The spatial distribution of sedimentary compounds and their environmental implications in surface sediments of Lake Khar Nuur (Mongolian Altai)

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Abstract
Lake sediments are valuable natural archives to reconstruct paleoclimate and paleoenvironmental changes which consist of inorganic and organic sediment compounds of allochthonous origin from the catchment and of autochthonous production in the lake. However, for robust paleo-reconstructions it is important to develop a better understanding about sedimentation processes, the origin of inorganic and organic sediment compounds and their distribution within the lake. In this context, modern process studies provide important insights, although environmental and anthropological changes can affect the spatial distribution of sediment compounds through time. Therefore, in this study the spatial distribution of grain size and geochemical proxies in 52 surface sediment samples from Lake Khar Nuur, a small high-altitude lake in the Mongolian Altai with a small and anthropogenically used hydrological catchment, is investigated. The results show a distinct sediment focussing in the two deep basins of the lake, which therefore act as accumulation zones. In those accumulation zones, total organic carbon (TOC), total nitrogen (N) and their isotopic composition (δ13C_TOC, δ15N) as well as n-alkanes indicate that organic sediment compounds are a mixture of both allochthonous and autochthonous origin. While the recent catchment vegetation consists of grasses/herbs and the shrub Betula nana (L.) with distinct differences in their n-alkane homologue patterns, those differences are not reflected in the sediment surface samples which rather indicates that grass-derived n-alkanes become preferentially incorporated in the lake. Extensive anthropogenic activity such as grazing and housing in the southern part of the catchment causes soil erosion which is well reflected by high TOC, N and sulphur (S) contents and 15N depleted δ15N values at the central southern shore, i.e. increased allochthonous sediment input by anthropogenically-induced soil erosion. Overall, the surface sediments of Lake Khar Nuur origin from allochthonous and autochthonous sources and are focussed in the accumulation zones of the lake, while their distribution is both environmentally and anthropogenically driven.

KEYWORDS
geochemistry, grain size, lake sediments, n-alkanes, sediment focussing, stable isotopes
Lake sediments are valuable natural archives providing long records of past limnological, hydrological, and anthropological change. However, robust reconstructions require an understanding of how sediments and the recent climate/environmental signals are incorporated into the sediment archive and how the sediment archive is distributed within the lake basin. Studies investigating the composition and spatial distribution of lake surface sediments have a long history in the field of limnology. Over the last decades, the spatial distribution of sedimentological, inorganic, organic and biogeochemical proxies (Anderson et al., 2008; Dearing, 1997; Ju et al., 2010; Kastner et al., 2010; Shuman, 2003; Thomas et al., 1972; Vogel et al., 2010; Wang et al., 2015; Yu et al., 2015; Yu et al., 2018), as well as heavy metals (Guo et al., 2018; Moore, 1980; Onyari & Wandiga, 1989), pollen (DeBusk, 1997; Li, 2018, 2019; Zhao et al., 2006), diatoms and o斯特s (Anderson, 1990; Martens & Tudorancea, 1991; Shah et al., 2017; Yu et al., 2019; Zalat & Vildary, 2005) was analysed in lake surface sediment samples by several studies. While most of these studies show that sediments and sedimentary compounds accumulate in the deep basins within lakes (i.e. sediment focussing), they mainly focussed on large hydrological settings with large lake surfaces. Therefore the spatial resolution of the analyses and the related process understanding are limited due to the low sample size compared to the lakes surface. Thus, there is a lack of studies investigating lake surface sediments in high spatial resolution to gain a better understanding about modern sedimentation processes, the origin of different sedimentary compounds and their distribution within the lake.

Sedimentary processes in hydrological systems can best be understood by the grain size and elemental (e.g. aluminium (Al), iron (Fe)) distribution which is directly related to transport mechanisms and transport energies and indicate allochthonous input into the lake (Håkanson & Jansson, 1983; Mclaren & Bowles, 1985; Wen et al., 2008; Wündisch et al., 2018). Both proxies can be used to trace sediment focussing in deeper parts of the lake below the mud deposition boundary (Anderson et al., 2008; Lehman, 1975; Wang et al., 2015). The origin of organic sedimentary compounds in lake sediments (allochthonous vs. autochthonous) can be disentangled by total organic carbon (TOC) and total nitrogen (N) contents, as well as their stable isotopes ($\delta^{13}$CTOC, $\delta^{15}$N) as well as n-alkanes.

We will investigate the origin and focussing of allochthonous and autochthonous inorganic and organic sediment compounds in the lake by grain size distribution, elemental composition (Al, Fe), TOC and N and their stable isotopes ($\delta^{13}$CTOC, $\delta^{15}$N) as well as n-alkanes.

We will investigate distinct differences in the vegetation types of the catchment by leaf wax n-alkane homologue patterns and if those vegetation types can also be detected in the lakes surface sediments.

We will identify traces of anthropogenic activity and soil erosion in the catchment in the lakes surface sediments by elemental sulphur (S) as well as TOC and N contents and their stable isotopes.

