Dear Frank Hagedorn,

we would like to thank you for your effort with our manuscript and the constructive comments/suggestions concerning the altitude discussion. Unfortunately, we have not measured the bulk $\delta^{13}C$ of topsoils and plants yet.

We have revised the manuscript according to the reviewers and your suggestions. Please find below our point-by-point reply and the manuscript with marked changes.

With best wishes on behalf of the co-authors,

Julian Struck

Anonymous Referee #1:

General comments

The study "Leaf wax n-alkane pattern and compound-specific d13C of plants and topsoils from semi-arid Mongolia" by Julian Struck and colleagues presents novel data on content and isotopic signatures of plant waxes along two climate transects in Mongolia. Generally, the study is written in a clear and understandable way. The methods used are standard within the respective research community and the execution from sampling to data analysis seems robust. Also, the presentation of data in terms of figures and tables is clear and straightforward understandable.

→ We are very happy about these positive comments on our manuscript.

A major issue with the language is that the clarity could be improved at instances when comparisons are made. Here, it is often just stated that something is "higher" or "larger", but it is often missing "compared to what". Most of the time this could be traced from the sentence before, but I would suggest to always add this information in the same sentence for clarity. Thus, please check again the whole manuscript for statements where comparisons are made.

→ We have checked the manuscript and adjusted sentences with comparisons for clarity.

Another major issue concerning the science is that (especially in the conclusions section) contradicting statements are made. Here, it is stated that n-alkane homologue patterns are not influenced by climatic parameters, although they are strongly correlated to OEP, which is actually a numerical representation of (changes in) the homologue patterns. Thus, I suggest to carefully review this section, since in the present form it is unclear and contradictory.

→ You are right, and we have to be more specific in the discussion that we have used the term *n*-alkane homologue patterns in terms of typical *n*-alkane distributions for different plants or plant-groups. E.g. typical n-alkane homologue pattern for grasses/ herbs show a dominance of *n*-C_{31} and *n*-C_{33} and deciduous trees/shrubs *n*-C_{27} and *n*-C_{29}.

Specific comments

1.) L2 There are previous studies, which looked at n-alkanes in (semi-)arid regions. Just as a singular example (there are others): Feakins and Sessions (2010 in Geochimica et Cosmochimica Acta). Thus, I suggest to remove or rephrase this bold statement.

→ We have rephrased this statement and refer especially to the semi-arid/arid regions of Mongolia.

2.)   L29 Please specify "additional paleoclimatic information".

→ We added 'about drought stress conditions'

3.)   L44-47 The paragraph does not fit very well here. Maybe it would be better to incorporate in the section L24-37?

→ We have incorporated the sentence dealing with the temperature correlation in the section L34-37.

4.)   L117 Please check equation 2 again. It is not clear what "v27" stands for.

→ We have corrected it to *n*-C27.

5.)   L168-170 How did Artemisia grow in your study area: herbaceous or woody shrub? Please specify.

→ Along transect II, Artemisia plants grow both as herbaceous plants and as perennial 'shrubby' plants with a woody base (e.g. Artemisia frigida). Our dataset includes Artemisia frigida samples and herbaceous Artemisia samples. Unfortunately, most samples could only be determined on a plant genus level (Artemisia spp.). We have included a specification in section 2.1 Geographical setting and sampling: *"Artemisia spp. summarizes different herbaceous species and perennial 'shrubby' species with a woody base (e.g. Artemisia frigida)."*

6.)   L189 Please give reference to the figure where the data is shown.

→ Done.

7.)   L190 Why "except Larix"? Please specify.

→ We have deleted this statement because Larix sp. is also within the typical range of C3 Plants.

8.)   L198-199 Please rephrase the sentence: "no significance" is redundant and it could be clearer

→ We have deleted the term significant and changed the sentence to *"While no differences are found between the $\delta^{13}C$ values of the grasses/herbs and woody shrubs, only Larix sp. is enriched up to 2‰, but still in the range of C_3 plants."*

9.)   L243-267 The whole paragraph shows an extensive use of the word "strong correlation". Could you please state (maybe already in the methods section), when you consider a correlation as "strong"? Is there a R2 threshold which you apply? Please specify.

→ We have rephrased this paragraph in terms of the word "strong correlation". We have not applied any R^2 threshold. The goodness of fit is based on the weighted R^2 values and comparisons were done relative to each other under consideration of the p-values.

10.)   L252 Please elaborate a bit more on the link with livestock grazing, since this is not obvious.

→ We agree with referee #1 that the former version was misleading. We have changed the structure of the paragraph. The link to livestock grazing is now described after the variations

of n-alkane concentrations. Due to the fact that biomass production decreases with increasing aridity, the n-alkane concentration in topsoils become reduced since less organic material is incorporated into the topsoil. *n*-Alkane concentrations in the topsoils become even more reduced with intensified livestock grazing, because less biomass will be incorporated.

11.) L265 I would remove the statement that the R2 of 0.683 "seems to be even stronger" than the R2 of 0.691. In my opinion they are similar.

→ We agree. The statement has been removed.

12.) L267 Please define WUE at some point (if not already done). I guess it is "water use efficiency".

→ It's defined in the introduction as water use efficiency.

13.) L288 It sounds contradicting that you state "the n-alkane homologue patterns from the topsoils are not influenced by climatic parameters, and thus the n-alkane ratio can reliably be used to detect and reconstruct differences between the vegetation forms of grasses and woody shrubs". First, in the sentence before you state that n-alkane concentrations and OEP values are significantly correlated to climatic parameters. Second, what do you mean with n-alkane homologue pattern (the sum of n-alkanes concentrations, ACL, OEP or n-alkane ratio)? Please check again, since in the present form the sentence is unclear and contradictory.

→ We have used the term *n*-alkane homologue patterns in terms of typical *n*-alkane distributions for different plants or plant-groups. E.g. typical n-alkane homologue pattern for grasses/ herbs show a dominance of n-C_{31} and n-C_{33} and deciduous trees/shrubs n-C_{27} and n-C_{29}. We have specified our statement and defined what we mean with n-alkane homologue patterns in this case.

14.) L294 Please specify what you mean with "detailed identification of plant species".

→ We agree and changed the sentence to "should include a detailed identification of plants regarding different species of each plant genus."

15.) L296 Again also here: It is contradictory when you state that "homologue patterns are not biased by climatic influences", although you show correlations of OEP with climate and describe these as "strong" in your discussion. In the end, OEP etc. are just numerical representations of the homologue pattern. Thus, please clarify the contradiction.

→ We have changes homologue patterns to ACL and *n*-alkane ratio

16.) L298 Maybe "can be potentially used", to weaken the conclusion a bit.

→ We agree and added potentially

17.) Figure 3 The sentence "Plants originate from transect II." Is redundant and can be removed.

→ This is correct, we removed it.

18.) Figure 6 Please indicate which regressions are linear and which are polynomial. Also I would suggest to add the p-values along with the R2, to clarify the significance of the fit.

→ We have added the p-values along with the R^2 values. Linear and non-linear regressions have now different colours and are described in the Fig caption. We have checked all regressions and based on the p-values we have chosen linear or non-linear regressions.

19.)   *Table 1 Check decimal places in last row*

→ We have changed it to three decimal places (0.000).

Technical corrections

Title "patterns" → corrected

L1 "patterns" → corrected

L11 "correlated" → corrected

L16 "are synthesized" → corrected

L21 check order of references and hyphenation → We checked the order of all references. Now they are all sorted alphabetically / Hyphenation is done by the Copernicus LATEX template

L67 check order of references → We checked the order of all references. Now they are all sorted alphabetically

L88 "accelerated" → corrected

L89 "dichloromethane" → corrected

L102 "Agilent" → corrected

L110 delete "sediment", since it is either soil or plant material → adjusted

L142 "Table 1 shows" → corrected

L249 Check brackets on reference → corrected

L250 Maybe better: ": : : OEP, which is strongly correlated : : :" → We agree

L269 "patterns" → corrected

L272 "patterns" → corrected

L277 Two points at end of sentence → corrected

L288 "decrease with increasing" → corrected

L288 "patterns" → corrected

L292 delete "for" → done

L294 Two points at end of sentence → corrected

Additional adjustments

L146 valuess → values

Figure caption of fig 1 → we changed black cycles to black/whithe cycles

Anonymous Referee #2:

The authors of this manuscript investigated the link between several environmental parameters (mean annual temperature, mean annual precipitation, and aridity index) and molecular and stable carbon isotope compositions of leaf wax n-alkanes extracted from modern higher plants and topsoils along 2 broad transects in Mongolia. The manuscript provides much needed molecular and stable isotope data for that area and will be of interest to biogeochemists, paleoecologists, and paleoclimatologists studying past climate change in the arid zones of central Eurasia. The manuscript fits within the scope of Biogeosciences Discussions and should be published in this journal provided the authors address the following issues:

→ We are very happy about this positive comment on our manuscript.

Major point to address

First, lumping topsoil n-alkane data when looking at Transects I and II

Transect II The data shown in Fig. 4 and Fig. 5 for Transect II has a lot of scatter. The transect includes 3 different areas A, B, and C, with area B corresponding to an altitudinal transect. Could this scatter be the result of additional factors controlling the molecular and d13C data along the altitudinal transect in addition to those that play role along the W-E transect (i.e. A, through B (only sites 22, 23, 24, 25) through C)? Could the altitudinal transect sites be plotting separately?

Transect II + Transect I A similar issue could be the reason for a large scatter in the d13C data in Fig. 6 (top 2 sections). There is a lot of scatter at ~ - 6C MAT, ~ 210 mm MAP, and ~ 0.28 AI. Could this be caused by multiple factors (in addition to those plotted along the X-axis) controlling the d13C values of n-alkanes along and within these transects? Can the data be plotted separately to provide a more nuanced assessment?

→ Thank you for this comment. The scatter you mentioned is not caused by the samples from the altitude transect (TSII-B), but by the samples from the Telmen catchment (TSII-C) (Fig.1, 2). Fig. 1 and 2 show exemplarily the correlation of $\delta^{13}C$ n-C_{29} with altitude, MAT and MAP, with colours/symbols indicating the different sites.

For all leaf wax proxies, we checked for altitude as a controlling factor and added an altitude correlation plot as separate figure (Fig. 2). There are significant correlations between altitude and Σ n-alkane (both, n-C_{23} – n-C_{35} and n-C_{25} – n-C_{35}), OEP and $\delta^{13}C$ (both, n-C_{29} and n-C_{31}). Correlation with ACL is weak and non-significant, and the n-alkane ratio shows no correlation at all. We will include a detailed description about altitude as a controlling factor within the figures and the discussion part.

