

1 **Response to the reviewers comments on manuscript cpd-10-4385-2014**

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3
4 We thank the two anonymous reviewers for their time reviewing this manuscript, for their
5 valuable comments and for raising interesting points of discussion. Our responses to the
6 specific comments are attached below, together with a list of other minor changes.
7

8 9 10 **1. Response to Rev#1:**

11
12 *Rev#1.: Overall, the study is well designed and executed. It would have been valuable to have*
13 *dDwax data from modern vegetation in the watershed, in order to constrain local*
14 *fractionation factors and reduce uncertainty in absolute values of reconstructed dDppt,*
15 *although this lack does little to diminish the significance of the study. One minor concern is*
16 *that the lower half of Figure 4b lacks scale bars and labels for the monthly mean climate*
17 *variables.*

18
19 Response: we agree that it would be valuable to have data from the vegetation in the
20 catchment, however the field work was limited to lake coring in this case. Labels and scale
21 bars were not included in the original version of Fig. 4b due to layout-reasons, but have now
22 been added.
23

24 25 26 **2. Response to Rev#2:**

27
28 Key-comment (1): temperature vs. precipitation:
29

30 *Rev#2: As soon as the abstract, it is stated: "Instrumental evidence and isotope-enabled*
31 *climate model experiments with the Laboratoire de Météorologie Dynamique Zoom model*
32 *version 4 (LMDZ4) demonstrate that dD values of precipitation in the region are influenced*
33 *by both temperature and precipitation amount. We find that those parameters are inversely*
34 *correlated on an annual scale; i.e. climate varies between cool/wet and dry/warm over the*
35 *last 50 years."*

36 *The instrumental evidence the authors refer to is a meteorological station (described in*
37 *chapter 5.2.1. and data shown in figure 4a). The isotopic values of rainfall shows a very clear*
38 *seasonality, with only 2 data points out of more than 20 significantly deviating from a nearly*
39 *perfect sine curve. Such a pattern clearly demonstrate that TEMPERATURE is by far the*
40 *first-order controlling knob of isotopes. Instead of interpreting the isotopic signal in terms of*
41 *temperature, the authors acrobatically state "However there are also small amount effect*
42 *observable", and they do list the couple of months that make the 2-years long record of*
43 *isotopic composition of rainfall deviating from the sine curve... Later in the article, they*
44 *tentatively interpret the isotopic ups and downs as going towards "warmer/drier" vs.*
45 *"cooler/wetter" conditions, which I find awkward given the apparently little effect of*
46 *seasonality of rainfall amount shown in Figure 4. The LMDZ, to me, can't be of great help to*
47 *justify the interpretation of the isotopic ratios as long as the model simulates an order of*
48 *magnitude more annual rainfall than the meteorological station (shown in Figure 5, the*
49 *feature being noticed by the authors themselves).*
50

1 *So why pushing precipitation in such case? It clearly obscures the discussion. This would*
2 *help, in my opinion, to clearly mention that rainfall may play a minor role in the isotopic*
3 *signal, but is very likely of second order. The discussion (and maybe interpretation) may then*
4 *be much more straightforward and convincing.*

5
6
7 Response: the question what -besides vapor source- actually influences the isotopic signal of
8 the precipitation is very important and answering this is not straightforward, hence we are
9 glad that the reviewer picks up this issue for discussion.

10
11 First of all the sinusoidal curve of isotopic data from the 2 years interval in Taxkorgan (Fig
12 4a) does imply a temperature control on monthly isotopic values. The question is if this also
13 implies a temperature control on annual average precipitation. Let's assume -for instance- two
14 years with equally warm summers, but the first being drier and the second wetter. From first
15 principals, and evidence for the amount effect in these latitudes, we anticipate that would be
16 accompanied by less negative δD -values in the first year and more negative δD values in the
17 second. We think there are indications for this effect from the short timeseries in Fig. 4a. As
18 described in the manuscript, June has the highest precipitation amount and an amount effect
19 on the order of -5 to -7.5‰ in $\delta^{18}O$ values, which lowers the annual mean precipitation
20 isotopic values. The amount effect is also supported by the LMDZ-derived correlations
21 between isotopes and climatic parameters, which in the summers show a significant negative
22 correlation with both temperature and precipitation (Fig. 6).

23
24 This, in our opinion, gives strong enough evidence that precipitation amount influences the
25 overall isotopic values and needs to be taken into account in the integrated signal as recorded
26 in sediments. However, we agree with the reviewer that the amount-effect is probably
27 secondary compared to the temperature control, especially if considering the very low
28 absolute changes of precipitation amount (as seen on the data from Taxkorgan in Fig. 5).

29
30 \Rightarrow We rephrased the last paragraph of section 5.2.1 and included this assumption here.

31
32 A second aspect to consider is the co-correlation of climatic parameters. Fig. 5 shows this for
33 the last 50 yrs on two spatial scales for temperature vs. precipitation: (1) locally i.e. point-data
34 from Taxkorgan climate station (3090m) which –even if located at ca. 500m lower and 80km
35 to the south- could be representative for the situation at Lake Karakuli. (2) regionally i.e.
36 LMDZ4-grid cells, which include the complete altitude range, hence higher altitudes with
37 higher precipitation. Both scales, i.e. (1) and (2) show a negative correlation between
38 temperature and precipitation (except for winters if taking the whole grid-cell/LMDZ-model).
39 Extending the timescale from 50 yrs to centennial-intervals than (3) proxy-data for the last
40 few hundred years also generally agree that the LIA was a cool/wet episode and that the MCA
41 was rather dry in large parts of arid Central Asia (Fig. 8c,e).

42
43 If temperature and precipitation are anti-correlated we can overcome the problem of
44 deciphering the dominant climatic parameter on δD by not distinguishing between what
45 *influences* the isotopic signal but what is *correlated* with the signal. Thus we are confident
46 that our assumptions are true that low δD -values indicate cooler/wetter conditions and that
47 higher values show warmer/drier conditions and that this holds for the whole time interval
48 covered by the record.

1 Key-comment (2): connection to large-scale climate dynamics

2
3 *Rev#2: Another thing I'd like the authors to avoid is to blindly connect their isotopic records*
4 *(along with the already published grain size fraction curve) with other reference curves, and*
5 *interpret their record within a broader context on the basis of similarities between all curves.*
6 *While the fact that there is no one-to-one coupling between their own records with other*
7 *curves taken from the North Atlantic, Greenland, etc., the authors still courageously drop a*
8 *line stating "Our interpretation of lower dD values indicating both relatively cool and wet*
9 *conditions fits well with results from other late Holocene records in arid Central Asia (Fig.*
10 *8)." Figure 8 indeed displays panels where many – if not all - curves just don't look like each*
11 *other (in terms of trends, rapid variability, etc.) They briefly deal with some discrepancies*
12 *invoking large-scale atmospheric patterns, but the reader's feeling of the tone employed by*
13 *the authors is that all those curves kind of tell the same story. While having a look at their*
14 *comparisons (figure 8) and reading the above-quoted sentence, I guess climate dynamicists*
15 *and high-res paleo-stat folks will just ignore your observations.*

16
17 *I understand it is important to connect one new climate record to other reference records to*
18 *better understand large-scale climate patterns. But central Asia is far enough from some*
19 *climate records shown in figure 8 to have a more descriptive discussion on what's happening*
20 *at the site prior to try connecting the dD record to other reference curves situated very far*
21 *away from central asia (and that don't really fit with your ones). And to be honest, even the*
22 *%silt and dD curve shown in Figure 7 are not really well fitting with each other (sometimes*
23 *they look negatively correlated!), unlike what is suggested in the text. The authors shouldn't*
24 *be shy and make everything to build their own "new reference curve" for the late Holocene*
25 *climate in central Asia.*

26
27
28 Response: we did not generally intend to suggest an one-to-one coupling between for instance
29 the Northern Atlantic or other parts of Central Asia and our study area. Instead we tried to
30 emphasize (e.g. in the conclusion) that responses of regional climate are complex due to the
31 interplay between the influencing atmospheric circulation systems. But, nevertheless, that
32 there is an imprint of changes in the dynamics and interplay of those systems (represented by
33 proxy data such as GISP K+ for the Siberian Anticyclone or NA hematite for Northern
34 Hemispheric circulations), observable in our record.

35
36 ⇒ While we feel that these issues have been discussed already in detail in section 5.4.
37 we rephrased parts of the conclusion and added one sentence to the abstract to
38 transport our message clearer.

39
40 The sentence *"Our interpretation of lower dD values indicating both relatively cool and wet*
41 *conditions fits well with results from other late Holocene records in arid Central Asia (Fig.*
42 *8)." refers mainly to the LIA, as compiled in the Central Asian Wetness index from Chen et*
43 *al. (8c) and also the Guliya accumulation rate (8e).*

44
45 ⇒ This is explained in the following paragraph, but to make this clearer we changed the
46 figure callout after the above sentence to (Fig. 8c, e and g). Here, as suggested by the
47 reviewer (see below) we also included a reference to the newly added compiled
48 temperature plot of Asia (Pages2k-Network, Nat Geo 2013), which is in Fig. 8g in the
49 revised version of the manuscript.