2 | STUDY SITE

Lake Khar Nuur is a small para-glacial lake located at 2,486 m above sea level (a.s.l) in the Mongolian Altai (48°37’N, 88°56’E) and has a small drainage basin (44.8 km$^2$) that covers altitudes from 2,486 to 3,156 m a.s.l. (Figure 1a. b). Glacial advances from the south-western slopes of the Tsengel Khairkhan Mountains left prominent moraine lobes during the last glaciation which blocked the valley and formed the endorheic Khar Nuur basin (Figure 1b) (Walther et al., 2017). The bedrock of most parts of the catchment is composed of friable black clay shales, whereas only the morainal deposits in the western parts of the catchment consist of granite. Steep slopes (>16°) occur in the northern, western and south-eastern parts of the catchment, but flatter areas are noticeable in the central southern part (Figures 1c, 2a). On the steep slopes of the catchment, very shallow Lephtosols occur, whereas Mollisols with humus rich topsoils (~50 cm) and underlying permafrost developed in the flatter areas of the central southern part of the catchment. At those flatter areas, several gullies exist (Figures 1b, 2c) and favour increased runoff and sediment transport into the lake during snowmelt and precipitation events. The other...
parts of the catchment only produce little episodic runoff and no perennial inflow into the lake exists (Figure 1b, c). Today, most parts of the catchment are anthropogenically used during the summer month, although the central southern part is preferential used by grazing and nomadic housing (Figure 1d). Additionally, a road was constructed around the southern part of the lake (Figure 2b).

At the nearest climate station Tolbo Sum (CDC-ID 44217), which is located ~90 km southeast of Lake Khar Nuur at an altitude of ~2,100 m a.s.l., mean annual precipitation sum was 145 ± 35 mm (2009–2017) and mean annual temperature was −2.4 ± 0.7°C (2009–2019) (DWD Climate Data Center, 2020). The number of ice days (maximum air temperature below 0°C) and freeze days (minimum air temperature < 0°C) were 140 and 247, respectively (DWD Climate Data Center, 2020). Since Lake Khar Nuur is located ~385 m higher than Tolbo Sum, even lower mean annual temperatures as well as more ice and freeze days can be expected. Consequently, the surface of Lake Khar Nuur is ice covered for eight to nine months per year (2016–2019; Planet Team, 2017). The harsh environmental conditions lead to the dominance of alpine grasses and herbs in the Khar Nuur catchment. Exceptional are small patches of 50 to 80 cm high Betula nana (L.) shrubs that covers a small area (~0.42 km²) with favourable edaphic conditions on a north-eastern slope (Figures 1d, 2b).

### MATERIAL AND METHODS

#### 3.1 Bathymetry

A bathymetric map of the lake floor was created using a Lowrance HPS 5 Fishfinder working with a frequency of 100 kHz. A ~240,000 depth-measurements were conducted and the respective drive-lines are shown in Figure 3(a). The map was created using the software Sonar Viewer 2.1.2 and Esri ArcGIS 10.5 applying the ‘spline with barriers’ interpolation tool.
Sampling and laboratory analyses

For this study, the sediment surface (0–1 cm) of the lake floor was sampled at 52 locations (for sample locations see Figure 1b) using a Van Veen Grab sampler (Lie & Pammat, 1965). All samples are from a water depth > 2 m (Figure 1a). Except for grain size analyses, samples were freeze-dried (−50°C, > 72 h), ground and sieved to < 40 μm.

Grain size analyses

For grain size analyses, sample aliquots were treated with hydrogen peroxide (H₂O₂, 15%, 30%) to remove organic matter and subsequently treated with hydrochloric acid (HCl, 15%) to remove carbonates. As a dispersant, 5 ml sodium-pyrophosphate (Na₄P₂O₇ · 10 H₂O) were added to each sample and shaken for > 2 h. Before the measurement, the fraction > 2 mm was removed from the samples by sieving due to technical reason. It has to be noted that the > 2 mm fraction was only present in the samples 38, 40, 45, 46, 48, 50 and 51 (grey sample points in Figure 3b), which, however, was not further quantified. The < 2 mm grain-size distribution was determined with a laser diffraction particle size analyzer (LS 13320, Beckman Coulter, Brea, California, USA). Samples were measured with the aqueous liquid module in several 60 s cycles until a reproducible signal was obtained. The ‘Fraunhofer’ optical model of light scattering was used for computing grain size distribution. In the following we will use Mode 1 which is the most frequently occurring particle size and was calculated with a modified version of the Software Gradistat 4.2 (Blott & Pye, 2001).

Elemental analyses

Al, Fe and S concentrations were measured with an ICP-OES 725-ES (VARIAN, Palo Alto, California, USA). Samples (0.2 g) were dissolved using a microwave-assisted modified aqua regia digestion of 2 ml HCl.
(32%), 4 ml nitric acid (HNO₃, 65%), 1 ml H₂O₂ (30%) and 6 ml deionized water. For error estimation, two samples (KN09, KN41) were measured in triplicates (relative error: Al, 1.6%; Fe, 1.6%; S, 11%). Additionally, the reference material LGC6 187 (river sediment, n = 8) was measured to calculate the relative analytical error, which was < 2.0%.