Nevertheless, altitude generally controls MAT ($R^2 = 0.624$) and MAP ($R^2 = 0.395$), and we think that all factors are important and influence the leaf wax signal. However, the scatter in our transect is rather the result of site-specific/micro-climatic characteristics and variations in plant physiology. We will strengthen the discussion and possible reasons for the scatter within the manuscript. In case of $\delta^{13}C$ (n-C_{29}), the scatter of TS II – C is caused by the occurrence of succulent plants using the CAM metabolism and are thought to be more enriched in ^{13}C.
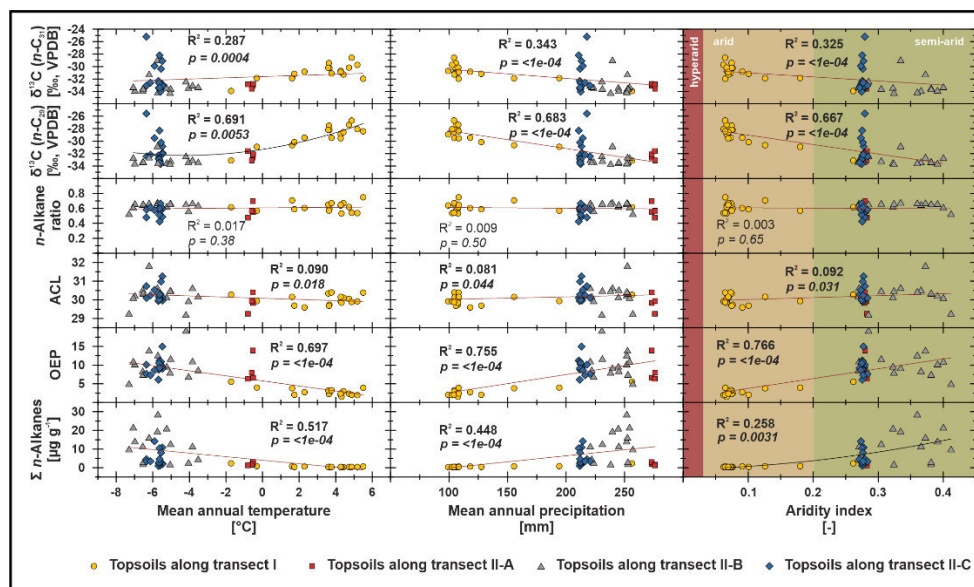
Figure 1. *n*-Alkane concentration, OEP, ACL, *n*-alkane ratio (n-C_{31}/(n-C_{29} + n-C_{31})) and compound-specific $\delta^{13}C$ (n-C_{29} and n-C_{31}) from Mongolian topsoils plotted against climatic parameters (MAP, MAT, AI). Red trend lines illustrate linear regressions, black lines polynomial regressions. Bold values indicate significance (α = 0.05).
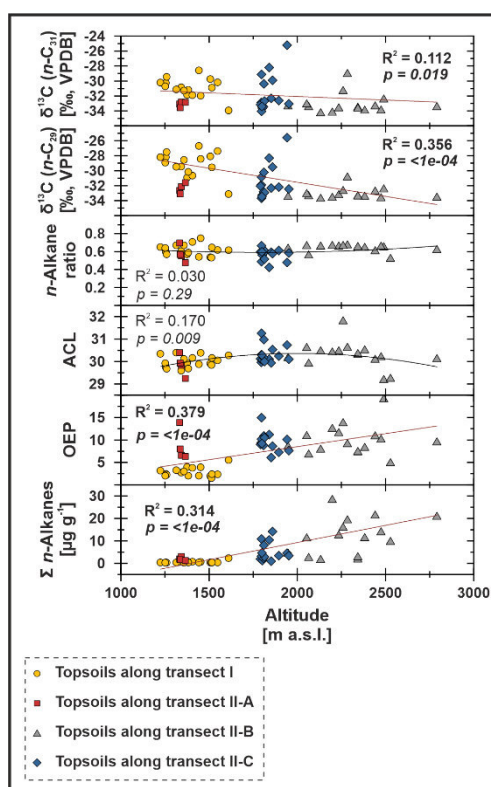

Figure 2. *n*-Alkane concentration, OEP, ACL, *n*-alkane ratio (n-C_{31}/(n-C_{29} + n-C_{31})) and compound-specific $\delta^{13}C$ (n-C_{29} and n-C_{31}) from Mongolian topsoils plotted against altitude (m a.s.l.). Red trend lines illustrate linear regressions, black lines polynomial regressions. Bold values indicate significance (α = 0.05).

Concerning Fig. 4 and Fig. 5 (in the manuscript), we see no real added value in separating the altitudinal transect. At this point, we simply separate the dataset in predominantly arid (transect

I) and predominantly semi-arid (transect II) to show differences between both environments. However, we understand your argumentation and we have divided the dataset for the scatterplots (Fig. 6 and 7) as shown here in figure 1 and 2.

Other minor issues

Line 34 (here and similar issues throughout the manuscript) "an enrichment of leaf wax d13C" d13C values are numbers. Values can't be enriched or depleted. Please re-phrase. 13C-enriched leaf wax perhaps?

➔ We have rephrased those sentences or change it to "^{13}C enriched".

Lines 82-83 "the topsoils were sampled together with the dominant plant species, which comprise the woody shrub Caragana spp. ..." How the dominance of these species was assessed? Is there any previous study concerning species distribution in the area covered by this project? Or is it a subjective assessment?

➔ This is a subjective assessment! Different plants were sampled around the soil sampling sites (~5 m²). Those plants were sampled individually and determined by a botanist at the Institute of Plant and Agricultural Sciences, Mongolian University of Life Sciences, Darchan, Mongolia.

Line 90 "Total lipids ... of plants" What part(s) of plant was(were) extracted? Just leaves or was it together with the stem and roots?

➔ For lipid extraction, just the leaves/needles were used, except for the grasses where we used the entire grass without roots.

Line 107 "were 0.1 per mil the standard deviation" Is there an "and" missing? Also, does it make sense to report d13C values in Suppl. Mat to the second digit after the decimal point, if the reported std. dev. is 0.1 per mil, i.e. no better than the first digit after the decimal point?

➔ This was truly a mistake! The std. dev. for the topsoils and plants was better than 0.66 per mil (0.7) for both compounds. We will change the sentence as follows:
➔ *The ~~average~~ standard deviation for the triplicate measurements were < 0.7‰ and the standard deviation for the alkane standards was better than 0.2‰ (n = 102).*

Lines 110-111 "n-alkane concentrations ... were calculated as the sum of n-C25 and n-C35" Why was n-C23 excluded? It is a major n-alkane in Larix sp.

➔ The sum of n-C_{25} to n-C_{35} typically comprises the n-alkanes within the leaf waxes of higher terrestrial plants. It is true that *Larix* has their dominance in n-C_{25} and n-C_{23}. However, differences between the concentration calculated from n-C_{25} - n-C_{35} and n-C_{23} - n-C_{25} are minor and n-C_{23} and n-C_{25} are not dominant in the respective topsoils, but we will implement this data within the supplements.
➔ We have added a statement within section 2.2.3 "Additionally, we tested the n-alkane concentrations as the sum of n-C_{23} to n-C_{35} because of the high proportions of n-C_{23} in *Larix sp.* However, differences between both n-alkane concentrations are minor thus, we present the n-alkane concentrations including n-C_{23} in the supplementary material."

Lines 118-119 "A normalised n-alkane ratio ... n-C29 and n-C31"; lines 177-178 Please explain the significance of this ratio. If this refers to trees/shrubs vs. grasses, why not to include n-C27 and n-C33, respectively?

➔ We have chosen this normalized n-alkane ratio, because n-C_{29} and n-C_{31} are the most dominant n-alkanes for the grasses as well as for the shrubs *Caragana spp.* and *Artemisia*

spp. For those plants, *n*-C_{27} and *n*-C_{33} are not the dominant chain-lengths and allow no separation in terms of grasses vs. shrubs, which you can see in the ACL that include *n*-C_{27} and *n*-C_{33}.

Line 165 "in line with previous regional studies" Specify what regions were covered by these studies previously? Is it similar to the region covered in this project?

→ Cheung et al. (2015) and Wang et al. (2018c) are located on the Tibetan Plateau, Liu et al. (2018) on the Chinese Loess Plateau and Bliedtner et al. (2018) in the Caucasus region. Concerning the environmental conditions, we think that the studies of Wang et al. (2018c) and Liu et al. (2018) are comparable to our study. Cheung et al. (2015) and Bliedtner et al. (2018) receives with 480 mm/a and up to 1800 mm/a higher amounts of precipitation compared to Mongolia. We will specify the regions of the previous studies in the manuscript.

→ We have changed the sentence to "findings are in line with previous regional studies from the Tibetan Plateau (Cheung et al., 2015; Wang et al., 2018c), the Chinese Loess Plateau (Liu et al., 2018) and the Caucasus region (Bliedtner et al., 2018a), which report *n*-C_{31} being mainly produced by grasses/herbs and *n*-C_{29} by deciduous trees/shrubs."

Line 169 "the findings of Wang et al. (2018b) from China" What part of China? It is a big country with multiple climatic and ecological zones.

→ The investigated transect of Wang et al. (2018b) covers the 400 mm isohyet in China. More specifically, it follows Inner Mongolia towards the Tibetian Plateau. We have changed it to "from China" to "along a transect from north-western to central China".

Line 202-233 Section "The leaf wax signal from plants to topsoils along transect II" This section could be broken down into several paragraphs to make it easier to follow. Also, why not to give a number for this and the next (starting on line 234) subsections? 4.3.1 and 4.3.2 perhaps?

→ We agree with this point and will implement this suggestion.

Lines 223-224 "Compared to the plants, … d13C isotopes of the topsoils are slightly more enriched" What does "d13C isotopes" mean? Please rephrase.

→ The isotopic $\delta^{13}C$ signature of the topsoils from transect II. Will be specified.

Also, I don't see this from the graphs in Fig. 5. The d13C values of n-C29 alkane in soils aren't really that different from those in Caragana and Larix spp. The absence of this "enrichment" is particularly evident when looking at the d13C values of n-C31.

→ Concerning the topsoils of transect II, differences between the topsoils (transect II) and plants reveal no statistical significance. However, the median of both compounds (*n*-C_{29} and *n*-C_{31}) is <u>slightly</u> enriched compared to the plants: For *n*-C_{29} it's up to 1.7‰ for *n*-C_{31} it's up to 1.5‰.

Line 237 "chapter 4.4" I'd call it a section rather than a "chapter".

→ We agree!

Lines 248-249 "are in agreement with climatic control and the fact that higher temperatures reduce the decarboxylation pathway and the formation of n-alkanes (Shepherd and Griffiths 2006)" I don't think this explanation works here. The cited paper evaluates the effect of stress on various factors that control leaf wax biosynthesis within plants. The subject matter of this section is n-alkane content of soils. There are multiple other reasons, in addition to the

biochemical ones within the plant, that could play a role in the distribution of n-alkanes along the transects studied.

➔ You are right, there are many other reasons playing a role, like *n*-alkane degradation, biomass productivity or livestock grazing, which we have discussed in this paragraph. Thus, we will delete this hypothesis to reduce confusion.

Lines 257-258 "In contrast, compound-specific d13C correlated significantly with climatic parameters." There is a lot of noise in the d13C data. Please discuss possible reasons for the scatter. See the MAJOR POINT above.

➔ Is discussed above.

Line 272 "Mongolian plants show" It is a peculiar way of referring to these plants. Using their species names and mentioning that they were sampled in Mongolia would be a better way of describing them.

➔ We agree and we will adapt this!

Lines 276-277 "for reconstructing vegetation changes in Larix sp." Sounds awkward. Please re-phrase.

➔ We will rephrase it as follows:
➔ *'However, Larix sp. produces only few amounts on n-alkanes and their dominance of mid-chain n-alkanes are not distinct in the respective topsoils. Thus, n-alkanes are not useful to reconstruct changes in the abundance of Larix sp.'.*

FIGURES

Figure 1 Please remind the reader what SRTM DEM stands for so that there is no need to look for this information in the text. Also, make the font and the arrows showing the location of Transect I and Transect II thicker to make them standout more.

