lake Karakuli, Northern Atlantic ice-rafting events (Fig 8j; Bond et al., 2001) and strengthening phases of the Siberian High (the anticyclonic high pressure ridge over Siberia), the latter recorded by [K^+] increases in the GISP2 ice core between ca. 3.5-2.8 and 0.5-0.2 kyrs BP (Fig 8i; Mayewski et al., 1997). Further, throughout the last ca. 1000 years, δD values of leaf waxes in Lake Karakuli are correlated with the mode of the North Atlantic Oscillation (NAO), showing more positive values during the current and medieval positive mode and more negative values during the LIA-negative mode (Fig. 8f; Trouet et al., 2009).

The interplay between the dominant atmospheric circulation systems in Central Asia – the Siberian High, the mid-latitude Westerlies and partly the Indian Summer Monsoon– as well as orographic influences, lead to complex climatic patterns. Trajectory studies in the modern atmosphere, as well as inventories of dust particles in ice cores, suggest the mid-latitude Westerlies as primary source of moisture during winter and spring, with the North Atlantic, the Mediterranean, the Black and Caspian Sea as possible regions of origin (Lei et al., 2014; Seong et al., 2009a and b; Wu et al., 2008). The Siberian High delivers cool but also relatively dry air during winter. The absence of sea-salt i.e. in the Muztagh Ata ice core (Aizen et al., 2001; Seong et al., 2009b) further gives evidence for a minor importance of the Indian Monsoon, and for mid-latitude Westerlies and local convection to be the most important moisture sources during the summer.

Even though Lake Karakuli receives some moisture in spring (Fig. 4), regions which are located as close as 190 km westwards at a similar altitude, such as Lake Sasi Kul and other parts of the central and western Pamirs receive much higher proportions and amounts of winter and spring precipitation (Lei et al., 2014; Miehe et al., 2001). Variations of strength and tracks of the Westerlies and related movement of the Polar Front (Machalett et al., 2008) could have influenced the amount of winter and spring moisture which has reached the Karakuli-region in the past. Lei et al. (2014) suggested that during negative NAO-modes (e.g. during the LIA) the storm tracks were moving further southwards, leading to wetter conditions in the Mediterranean and higher amounts of moisture been transported into Central Asian realms of the same latitude. In contrast other authors proposed a more complex interplay between the Eurasian and Pacific circulation systems on basis of modelling data, and a generally higher delivery of moisture into Central Asia during episodes of strengthened Westerlies (i.e. positive NAO-modes) (Syed et al., 2010; Syed, 2011). Recently, a possible negative correlation between lower winter precipitation in the Mediterranean (positive NAO-mode) and higher winter precipitation at Son Kol (central Tien Shan; ca. 400 km north of Lake Karakuli) was also suggested by Lauterbach et al. (2014) on basis of δ^{15}N-data on total nitrogen (Fig. 8d).

Based on our data, we hypothesize that the relatively wet episodes recorded in our sediment core from Lake Karakuli were mainly caused by increased late-winter and spring precipitation derived from mid-latitude Westerlies. Cooling/wettening periods at 3.5 cal. ka BP and between 1.9 and 1.5 kyrs BP (DACP) are simultaneous with increased winter precipitation at Son Kol (Fig. 8d), indicating common climatic variations in the eastern Pamirs and the central Tien Shan. For the LIA, this connection is less pronounced. Instead, for the last ca. 1.5 kyrs BP, we see a close similarity to isotopic trends in the central Pamirs (Fig. 8b), which in turn drift apart between 1.5 and 2.5 kyrs BP. An explanation for this could be the increased influence of the significantly strengthened Siberian High during the LIA (Fig. 8i). This possibly weakened the mid-latitude Westerlies or pushed their tracks further to the south, resulting in comparably drier conditions at more northern regions such as the Tien Shan, but wetter conditions in the central and eastern Pamirs (Lei et al., 2014). A similar mechanism could explain the climatic pattern in the eastern Pamirs at present, with low winter and spring precipitation at low altitudes during the current positive NAO-mode and Westerlies penetrating more to the North, while the central Pamirs still receive high winter precipitation.