3.5 Carbon and nitrogen analyses and bulk isotopic analyses

Carbon and nitrogen contents and their stable isotopes were analysed with an Elementar Analyser (vario EL cube) coupled to an isotope ratio mass spectrometer (Isoprime precision). ~30 mg of untreated sediment per sample was weighed into tin boats (Elementar, 6 × 6 × 12) and measured for total carbon (TC), total nitrogen (N) and the nitrogen isotopic composition (δ¹⁵N). Prior to the analyses of TOC and the carbon isotopic composition (δ¹³C), carbonates were removed from the samples with 1 N HCl at 60°C for 8 h. Samples were subsequently washed with ultrapure water to pH neutrality and ~30 mg were weighed into tin boats. The analytical precision of the δ¹⁴N and δ¹³C analyses was checked against certified standards (L-Prolin, EDTA and USGS65), and gave an analytical error < 0.25‰ and < 0.05‰, respectively. The δ¹⁴N and δ¹³C are given in their delta notation against Air and the Vienna Pee Dee Belemnite (VPDB).

Total inorganic carbon (TIC) was calculated by subtracting TOC from TC. The molar ratio of TOC and N was calculated based on their respective molecular weights (Equation (1)). Relative errors were calculated based on triplicate measurements (TC: < 0.1%, N: < 0.9%, TOC: < 0.8%).

\[
\text{TOC/N[molar]} = \frac{\text{TOC} \times 12.0107 \times 14.0067}{\text{g} \cdot \text{mol}^{-1}/\text{N} \times \%}
\]

3.6 Leaf wax analyses

Total lipids of 49 surface sediment samples, 13 topsoil samples (5–28 g, depending on bulk density and TOC content) and seven plant samples (0.8–1.1 g) were extracted with 40 ml dichloromethane (DCM) and methanol (MeOH) (9:1, v/v). According to Bieldtner et al. (2018), each sample was ultrasonically extracted over three 15 min cycles. The total lipid extract was separated by solid phase extraction using aminopropyl silica gel (Supelco, 45 μm, Bellefonte, Pennsylvania, USA) as a stationary phase. The n-alkanes were eluted with 4 ml hexane and further purified over coupled silver nitrate (AgNO₃)-zeolite (Geokleen, Beibington, Merseyside, UK) pipette columns. The n-alkanes trapped in the zeolite were subsequently dissolved in hydrofluoric acid and recovered by liquid-liquid extraction using hexane.

An Agilent 7890B gas chromatograph equipped with an Agilent HP5MS column (30 m, 320 μm, 0.25 μm film thickness) and a flame ionization detector (FID) was used for identification and quantification of the n-alkanes, relative to external n-alkane standards (n-alkane mix nC₂₁–nC₄₀, Supelco, Bellefonte, Pennsylvania, USA). Precision of compound quantification was about 1.2% based on multiple measurements of the external standard (n = 10).

n-Alkane concentrations (Σ n-alkanes) are given in μg g⁻¹ dry weight and were calculated as the sum of nC₂₁ to nC₃₅. The odd-over-even predominance (OEP) was calculated according to Hoefs et al. (2002) (Equation 2) and serves as a proxy for degradation with values below five indicating enhanced n-alkane degradation (Bugle et al., 2010; Zech et al., 2010).

\[
\text{OEP} = \frac{\sum_{nC_{27}} + nC_{29} + nC_{31} + nC_{33}}{nC_{25} + nC_{27} + nC_{29} + nC_{31}}
\]

We used a normalized n-alkane ratio that we modified according to the differences between the two prominent vegetation forms in the Khar Nuur catchment and that we calculated to detect possible contributions from those vegetation forms in the surface sediments. Differences in the respective vegetation forms are because nC₃₁ is the most abundant n-alkane of grasses/herbs and nC₂₇ of the shrub Betula nana (L.) (see Results 4.3; Equation 3).

\[
\text{n-alkane ratio} = \frac{nC_{31}}{nC_{27} + nC_{31}}
\]

3.7 Data analyses

The hydrological Khar Nuur catchment and its geomorphological attributes were derived from a digital elevation model (SRTM) with a spatial resolution of ~30 m × 30 m. Based on the sedimentological and geochemical results of the surface sediment samples, spatial distribution maps were generated using the ‘spline with barriers’ tool in Esri ArcGIS 10.5. Pearson’s r-values were calculated to detect correlations of the analysed proxies and a student’s t-test was performed to determine significant differences (α = 0.05). All statistical analyses were performed using the statistical software OriginPro 2017.

4 RESULTS

4.1 Bathymetry and grain size distribution

The bathymetric map of Lake Khar Nuur shows two basins within the lake. Following the morphology of the catchment, the first basin
in the northeast of the lake (basin A; Figures 1b, 3a) shows sub-aquatic steep slopes and a large plain in the centre with the deepest part 49.4 m below lake level (Figures 1b, 3a). In the southwest of the lake, a second basin is present (basin B, Figures 1b, 3a) and the sub-aquatic slopes also follow the catchment’s morphology with steep slopes on the northern shore and distinctly flatter slopes on the southern shore. Both basins are separated by a sub-aquatic ridge that follows the distinct ridge in the north-western part of the catchment (Figures 1b, c, 3a).

The Mode 1 of the grain size distribution shows highest values (coarse silt to fine sand) at the shore of basin A, but also below the steep slopes in basin B (Figure 3b). The finer grain size fractions (smaller than coarse silt) as indicated by low Mode 1 values accumulate in the centre of both basins (Figure 3b). This is well reflected by a significant anti-correlation of Mode 1 and water depth ($r = -0.45; \alpha < 0.05$; Table 1), indicating fine grained sediments at greater water depth and vice versa. We have to note that particles > 2 mm were detected within samples 38, 40, 45, 46, 48, 50 and 51 (grey sample points in Figure 3b), but not further quantified. So, Mode 1 values are only representative for the < 2 mm fraction.