➔ We have changed it to SRTM digital elevation model.
➔ Fonts and arrow thickness will be adapted

Figure 2 Make the bar representing the scale (0-500) less prominent. That's one of the first things that draws reader's attention when you look at the map. Instead, highlight A, B, C better, so that the title of each map is not hidden among all the other text on the maps.

➔ We agree with this point and will change it accordingly.

Figure 3 Please specify whether the n-alkane data shown in the bar graphs represents all the plants collected along the Transects I and II or only a subset. Figure 4 Remind the reader what kind of "n-alkane ratio" is plotted here.

➔ All the plants were sampled along transect II. We will highlight this information within the figures caption.
➔ We will add the equation of the n-alkane ratio within the figure.

Figure 5 Specify that "compound-specific" refers to n-C29 and n-C31 alkanes. / Figure 6 Which homologues were included in the calculation of n-alkanes concentrations? What ratio of n-alkanes are the authors referring to?

➔ We have added this information within the figure caption.

Leaf wax *n*-alkane patterns and compound-specific $\delta^{13}\text{C}$ of plants and topsoils from semi-arid/aridsemi-arid Mongolia

Julian Struck[1], Marcel Bliedtner[1,2], Paul Strobel[1], Jens Schumacher[3], Enkhtuya Bazarradnaa[4], and Roland Zech[1]

[1]Institute of Geography, Friedrich-Schiller-University Jena, Löbdergraben 32, 07743 Jena, Germany
[2]Institute of Geography and Oeschger Centre for Climate Change Research, University of Bern, Hallerstrasse 12, 3012 Bern, Switzerland
[3]Institute of Mathematics, Friedrich-Schiller-University Jena, Ernst-Abbe-Platz 2, 07743 Jena, Germany
[4]Institute of Plant and Agricultural Sciences, Mongolian University of Life Sciences, Mongolia

Correspondence: Julian Struck (julian.struck@uni-jena.de)

Abstract. Leaf wax *n*-alkanen-Alkane patterns and their compound-specific $\delta^{13}\text{C}$ signatures are valuable proxies for paleoenvironmental reconstructions. So far, their potential has not been investigated in semi-arid to arid regions of Mongolia. We have therefore analysed the leaf wax *n*-alkanes and their compound-specific $\delta^{13}\text{C}$ signature of five plant species (*Poaceae, Cyperaceae, Artemisia spp., Caragana spp.* and *Larix sp.*) , and topsoils ($0 - 5\,\text{cm}$) along two transects in central and southern
5 Mongolia.

Grasses showdepict a distinct dominance of the *n*-C_{31} homologue, whereas the shrubs Caragana spp. and Artemisia spp. are dominated by *n*-C_{29}. Larix sp. is characterized by the mid-chain *n*-alkanes *n*-C_{23} and *n*-C_{25}. From plant to topsoilsoil, *n*-alkane homologue pattern show the potential to differentiate between grass covered sites from those covered by Caragana spp. *n*-Alkane concentrations and odd-over-even predominance (OEP)OEP values of the topsoils are distinctly influenced
10 by mean annual temperature, mean annual precipitation and aridity, likely reflecting the degree of *n*-alkane degradation and biomass production. In contrast, the *n*-alkane average chain-length ACL and the *n*-alkane ratio (*n*-C_{31}/*n*-C_{29}+ *n*-C_{31}) are not affected by climatic parameters and thus, not biased by climate. The compound-specific $\delta^{13}\text{C}$ signatures are strongly correlated to climate, showing a significant enrichment with increasing aridity, indicating the effect of water use efficiency. Our calibration results suggest that long-chain *n*-alkanes and their compound-specific $\delta^{13}\text{C}$ signatures have great potential to
15 reconstruct paleoenvironmental and -climatic conditions when used in sediment archives from Mongolia.

1   Introduction

Leaf wax biomarkers such as long-chain *n*-alkanes (*n*-C_{25} - *n*-C_{35}) are produced in the plant cuticle as a protection layer against environmental stress and are synthesized by the polyketide biosynthetic pathway resulting in a distinct odd-over-even predominance (OEP) (Eglinton and Hamilton, 1967; Shepherd and Wynne Griffiths, 2006). Due to their water insolubility,
20 chemical inertness and relative resistance against biochemical degradation, leaf wax *n*-alkanes stay well preserved in sediment archives over geological timescales and serve as valuable biomarkers for former environmental conditions (Eglinton and Eglin-

ton, 2008). During the last decades, leaf wax *n*-alkanes have increasingly been used for paleoenvironmental reconstructions in lake sediments (Aichner et al., 2017; Rach et al., 2017; Schwark et al., 2002; Sun et al., 2016), marine sediments (Castañeda et al., 2009; Rommerskirchen et al., 2006; Schefuss et al., 2005), loess-paleosol sequences (Häggi et al., 2019; Schäfer et al., 2018; Zech et al., 2013) and fluvial sediment-paleosol sequences (Bliedtner et al., 2018b).

The relative homologue distribution of leaf wax *n*-alkanes has been used as a chemotaxonomic marker to differentiate among vegetation forms and thus reconstruct paleovegetation: the *n*-alkanes n-C_{27} and n-C_{29} are thought to be mainly produced by deciduous trees/shrubs, whereas n-C_{31} and n-C_{33} are mainly produced by grasses/herbs (Bliedtner et al., 2018a; Schäfer et al., 2016; Vogts et al., 2009). The compound-specific $\delta^{13}C$ signatureisotopes of leaf wax *n*-alkanes have also been used for reconstructing changes in the vegetation composition of C_3 (-20 to $-35‰$) and C_4 (-10 to $-14‰$) plants (Castañeda et al., 2009; Lane, 2017; Rao et al., 2016; Rommerskirchen et al., 2006), and give additional paleoclimatic information about drought stress and arid conditions for C_3 plants (Aichner et al., 2010a, b; Schäfer et al., 2018). Several studies have shown a strong correlation between the $\delta^{13}C$ leaf wax signal of C_3 plants and water use efficiency (WUE) that is influenced by precipitation, temperature and evapotranspiration and describes the stomata conductance of a plant to avoid water loss (Diefendorf and Freimuth, 2017; Farquhar et al., 1982; Rao et al., 2017). Along a 400 mm isohyet in China, Wang et al. (2018b) have been shown that the compound-specific $\delta^{13}C$ signature of leaf wax *n*-alkanes are strongly correlated to temperature, with strongest correlations observed for the average temperature of June, July and August (Wang et al., 2018b). Thus, warmer/dryer conditions cause an increase of WUE resulting in an ^{13}C enrichment of leaf wax $\delta^{13}C$ and cooler/wetter conditions *vice versa* (Aichner et al., 2015; Castañeda et al., 2009; Diefendorf and Freimuth, 2017; Wang et al., 2018b).

Although leaf wax *n*-alkane patternshomologue distributions and compound-specific $\delta^{13}C$ isotopes have been increasingly used in sediment archives for paleoenvironmental reconstructions within the last decadesyears, they need to be calibrated regionally on recent reference material before any paleoenvironmental reconstruction can be made. The need for regional calibrations has been emphasized by the fact that Bush and McInerney (2013) questioned whether leaf wax *n*-alkane homologue patterns can discriminate between modern vegetation forms on a global scale while several regional studies found them discriminating on a regional scale. Although the most abundant homologues differ from region to region, a good discrimination power has been reported from Europe (Schäfer et al., 2016; Zech et al., 2009, 2010), the Caucasus region (Bliedtner et al., 2018a), North- and South America (Diefendorf et al., 2015; Feakins et al., 2016; Lane, 2017) and the African rain forest and savanna (Vogts et al., 2009).

However, when interpreting leaf wax *n*-alkanes and their compound-specific $\delta^{13}C$ signatureisotopes several potential pitfalls such as species-specific and intra-leaf variations (Diefendorf et al., 2011; Gao et al., 2015), the influence of environmental and climatic factors (Carr et al., 2014; Diefendorf and Freimuth, 2017; Farquhar et al., 1982; Hoffmann et al., 2013; Rao et al., 2017; Tipple et al., 2013), the dependency on altitude (Feakins et al., 2018; Hultine and Marshall, 2000), and *n*-alkane degradation (Brittingham et al., 2017; Buggle et al., 2010; Li et al., 2018a) have to be considered and accounted for. So far, such regional calibration studies on recent leaf wax *n*-alkane patterns, andhomologue pattern compound-specific $\delta^{13}C$ isotopes and as well as potential climatic biases onof the vegetational/climatic leaf wax signal are scarce for semi-arid regions and do not exist for Mongolia.

Thus, this~~This~~ study investigates leaf wax *n*-alkane ~~homologue~~ patterns and compound-specific δ^{13}C ~~isotopes~~ of modern plants and topsoils from semi-arid/arid~~semi-arid~~ Mongolia to evaluate their potential for regional paleovegetation and -climate reconstructions. More specifically, we tested the chemotaxonomic potential of leaf wax *n*-alkane~~homologue~~ patterns from

60 five dominant plant species and whether their homologue distribution can be used to discriminate between woody shrubs and grasses/herbs on a regional scale. Moreover, we investigate differences in the compound-specific δ^{13}C signature of leaf waxes from plants and topsoils, and how the plant signal is incorporated into the topsoil. Since only the topsoils represent an averaged decadal leaf wax signal, we check for potential environmental/climatic influences by correlating the leaf wax *n*-alkane patterns~~homologue pattern~~ and δ^{13}C with altitude, mean annual temperature (MAT), mean annual precipitation (MAP) and the

65 aridity index (AI). While we test that the *n*-alkane patterns~~homologue pattern~~ are not biased by climatic influences, we test the potential of leaf wax δ^{13}C to reflect on MAT, MAP and aridity ~~, and thus climate~~. Additionally, we test for a potential bias from altitude on leaf wax *n*-alkane patterns and the δ^{13}C signal. Therefore, our calibration results will be a base for future robust paleovegetational and -climate reconstructions in semi-arid/arid~~semi-arid~~ Mongolia using leaf wax *n*-alkanes from regional sediment archives. Such lacustrine, loess-paleosol and colluvial sediment archives have been~~were~~ increasingly investigated in

70 semi-arid/arid~~semi-arid~~ Mongolia and could potentially be used for future leaf wax based paleoenvironmental reconstructions (Klinge et al., 2017; Peck et al., 2002; Prokopenko et al., 2007; Rudaya and Li, 2013; Wang et al., 2011).

2 Material and methods

2.1 Geographical setting and sampling

Semi-arid/arid~~semi-arid~~ Mongolia is a highly continental region characterized by harsh/long winters and hot/short summers

75 (Dashkhuu et al., 2015). Mongolia is located at the interface of three major atmospheric circulation systems controlling the regional climate (Fig. 1). The summer climate is dominated by the East Asian Summer Monsoon (EASM) and the Westerlies that provide most of the yearly moisture and precipitation during the summer month, i.e. 75% of the annual precipitation falls in June, July and August (Rao et al., 2015; Wang and Feng, 2013). The dry and cold winter climate is dominated by the Siberian high that mostly blocks the moisture supply from the Westerlies during winter (Peck et al., 2002). The Mongolian

80 climate has a north-south gradient in MAT and MAP, with increasing MAT from north to south and MAP *vice versa* (Harris et al., 2014, Fig. 2A, B). This north-south gradient in temperature and precipitation is further reflected by the AI (Fig. 2C) and in the distribution of regional vegetation biomes with taiga and mountain/forest steppe in nN~~n~~orthern and cC~~c~~entral Mongolia and steppe/desert steppe in sS~~s~~outhern Mongolia (Hilbig, 1995; Klinge and Sauer, 2019).