Despite a slight increase in total precipitation amount over the last 50 years in the dry areas of Western China (Yao et al., 2012; Zhang and Cong, 2014), effective moisture in our study area has decreased due to rising temperatures. The two closed ponds and Lake Karakuli itself show clear geomorphological evidence for recent shrinking (field observations) and isotopic evidence for evaporative enrichment above meteoric waters (Table 1). This is in contrast to several endorheic lakes in Central Asia, whose lake levels are rising due to the currently increased meltwater input from receding glaciers (e.g. Bosten Lake; Wünnemann et al., 2006; or large Lake Karakul in Tajikistan, Mischke et al., 2010).

6 Conclusion

The biomarker isotopic record from Lake Karakuli, eastern Pamirs, shows distinct episodes of relatively cool/wet and warm/dry climate over the last 4200 years. Variations in the North Atlantic conditions and Siberian High both appear to show similarities with variations captured in our biomarker isotopic record, including notable excursions associated around 3.5 kyrs, the MCA, and the LIA. However, there are also indications for complex responses of regional climate, i.e. different responses between the western (e.g. western and central Pamir), eastern (e.g. eastern Pamir) and northern (e.g. Tien Shan) parts of Central Asia. These regional differences are thought to arise from changes in the dynamics and interplay of the involved large scale atmospheric circulation systems, especially the strengths and pathways of the Westerlies. Our data provide evidence that the transition between regions of summer-only and winter/spring dominated precipitation could have been a key factor for local climate in the past. They further show a rapid aridification in the eastern Pamir during the last 50-100 years.

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Table 1: $\delta^{18}O$ and δD values of water samples collected in September 2008 at Lake Karakuli, its inflows and nearby ponds.

Tables

Table 1: $\delta^{18}O$ and δD values of water samples collected in September 2008 at Lake Karakuli, its inflows and nearby ponds.

Latitude [°N]	Longitude [°E]	Altitude [m]	Description	$\delta^{18}O$ [‰]	1 σ	δD [‰]	1 σ
38.41933	75.05995	3684	inflow1	-12.1	0.05	-83.2	0.2
38.42021	75.05008	3688	inflow2	-12.1	0.01	-84.4	0.2
38.43968	75.05725	3657	Karakuli - core position surface water	-9.4	0.04	-67.8	0.2
38.43968	75.05725	3657	Karakuli - core position above sediment	-9.2	0.03	-67.2	0.2
38.46294	75.02928	3658	pond near Karakuli	5.45	0.02	13.5	0.4
38.46334	75.04267	3676	pond near Karakuli	-3.6	0.02	-37.1	0.5