4.2 | Elemental distribution, carbon and nitrogen contents and bulk isotopic composition

The elemental distribution in Lake Khar Nuur surface sediments shows highest concentrations of Al (42,900 ± 3,900–58,700 ± 4,500 ppm) and Fe (32,500 ± 1,400–51,000 ± 2,200 ppm) in the central and deepest parts of basins A and B, whereas the nearshore samples show lowest concentrations (Al: < 40,900 ± 3,300 ppm; Fe: < 27,000 ± 1,200 ppm) (Figure 4a, b). Sulphur (S) also shows high concentrations in the deepest parts in both basins, but highest concentrations occur at the central southern shore (4,900 ± 1,500 ppm). All other nearshore samples and samples at the sub-aquatic ridge show low concentrations (< 1,000 ± 100 ppm) (Figure 4c).

A similar spatial distribution to S is shown by TOC and N with highest concentrations in the deepest parts in both basins A and B (TOC: 3.8–4.7%, N: 0.6–0.7%) and high concentration at the central southern shore. Low concentrations are shown by the nearshore samples and samples at the sub-aquatic ridge (TOC: < 2.01 ± 0.01%; N: < 0.44 ± 0.01%) (Figure 4d, e). The spatial distribution of TIC shows the highest concentrations in basins A and B and at the central southern shore (2.21 ± 0.04 to 3.11 ± 0.06%), whereas the remaining nearshore samples and samples at the sub-aquatic ridge show low concentrations (< 2.06 ± 0.04%) (Figure 4f).

The isotopic composition of TOC ($\delta^{13}C_{TOC}$) ranges from −29.0% to −19.5% (Figure 4g). For most of the lake, the samples show $^{13}C$ depleted $\delta^{13}C_{TOC}$ values, whereas $\delta^{13}C_{TOC}$ values are enriched in $^{13}C$ at the northern shoreline and for two samples at the southern shoreline (KN19 and KN52, Figure 4g). The nitrogen isotopic composition ($\delta^{15}N$) ranges between 1.7% and 4.8% (Figure 4h). While most parts of the lake are enriched in $^{15}N$, samples are remarkably depleted in $^{15}N$ at the central southern shore (Figure 4h). The molar TOC/N ratio shows higher values in both basins A and B compared to the nearshore samples (Figure 4i). However, the molar TOC/N ratio ranges between 1.22 and 6.60 and reflects only a very small range.

The spatial distribution of Al, Fe and TOC/N is also reflected in significant correlations with water depth ($r$: Al = 0.87, Fe = 0.81 and TOC/N = 0.59; $\alpha < 0.05$; Table 1). Although TOC, N and TIC are also significantly correlated with water depth ($r$ between 0.56 and 0.75; $\alpha < 0.05$; Table 1) and Al ($r$ between 0.61 and 0.80; $\alpha < 0.05$; Table 1), they correlate significantly with S and with each other ($r$ values between 0.63 and 0.96; $\alpha < 0.05$; Table 1). The $\delta^{13}C_{TOC}$ and water depth significantly anti-correlate ($r = -0.72; \alpha < 0.05$; Table 1) indicating $^{13}C$ depleted $\delta^{13}C$ values at greater water depth and vice versa.

4.3 | n-Alkanes

The n-alkanes from plant and topsoil samples from sites with grasses/ herbs and Betula nana (L.) in the Khar Nuur catchment show distinct differences in their chain-length distribution pattern (Figure 5). Sites with grasses/herbs show a $nC_{29}$ dominance for both the plants and the respective topsoils (Figure 5a). The n-alkane concentration of the grass samples ranges from 440 to 3,260 $\mu$g $^{-1}$ and is distinctly lower for the topsoils (17 ± 14 $\mu$g $^{-1}$). The grass samples show higher OEP values (24.5 ± 7.7) compared to the respective topsoil samples (8.4 ± 1.8). In contrast, sites covered with the shrub Betula nana (L.) show a distinct nC$_{27}$ dominance (Figure 5b). The n-alkane concentration of the two Betula nana (L.) samples is 500 ± 20 $\mu$g $^{-1}$ and 19 $\mu$g $^{-1}$ for the topsoil sample. Higher OEP values are shown by the Betula nana (L.) samples (27.0 ± 1.2) compared to the respective topsoil sample (14.5).

Based on our findings, we identified nC$_{29}$ and nC$_{27}$ as two endmembers for the vegetation forms of grasses/herbs and shrubs in the Khar Nuur catchment, respectively, and calculated a modified n-alkane ratio to distinguish between both vegetation forms (cf. section 3.6). The n-alkane ratio of the sites with grasses/herbs is 0.91 ± 0.07 (plants) and 0.74 ± 0.08 (topsoils), and 0.12 ± 0.03 (plants) and 0.27 (topsoil) for sites with Betula nana (L.).