Topsoils $(0-5\,\mathrm{cm})$ were sampled~~For this study, we sampled topsoils $(0-5\,\mathrm{cm})$~~ along a north-south transect (transect I, see

85 Fig. 1 for location) in June 2016, and~~as well as~~ plants and topsoils $(0-5\,\mathrm{cm})$ along an east-west transect (transect II, see Fig. 1 for location) in July/August 2017. Along transect II, the topsoils were sampled together with the dominant plant species $(5\,m^2$ around the topsoil sampling site), which comprise the woody shrub *Caragana spp.*, the grasses/herbs *Poaceae, Cyperaceae* and *Artemisia spp.*, and *Larix sp.* as coniferous tree ~~Larix sp.~~. *Artemisia spp.* summarizes different herbaceous species and perennial 'shrubby' species with a woody base (e.g. *Artemisia frigida*). Transect I covers an altitudinal range between 1224 and 1611

90 m a.s.l., ~~a~~ MAT and MAP range from -1.7 to $5.5\,°C$ and 99 to $276\,mm/a$~~, whereas~~ Transect II covers ~~a~~ an altitudinal range between 1333 and 2792 m a.s.l., and a range from -7.3 to $-0.5\,°C$ and 210.8 to $276.2\,mm/a$ in MAT and MAP, respectively and is separated in the Ugii Nuur catchment (TS II-A), an altitudinal transect (TS II-B) and the Telmen Nuur catchment (TS II-C) (Fig. 2; Fick and Hijmans, 2017).

2.2 Leaf wax analysis

2.2.1 Leaf wax extraction and ~~quantification~~chromatography

Total lipids of the topsoils ($\sim 35\,g$) from transect I were extracted at the University of Bern, Switzerland, using accelerated solvent extraction (Dionex ASE 200: $6.9\,MPa, 100\,°C$) with $60\,ml$ dichloromethane (DCM):methanol (MeOH) (9:1, v/v) over three extraction cycles as described by Bliedtner et al. (2018b) ~~Schäfer et al. (2016)~~. Total lipids of topsoils ($\sim 10\,g$) and plant leaves ($\sim 1\,g$) from transect II were extracted at the Friedrich-Schiller-University of Jena, Germany, using ultrasonic extraction
100 with $20\,ml$ DCM:MeOH (9:1, v/v) over three cycles as described by Bliedtner et al. (2018a).

The total lipid extract from both transects was separated over aminopropyl pipette columns (Supelco, $45\,\mu m$) into (i) an apolar fraction including the n-alkanes, (ii) a more polar fraction and (iii) an acid fraction. ~~Subsequently, n-Alkanes~~n-alkanes were eluted with $\sim 4\,ml$ hexane and additionally cleaned over coupled silver-nitrate ($AgNO_3$) – zeolite pipette columns. n-Alkanes were subsequently dissolved in hydrofluoric acid and liquid–liquid recovered with ~~n-~~hexane. Identification and quantification
105 of the n-alkanes was performed on an Agilent 7890B gas chromatograph equipped with an Agilent HP5MS column ($30\,m \times 320\,\mu m \times 0.25\,\mu m$ film thickness) and a flame ionization detector (GC-FID). For identification and quantification, external n-alkane standards with a known concentration (n-alkane mix n-$C_{21} - n$-C_{40}, Supelco) were run with each sequence.

2.2.2 Compound-specific $\delta^{13}C$ analysis

Compound-specific carbon isotopes were measured for the most abundant n-alkanes n-C_{29} and n-C_{31}. Isotope measurements
110 were performed on an Isoprime Vision isotope ratio mass spectrometer coupled to a gas chromatograph (Agilent 7890B GC) equipped with an Agilent HP5GC column ($30\,m \times 320\,\mu m \times 0.25\,\mu m$ film thickness) via a GC5 pyrolysis/combustion interface. The GC5 was operating in combustion mode with a CuO reactor at $850\,°C$. Samples were injected in splitless mode and measured in triplicates. n-Alkane standards (n-C_{27}, n-C_{29} and n-C_{33}) with known isotopic composition (Schimmelmann standard, Indiana) were measured as duplicates after every third triplicate. The ~~average~~ standard deviation for the triplicate
115 measurements were $< 0.7‰$, and the standard deviation for the standards was ~~better than~~ $< 0.2‰$ ($n = 102$). Carbon isotopic composition is given in the delta notation ($\delta^{13}C$) versus the Vienna Pee Dee Belemnite standard (VPDB).

2.2.3 Data analysis

n-Alkane concentrations ($\sum n$-Alkane) are given in $\mu g\,g^{-1}$ dry weight and were calculated as the sum of n-C_{25} to n-C_{35}. Additionally, we tested the n-alkane concentrations as the sum of n-C_{23} to n-C_{35} because of the high proportions of n-C_{23} in
120 *Larix sp.* However, differences between both n-alkane concentrations are minor and thus, we present the n-alkane concentra-

tions including n-C$_{23}$ in the supplementary material. The OEP was calculated according to Hoefs et al. (2002) and serves as a proxy for degradation with values below five indicating enhanced n-alkane degradation (Buggle et al., 2010; Zech et al., 2009, 2010).

$$\text{OEP} = \frac{n\text{-C}_{27} + n\text{-C}_{29} + n\text{-C}_{31} + n\text{-C}_{33}}{n\text{-C}_{26} + n\text{-C}_{28} + n\text{-C}_{30} + n\text{-C}_{32}} \tag{1}$$

125   The average chain length (ACL) was determined after Poynter et al. (1989) and is ~~commonly~~ used to distinguish between leaf waxes predominantly produced by deciduous trees and shrubs (n-C$_{27}$ and n-C$_{29}$) and grasses and herbs (n-C$_{31}$ and n-C$_{33}$) (Bliedtner et al., 2018a).

$$\text{ACL} = \frac{27 \cdot n\text{-C}_{27} + 29 \cdot n\text{-C}_{29} + 31 \cdot n\text{-C}_{31} + 33 \cdot n\text{-C}_{33}}{n\text{-C}_{27} + n\text{-C}_{29} + n\text{-C}_{31} + n\text{-C}_{33}} \tag{2}$$

A normalized n-alkane ratio was calculated for the most abundant n-alkanes n-C$_{29}$ and n-C$_{31}$.

130    $$n\text{-Alkane ratio} = \frac{n\text{-C}_{31}}{(n\text{-C}_{29} + n\text{-C}_{31})} \tag{3}$$

2.3   Statistical analysis

Differences in n-alkane concentration, OEP, ACL, n-alkane ratio and compound-specific δ^{13}C among plant species and between topsoils and plants were analysed using analysis of variance (ANOVA) followed by pairwise comparisons based on Tukey's honestly significant difference. Since the relative homologue distribution of the n-alkanes is important for the discrimination

135   between plant species, the n-alkane ~~homologue~~ patterns were analysed as compositional data according to Aitchison (2003). Correlations of n-alkane concentration, OEP, ACL, n-alkane ratio and compound-specific δ^{13}C with climatic~~environmental~~ parameters and altitude were tested using weighted linear or polynomial regression. The environmental parameters MAT and MAP were derived from the WorldClim 2.0 dataset (1970-2000, 30s resolution, Fick and Hijmans, 2017) and the AI from the Global Aridity Index and Potential Evapo-Transpiration (ET0) Climate Database v2 (1970-2000, 30s resolution, Trabucco and

140   Zomer, 2019). The altitude was extracted from the Shuttle Radar Topography Mission (SRTM) data. Regressions were tested only for the topsoils because they represent an averaged leaf wax signal over some decades (Angst et al., 2016) and can thus be correlated with the WorldClim and the Global Aridity Index data that represents averaged climate data over 30 years. In contrast, plants only reflect an annual signal of the sampling year 2017 and cannot be correlated with the available climatic parameters from the WorldClim and the Global Aridity Index dataset, and also annual climate data for the sampling year 2017

145   are not available. Model selection was based on hierarchical comparison of models with increasing polynomial order using F ratios. Goodness of fit of the final models was assessed using weighted R^2 values. All statistical analyses were done using the statistical software system R (R Core Team, 2019), and the package compositions (Boogaart, 2013) for compositional data analysis.

3 Results

3.1 *n*-Alkane patterns in plants and topsoils

Leaf wax *n*-alkanes are present in all analysed plants and topsoils and show a distinct OEP (Fig. 3). For the analysed plants from transect II, the most abundant *n*-alkane homologues vary among the plant species: *Poaceae* and *Cyperaceae* tend to be dominated by *n*-C_{31}, and *Artemisia spp.* and *Caragana spp.* by *n*-C_{29}. *Larix sp.* has its dominance on the mid-chain *n*-alkane *n*-C_{25}. The topsoils from both transects are mostly dominated by *n*-C_{31} (Fig. 3). Figure 4 illustrates differences in *n*-alkane concentrations, OEP, ACL and the *n*-alkane ratio of the analysed plant species and topsoils, and table 1 shows the corresponding level of significance. *n*-Alkane concentrations are significantly higher in plants than topsoils (p $= 5.1e^{-07}$). Plants range from 9 to 2508 µg g^{-1}, with *Caragana spp.* having significantly the highest concentrations, and *Larix sp.* having the lowest concentrations (Fig. 4A, Tab. 1). *n*-Alkane concentrations in topsoils range from 0.2 to 59 µg g^{-1} with transect I having much lower concentrations than transect II (Fig. 4A). Plants show a wide OEP with values ranging between 4 and 39. Highest OEP values are observed for *Caragana spp.*, followed by *Poaceae, Cyperaceae, Artemisia spp.* and *Larix sp.* Topsoils have generally lower OEP values than plants ranging from 1.5 up to 5.5 for transect I and from 4.8 to 19 for transect II, respectively (Fig. 4B). ACL values of plants range from 28.3 to 30.8 with *Larix sp.* showing significantly lower ACLs compared to the other plants (Tab. 1). ACL values for topsoils are higher but in the same range as most plant species, ranging from 29.6 to 30.4 along transect I and between 29.2 and 31.8 along transect II (Fig. 4C). The *n*-alkane ratio show values between 0.16 and 0.8. *Poaceae* and *Cyperaceae* tend to have the highest values, *Caragana spp.* and *Larix sp.* the lowest. The topsoils range from 0.42 to 0.75 for both transects (Fig. 4D).

3.2 Compound-specific δ^{13}C

Compound-specific δ^{13}C ~~isotopes~~ were measured for the most abundant *n*-alkanes *n*-C_{29} and *n*-C_{31} (Fig. 5). Plants show ~~consistent~~ δ^{13}C values between -36‰ to -29.5‰ for $\delta^{13}C_{29}$, and between -35.8‰ and -30.3‰ for $\delta^{13}C_{31}$, with *Larix sp.* being ~~having the~~ most enriched in leaf wax ^{13}C ~~values~~ among all plant~~s~~ species. In comparison to plants, topsoils tend to be more enriched in ^{13}C with a larger scatter and range ~~and scattered, ranging~~ from -33.8‰ to -25.6‰ for $\delta^{13}C_{29}$, and from -34.3‰ to -25.2‰ for $\delta^{13}C_{31}$ with a distinct enrichment in ~~δ^{13}C~~ along transect I (Fig. 5). Compound-specific δ^{13}C ~~isotopes~~ of *n*-C_{29} and *n*-C_{31} differ significantly between topsoils from transect I and transect II, but not significantly between plant species and between plant species and topsoils from transect II (Tab. 1).