1 Although there is no 100% 1:1-similarity between the Karakuli silt and δD curves we still
2 think that the general ups and downs of the two proxy curves (represented by non-shaded and
3 shaded areas) vary somewhat consistently.
4

5
6
7 Further comments:
8
9

10 *Rev#2: If they want to invoke large-scale atmospheric rearrangements for interpreting their*
11 *isotopic signals in light of other studies, they should at least acknowledge that a change in*
12 *moisture source (local vs. remote) can drive a change in the isotopic composition of rainfall*
13 *without a corresponding change in temperature (and precipitation rate)*
14

15 Response: potential changes of moisture sources always need to be considered when
16 interpreting isotopic data. In our manuscript this is mainly handled by discussing shifts of the
17 paths of Westerlies throughout section 5.4.
18

19 ⇒ Further one additional sentence has been added to the first paragraph of section
20 5.2.1.
21
22
23

24 *Rev#2: If the authors decide to opt for a "temperature-driven" isotopic record, then they*
25 *should not miss the occasion to compare their results to the temperature results published in*
26 *the PAGES2k consortium paper (2013, Nature Geoscience) where a large set of data from*
27 *Asia were used to derive a continental-scale temperature record.*
28

29 Response: this was an oversight and we have now compared our results to PAGES2k and the
30 comparison is shown in Fig. 8g of the revised version of the manuscript and associated text.
31
32

33 *Rev#2: I understand why the authors opt to use the C26 and C28 for the dD. Still, the*
34 *supplementary information figure S3 shows some significant shifts in the d13C of those acids*
35 *that are paralleled, in particular for the 4-2 ka time interval, than find some echoes in the*
36 *dD, which suggests there were some contributions from different plant types to those d13C*
37 *curves that affected the dD as well.*

38 *Hence I would have liked to see a figure with temporal changes in the d13C and dD of all*
39 *individual fatty acids in the main article (not in the supplement), along with their own*
40 *respective concentrations. This would help convincing more the reader that shifts in*
41 *vegetation types does not significantly complicate the interpretation of dD that can have had*
42 *been obscured by changes in the C3/C4 contributions (having different fractionations on the*
43 *dD) of the different fatty acids homologues.*
44
45

46 Response: The reviewer raises an interesting concern about C₃ versus C₄ plant types. We had
47 previously assumed that C₄ plants were unimportant in the catchment and region. However in
48 response to the reviewers point we have search more deeply and found that there at least
49 partly C₄ plants of the taxa *Chenopodiaceae* could occur in some high altitude mountainous
50 deserts of the eastern Pamirs (newly included reference Sage et al., 2011), and thus a
51 contribution of C₄-derived lipids cannot be totally excluded. Further there is a possibility of

1 low levels of distal transport of C₄ waxes from surrounding Central Asian deserts; however,
2 this does indeed seem unlikely to contribute significant amounts to the sedimentary lipid pool.

3
4 Nevertheless we have acquired a δ¹³C record that allows us to explore the influence of
5 vegetation changes on the δD wax record. In particular we were more concerned about the
6 potential changes between aquatic and terrestrial sources, rather than the C₄ contribution.
7 However, in contrast to other densely macrophyte covered high-altitude lakes on the Tibetan
8 Plateau, that I have studied (Aichner et al., OG 2010), there are few macrophytes observable
9 in Lake Karakuli and thus the aquatic contribution is currently minimal in Lake Karakuli.
10 Furthermore, given the relative abundance distribution of fatty acids in an aquatic plant
11 collected close to the shore (Fig. S4), we would expect much higher abundances of C₁₆ and
12 C₁₈ in our sediments if the aquatic contribution is high. But this is not the case throughout the
13 whole sediment core (Fig. S2).

14
15 We agree that the slightly enriched δ¹³C-values (i.e. reaching values higher than -30‰) of C₂₈
16 during the period 4.0-3.5 kyrs BP must be either due to enhanced C₄ input or macrophyte
17 productivity could have been increased; and this might have additionally enhanced the
18 corresponding δD-C₂₈-values during this interval because of the different fractionation factors
19 for C₄-plants and/or different source water. However, this doesn't change anything about the
20 overall interpretation that this was a relatively dry and warm episode because this inference
21 can be made from both δD and δ¹³C (even though the δD-amplitude might be biased towards
22 more positive values for a few permil).

23
24 ⇒ We have now adjusted the discussion to develop both of these points (C₄ and
25 aquatic) further (new last paragraph in section 5.1.2; slight modification in section
26 5.1.3; additional sentence in the first paragraph of 5.3). We have also added another
27 figure to the supplement (S4) to illustrate the relatively low abundance of aquatic
28 plants in the lake at present which is limited to scattered patches close to the shore.

29
30 Concerning the potential changes of aquatic vs. terrestrial input, or C₄ vs. C₃ plants, the δ¹³C
31 values give the most important information which makes compound concentrations obsolete
32 (also the latter do not show many changes throughout the whole record and thus do not add a
33 new perspective to the story). The rather stable δ¹³C-values of C₂₈ (except for the above
34 mentioned interval between 4.0 and 3.5 kyrs BP) speak for relatively constant terrestrial C₃-
35 contribution to this compound. Slight ¹³C-enrichments of C₂₆, e.g. in the lower and middle
36 core section, could speak for a bias due to enhanced aquatic or C₄ input here. But since the
37 δD-curves of C₂₆ and C₂₈ run mostly parallel (except for the interval ca. 2500-2000 yrs BP),
38 we assumed this to be of minor relevance for the overall interpretation.

39
40 We have produced a study that takes a variety of approaches including proxy data, model
41 experiments and instrumental data. We attempt to communicate the main findings in the main
42 paper, and then to provide ancillary information in the supplement, as a resource to those
43 interested to delve deeper. The nuances of the different information contained in the various
44 chain lengths can be interpreted with knowledge of the biomarker production and carbon
45 isotopic compositions characteristic of different sources, and this is of interest to organic
46 geochemistry readers. But for the readers of *Climates of the Past*, the paleoclimate story that
47 emerges from the long chain hydrogen isotope record is most of interest for the research
48 question of climatic reconstructions.

1 **3. Other changes:**

2
3 p. 4389, 1.5 (section 2): changed time range of Bulun Kul climate data from 1956-1986 to =>
4 1956-1968

5
6 p. 4395, 1.25-28 (last sentence of paragraph 5.1.2): we removed the last sentence “We
7 estimate the macrophyte contribution....(Fig.7).” and the resulting scale bar in Fig. 7
8 showing the estimated quantified macrophyte contribution. Instead we added a relative scale
9 bar illustrating higher/lower macrophyte productivity and/or C₄-contribution. As described
10 above, we added another paragraph discussing the potential relevance of C₄-contribution to
11 the lipid pool of our studied lake.

12
13 p.4397, 1.4: changed year of reference Bowen and Revenaugh, 2013 => 2003

14
15 p.4398, 1.19: changed reference “Bowen, 2014” to “Bowen and Revenaugh, 2003”.

16
17 p.4406, (conclusions): several “Eastern”, “Western” and “Central” have been decapitalized
18 => “eastern”, “western”, “central”, except for “Central Asia”

19
20 p.4406, (acknowledgements): the acknowledgements have been slightly modified

21
22 There are a few other minor improvements/corrections of language/grammar which are not
23 listed here but which are annotated in the attached revised version of the manuscript.

24
25 Newly included references:

26 Sage, R.F., Kocacinar, F., Kubien, D.S.: C₄ photosynthesis and temperature, in: C₄
27 Photosynthesis and Related CO₂ Concentrating, Advances in Photosynthesis and Respiration
28 Mechanisms, 32, Springer Netherlands, doi: 10.1007/978-90-481-9407-0_10, 161-195, 2011

29 PAGES 2k Network.: Continental-scale temperature variability during the last two millennia.
30 Nat. Geosci., 6, 339–346, doi:10.1038/ngeo1797, 2013.