In the lake, the n-alkane concentration is the highest in both basins with values up to 12.7 $\mu$g $^{-1}$ in basin A and values up to 11.5 $\mu$g $^{-1}$ in basin B (Figure 6a). All other samples show concentrations < 9.7 $\mu$g $^{-1}$ with the lowest concentrations in the nearshore samples (Figure 6a). Both n-alkanes nC$_{29}$ and nC$_{27}$ show highest concentrations in the centre of basins A and B and follow the same pattern as the total n-alkane concentration (Figure 6b, c). It is notable that the concentration of nC$_{29}$ (0.01–1.2 $\mu$g $^{-1}$) is distinctly lower compared to nC$_{27}$ (0.11–2.4 $\mu$g $^{-1}$) (Figure 5b, c). The $P_{\text{ext}}$ ranges between 0.13 and 0.42 and shows highest values in the two basins with values between 0.36 and 0.42. All other samples show values < 0.32 (Figure 6e). The OEP ranges between 5.2 and 8.0 and shows low values for the samples located in the two basins (Figure 6d). High values are present in some samples located at the south-eastern, north-eastern and north-western shore (Figure 6d). The modified n-alkane ratio with regional end-members ranges between 0.53 and 0.71. The lowest values occur at the central southern shore and in both basins A and B (< 0.62), whereas highest values (0.65–0.71) are present in samples located at the shorelines of the lake. Exceptional with lower values is one shoreline sample west of the sub-aquatic ridge (Figure 6f).
### TABLE 1  Correlation matrix (Person’s r) of the investigated proxies

|          | WD    | Mode 1 | Al    | Fe    | S     | TOC   | $\delta^{13}$C | N     | $\delta^{15}$N | TIC   | TOC/N | $C_{21}$ | $C_{31}$ | n-Alkane-concentration | OEP | n-Alkane-ratio |
|----------|-------|--------|-------|-------|-------|-------|----------------|-------|----------------|-------|-------|---------|---------|------------------------|-----|---------------|
| Mode 1   | −0.45 |        |       |       |       |       |                |       |                |       |       |         |         |                        |     |               |
| Al       | 0.87  | −0.49  |       |       |       |       |                |       |                |       |       |         |         |                        |     |               |
| Fe       | 0.81  | −0.55  | 0.86  |       |       |       |                |       |                |       |       |         |         |                        |     |               |
| S        | 0.23  | −0.17  | 0.28  | 0.25  |       |       |                |       |                |       |       |         |         |                        |     |               |
| TOC      | 0.75  | −0.39  | 0.80  | 0.67  | 0.66  |       |                |       |                |       |       |         |         |                        |     |               |
| $\delta^{13}$C | −0.72 | 0.40   | −0.68 | −0.62 | −0.19 | −0.64 |                |       |                |       |       |         |         |                        |     |               |
| N        | 0.71  | −0.39  | 0.75  | 0.67  | 0.76  | 0.96  | −0.60         |       |                |       |       |         |         |                        |     |               |
| $\delta^{15}$N | 0.35  | −0.17  | 0.24  | 0.36  | −0.47 | −0.08 | −0.41         | −0.14 |                |       |       |         |         |                        |     |               |
| TIC      | 0.56  | −0.28  | 0.61  | 0.52  | 0.63  | 0.69  | −0.40         | 0.79  | −0.33         |       |       |         |         |                        |     |               |
| TOC/N    | 0.59  | −0.35  | 0.64  | 0.53  | 0.39  | 0.83  | −0.64         | 0.70  | 0.08          | 0.33  |       |         |         |                        |     |               |
| $C_{21}$ | 0.75  | −0.28  | 0.78  | 0.56  | 0.40  | 0.85  | −0.66         | 0.78  | −0.01         | 0.66  | 0.67  |         |         |                        |     |               |
| $C_{31}$ | 0.75  | −0.35  | 0.81  | 0.64  | 0.44  | 0.88  | −0.71         | 0.83  | 0.03          | 0.67  | 0.73  | 0.94     |         |                        |     |               |
| n-Alkane-concentration | 0.76  | −0.33  | 0.81  | 0.62  | 0.47  | 0.90  | −0.70         | 0.85  | −0.01         | 0.69  | 0.72  | 0.97     | 0.99    |                        |     |               |
| OEP      | −0.62 | 0.37   | −0.66 | −0.58 | −0.22 | −0.58 | 0.35          | −0.59 | −0.08         | −0.62 | −0.37 | −0.62    | −0.55    | −0.60                   |     |               |
| Alkane-ratio | −0.23 | 0.12   | −0.26 | −0.20 | −0.74 | −0.63 | 0.27          | −0.70 | 0.44          | −0.56 | −0.45 | −0.43    | −0.39    | −0.45                   | 0.47 |               |
| $P_{\text{aq}}$ | 0.67  | −0.31  | 0.70  | 0.54  | 0.53  | 0.84  | −0.58         | 0.82  | −0.19         | 0.75  | 0.63  | 0.85     | 0.77     | 0.83                   | −0.76 | −0.73        |

Bold values indicate significance ($z = 0.05$). WD, water depth; Al, aluminium; Fe, iron; S, sulphur; TOC, total organic carbon; $\delta^{13}$C, carbon isotopic composition; N, total nitrogen; $\delta^{15}$N, nitrogen isotopic composition; TIC, total inorganic carbon; OEP, odd-over-even predominance.
Significant correlations exist between the spatial distribution of the \( n \)-alkane concentration, the \( n \)-alkanes \( nC_{21}, nC_{31} \) and the \( P_{aq} \) \( (r \) between 0.67 and 0.99; \( \alpha < 0.05; \) Table 1). While the OEP is significantly anti-correlated with water depth \( (r = -0.62; \alpha < 0.05) \), there exists no significant correlation between the modified \( n \)-alkane ratio and water depth \( (r = -0.23; \alpha > 0.05; \) Table 1). Moreover, there is a significant anti-correlation between the \( P_{aq} \)' and the modified \( n \)-alkane ratio \( (r = -0.73; \alpha < 0.05; \) Table 1).