4 Discussion

4.1 *n*-Alkane patterns in plants

The plant species from Mongolia show distinct differences in their relative *n*-alkane homologue pattern (Fig. 3). The grasses *Poaceae* and *Cyperaceae* are dominated by *n*-C_{31}, whereas the woody shrub *Caragana spp.* is dominated by *n*-C_{29}. Those

findings are in line with previous regional studies from the Tibetan Plateau (Cheung et al., 2015; Wang et al., 2018c), the Chinese Loess Plateau (Liu et al., 2018) and the Caucasus region (Bliedtner et al., 2018a), which report n-C_{31} being mainly produced by grasses/herbs and n-C_{29} by deciduous trees/shrubs~~deciduous trees and shrubs~~. Although *Artemisia spp.* could be expected to be herbaceous with a dominance in n-C_{31}, our results show a distribution maximum at n-C_{29}, i.e. it is more similar to the woody shrub *Caragana spp.* than to the grass species. This does not necessarily be contradictory and corroborates the findings of Wang et al. (2018b) ~~from~~ along a 400 mm isohyet transect from north-western to central China who reports that *Artemisia* can grow both as herbaceous and as a woody shrub (e.g. *Artemisia frigida*). In contrast to the other plant species, the coniferous tree *Larix sp.* is dominated by the mid-chain n-alkanes n-C_{23} and n-C_{25}, resulting in significant lower ACL~~s~~ (Fig. 4C, Tab. 1). However, statistically significant differences between the ACL of the other plant species are not evident, although the relative homologue pattern reveal differences among them (Tab. 1, Fig. 3 and 4C). The ACL of the grasses/herbs *Poaceae* and *Cyperaceae* and *Artemisia spp.* and the woody shrub *Caragana spp.* have only slight differences and a small range between 29.6 and 29.9. This is due to a strong ACL scattering of the grasses/herbs that overlap the ACL of the woody shrubs (Fig. 4C). Thus, a clear chemotaxonomic discrimination between grasses/herbs and woody shrubs is not given by the ACL for the investigated modern plants from Mongolia. A better chemotaxonomic discrimination is provided by the n-alkane ratio n-$C_{31}/(n$-$C_{29} + n$-$C_{31})$ that is based on the most abundant homologues n-C_{29} and n-C_{31}. The n-alkane ratio significantly separates the grasses *Poaceae* and *Cyperaceae* from the woody shrub *Caragana spp.* and the coniferous tree *Larix sp.* (Fig. 4D). The n-alkane ratio of *Artemisia spp.* lies in-between the woody shrubs and grasses, most likely because of their ability to grow as both herbaceous and woody shrubs (Fig. 4D). This is further expressed statistically as the n-alkane ratio of *Artemisia spp.* is equal to those calculated from both *Caragana spp.* and the grass species (Tab. 1). Beside *Artemisia spp.*, the n-alkane ratio has the chemotaxonomic potential to discriminate significantly between *Larix sp.* and grasses as well as between *Caragana spp.* and grasses. However, the n-alkane ~~homologue~~ pattern from *Larix sp.* with their mid-chain dominance has to be interpreted with caution when comparing species-specific differences to the long-chain dominated plant species, that is mostly due to the fact that *Larix sp.* only produce small amounts of n-alkanes. When incorporated into the soil, the coniferous n-alkane signal from *Larix sp.* should become overproportional overprinted by the undergrowth of the grasses/herbs (Diefendorf et al., 2011; Schäfer et al., 2016).

4.2   Compound-specific δ^{13}C of plants

The compound-specific δ^{13}C values of the n-alkanes from our investigated plants from transect II show consistent δ^{13}C values among the plant species, except *Larix sp.*, and are in a typical range of C_3 plants (Fig. 5; Tipple and Pagani, 2007). Although Pyankov et al. (2000) have reported C_4 plants in Mongolia among 16 plant families including *Poaceae*, those are not evident along our sampled plant transect. While some C_4 plants have been found in the Khangai Mountains, their distribution is mainly limited to the semiarid-steppe and semi-desert areas in southern Mongolia and the Gobi desert, i.e. beyond our plant sampling sites (Pyankov et al., 2000; Su et al., 2011). Statistically significant differences did not exist between the most abundant homologues n-C_{29} and n-C_{31} ($p = 1$), indicating that no different fractionation has occured during biosynthesis (Wang et al., 2018b). Consistent δ^{13}C values between the most abundant homologues are in good agreement with compound-specific δ^{13}C

analyses of~~on~~ three *Artemisia* species (*Artemisia argyi, capilares* and *scoparia*) along a $400\,\text{mm}$ isohyet in China (Wang et al., 2018b). While no differences are found between the $\delta^{13}C$ values of the grasses/herbs and woody shrubs, only *Larix sp.* is enriched up to 2‰, but still in the range C_3 plants (Fig. 5, Tab. 1). Such an enrichment of coniferous trees compared to other plants might be explained by differences in species-specific fractionation (Diefendorf et al., 2015).

4.3 Comparing *n*-alkane pattern~~s~~ and compound-specific $\delta^{13}C$ of plants versus topsoils

4.3.1 The leaf wax signal from plants to topsoils along transect II

Along transect II, modern plants have higher *n*-alkane concentration than the topsoils, with *Artemisia spp.* and *Caragana spp.* having significantly higher *n*-alkane concentrations than the respective topsoils (Fig. 4A, Tab. 1). Thus, ~~the~~ lower *n*-alkane concentration in the topsoils indicate that *n*-alkanes become diluted during the incorporation from plant biomass into the topsoil (Fig. 4A). Likewise, the OEP~~odd-over-even predominance~~ decreases from plants to topsoil and indicate enhanced organic matter degradation (Buggle et al., 2010; Schäfer et al., 2016) and microbial alteration (Schulz et al., 2012) (Fig. 4B). Despite possible degradation effects during soil developement, the topsoils show distinct OEP values between 4.8 and 19, still indicating a good preservation (Zech et al., 2009). Along transect II, one exception ~~One exception along transect II~~ in terms of higher *n*-alkane concentration and OEP is TSC10 Ah1, showing $59\,\mu\text{g}\,\text{g}^{-1}$ and 19, respectively (Fig. 1, sampling site 25). Site TSC10 is characterized by stagnating soil conditions with a distinct organic rich topsoil, limiting organic matter degradation and microbial alteration of *n*-alkanes (Hoefs et al., 2002). Thus, TSC10 Ah1 remains exceptional and not comparable to the other topsoils from transects II. Overall, decreasing concentrations and OEP values from plants to topsoils are in good agreement with other regional studies (Bliedtner et al., 2018a; Howard et al., 2018; Li et al., 2018b; Schäfer et al., 2016; Zech et al., 2009).

For the topsoils, n-C_{31} is on average the most abundant *n*-alkane homologue, indicating a typical *n*-alkane pattern produced by grasses (Bliedtner et al., 2018a, Fig. 3). The only exceptions are sites covered with *Caragana spp.* ($n = 8$) where higher amounts of n-C_{29} are evident within the respective topsoils, and the two *Caragana* covered topsoils TLC4 Ah1 and TLC6 Ah1 (Fig. 1, sampling sites 40 and 42) even show a dominance of n-C_{29} (Suppl. Mat.). Thus, the dominant n-C_{29} signal produced by the woody shrubs is also reflected in the respective topsoils. This is further expressed by lower ACLs and *n*-alkane ratios for those topsoils, which explains the scattering towards n-C_{29} in ACL and 0.4 for the *n*-alkane ratio, respectively (Fig. 4C, D). At sites covered with *Larix sp.*, the mid-chain length dominance of *Larix sp.* is not reflected in the respective topsoils, which are mainly dominated by n-C_{31} *n*-alkanes. Thus, *n*-alkanes from *Larix sp.* must become strongly diluted from plant to topsoil and the topsoils reflect mostly the *n*-alkanes from the grassy undergrowth like previously shown by Schäfer et al. (2016) for several coniferous sites including *Larix, Picea, Abies and Pinus*. Compared to the plants, ~~compound-specific $\delta^{13}C$ isotopes~~ the isotopic signature of the topsoils is~~are~~ slightly more enriched but reveal no statistical significance (Fig. 5, Tab. 1), which is in line with previous studies and might reflect an enrichment by diagenesis from litter to topsoil or a change in vegetation composition (Wu et al., 2019, and references therein).

Environmental information of the plants compound-specific $\delta^{13}C$ signal only reflects one vegetation period, whereas the topsoils compound-specific $\delta^{13}C$ signal reflects environmental variability on decadal timescales, which might explain the topsoil

δ^{13}C enrichment. However, one topsoil (TLC4 Ah1) shows a strong ^{13}C enrichment~~enrichmentstrongly enriched δ^{13}C values~~ up to $\sim -25‰$. Such an enrichment might be explained by n-alkane contributions from succulent plants, which tend to be~~have~~ more enriched in δ^{13}C ~~values~~ within the range of C_3 plants (Boom et al., 2014). Succulents were growing on stone rich, thin topsoils in the catchment of Lake Telmen (Fig. 1, TS II-C). For comparison, we sampled the succulent *Orostachys malaco-* *phylla* from the Telmen catchment and analysed their compound-specific δ^{13}C isotopes that yield $-24.7‰$ for n-C_{29} ($n = 1$) and $-25.03‰$ for n-C_{31} ($n = 1$). Thus, increased inputs of succulent δ^{13}C might best~~be able to~~ explain the ~~more~~ enriched isotopes~~values~~ in the Telmen catchment and the extreme value of $\sim -25‰$ from site TLC4.

4.3.2 The leaf wax signal of the topsoils along both transects

The topsoils of both Mongolian transects show distinct differences in n-alkane concentration and OEP, which are higher along transect II and decrease along transect I. This is mostly due to the fact that n-alkane production and degradation is influenced by the climatic gradient along transect I (see section~~chapter~~ 4.4 and Fig. 2 for more detailed discussion). Beside some n-C_{29} dominated sites with *Caragana spp.*, the ACL and the n-alkane ratio show the dominance of n-C_{31} which indicate the n-alkane ~~n-alkanes which indicate their~~ origin from the grasses *Poaceae* and *Cyperacea* (Fig. 4, Bliedtner et al., 2018a; Schäfer et al., 2016; Vogts et al., 2009; Zech et al., 2010). This is further expressed by the results of ANOVA, because the ACL and n-alkane ratios from topsoils and grasses are not statistically different (Tab. 1).