31
32 Newly included supplement:

33 A .kml-file showing positions of coring site and climate stations have been included as
34 supplement S7.

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 trend 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

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

4
5 Keywords: Pamir, Tibetan Plateau; Muztagh Ata, paleolimnology; biomarker; climate model;
6 LMDZ4

8 1 Introduction

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

20 While a large number of studies have analysed climate dynamics in monsoonal eastern Asia
21 and the north- and southeastern Tibetan Plateau (e.g. reviewed in Morill et al., 2003, An et al.,
22 2006 and Herzschuh, 2006), the density of paleoclimate records in continental Central Asia
23 remains comparably low. Central Asian records include studies of glacial extent in the Pamir
24 (e.g. Narama, 2002a and b) and tree-ring width reconstructions (e.g. Esper et al., 2002;
25 Treydte et al, 2006). Lacustrine sedimentary archives exist from Kyrgyzstan (Ricketts et al.,
26 2001; Lauterbach et al., 2014, Mathis et al., 2014), the Aral Sea (Sorrell et al., 2007a and b;
27 Boomer et al., 2009; Huang et al., 2011), the Western and Southern Tarim Basin (Zhao et al.,
28 2012; Zhong et al., 2007), and the Pamirs/Tajikistan (Mischke et al. 2010c; Lei et al., 2014)
29 (Fig 1b). Only one of those studies has included compound specific hydrogen isotopic
30 analyses (Lauterbach et al., 2014), which have elsewhere in Asia shown ~~its~~ potential to
31 provide information about moisture sources, precipitation amount and temperature (Mügler et
32 al.; 2010, Aichner et al., 2010c; Liu et al., 2008).

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

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

22

23 **2 Study site**

24 Lake Karakuli (also: Lake Kala Kule) is a small lake (ca. 1 x 1.5km) located at the
25 westernmost edge of Xinjiang Province (PR China) at an altitude of 3650 m, between the
26 massifs of Kongur Shan and Muztagh Ata, both exceeding 7500 m (Fig. 1a). Those mountains
27 which form the eastern edge of the Pamir plateau and the very westernmost edge of the
28 Tibetan Plateau are directly adjacent to the mountain ranges of Karakorum and Tien Shan.
29 The climate in this high altitude region is cold and dry. At Taxkorgan climate station, 80 km
30 south of Lake Karakuli (3090 m), average annual temperatures and precipitation amounts are
31 3.2°C and 69 mm, respectively (1957-1990; Miehe et al., 2001) with June and July being the
32 wettest months. Climatic data from Bulun Kul (3310 m), 30 km north of our study area, are in
33 a similar range (0.6 °C and 127 mm) with a precipitation maximum during spring and summer

1 (1956-1968; Miehe et al., 2001). At higher altitudes, precipitation amounts increase by
2 orographic forcing. At the Muztagh Ata, annual rain- and snowfall was estimated to account
3 for about 300 mm at the glacier accumulation zone (at 5919 m; Seong et al., 2009a) while
4 other studies estimated a water equivalent depth of 605 mm for snow accumulation at 7010 m
5 (Wu et al., 2008).

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

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

18

19 **3 Material and methods**

20 **3.1 Coring and chronology**

21 A sediment core with a composite length of ca. 820 cm was taken in September 2008 at
22 38.43968 °N and 75.05725 °E from a water depth of 16 m, using an UWITEC coring system
23 and a floating platform (the coring position is shown in supplement S7). The chronology was
24 based-on seventeen radiocarbon ages derived from ¹⁴C AMS dating conducted on total
25 organic carbon (TOC) (Liu et al., 2014). The 0 cal. yr BP (1950 A.D.) was derived from
26 ²¹⁰Pb/¹³⁷Cs dating and appeared at ca. 10.5 cm depth. A reservoir-effect of 1880 years was
27 extrapolated from dating of core-top samples and assumed to be constant throughout the core.
28 The ¹⁴C-ages indicate a nearly constant sedimentation rate across 4.3 kyr. For calibration of
29 the ages and construction of the age-depth model the IntCal09 dataset was used (Reimer et al.,
30 2009) applying a Bayesian method (Blaauw and Christen, 2011); for details see Liu et al.
31 (2014).

1 **3.2 Lab chemistry**

2 Sediments were extracted with Accelerated Solvent Extraction system (ASE 350; Dionex),
3 under high pressure (1500psi) and temperature (100°C) and using DCM/MeOH (9:1) as
4 solvent. Alkanoic acids were separated from the total lipid extract using column
5 chromatography (5 cm x 40 mm Pasteur pipette, NH₂ sepra bulk packing, 60 Å), eluting with
6 2:1 DCM/isopropanol, followed by 4% formic acid in diethylether, yielding neutral and acid
7 fractions respectively. The acid fraction was esterified with 5% HCl and 95% methanol (of
8 known isotopic composition) at 70°C for 12 h to yield corresponding fatty acid methyl esters
9 (FAMES). Lipids were obtained by liquid-liquid extraction using hexane as the non-polar
10 solvent, and dried by passing through a column of anhydrous Na₂SO₄. They were further
11 purified using column chromatography (5 cm x 40 mm Pasteur pipette, 5% water-deactivated
12 silica gel, 100–200 mesh), eluting with hexane, followed by FAMES eluted with DCM.

13 **3.3 Biomarker isotopic analysis**

14 Compound specific isotopic values were obtained using gas chromatography isotope ratio
15 mass spectrometry (GC-IRMS). We used a Thermo Scientific® Trace gas chromatograph
16 equipped with a Rxi-5ms column (30 m x 0.25 mm, film thickness 1µm) and a programmable
17 temperature vaporizing (PTV) injector operated in solvent split mode with an evaporation
18 temperature of 60°C. The GC was connected via a GC Isolink with pyrolysis/combustion
19 furnace (at 1400/1000 °C) and a Conflo IV interface to a DeltaV_{plus} isotope ratio mass
20 spectrometer. The H₃⁺-factor (Sessions et al., 2001) was determined daily to test
21 measurement-linearity of the system and accounted for 5.8 ppm mv⁻¹ on average. Reference
22 peaks of H₂/CO₂ bracket *n*-alkanoic acid peaks during the course of a GC-IRMS run; two of
23 these peaks were used for standardization of the isotopic analysis, while the remainders were
24 treated as unknowns to assess precision. Except for the case of co-elution, precision of these
25 replicates was better than 0.6‰.

26 Data were normalized to the Vienna Standard Mean Ocean Water (VSMOW)-Standard Light
27 Antarctic Precipitation (SLAP) hydrogen isotopic scale and to Vienna Pee Dee Belemnite
28 (VPBD) carbon isotopic scale by comparing with an external standard containing 15 *n*-alkane
29 compounds (C₁₆ to C₃₀) of known isotopic composition (obtained from A. Schimmelmann,
30 Indiana University, Bloomington). The RMS error of replicate measurements of the standard
31 across the course of analyses was below 5‰ (hydrogen) and 0.7‰ (carbon) For hydrogen
32 isotopes we further monitored for instrument drift by measuring the δD values of a C₃₄ *n*-

1 alkane internal standard co-injected with the sample ($-240.6 \pm 3.0\%$; $n=105$). The isotopic
2 composition of H and C added during methylation of alkanolic acids was estimated by
3 methylating and analyzing phthalic acid as a dimethyl ester (isotopic standard from A.
4 Schimmelmann, University of Indiana) yielding $\delta D_{\text{methanol}} = -198.3 \pm 3.9\%$, $\delta^{13}C_{\text{methanol}} = -$
5 $25.45 \pm 0.42\%$ ($n=7$). Correction for H and C added by methylation was then made by way of
6 mass balance.

7 **3.4 LMDZ4 simulations**

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

20

21 **4 Results**

22 **4.1 Lipid concentrations**

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

30 **4.2 δD and $\delta^{13}C$ values of leaf wax lipids and water samples**

1 In total, we measured 125 core samples for hydrogen isotopic composition and 66 samples for
2 carbon isotopic composition (Tables S6). Samples contained C₁₆- C₂₈ *n*-alkanoic acids with an
3 even:odd chain length preference. We report isotopic results for the C₂₄, C₂₆ and C₂₈ *n*-
4 alkanolic acids as these are target long chain compounds within the dynamic range of isotopic
5 measurement capabilities (Tables S6, Fig. S3).

6 $\delta^{13}\text{C}$ values are generally more depleted with increasing chain-length, with C₂₄ averaging to -
7 27.9±1.4‰, C₂₆ to -29.3±1.0‰, and C₂₈ *n*-alkanoic acids to -31.0±0.9‰ (Figs. 2 and S3). For
8 the C₂₈ we find no significant downcore trend. C₂₄ shows the largest variations in $\delta^{13}\text{C}$ values
9 with generally more ¹³C-depleted values in the middle of the core (min: -30.7‰) compared to
10 the core-base and core-top (max: -24.3‰) (Fig. S3). For hydrogen isotopes, compounds are
11 also more D-depleted with increasing chain-length (C₂₄: -173±6‰; C₂₆: -182±7‰; C₂₈: -
12 185±6‰; Fig. 2). We observe downcore variations in δD values for C₂₆ and C₂₈ ranging from
13 -196 to -167‰.