**5 | DISCUSSION**

### 5.1 Allochthonous versus autochthonous sediment and inorganic and organic sediment compound origin in Khar Nuur sediments

Lake sediments and their inorganic and organic sediment compounds are a mixture of allochthonous material from the...
catchment and autochthonous material produced within the lake (Meyers & Ishiwatari, 1993). Therefore, it is an important precondition to disentangle both signals to robustly interpret transport processes, inorganic and organic sediment compound origin/sources and the environmental conditions under which they accumulate.

5.1.1 Grain size and elements

A fundamental proxy to differentiate sediment transport energy and transport regimes in lake sediments is the grain size distribution. The most prominent sediment transport regimes can be identified by the

\[ nC_{31} / (nC_{27} + nC_{31}) \]

where the lower and upper scale limit is based on the lowest and highest measured values. Also shown are the slope and the catchment of Lake Khar Nuur [Colour figure can be viewed at wileyonlinelibrary.com]
Mode 1 which describes the most frequently occurring particle size in each sample (Blott & Pye, 2001). The grain size distribution in the surface sediments of Lake Khar Nuur exhibits high Mode 1 values in the nearshore samples compared to low values in the two basins (Figure 3b). Consequently, the grain size distribution follows the direction of sediment transport into the lake, indicating a higher energy regime near the shore and a lower energy regime towards the central part of the lake where sediments are focussed (Håkanson & Jansson, 1983; Mclaren & Bowles, 1985; Wang et al., 2015). This sediment focussing is likely caused by higher transport energies near the several small periodically inflows which mainly occur along the depth contours of the catchment during precipitation events and snow melt (Figure 1b). The clay and silt fractions are transported towards the lake centre of basins A and B where they can settle at calmer water conditions. Sediment focussing of the finer fractions in deeper waters of Lake Khar Nuur is further supported by the significant anti-correlation of water depth and Mode 1 ($r = -0.45$, $\alpha < 0.05$; Table 1; Figure 3b). This typical sediment distribution was also found in previous studies (Anderson et al., 2008; Thomas et al., 1972; Wang et al., 2015). It has to be noted that the > 2 mm fraction was removed before grain size measurements due to technical reasons. While the > 2 mm fraction was not quantified but present in some samples (grey sample points in Figure 3b), it is likely that the > 2 mm fraction would lead to higher Mode 1 values in those samples. However, the > 2 mm fraction was exclusively found in the nearshore samples which supports our findings that high transport energy regimes occur near the periodical inflows at the shoreline.

The decrease in grain size from the shoreline to the basins is accompanied by an increase in the concentration of the allochthonous elements Al and Fe. Both elements are typically bound to the fine grain size fraction (i.e. clay and feldspars) (e.g. Wen et al., 2008; Wündsch et al., 2018) and therefore significantly anti-correlate with the Mode 1 in Lake Khar Nuur ($r = -0.49$ and $-0.55$, respectively; $\alpha < 0.05$; Table 1; Figures 3b, 4a, b). Since Al and Fe are thought to be of allochthonous origin in lacustrine systems (Wen et al., 2008; Wündsch et al., 2018), their significant correlation with water depth ($r = 0.87$ and 0.81, respectively; $\alpha < 0.05$; Table 1) indicates the high accumulation of allochthonous material within the deep zones of basins A and B. According to Anderson et al. (2008, and references cited therein), sediment accumulation mostly occurs below a mud deposition boundary depth which is further described by Wang et al. (2015) as the sediment accumulation zone that is least influenced by wind-driven turbulences and other processes. Thus, this accumulation zone provides the best suited sediments for paleoenvironmental studies, and for Lake Khar Nuur, a distinct sediment focussing occurs in the accumulation zones in the two basins A and B (Anderson et al., 2008; Lehman, 1975; Wang et al., 2015).

### 5.1.2 Carbon and nitrogen contents and bulk isotopic composition

The accumulation zones in the two basins in Lake Khar Nuur are also evident in the characteristic spatial distribution of inorganic and organic sediment compounds. The highest TOC and N concentrations, as well as highest molar TOC/N ratios occur in both basins, resulting in significant correlations of TOC, N and TOC/N ratio with water depth ($r = 0.75$, 0.71 and 0.59, respectively; $\alpha < 0.05$; Table 1; Figure 4d, e, i). Low molar TOC/N ratios in the two basins would classically indicate a predominantly autochthonous origin of organic material since values $< 10$ are thought to be representative for aquatic plants (Meyers, 1994). However, TOC, N and their molar ratio also significantly correlate with the allochthonous elements Al and Fe (Table 1) and indicate that organic matter cannot be exclusively of autochthonous origin in the accumulation zones. Those discrepancies could be possibly explained by (i) the heterogenic nature of organic matter that often comprises a mixed signal from allochthonous and autochthonous sources in lake sediments and (ii) the very low TOC and N concentrations of the nearshore samples that might bias the respective molar TOC/N ratios (Woszczyk et al., 2011). However, it is notable that hotspots with high TOC and N values are not only evident in the two basins, but also occur in the central southern part of the lake. Here, the site-related sub-catchment is less steep and the well-developed soils favour the settlement of nomadic families and livestock grazing during summer. Thus, this observed hotspot of organic matter in the lake likely indicates increased input by erosion, mainly due to anthropogenic activity (cf. section 5.3).