Figures

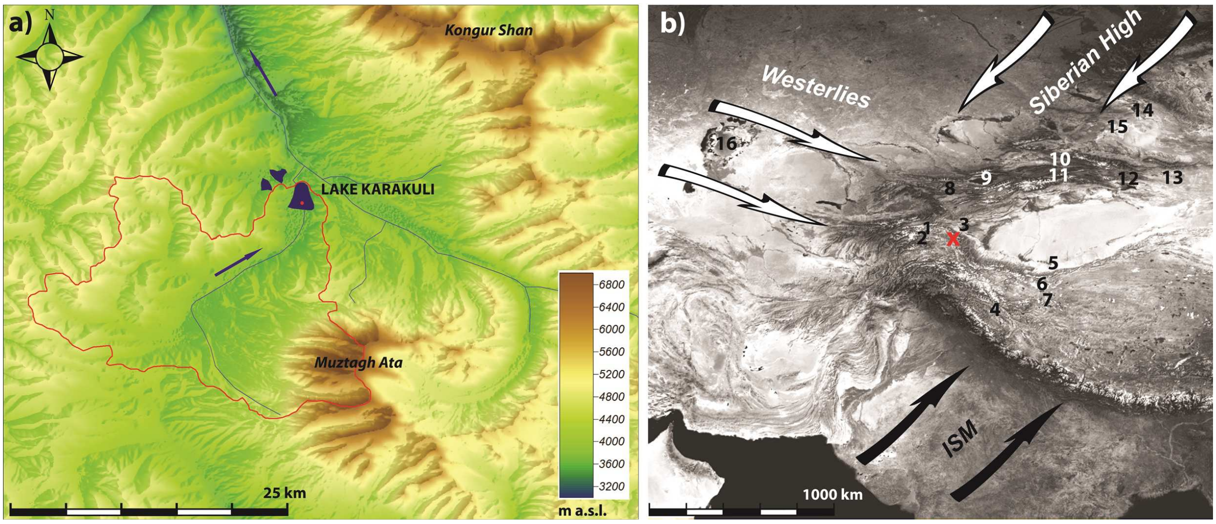


Fig. 1: (a) Catchment of Lake Karakuli and coring position (red dot). (b) Location of our study area (red cross) and other paleoclimatic records mentioned in the text. 1: large Lake Karakul, Tajikistan (Mischke et al., 2010); 2: Lake Sasi Kul (Lei et al., 2014); 3: Kashgar (Zhao et al., 2012); 4: Tso Kar (Wünnemann et al., 2010); 5: Southern Tarim Basin (Zhong et al., 2007); 6: Guliya Ice Core (e.g. Thompson et al., 1997); 7: Lake Bangong (Gasse et al., 1996); 8: Son Kol (Lauterbach et al., 2014, Mathis et al., 2014), 9: Issyk Kul (Ricketts et al., 2001); 10: Yili section (Li et al., 2011); 11: Kesang Cave (Cheng et al., 2012); 12: Boston Hu (Wünnemann et al., 2006); 13: Lake Balinkun (An et al., 2012); 14: Ulungur Hu (Liu et al., 2008); 15: Lake Manas (Rhodes et al., 1996); 16: Aral Sea (Sorrell et al., 2007a and b; Boomer et al., 2009; Huang et al., 2011).

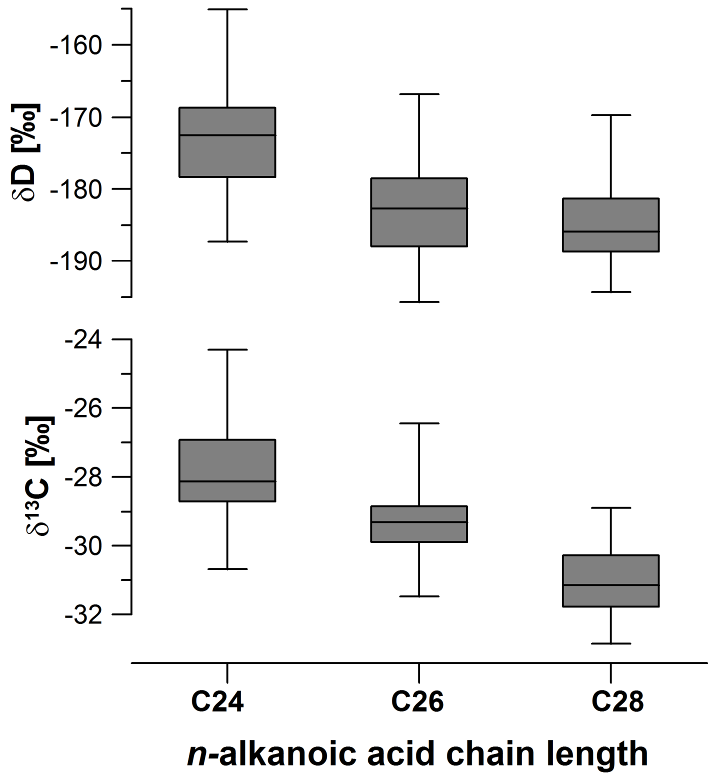


Fig. 2: Box and whisker plots of δD and $\delta^{13}C$ values in sediment samples by chain length.