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

19

20 **5 Discussion**

21 **5.1 Origin of organic compounds and implications for source water**

22 **5.1.1 Molecular abundance distribution**

23 Organic compounds in lake sediments originate from a mixture of terrestrial and aquatic
24 organisms, with molecular abundance distributions and isotopic compositions that may be
25 diagnostic of source. Most plants contain a broad range of biomarkers (e.g. *n*-alkanes or
26 fatty acids) but the fingerprints of the different compound classes are mainly dominated by
27 compounds of a specific chain-length. Terrestrial and emergent aquatic plants for instance
28 produce higher proportional abundances of long-chain *n*- alkanes (e.g. C₂₉ and C₃₁) while
29 submerged macrophytes contain higher amounts of mid-chain *n*-alkanes (e.g. C₂₃ and C₂₅)
30 (Ficken et al., 2000; Aichner et al., 2010b). *n*-Alkanoic acids show a less distinct pattern
31 (Ficken et al., 2000), but also here long-chain compounds (e.g. C₂₈-FAs) are mostly

1 interpreted to be originated from terrestrial sources (e.g. Kusch et al., 2010; Feakins et al.,
2 2014).

3 In the sediments of Lake Karakuli the contribution of aquatic plants to the lipid pool is
4 considered to be relatively low compared to other Tibetan high altitude lakes. A **submerged**
5 **aquatic** plant sample collected close to the shore-line (ca. 20 cm water depth) shows a **strong**
6 dominance of C₁₆ and C₁₈-FAs and minor relative amounts of C₂₀ to C₃₀ even-chain FAs (see
7 Fig. S4). This fatty acid-pattern is in agreement with published fingerprints of other aquatic
8 plants collected on the Tibetan Plateau (Wang and Liu, 2012). Hence, the low relative
9 abundance of C₁₆ and C₁₈-FAs in our sediment samples suggests a relatively low contribution
10 of plant material derived from aquatic macrophytes to the **sedimentary** organic matter in Lake
11 Karakuli, ~~at least at the position where the sediment core was taken.~~

12 **5.1.2 Carbon isotopic signal**

13 Additional indication for the source of compounds comes from their carbon isotopic
14 signature. Lipids of terrestrial C₃-plants usually show values around -30 to -35‰, while
15 compounds derived from terrestrial C₄-plants and from submerged aquatic macrophytes can
16 reach significantly more enriched values in the range -15 to -20‰ (Chikaraishi and Naraoka
17 2005; Aichner et al, 2010a). The difference between C₃ and C₄-plants can be explained by
18 different isotopic fractionation in carbon assimilation of those two plant types, while the
19 enriched values of submerged aquatic plants are due to the uptake of different carbon sources
20 i.e. isotopically enriched bicarbonate instead of dissolved CO₂ (Allen and Spence, 1981; Prins
21 and Elzenga, 1989).

22 ~~As a consequence, in regions where C₄-vegetation is widely absent, the carbon isotopic~~
23 ~~signature of biomarkers can be applied to distinguish between aquatic and terrestrial sources~~
24 ~~(Aichner et al., 2010a and b).~~ In our sediment core from Lake Karakuli δ¹³C-values of the
25 C₂₈-FA are similar to that of terrestrial C₃-plants without a clear trend (Fig. 2; Fig. S3). Thus,
26 we conclude that this compound is predominantly derived from terrestrial C₃ grasses in the
27 lake catchment. δ¹³C values of C₂₄ and C₂₆-*n*-alkanoic acids are slightly higher than for C₂₈,
28 indicating an increasing contribution of submerged aquatic plant material **and/or lipids**
29 **derived from C₄-plants** with decreasing chain lengths.

30 δ¹³C values of C₂₄ *n*-alkanoic acids are controlled by relative contributions of aquatic
31 macrophytes and/or macrophyte productivity, with higher productivity leading to higher δ¹³C
32 values (Aichner et al., 2010b). We hypothesize that **a** higher proportional input of aquatic
33 material to the sedimentary organic matter is indicative of warmer and possibly also drier

1 conditions. Longer ice-free periods and a lower lake level could be the driving factors behind
2 enhanced macrophyte growth during warmer years. ~~We estimate the macrophyte contribution~~
3 ~~to the lipid pool based on a simple binary isotopic model with 19‰ as average end member~~
4 ~~value for aquatic lipids (Aichner et al., 2010a) and 33‰ for terrestrial lipids (Fieken et al.,~~
5 ~~2000) (Fig. 7).~~

6 C₄-plants are widely absent on the central and eastern Tibetan Plateau at present, but they are
7 wide-spread in Central Asian deserts and some *Chenopodiaceae* which use the C₄-pathway
8 have occasionally been observed at high altitude alpine deserts of the Pamir (Sage et al.,
9 2011). Thus we cannot totally exclude the contribution of C₄-derived lipids to the sedimentary
10 organic matter of Lake Karakuli, however, we consider these sources as of secondary
11 importance. Nevertheless, if we have underestimated the input of alkanolic acids derived from
12 C₄-plants this would not bias the overall interpretation, because higher abundances of C₄-
13 plants resulting in higher sedimentary $\delta^{13}\text{C}$ would indicate a drier/warmer climate, which is
14 similar to the hypothesis that drying/warming leads to increased macrophyte productivity.

15 5.1.3 Hydrogen isotopic signal

16 Hydrogen isotopes provide further evidence for the origins of C₂₄ and C₂₆ or C₂₈ *n*-alkanoic
17 acids. The average δD values of C₂₄ are ca. 9-12‰ higher than that of C₂₆ and C₂₈ (Fig. 2). A
18 different water source i.e. isotopically enriched lake water (see Tab. 1) instead of water
19 derived from precipitation or snow-melt could explain this. We assume that C₂₄ is derived
20 from a mixed aquatic and terrestrial sources ~~which could be both aquatic and terrestrial~~, while
21 C₂₈ and also C₂₆ can be considered as of mainly terrestrial origin.

22 The δD -values of these terrestrial biomarkers is representative of the hydrogen isotopic
23 composition of the source water which –for terrestrial plants– could be expected to be spring
24 and summer precipitation during the growing season (Sachse et al., 2012), although a
25 contribution of D-depleted melt-water from snow in the early spring growth period is highly
26 likely (Fan et al., 2013). The fractionation factors between source water (~~i.e. leaf water~~) and
27 lipids are variable but previous studies found that for terrestrial C₃-grasses they average to -
28 $149\pm 28\text{‰}$ (n=47) for the C₂₉ *n*-alkane, while they are ca. $-134\pm 28\text{‰}$ (n=53) for C₄-grasses
29 and in similar range for forbs (Sachse et al., 2012). In arid ecosystems, soil-water evaporation
30 (for grasses; Smith and Freeman, 2006) and transpiration from the leaf, lead to isotopic
31 enrichment of leaf water above the meteoric water (Feakins and Sessions, 2010; Kahmen et
32 al., 2013a and b). Recent results from the central Tibetan Plateau, ~~which is~~ a similar
33 environmental setting to our study, quantified the apparent isotopic fraction between meteoric

1 water and *n*-alkanes to be ca. -95‰ due to ca. +70‰ evapotranspirational isotopic enrichment
2 above meteoric water (Günther et al., 2013). This is in agreement with the average
3 fractionation from Feakins and Sessions (2010) who suggested ca. -95‰ as net fractionation
4 factor between meteoric water and leaf wax *n*-alkanes in an arid ecosystem (southern
5 California), and found similar values for *n*-alkanoic acids in a later study from that region
6 (Feakins et al., 2014).

7 While the fractionation was not directly determined on modern plant *n*-alkanoic acids in this
8 catchment, based on core-top δD_{lipid} -values of ca. -190‰ and knowledge of hydrogen isotope
9 values of modern precipitation and waters in the catchment we can infer a reasonable
10 catchment average apparent fractionation (Fig. 3). Summer precipitation in the catchment
11 averages ca. -45‰ at Lake Karakuli, compared to mean annual precipitation average of ca. -
12 90‰ (derived from the Online Isotopes in Precipitation Calculator, OIPC; Bowen and
13 Revenaugh, 2003.; Fig. 4b). Assuming If the summer precipitation (-45‰; OIPC) is
14 indicative of source water, and then-given the OIPC summer precipitation δD value of -45‰
15 and the measured sedimentary value of C_{28} *n*-alkanoic acids (-190‰) we would compute an
16 apparent fractionation of ca. -150‰ (see supplement S5 for formula to calculate isotopic
17 fractionation factors). Whereas if we use mean annual precipitation (ca. -90‰; OIPC) then the
18 calculated apparent fractionation would be ca. -110‰ which is closer to the reported
19 fractionation factors for arid ecosystems (Feakins and Sessions, 2010; Günther et al., 2013).

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

32 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

1 To further establish the connections between climate anomalies and isotopic signatures of
2 precipitation in Central Asia, we compare instrumental data and climate model simulations.
3 At Taxkorgan meteorological station we find a negative correlation between annual
4 temperature and precipitation amount over a period of 43 yrs (1957-2000; Fig. 5; data
5 provided from Tian et al., 2006). Similar trends can be observed when comparing simulated
6 data over a period of 50 yrs (1958-2009; Fig. 5). We use the LMDZ4 climate model (Hourdin
7 et al., 2006) to characterize the climatic processes in our study area (as described in Lee et al.
8 2012). We find higher annual precipitation amounts in the LMDZ4 model simulations
9 compared to instrumental observations at Taxkorgan meteorological stations. This is related
10 to the scale of the model resolution of $3.75^\circ \times 2.5^\circ$ (Lee et al., 2012) which includes the
11 relatively high precipitation amounts in higher altitudes during winter (Seong et al., 2009a and
12 b; Wu et al., 2008) within the grid box. Significant negative correlations ($r=0.58$; $p<0.0001$)
13 between temperature and precipitation amount can be inferred for the summer months (April-
14 September), while comparisons over the winter or whole year deliver non-significant
15 correlations ($p>0.01$; Fig. 5).