Additional information about organic matter origin and environmental conditions under which the sediments become deposited might be provided by $\delta^{13}\text{CTOC}$ and $\delta^{15}\text{N}$, although different fractionation factors can complicate the clear interpretation and source assignment of both isotopes (Meyers, 2003). The $\delta^{13}\text{CTOC}$ values from Lake Khar Nuur are depleted in $^{13}\text{C}$ ($\approx 28.7$ to $-24.2\%$) for most parts of the lake, except for the northern and central southern shorelines where $\delta^{13}\text{CTOC}$ values are more enriched in $^{13}\text{C}$ ($\approx 23.9$ to $-19.6\%$). Differences in $\delta^{13}\text{CTOC}$ are often explained by the different photosynthetic pathway of C3 and C4 plants, but in the Khar Nuur catchment only plants using the C3 photosynthetic pathway (Calvin–Benson pathway) grow, typically showing $\delta^{13}\text{C}$ values from $\approx 33$ to $-22\%$ (Meyers, 1994; Pyankov et al., 2000). Changes in $\delta^{13}\text{C}$ of C3 plants indicate variations in plant water use efficiency and thus drought stress (Rao et al., 2017; Struck et al., 2020), but those changes in water use efficiency would only explain a few % differences and not the large differences at the northern and central southern shoreline. Another explanation are aquatic plants that can have $\delta^{13}\text{CTOC}$ values in the isotopic range of C3 plants ($\approx 30$ to $-25\%$) when they use dissolved carbon dioxide ($\text{CO}_2$) from the water column which is in isotopic equilibrium with the atmosphere (Meyers, 1994). However, when the availability of dissolved atmospheric CO2 ($\delta^{13}\text{C} = -7\%$) is limited and aquatic plants begin to use dissolved bicarbonate ion ($\text{HCO}_3^-$) ($\delta^{13}\text{C} = 1\%$), their isotopic composition becomes enriched in $^{13}\text{C}$, resulting in up to 10% more positive $\delta^{13}\text{CTOC}$ values (Bradley, 2015; Yu et al., 2015). The $\text{HCO}_3^-$ origin in the lake from formerly deposited back-dissolved TIC can be of autochthonous origin by evaporitic carbonate precipitation and/or biogenic incorporation (formation of carbonate shells by plants and animals) (Jones & Bowser, 1978; Ju et al., 2010; Kelts & Hsü, 1978; Yu et al., 2018). Additionally, TIC can derive from allochthonous sources such as limestones in the catchment and/or aeolian dust which is an important carbon source in semi-arid regions (Jones & Bowser, 1978; Grunert & Lehmkuhl, 2004; Kelts & Hsü, 1978). Although we cannot completely rule out the contribution of allochthonous carbonates in the Khar Nuur sediments, they are rather unlikely because the bedrock of the catchment is not made of calcareous bedrock which is supported by modern $^{13}\text{C}$-ages of a dated
water plant and a surface sediment (see Supporting Information, Table S1). Thus, since the so-called ‘hardwater effect’ from old calcareous bedrock is not evident in the Khar Nuur catchment, TIC should mainly be derived from carbonate precipitation and biogenic sources. However, samples with 13C enriched δ13C TOC values along the northern shore have likewise low TIC concentration. Therefore, we suggest that the strong enrichment is due to a dissolution of the formerly deposited inorganic carbon in the lake water and that the southward exposure of the northern shore enables enhanced aquatic productivity in the lake, also below the ice cover (eight to nine months) (Gibson et al., 1999; Loose et al., 2009). However, we have to mention that a more detailed picture of the TIC sources would be derived by further mineralogical analyses.

The δ15N signal can also be used to investigate organic matter sources and biogeochemical processes in lake sediments (Meyers, 2003; Talbot, 2001; Zech et al., 2011). Aquatic plants often have more positive values (+8.5‰) compared to terrestrial plants (+0.5‰), because dissolved inorganic nitrogen is 7–10‰ more positive than atmosphere-derived nitrogen (Peterson & Howarth, 1987; Peters et al., 1978; Meyers, 2003). The δ15N values in Lake Khar Nuur are mostly between 3 and 5‰, indicating a mixture of autochthonous and allochthonous organic matter. Distinctly low values only occur near the central southern shore and to some lesser degree close to the other shores, indicating a larger proportion of allochthonous organic matter in those samples. This is in line with the observed anthropogenic usage of this part of the catchment and the gully erosion (Figure 2c). While the δ13C TOC signal is not able to differentiate between the allochthonous input of C3 plants and autochthonous production from aquatic algae, the δ15N signal identifies this hotspot of increased allochthonous input at the central southern shoreline.