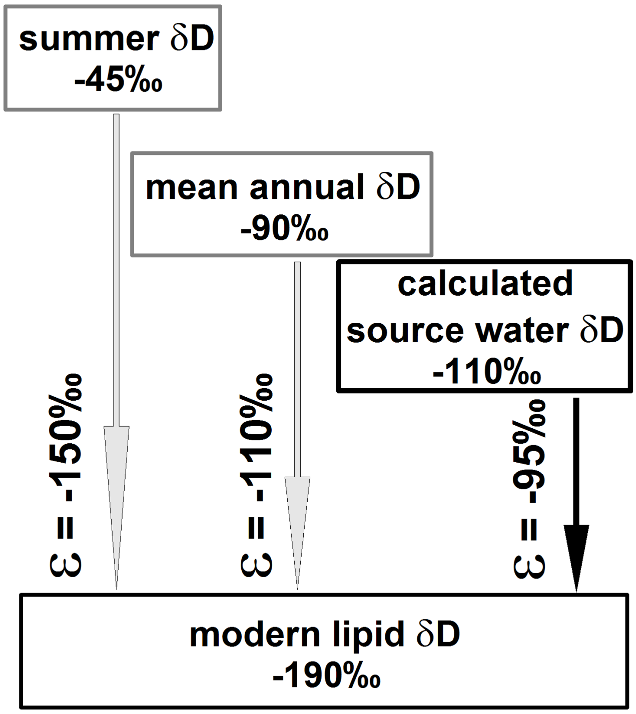


Fig. 3: Calculated isotopic fractionation factors (ε) between summer and mean annual precipitation and modern lipids, as well as calculated source water δD on basis of published fractionation factors in arid ecosystems (ca. -95‰ according to Feakins and Sessions, 2010; Günther et al., 2013).

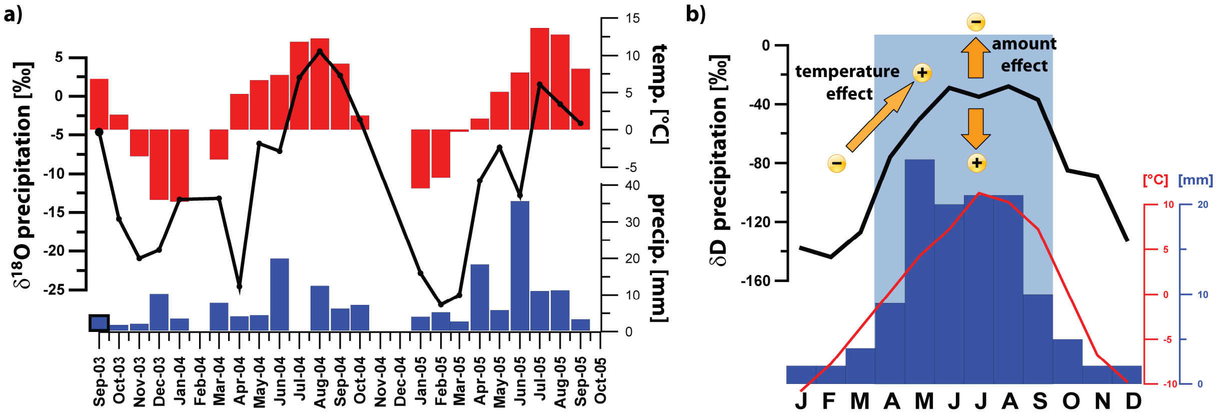


Fig.4: (a) Monthly isotopic and climate data from Taxkorgan climate station (Yao et al., 2013), located ca. 80 km south of Lake Karakuli (altitude ca. 3100 m). (b) Average monthly climate (Miehe et al., 2001) and isotopic (OIPC; Bowen and Revenaugh, 2003) data from Bulun Kul climate station located ca. 30 km northeast of Lake Karakuli (altitude ca. 3300 m). Shaded area indicates summer/wet season.