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

28 In a recent study Tian et al. (2006) found a positive correlation between $\delta^{18}\text{O}$ in the local
29 Muztagh Ata ice core (which covers the period 1957-2003) and annual temperatures from
30 Taxkorgan climate station. In contrast they found no significant relationship between ice-core
31 $\delta^{18}\text{O}$ and annual precipitation amount at Taxkorgan (Tian et al., 2006). Different precipitation
32 dynamics between middle and high altitudes, and/or seasonal differences, as supported by our
33 LMDZ4-data, could explain this discrepancy. Elevation differences may play a role in

1 different precipitation patterns and these may be associated with isotope effects. The Muztagh
2 Ata glacier accumulation zone receives higher annual precipitation amounts and also a higher
3 proportional input from winter precipitation compared to lower altitudes (Seong et al., 2009a
4 and b). Whilst instrumental and modelling data inferred a slight increase of precipitation
5 amount throughout the last 50 years in the westernmost part of China (Yao et al., 2012; Zhang
6 and Cong, 2014), a decreasing accumulation rate at the Muztagh Ata ice core since 1976 was
7 measured by Duan et al. (2007). Even if the instrumental data from Taxkorgan do not show a
8 significant trend in precipitation amount between 1957 and 2000, this does not rule out
9 changes of snowfall at higher altitudes. Increasing temperatures could have further
10 contributed to the lower observed accumulation rates.

11 ~~According to the interpretations given above, there are three main factors which potentially~~
12 ~~influence δD values of biomarkers in our sediment core: a) temperature; b) precipitation~~
13 ~~amount and c) the proportional uptake of D-depleted source water in the early vegetation~~
14 ~~period, which is derived from snow-melt and/or early spring precipitation.~~ Since temperature
15 and precipitation amounts are anti-correlated on an interannual timescale (Fig. 5), we interpret
16 low δD values to indicate both relatively cool and wet conditions. In addition to fluctuations
17 in mean annual precipitation isotopes, snow-melt and delivery to plants may vary. We suggest
18 that a high proportional contribution of water derived from snow-melt, after relatively long
19 and wet winters with high amounts of snowfall, can further lead to more negative δD leaf wax
20 values.

21 **5.3 Paleoclimatic interpretation of downcore data**

22 δD and $\delta^{13}C$ values from Lake Karakuli sediment core suggest relatively warm and dry
23 conditions between ca. 4-3.5 kyrs BP (Fig. 7). $\delta^{13}C$ values are highest for C_{24} during this
24 interval and even C_{28} shows slightly enriched values ($>-30\text{‰}$; Fig. S3). Also δD shows
25 maximum values during this episode. Even though an increased input from C_4 -plants or
26 enhanced productivity of aquatic macrophytes could slightly have biased δD -values towards a
27 more positive signal, we infer that this period probably was the warmest/driest in our studied
28 time-interval. After 3.5 kyrs a gradual cooling trend started (interrupted by a warmer/drier
29 period between ca. 3.0 and 2.7 kyrs BP), peaking in coolest and wettest conditions around 2.5
30 kyrs BP. Between ca. 2.5 and 1.9 kyrs BP we observe a reversal to a slightly warmer and drier
31 climate, based on δD evidence. We note that the $\delta^{13}C$ values are rather variable and
32 inconclusive in this core-section, and we observe an offset between $\delta D-C_{26}$ and $\delta D-C_{28}$ (these
33 are normally within analytical error of each other). We hypothesize that a warming influenced

1 precipitation isotopes but that the change wasn't intense and stable enough to trigger a large-
2 scale ecosystem response to be recorded in the $\delta^{13}\text{C}$ values. Between ca. 1.9 and 1.4 kyrs BP,
3 cool and wet conditions occurred again before returning to a warm and dry episode from ca.
4 1.4 to 0.6 kyrs BP (possibly interrupted by a cooling event around 1 kyrs BP). The last 0.6
5 kyrs have been mainly cool and wet again, except for the last ca. 100 years, where the
6 topmost three samples of the sediment core indicate another reversal to relatively warm and
7 dry conditions.

8 Enhanced precipitation, rather than lower temperatures, has been argued to be the main
9 driving force behind growth of glaciers in Asian high-altitude regions (Seong et al., 2009b).
10 The cool/wet episodes deduced from our organic geochemical record match relatively well to
11 reconstructed glacial advances at Mts. Muztagh Ata and Kongur Shan. On basis of ^{10}Be -
12 dating of erratic boulders Seong et al. (2009a) estimated maximal glacial advances at 4.2 ± 0.3
13 kyrs, 3.3 ± 0.6 kyrs, 1.4 ± 0.1 kyrs, and a few hundred years before present (Fig. 7). Further, the
14 δD -data are in good agreement with silt-contents in the same sediment core (Fig. 7). These
15 have been interpreted to be influenced by glacial input and thus higher contents indicating
16 cooler/wetter conditions (Liu et al., 2014).

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

28 Similarly a number of records have shown a pronounced warm/dry period during the
29 Medieval Climate Anomaly (MCA; Fig 8 c,e; Chen et al., 2010; Lauterbach et al., 2014;
30 Esper et al., 2002) also seen in our record from Late Karakuli. At ca. 1 cal. ka BP we observe
31 a ca. 100-year interruption of this event indicated by three samples with lower δD -values.
32 Recently, Lei et al. (2014) observed a similar spike in carbonate $\delta^{18}\text{O}$ values from Lake Sasi
33 Kul, which is located ca. 190 km west of our study site (Fig. 8b). Thus we hypothesize that

1 this interruption was not just a local phenomenon. Warm and dry conditions during the MCA
2 have also been observed at Kashgar (western Tarim Basin; just ca. 150 km north of Lake
3 Karakuli; Zhao et al., 2012), and from from Lakes Bangong Co on the western Tibetan
4 Plateau (Gasse et al., 1996) and large Karakul in the Tajik Pamir (Mischke et al., 2010).

5 Applying these findings to the complete record we see fluctuating climatic conditions
6 throughout the late Holocene with clearly identifiable warmer/drier and cooler/wetter
7 episodes (Fig. 8). During the oldest section of our record (ca. 4.2-3.4 kyrs BP) average
8 conditions appeared having been warmer and drier than during the medieval and today,
9 followed by a general (even though non-continuous) cooling trend until ca. 2.4 kyrs BP. A
10 cool and wet phase of roughly 1000 years starting at ca. 3.5 kyrs BP has been observed in
11 numerous global climate records (Mayewski et al., 2004). At the nearby oasis of Kashgar,
12 conditions prevailed relatively wet from ca. 4.0 until ca. 2.6 kyrs BP (Zhao et al., 2012). At
13 the large Lake Karakul in Tajikistan a rapid drop of TOC-contents occurred at ca. 3.5 cal. ka
14 BP, indicating a drop of lake productivity probably induced by low-temperatures and
15 eventually associated with shorter ice-free periods in the summer (Mischke et al., 2010; Fig.
16 8h). At Lake Balikun (northeastern Xinjiang) a reversal to wetter conditions occurred after a
17 pronounced dry event lasting from 4.3-3.8 kyrs BP (An et al., 2012). In Lake Manas (northern
18 Xinjiang) a wet episode was reconstructed for 4.5-2.5 kyrs BP, interrupted by a short dry
19 period between 3.8-3.5 kyrs BP (Rhodes et al., 1996). Low $\delta^{18}\text{O}$ -values in the Guliya ice core
20 between 3.5 and 3.0 kyrs BP also give evidence for low temperatures on the northwestern
21 Tibetan Plateau (Thompson et al., 1997) while in the southern Tarim Basin a rapid shift to
22 wetter conditions at ca. 3.0 kyrs BP have been observed (Zhong et al., 2007).

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

30 **5.4 Implications for Central Asian climate dynamics**

31 The sequence of relatively cool/wet and warm/dry episodes displays coherency with other
32 records of Northern Hemisphere climate records. There is a similarity between cyclicality of

1 cooling events at Lake Karakuli, Northern Atlantic ice-rafting events (Fig 8j; Bond et al.,
2 2001) and strengthening phases of the Siberian High (the anticyclonic high pressure ridge
3 over Siberia), the latter recorded by $[K^+]$ increases in the GISP2 ice core between ca. 3.5-2.8
4 and 0.5-0.2 kyrs BP (Fig 8i; Mayewski et al., 1997). Further, throughout the last ca. 1000
5 years, δD values of leaf waxes in Lake Karakuli are correlated with the mode of the North
6 Atlantic Oscillation (NAO), showing more positive values during the current and medieval
7 positive mode and more negative values during the LIA-negative mode (Fig. 8f; Trouet et al.,
8 2009).