4.4 Climatic influences on topsoil n-alkane patterns and compound-specific δ^{13}C

To test potential climatic influences on our n-alkane proxies, we correlate them with MAT, MAP and AI (Fig. 6). The n-alkane patterns show that n-alkane concentrations in topsoils and their preservation (OEP) are correlated to climatic parameters, but also with altitude, indicating higher n-alkane concentrations and a better n-alkane preservation above 2000 m a.s.l. (Fig. 7, transect II-B). Since altitude generally controls MAT ($R^2 = 0.624$) and MAP ($R^2 = 0.395$), we suggest that variations along the investigated transects are primarily climate induced. Thus, we detected MAT ($R^2 = 0.517, p =< 1e-04$) as main climatic control parameter on n-alkane concentrations for the topsoils from both transects. Correlations of n-alkane concentration with MAP ($R^2 = 0.448, p =< 1e-04$) and AI ($R^2 = 0.258, p = 0.0031$) are likewise significant indicating a clear correlation (Fig. 6). ~~The negative correlation with MAT and positive correlations with MAP and AI are in agreement with climatic control and the fact that higher temperatures reduce the decarbonylation pathway and the formation of n-alkanes Shepherd.2006.~~ We ~~Thus, we~~ conclude that lower n-alkane concentrations probably indicate reduced biomass production and enhanced n-alkane degradation in the topsoils. The former even intensifies when combined with livestock grazing (Kölbl et al., 2011, and references therein), especially along transect I (sampling sites $1 - 17$) where biomass production is reduced and overgrazing occur extensively (Fig. 6). In contrast, sites along transect II-B (above 2000 m a.s.l.) describe a favourable area for plant growth and thus an increase in n-alkane concentrations and OEP (Fig. 7). However, especially transect II-B and II-C are characterized by a distinct scatter, which is mainly the result of variations in plant physiology and site-specific/micro-climatic characteristics which are not covered by the reanalysis data.

Previous studies have shown correlations between the production of the most abundant homologues with~~and~~ climatic parameters and altitude, i.e. common vegetation proxies such as the ACL and n-alkane ratio could~~should~~ reflect changes in MAT since plants tend to produce longer n-alkanes as a protection against water loss (Bush and McInerney, 2013; Feakins et al., 2016; Sachse et al., 2006; Tipple et al., 2013; Wang et al., 2018b, a). However, this~~This~~ relation could not be observed by the ~~on~~ n-alkanes ~~extracted~~ from Mongolian topsoils, since the ACL and the n-alkane ratio as common vegetation proxies show no correlations with MAT, MAP, AI or altitude (Fig. 6, 7).

In contrast, compound-specific δ^{13}C of the topsoils correlate significantly with climatic parameters. Our results show an enrichment in δ^{13}C with increasing temperature, aridity and decreasing precipitation (Fig. 6). This climate induced enrichment in ~~compound-specific δ~~ ^{13}C follows mainly the north-south gradient in decreasing MAP and increasing MAT along transect I from central Mongolia into the Gobi Desert. The only exceptions are the extreme values near Lake Telmen, which are mostly due to the input of the ^{13}C enriched succulent *Orostachys malacophylla* with ~~^{13}C enriched leaf waxes which explain the overall scatter in δ^{13}C along transect II-C.~~ As already proposed by Diefendorf et al. (2010), MAP is an intense predictor on δ^{13}C (n-C$_{29}$: $R^2 = 0.683, p = < 1e-04$, n-C$_{31}$: $R^2 = 0.343, p = < 1e-04$), which is further expressed in distinct linear correlations with the AI (Fig. 6). However, δ^{13}C correlates also with altitude, i.e. leaf wax ^{13}C shows significantly depletion with increasing altitude. Previous studies have shown a strong positive correlations between δ^{13}C and altitude, indicating enhanced ^{13}C enrichment due to an altitude effect (Feakins et al., 2018) and/or environmental/climatic plant physiological adaptions like a decrease in stomatal conductance with increasing altitude (Hultine and Marshall, 2000, and references therein). However, our data shows the opposite behaviour, indicating that the observed ^{13}C enrichment is based on stomata conductance induced by climate (water stress) and not by altitude (Fig. 6, 7). Thus, the ^{13}C enrichment basically indicates the photorespiration of C$_3$ plants, affected by water availability and evapo-transpiration and thus, the WUE (Tipple and Pagani, 2007; Diefendorf and Freimuth, 2017).

5   Conclusions

This study investigates leaf wax n-alkane ~~homologue~~ patterns and compound-specific δ^{13}C of modern plants and topsoils from semi-arid/arid~~semi-arid~~ Mongolia to test their chemotaxonomic potential and dependency on climate. Our results provide the first regional calibration of leaf wax n-alkanes for semi-arid/arid~~semi-arid~~ Mongolia with the following results:

i. *Caragana spp., Artemisia spp.* and grasses (*Poaceae* and *Cyperacea*) from semi-arid/arid Mongolia~~Mongolian plants~~ show distinct differences in their relative n-alkane ~~homologue~~ patterns. n-Alkanes from the grasses ~~(*Poaceae* and *Cyperaceae*)~~ are clearly dominated by n-C$_{31}$, whereas the woody shrub *Caragana spp.* is dominated by n-C$_{29}$. Since *Artemisia* species can grow both as herbaceous and woody shrubs, *Artemisia spp.* shows not a typical n-C$_{31}$ dominance but is rather more equal to *Caragana spp.* with a dominance in n-C$_{29}$. *Larix sp.* is dominated by the mid-chain n-alkanes n-C$_{23}$ and n-C$_{25}$. However, *Larix sp.* produces only few amounts of n-alkanes and their dominance of mid-chain n-alkanes are not distinct in the respective topsoils. Thus, n-alkanes are not useful to reconstruct changes in the abundance of *Larix sp.* ~~Since, *Larix sp.* produces only very few amounts of n-alkanes they are not useful for reconstructing vegetation changes in *Larix sp.*~~ Although the ACL reveal no potential to discriminate between plant species, the most abundant n-alkanes

n-C_{29} and n-C_{31} allow to discriminate between woody shrubs and grasses, which is expressed in the n-alkane ratio n-C_{31}/n-$C_{29} + n$-C_{31}.

ii. From plants to topsoils of transect II, the decrease of n-alkane concentrations and OEP values indicate n-alkane dilution with mineral soil components and ongoing n-alkane degradation. The n-alkane pattern of the topsoils are mainly characterized by a dominance of n-C_{31}, indicating dominant input from grasses. *Caragana* covered sites tend to reflect the homologue pattern of *Caragana spp.*, with n-C_{29} being the most dominant n-alkane. Topsoils under *Larix sp.* are dominated by the input from the grassy undergrowth. There are no significant differences in compound-specific δ^{13}C between plant species and topsoils. Topsoils tend to be 2‰ enriched compared to the plants, indicating diagenesis from litter to topsoil and the climatic influence.

iii. n-Alkane concentrations and OEP values from Mongolian topsoils are significantly correlated to climatic parameters and decrease with increasing MAT and decreasing MAP. In contrast, our data indicate that the n-alkane homologue patterns from the topsoils (ACL, n-alkane ratio) are not influenced by climatic parameters and thus, the n-alkane ratio can reliably be used to detect and reconstruct differences between the vegetation forms of grasses and woody shrubs. Although increasing altitude correlates with increasing n-alkane concentrations and OEP, altitude has no influences on ACL and n-alkane ratio. For compound-specific δ^{13}C of the topsoils, strong correlations exist with increasing MAT and decreasing MAP, indicating an enhanced enrichment in δ^{13}C with increasing aridity and drought stress. Although, δ^{13}C and altitude are also significantly negative correlated, the influence of altitude is negligible since MATMAT decrease and MAP increase with altitude. Thus, leaf wax δ^{13}C is a valuable proxy for changes in climate and water use effciency in semi-arid/arid Mongolia..

Our results show, that the n-alkane homologues n-C_{29} and n-C_{31} have the chemotaxonomic power to differentiate between grasses and the woody shrub *Caragana spp.* Future studies on plant n-alkane patternshomologues should include a detailed identification of plants regarding different species of each plant genus to reveal the full power of the n-alkane ratio as a vegetation proxy. This is particularly the case for different *Artemisia* species, which can so far not be separated from grasses and woody shrubs. While the n-alkane patternsACL and the n-alkane ratio are not biased by climatic influences, compound-specific δ^{13}C indicate a strong climatic dependency. Thus, n-alkanes and their compound-specific δ^{13}C signatures can be potentiallyused as valuable proxies for future leaf wax based paleoenvironmental reconstruction in sediment archives from semi-arid/aridsemi-arid Mongolia.