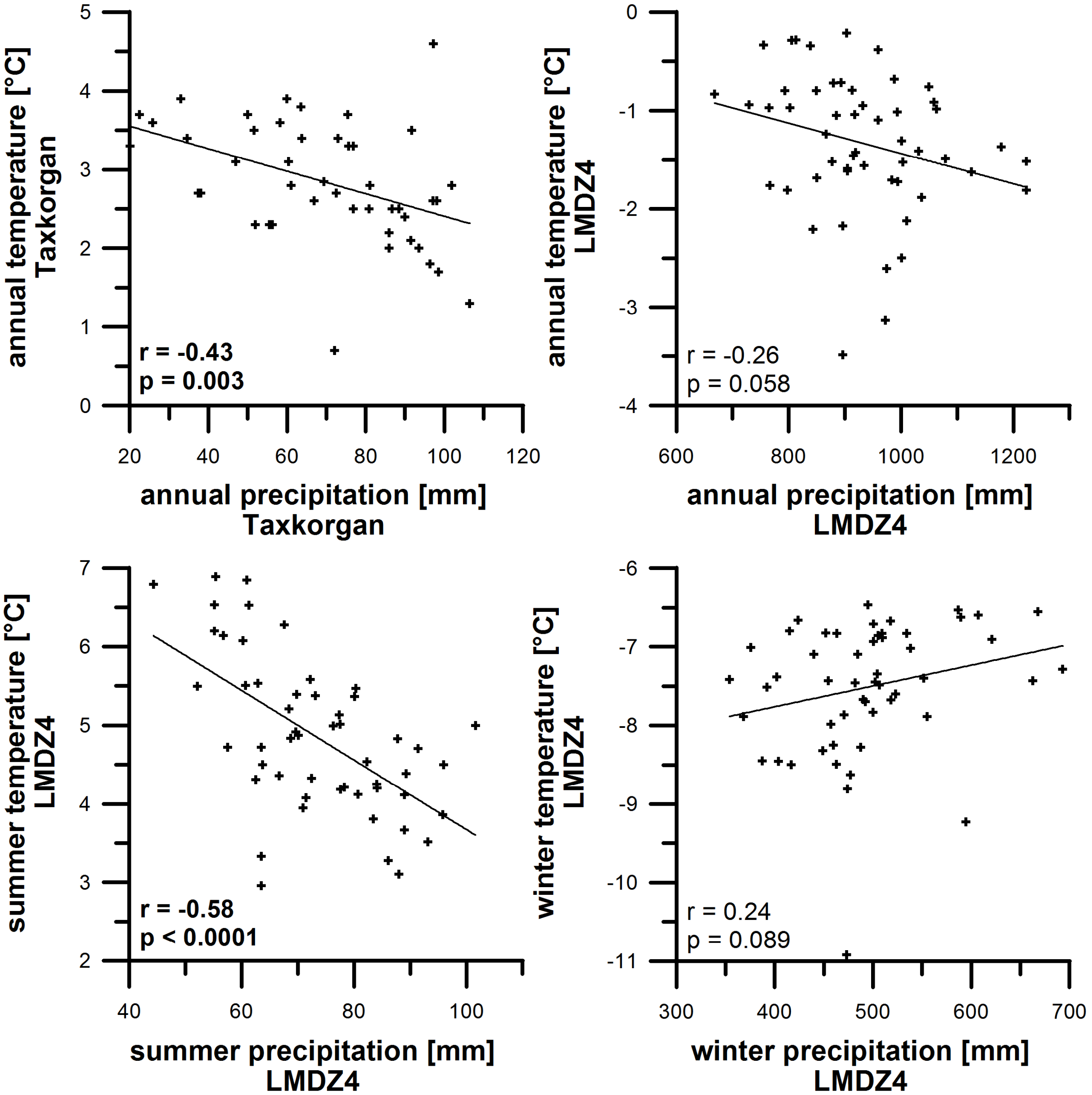


Fig. 5: Correlations of temperatures with precipitation amounts based on instrumental data from Taxkorgan meteorological station (1957-2000; annual averages) and model data using LMDZ4 simulations (1958-2009; summer: April-September; winter: October-March). Bold correlation coefficients are significant at the 0.01-level.