9 The interplay between the dominant atmospheric circulation systems in Central Asia – the
10 Siberian High, the mid-latitude Westerlies and partly the Indian Summer Monsoon– as well as
11 orographic influences, lead to complex climatic patterns. Trajectory studies in the modern
12 atmosphere, as well as inventories of dust particles in ice cores, suggest the mid-latitude
13 Westerlies as primary source of moisture during winter and spring, with the North Atlantic,
14 the Mediterranean, the Black and Caspian Sea as possible regions of origin (Lei et al., 2014;
15 Seong et al., 2009a and b; Wu et al., 2008). The Siberian High delivers cool but also relatively
16 dry air during winter. The absence of sea-salt i.e. in the Muztagh Ata ice core (Aizen et al.,
17 2001; Seong et al., 2009b) further gives evidence for a minor importance of the Indian
18 Monsoon, and for mid-latitude Westerlies and local convection to be the most important
19 moisture sources during the summer.

20 Even though Lake Karakuli receives some moisture in spring (Fig. 4), regions which are
21 located as close as 190 km westwards at a similar altitude, such as Lake Sasi Kul and other
22 parts of the central and western Pamirs receive much higher proportions and amounts of
23 winter and spring precipitation (Lei et al., 2014; Mische et al., 2001). Variations of strength
24 and tracks of the Westerlies and related movement of the Polar Front (Machalett et al., 2008)
25 could have influenced the amount of winter and spring moisture which has reached the
26 Karakuli-region in the past. Lei et al. (2014) suggested that during negative NAO-modes (e.g.
27 during the LIA) the storm tracks were moving further southwards, leading to wetter
28 conditions in the Mediterranean and higher amounts of moisture been transported into Central
29 Asian realms of the same latitude. In contrast other authors proposed a more complex
30 interplay between the Eurasian and Pacific circulation systems on basis of modelling data, and
31 a generally higher delivery of moisture into Central Asia during episodes of strengthened
32 Westerlies (i.e. positive NAO-modes) (Syed et al., 2010; Syed, 2011). Recently, a possible
33 negative correlation between lower winter precipitation in the Mediterranean (positive NAO-

1 mode) and higher winter precipitation at Son Kol (central Tien Shan; ca. 400 km north of
2 Lake Karakuli) was also suggested by Lauterbach et al. (2014) on basis of $\delta^{15}\text{N}$ -data on total
3 nitrogen (Fig. 8d).

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

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

27

28 **6 Conclusion**

29 The biomarker isotopic record from Lake Karakuli, eastern Pamirs, shows distinct episodes of
30 relatively cool/wet and warm/dry climate over the last 4200 years. Variations in the North
31 Atlantic conditions and Siberian High both appear to show similarities with variations
32 captured in our biomarker isotopic record, including notable excursions associated around 3.5
33 kyrs, the MCA, and the LIA. However, there are also indications for complex responses of

1 regional climate, i.e. different responses between the western (e.g. western and central Pamir),
2 eastern (e.g. eastern Pamir) and northern (e.g. Tien Shan) parts of Central Asia. These
3 regional differences are thought to arise from changes in the dynamics and interplay of the
4 involved large scale atmospheric circulation systems, especially the strengths and pathways of
5 the Westerlies. Our data provide evidence that the transition between regions of summer-only
6 and winter/spring dominated precipitation could have been a key factor for local climate in
7 the past. They further show a rapid aridification in the eastern Pamir during the last 50-100
8 years.

9

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21

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

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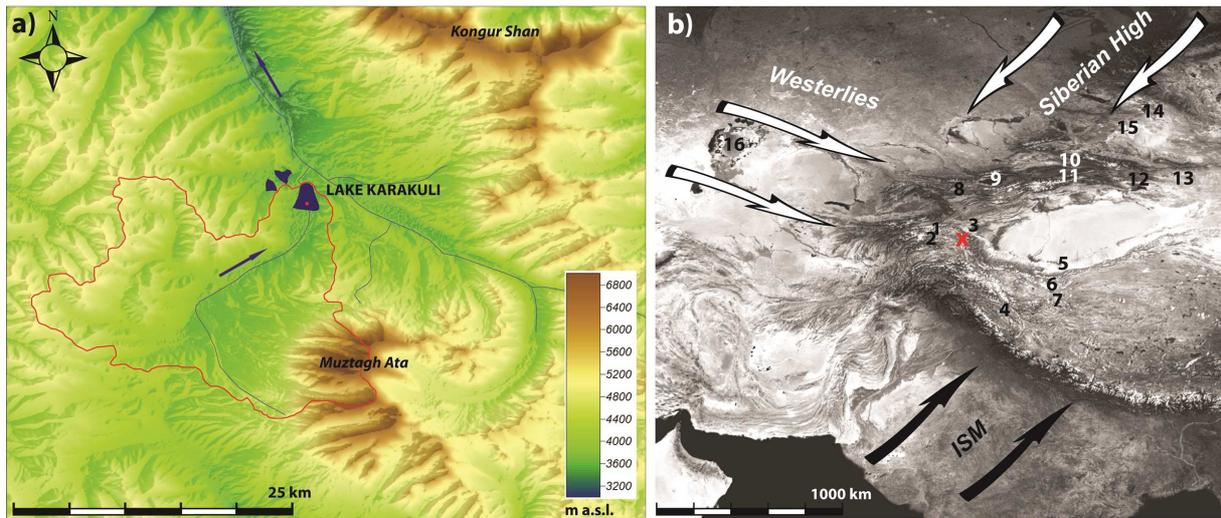
Tables

Table 1: $\delta^{18}\text{O}$ 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	$\delta^{18}\text{O}$ [‰]	1σ	δD [‰]	1σ
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

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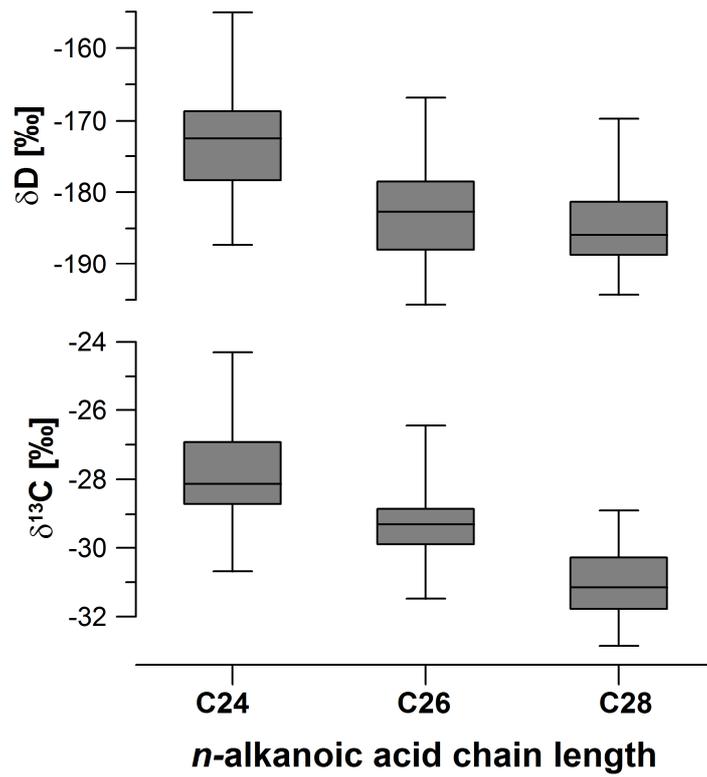
1 **Figures**



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

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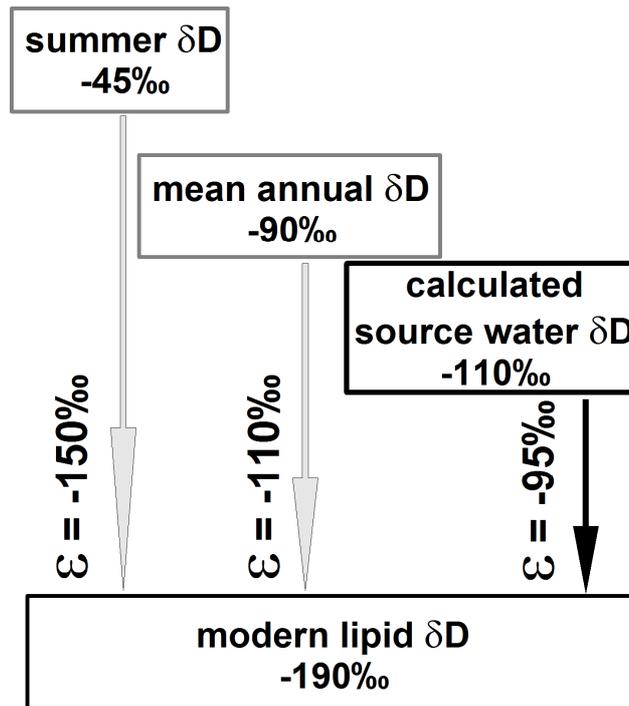