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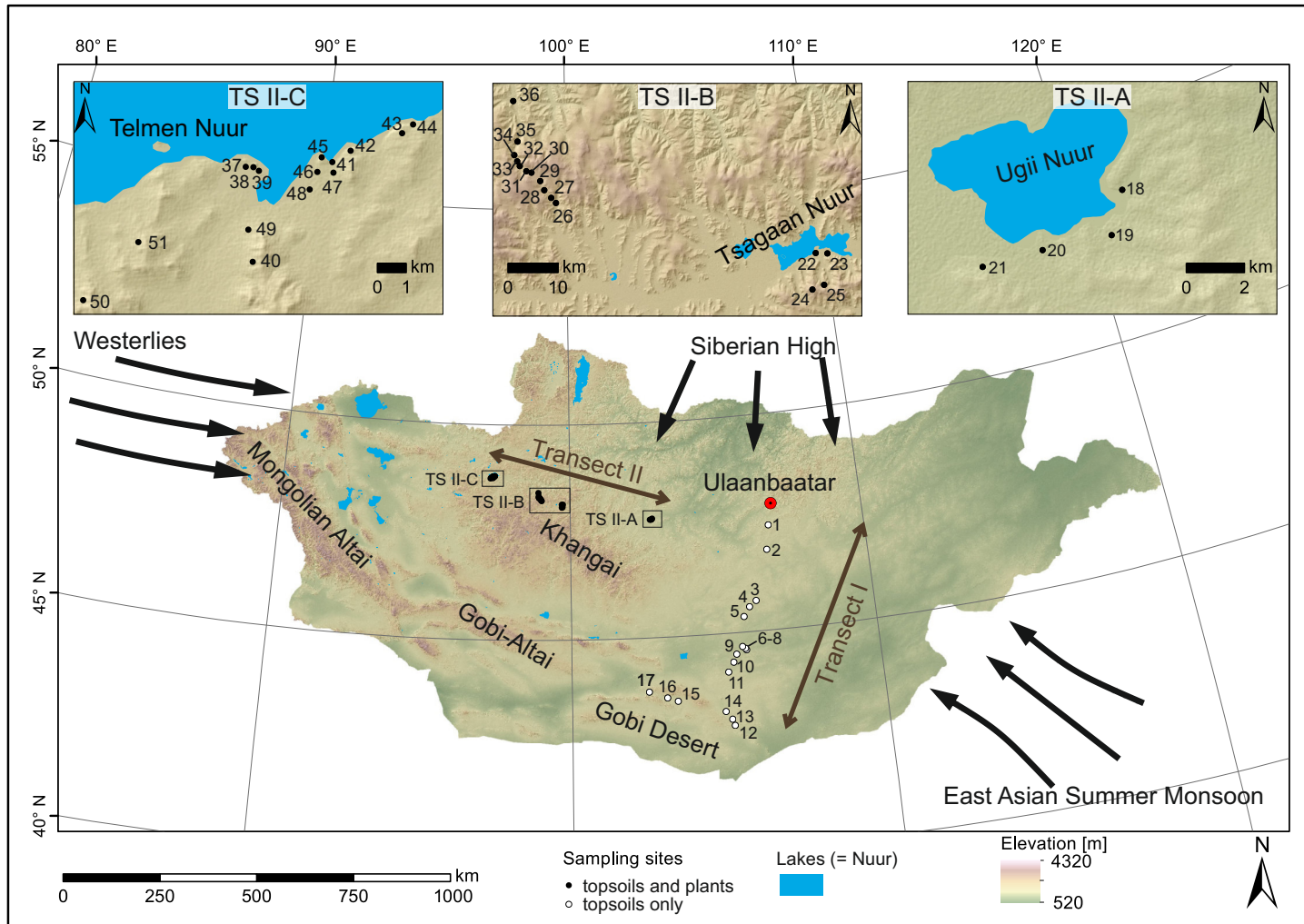
Figure 1. Map of Mongolia (SRTM digital elevation modelDEM). The black/white circles mark the sampling sites along transect I and II. Black arrows indicate the influence of three major atmospheric circulation systems: the Westerlies, the East Asian Summer Monsoon and the Siberian High. Submaps show (TS II-A) the Ugii Nuur catchment, (TS II-B) an altitude transect near the Tsagaan Nuur catchment and (TS II-C) the Telmen Nuur catchment in more detail.
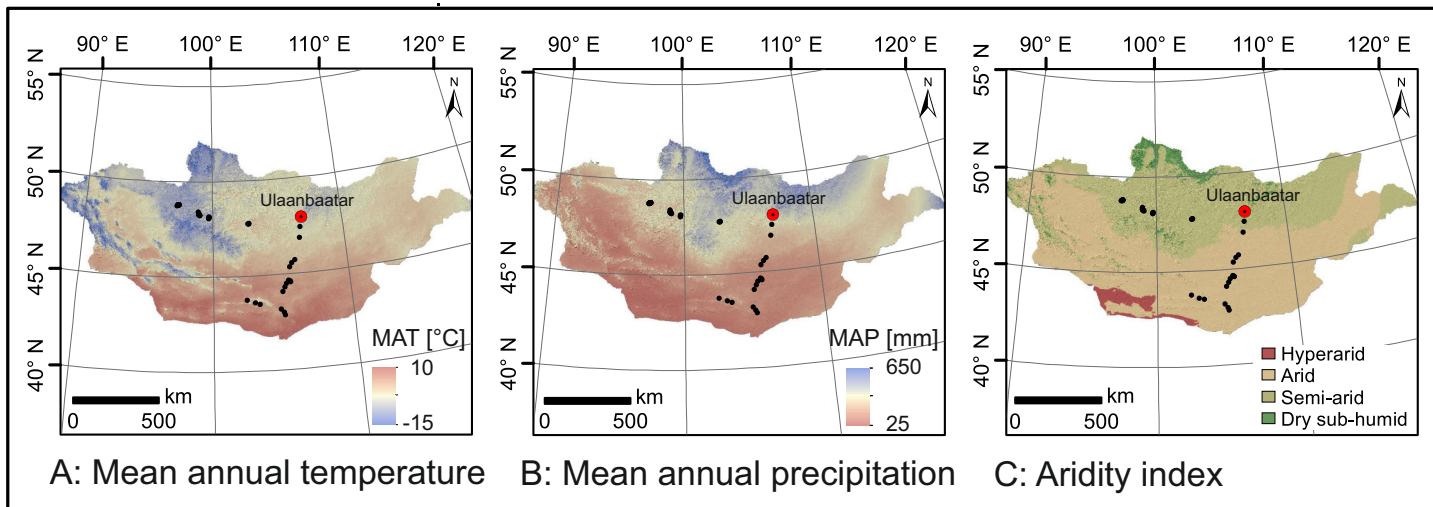
Figure 2. Climate and environmental conditions of Mongolia. Mean annual temperature (A), mean annual precipitation (B) and the aridity index (C). MAT and MAP are based on the WorldClim Dataset of Fick and Hijmans (2017), the AI is based on the Global Aridity Index and Evapo-Transpiration Climate Database v2 of Trabucco and Zomer (2019).
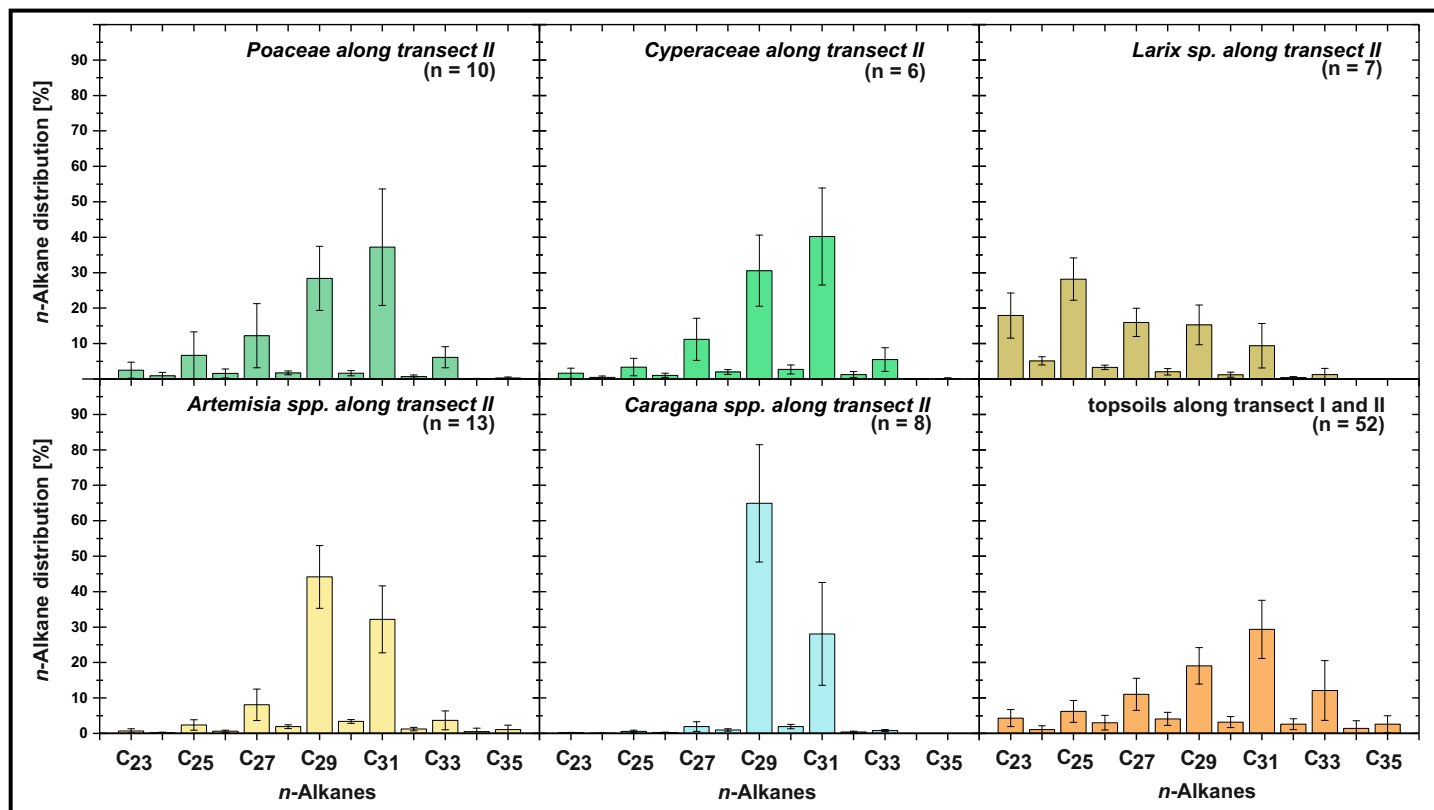
Figure 3. *n*-Alkane patternshomologues pattern of plants (transect II) and topsoils (transect I and II) from Mongolia. The bars show the mean values ± standard deviations.
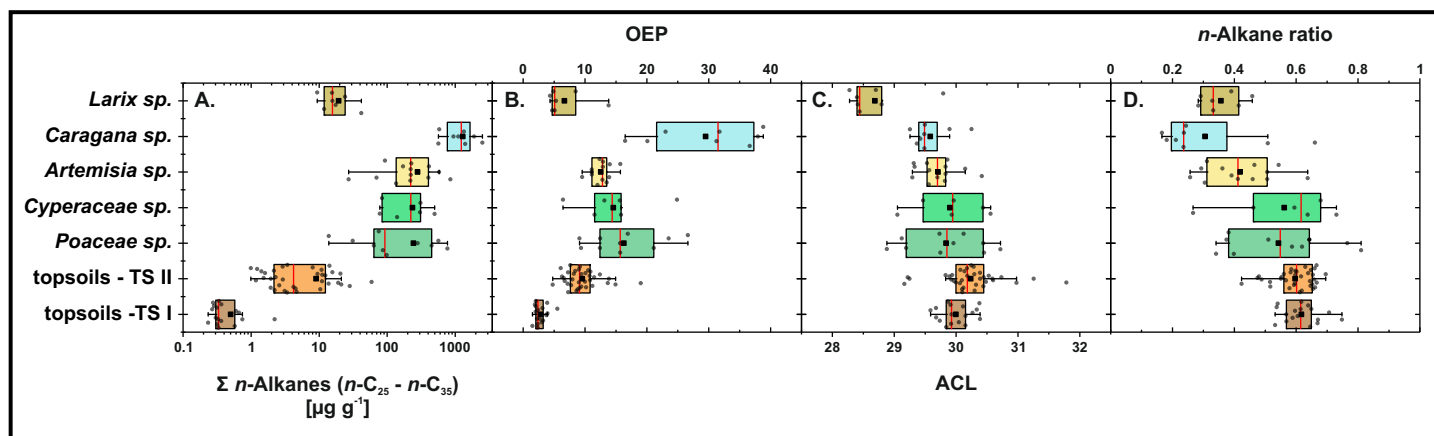
Figure 4. *n*-Alkane concentration (n-C_{25} - n-C_{35}) (A.), OEP (B.), ACL (C.) and *n*-alkane ratio (n-C_{31}/(n-C_{29} + n-C_{31})) (D.) of plants and topsoils from Mongolia (n = *Larix sp.* = 7, *Cyperaceae* = 6, *Poaceae* = 10, *Caragana spp.* = 8, *Artemisia spp.* = 13, topsoils TS I = 17, topsoils TS II = 35). The boxplots indicate median values (red lines), mean values (black squares), interquartile ranges with lower (25%) and upper (75%) quartiles (box), outlayers (whiskers) and investigated samples (grey circles). See Tab. 1 for statistics.
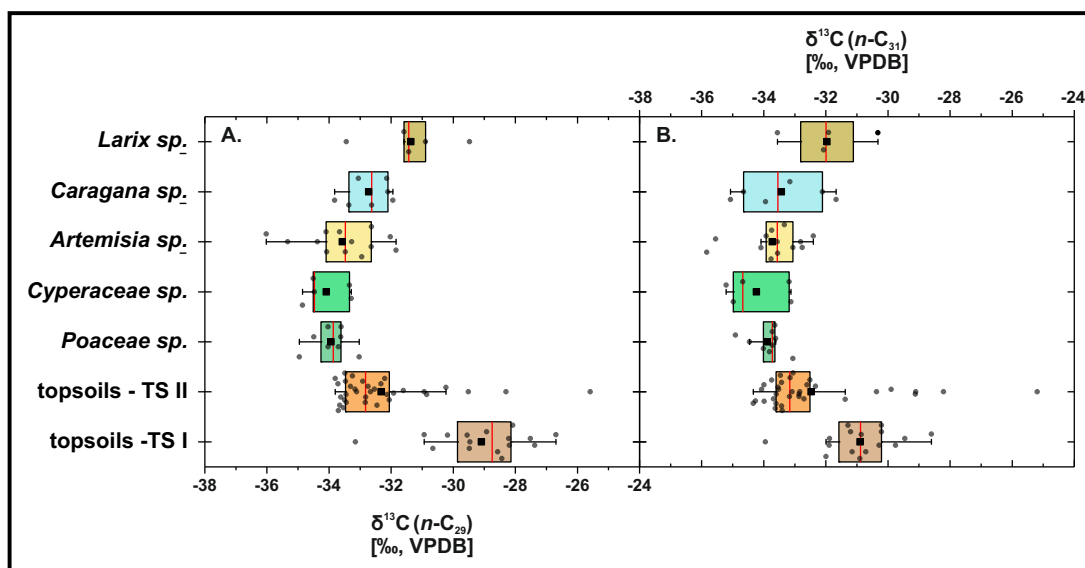
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Figure 5. Compound-specific $\delta^{13}C$ of plants and topsoils from Mongolia. (A.) Compound-specific $\delta^{13}C_{29}$ (n: *Larix sp.* = 5, *Cyperaceae* = 5, *Poaceae* = 8, *Caragana spp.* = 7, *Artemisia spp.* = 13, topsoils TS I = 16, topsoils TS II = 34). (B.) $\delta^{13}C_{31}$ (n: *Larix sp.* = 4, *Cyperaceae* = 5, *Poaceae* = 9, *Caragana spp.* = 6, *Artemisia sp.* = 13, topsoils TS I = 16, topsoils TS II = 34). The boxplots indicate median values (red lines), mean values (black squares), interquartile ranges with lower (25%) and upper (75%) quartiles (box), outlayers (whiskers) and investigated samples (grey circles). See Tab. 1 for statistics.
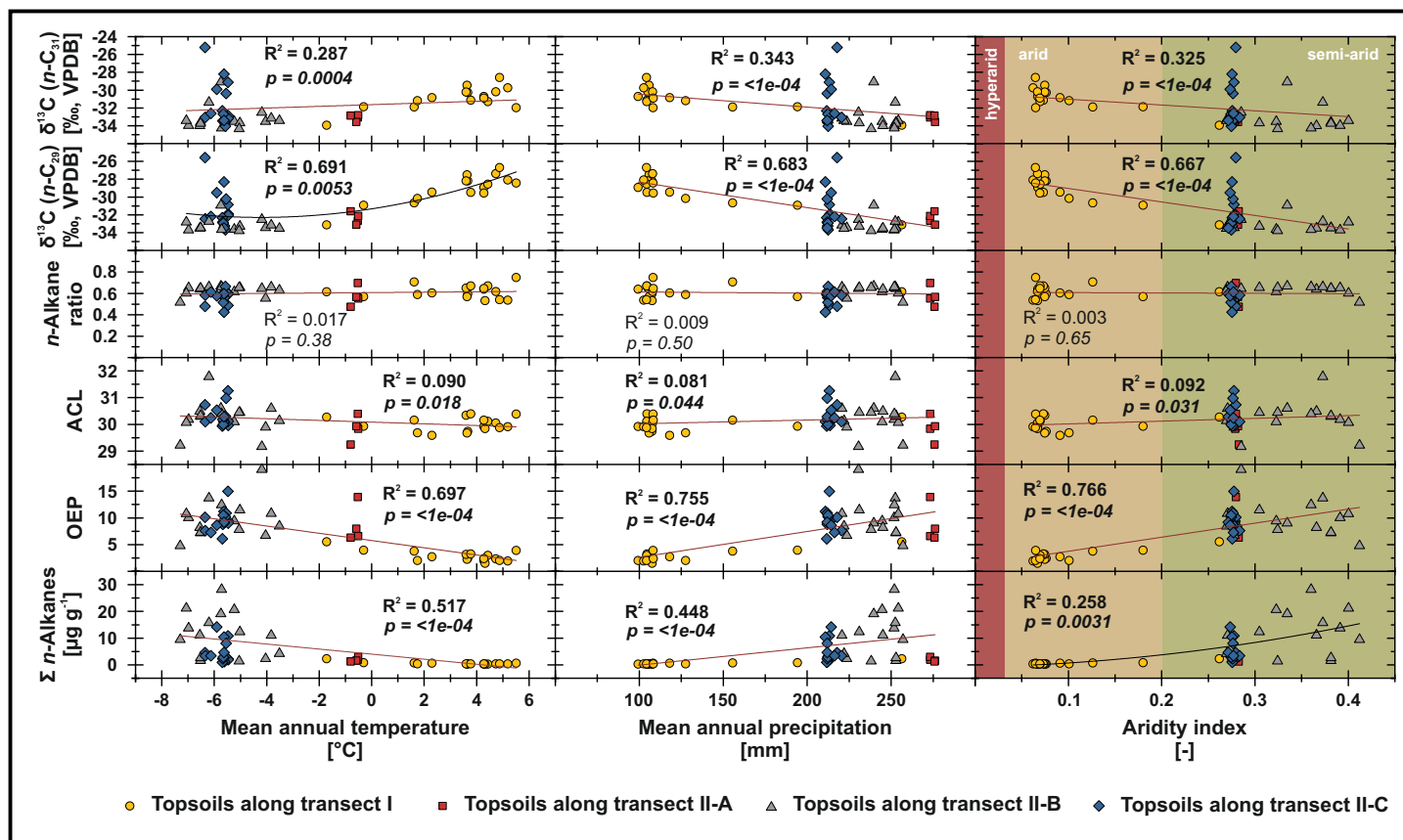
Figure 6. *n*-Alkane concentration (n-C$_{25}$ - n-C$_{35}$), OEP, ACL, *n*-alkane ratio (n-C$_{31}$/(n-C$_{29}$ + n-C$_{31}$)) and compound-specific δ^{13}C (n-C$_{29}$ and n-C$_{31}$) from Mongolian topsoils plotted against climatic parameters (MAP, MAT, AI). Red trend lines illustrate linear regressions, black lines polynomial regressions. Bold values indicate significance ($\alpha = 0.05$).
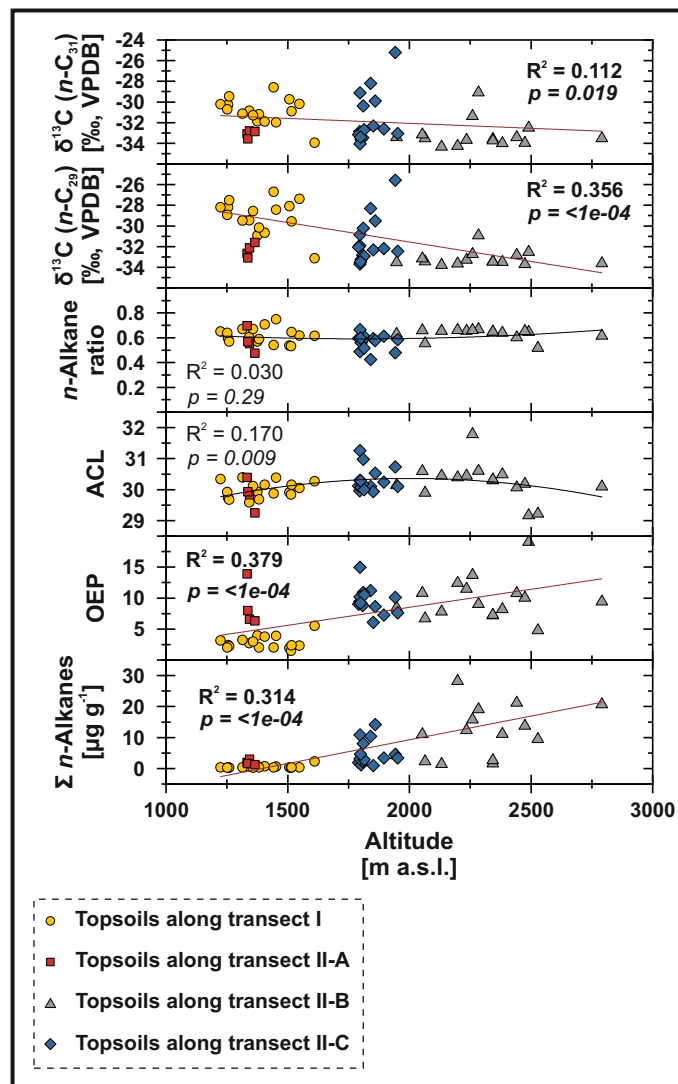
Figure 7. *n*-Alkane concentration (n-C_{25} - n-C_{35}), OEP, ACL, *n*-alkane ratio (n-C_{31}/(n-C_{29} + n-C_{31})) and compound-specific $\delta^{13}C$ (n-C_{29} and n-C_{31}) from Mongolian topsoils plotted against altitude (m a.s.l.). Red trend lines illustrate linear regressions, black lines polynomial regressions. Bold values indicate significance ($\alpha = 0.05$).