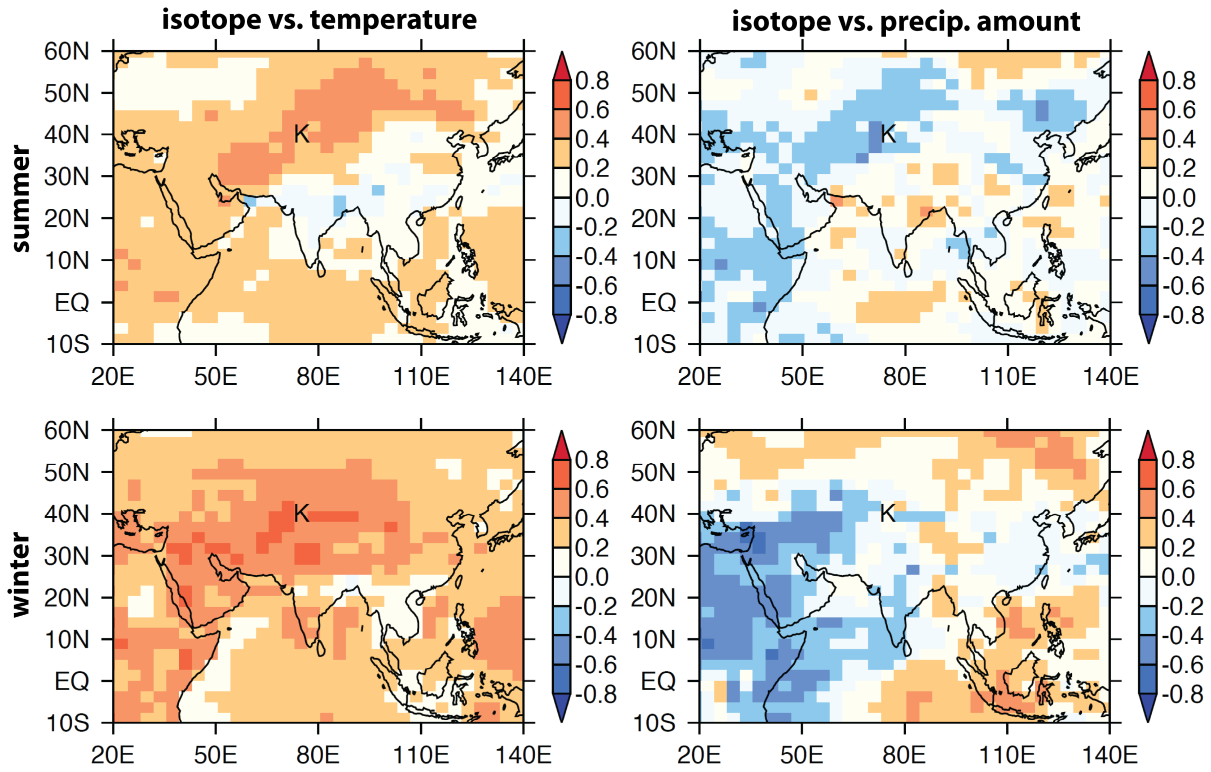


Fig. 6: Spatial correlation coefficient (*r*) summer (April-September) and winter (October-March) $\delta^{18}O$ of precipitation at the Karakuli site (marked as K in the plots) with temperatures and precipitation amounts at each grid point from 1958 to 2009 using LMDZ4 simulations (Lee et al., 2012).