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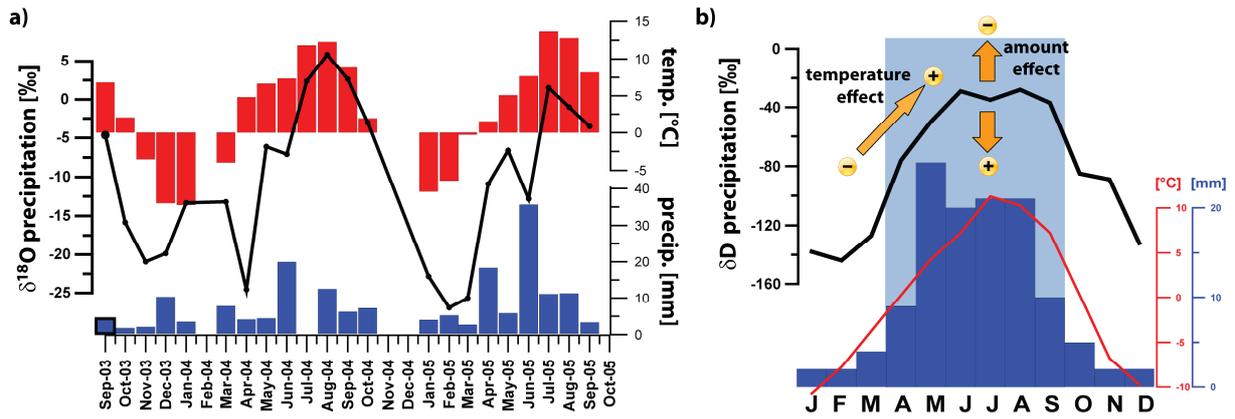
3 Fig. 2: Box and whisker plots of δD and $\delta^{13}C$ values in sediment samples by chain length.

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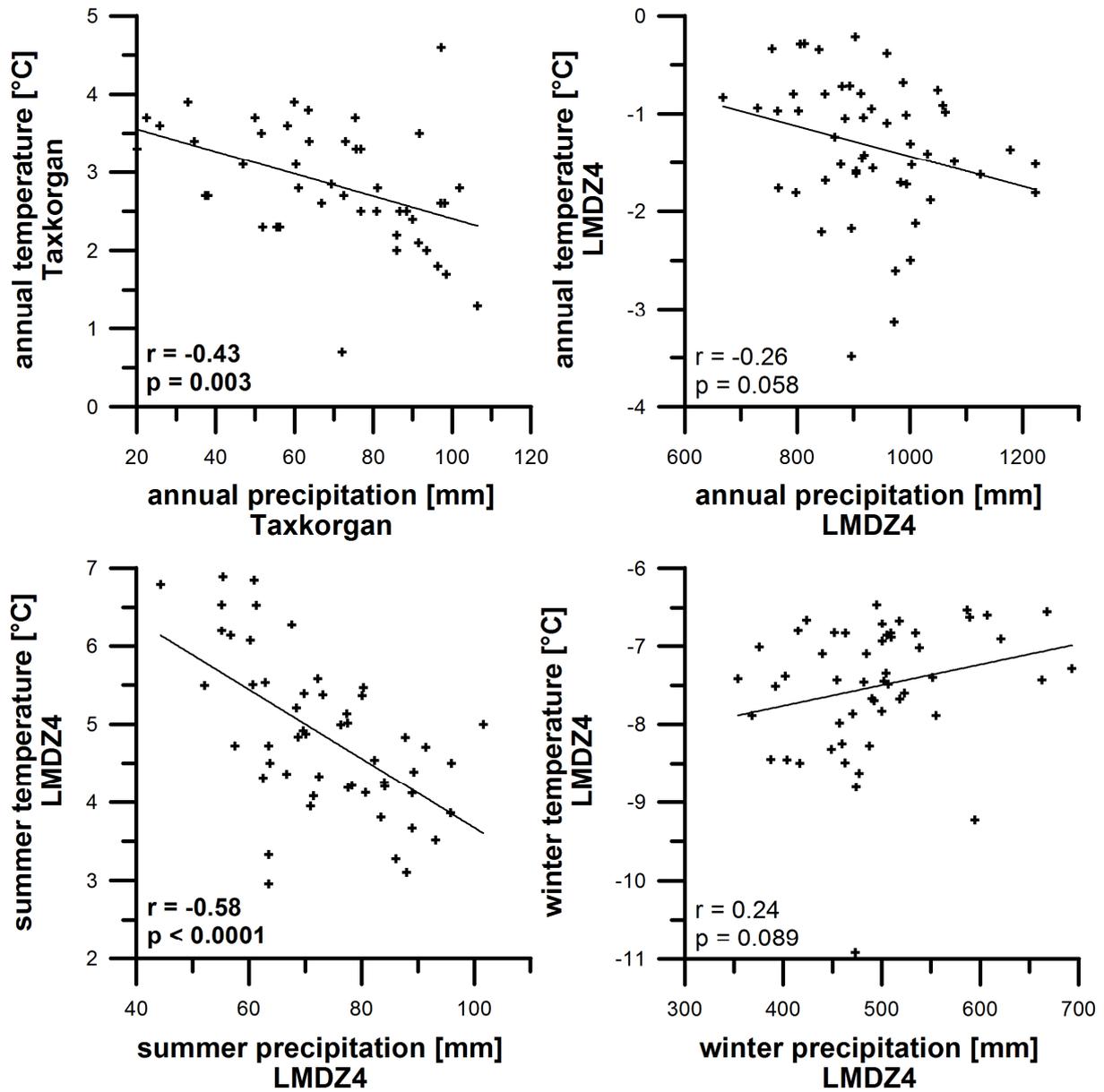
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Fig. 3: Calculated isotopic fractionation factors (ϵ) between summer and mean annual precipitation and modern lipids, as well as calculated source water δD on basis of published fractionation factors in arid ecosystems (ca. -95‰ according to Feakins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



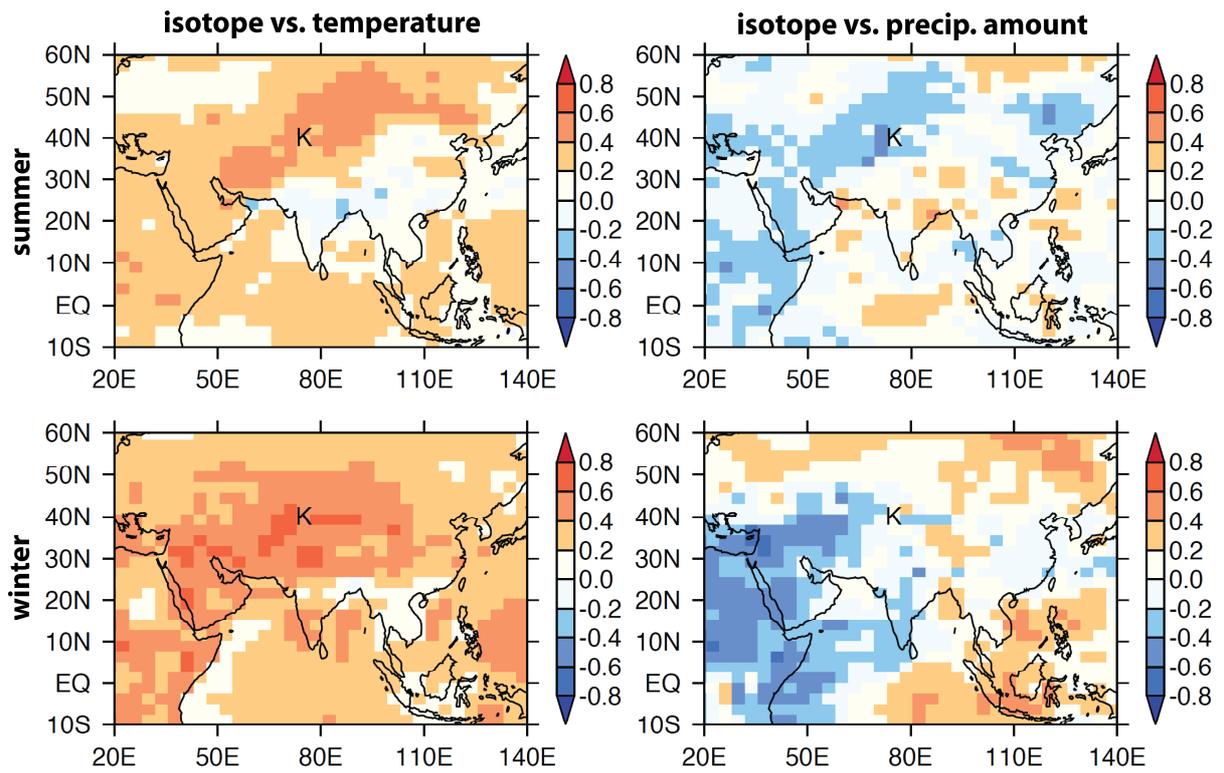
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Fig.4: (a) Monthly isotopic and climate data from Taxkorgan climate station (Yao et al., 2013), located ca. 80 km south of Lake Karakuli (altitude ca. 3100 m). (b) Average monthly climate (Miehe et al., 2001) and isotopic (OIPC; Bowen and Revenaugh, 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.