Table 1. ANOVA p-values indicating differences among plant species and between topsoils and plants for n-alkane pattern and compound-specific $\delta^{13}C$ (n-C_{29} and n-C_{31}). Bold values indicate significance ($\alpha = 0.05$). TS = topsoils.

	$\sum n$-Alkane (n-C_{25} - n-C_{35})	OEP	ACL	n-Alkane ratio	$\delta^{13}C_{29}$	$\delta^{13}C_{31}$
Poaceae - Cyperaceae	1.000	0.979	1.000	0.999	1.000	0.999
Larix sp. - Cyperaceae	0.505	**0.032**	**0.000**	**0.010**	0.203	0.315
Poaceae - Artemisia spp.	0.999	0.392	0.986	0.069	0.998	1.000
Poaceae - Caragana spp.	**0.000**	**0.000**	0.861	**0.000**	0.814	0.995
Poaceae - Larix sp.	0.325	**0.001**	**0.000**	**0.007**	0.164	0.381
Cyperaceae - Artemisia spp.	0.999	0.948	0.959	0.079	0.995	0.991
Cyperaceae - Caragana spp.	**0.000**	**0.000**	0.802	**0.000**	0.812	0.965
Caragana spp. - Artemisia spp.	**0.000**	**0.000**	0.991	0.185	0.928	0.999
Larix sp. - Artemisia spp.	0.137	0.083	**0.000**	0.806	0.230	0.425
Larix sp. - Caragana spp.	**0.000**	**0.000**	**0.004**	0.946	1.826	0.734
TS Transect II - Poaceae	0.063	**0.000**	0.204	0.801	0.085	0.198
TS Transect II - Cyperaceae	0.259	0.075	0.658	0.99	0.162	0.219
TS Transect II - Artemisia spp.	**0.006**	0.251	**0.012**	**0.000**	0.132	0.181
TS Transect II - Caragana spp.	**0.000**	**0.000**	**0.009**	**0.000**	0.994	0.799
TS Transect II - Larix sp.	1.000	0.573	**0.000**	**0.000**	0.818	0.996
TS Transect I - TS Transect II	**0.003**	**0.000**	0.082	0.273	**0.000**	**0.006**