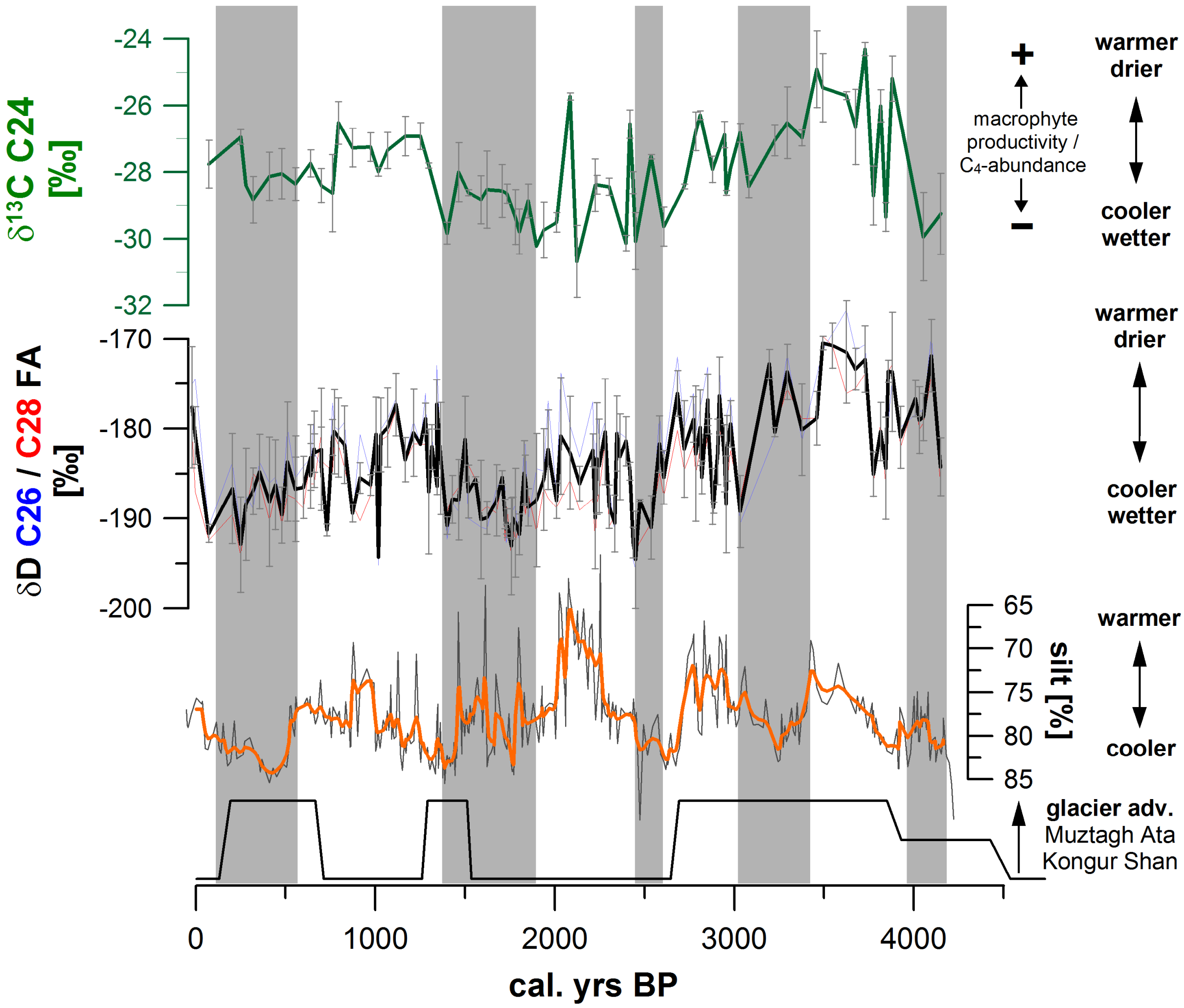
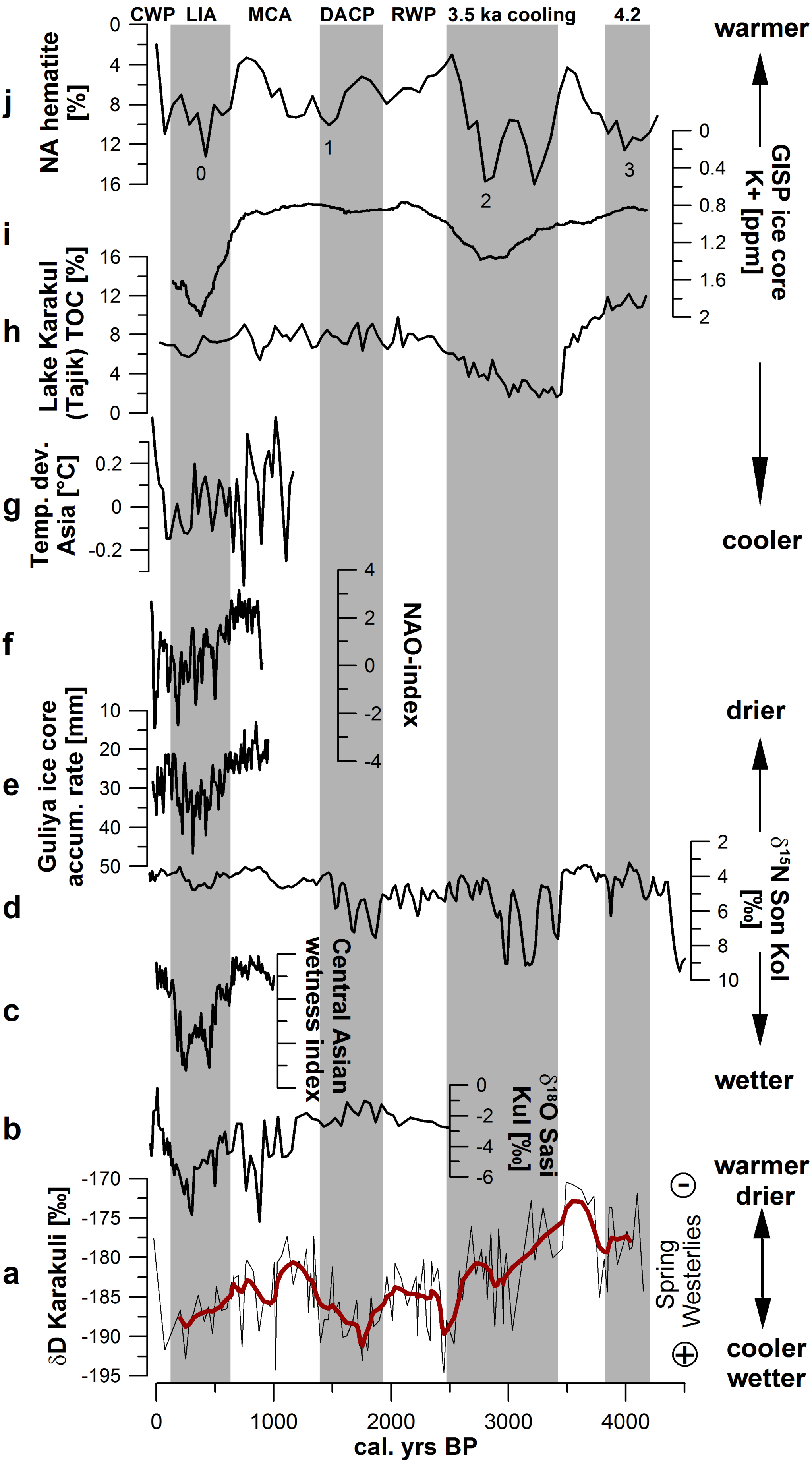