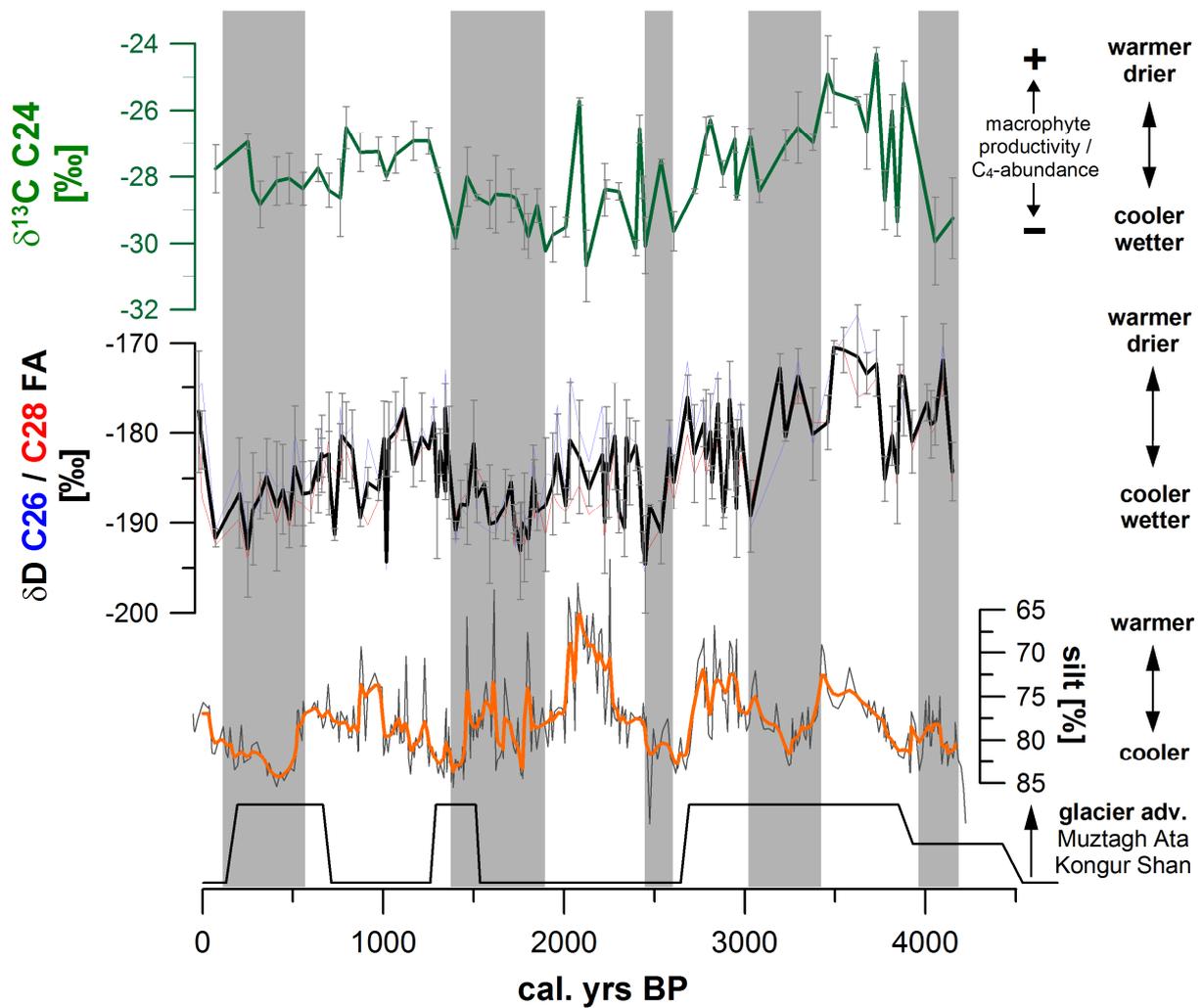


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 2 Fig. 5: Correlations of temperatures with precipitation amounts based on instrumental data
 3 from Taxkorgan meteorological station (1957-2000; annual averages) and model data using
 4 LMDZ4 simulations (1958-2009; summer: April-September; winter: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

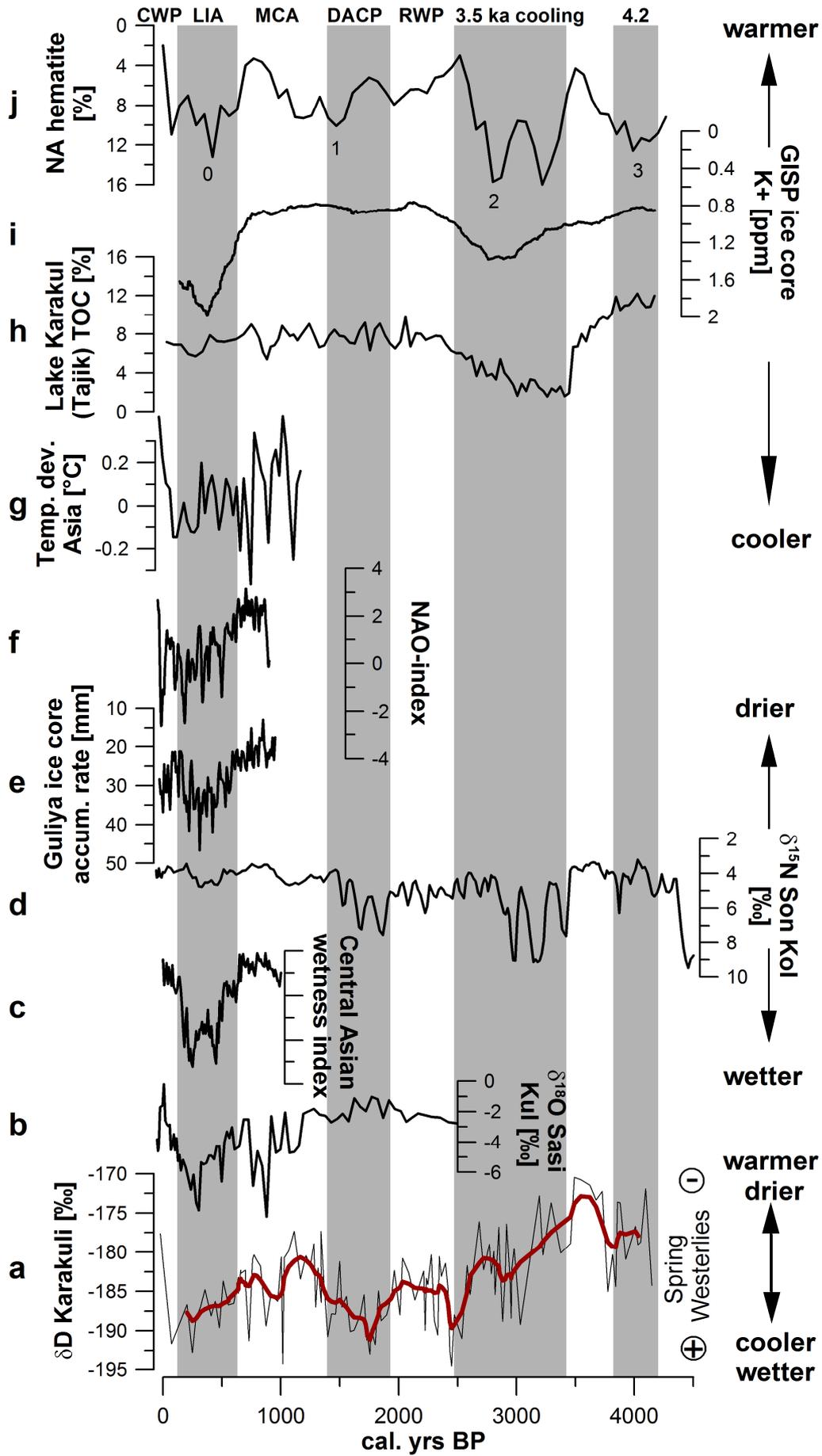
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 2 Fig. 6: Spatial correlation coefficient (r) summer (April-September) and winter (October-
 3 March) $\delta^{18}\text{O}$ of precipitation at the Karakuli site (marked as K in the plots) with temperatures
 4 and precipitation amounts at each grid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



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



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

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