Fig. 7: Summary of organic geochemical results from this study in context with silt contents of the same sediment core (Liu et al., 2014;orange line: 5-point weighted average) and data of local glacier advances on basis of ^{10}Be-dating (Seong et al., 2009a; centers and widths of boxes mark the mean age and the error ranges of the events). Biomarker hydrogen isotopic data are presented as mean of triplicate measurements for the C_{26} (blue line) and C_{28} *n*-alkanoic acids (red line) as well as unweighted average of the two (thick black line, with 1σ error bars). Shaded areas are relatively cool and wet episodes, based on leaf wax isotopic data.



CWP
LIA
MCA
DACP
RWP
3.5 ka cooling
4.2
warmer
cooler
drier
wetter
warmer
drier
cooler
wetter
j
NA hematite [%]
i
GISP ice core K+ [ppm]
h
Lake Karakul (Tajik) TOC [%]
g
Temp. dev. Asia [°C]
f
NAO-index
e
Guliya ice core accum. rate [mm]
d
δ15N Son Kol [‰]
c
Central Asian wetness index
b
δ18O Sasi Kul [‰]
a
δD Karakuli [‰]
Spring Westerlies
cal. yrs BP

Fig. 8: Comparison to local and Northern Hemispheric paleorecords. Shaded areas indicate relatively cool/wet episodes at Lake Karakuli; (a) δD of C_{26} and C_{28} *n*-alkanoic acids Lake Karakuli (this study); average values as in Fig. 8, red line: 5-point weighted average. (b) $\delta^{18}O$ Sasi Kul, Pamir, Tajikistan (Lei et al., 2014). (c) Central Asian wetness index (Chen et al., 2010). (d) $\delta^{15}N$ TN, Son Kol, Central Tien Shan, Kyrgyzstan (Lauterbach et al., 2014). (e) Guliya ice core accumulation rate (Thompson et al., 1997). (f) North Atlantic Oscillation index (Trouet et al., 2009). (g) 30-year average of compiled temperature deviations in Asia (Pages 2k Network, 2013). (h) TOC-contents large Lake Karakul, Pamir, Tajikistan (Mischke et al., 2010). (i) K^+ GISP2 ice core (Mayewski et al., 1997). (j) Northern Atlantic Hematite grains indicate Northern Hemispheric cooling events "Bond-events" (Bond et al., 2001).