5.1.3 | Leaf wax n-alkanes

A more homogenic part of the organic matter pool in lake sediments are n-alkanes. Based on their chain-length distribution one can differentiate whether they have a allochthonous origin from the leaf waxes of terrestrial plants (nC27–nC35) or a autochthonous origin from aquatic (submerged) macrophytes (nC21 and nC23; Bliedtner et al., 2018; Eglinton & Eglinton, 2008; Ficken et al., 2000; Sachse et al., 2004; Struck et al., 2020). In Lake Khar Nuur, the spatial distribution of the most abundant autochthonous (aquatic; nC21) and allochthonous (terrestrial; nC23) n-alkanes show highest concentrations in the accumulation zones in the deep basins and thus, underline the sedimentary focussing in the basins (Figure 6b, c). Compared with each other, allochthonous-derived nC23 show distinctly higher concentration in those accumulation zones than the autochthonous-derived nC21. Consequently, the PnC21 shows lower values of 0.12 to 0.42 over the whole lake, indicating a variable mixture of allochthonous and autochthonous n-alkanes (Figure 6e). Although autochthonous n-alkanes dominate throughout the lake, notable autochthonous n-alkane contributions are present in the two basins of the lake (Figure 6e). Relatively high PnC21 values close to the central southern shore can be explained by enhanced autochthonous productivity due to anthropogenic activity and input of nutrients (see section 4.3). Overall, the n-alkane preservation is good with OEP values above 5, and higher values at some sites close to the shore probably document input of poorly degraded allochthonous n-alkanes.

It is important to note that n-alkanes represent only a small fraction of the total organic matter pool in lake sediments and are possibly less influenced by degradation effects on shorter timescale compared to TOC, N and their stable isotopes (Eglinton & Eglinton, 2008; Meyers, 2003). Nevertheless, this does not hamper the overall picture that the heterogenic organic matter pool as well as the more homogenic n-alkanes comprise a mixture of autochthonous and allochthonous organic matter sources.

5.2 | The vegetation distribution in Khar Nuur sediments using leaf wax n-alkanes

Long-chain n-alkanes have the potential to differentiate between vegetation types by their homologue patterns (Bliedtner et al., 2018; Marseille et al., 1999; Schwark et al., 2002). The fundamental assumption of this approach is that the n-alkane chain-lengths C27 and C29 are thought to be produced by deciduous trees/shrubs and the chain-lengths C31 and C33 by grasses/herbs (Bliedtner et al., 2018; Marseille et al., 1999; Schwark et al., 2002). This holds also true for the Khar Nuur catchment, where distinct differences in the leaf wax patterns are apparent for sites dominated by grasses/herbs and sites dominated by the shrub Betula nana (L.) (Figure 2b). Plant samples and their respective topsoils show that sites with grasses/herbs are dominated by nC31, whereby sites with the shrubby Betula nana (L.) are dominated by nC27 (Figure 5). Those findings are underlined by recent calibration studies from Mongolia and China, where grasses/herbs show a nC31 predominance and Betula platyphylla show a nC27 predominance (Wang et al., 2018; Struck et al., 2020). Thus, leaf wax n-alkane patterns have the potential to differentiate between both vegetation types in the Khar Nuur catchment and contributions from the respective vegetation types should be differentiated in the Khar Nuur sediment surface samples (see section 4.3; Figure 5). To disentangle both vegetation types, we applied a modified normalized n-alkane ratio (cf. section 2.3.4; Equation 3; Figure 6f). However, the distinct differences of the n-alkane ratio for grasses/herbs (values > 0.66) and Betula nana (L.) (values < 0.27) found in modern plants and topsoils are not reflected in the lake surface samples (0.52–0.71) that show rather a grass/herb signal for the whole lake than a Betula nana (L.) signal. These findings are partly unexpected for the nearshore samples KN28 and KN35 in basin A (see Figure 1b, c), because Betula nana (L.) is present in the respective sub-catchments of those samples and could possibly yield lower values of the n-alkane ratio. Instead, those nearshore surface sediment samples reflect a grass signal rather than a Betula nana (L.) signal. Because both vegetation types produce similar amounts of n-alkanes in the Khar Nuur catchment (except one grass sample from the southern part of the catchment, see Table S2), we suggest that n-alkanes from the grass sites and the grassy undergrowth of the Betula nana (L.) sites in this sub-catchment are apparent in greater amounts and contribute to the grass dominated n-alkane ratio in those nearshore samples. The other surface sediment samples reflect the grass/herb signal of their respective sub-catchments, however, lower values from the n-alkane ratio are present at the central southern shore (0.52–0.55). Here, the aforementioned allochthonous input of eroded topsoil material possibly leads to higher production.
rates of aquatic (algae) n-alkanes via nutrient loading, which can also produce to some amounts the chain-length nC27 (Ficken et al., 2000) and possibly contribute to the n-alkane ratio. In this context, it is notable that the Pepe and the n-alkane ratio are significantly anti-correlated ($r = -0.73; \alpha < 0.05$; Table 1) in the Khar Nuur sediments, indicating possible contributions from aquatic plants also on the longer chain nC27. Thus, possible aquatic biases to the longer chains would complicate a clear interpretation of the n-alkane ratio in terms of vegetation types, although the grass/herb-derived proportion of nC27 dominates in all samples. Overall, the n-alkane ratio enables differentiation between the vegetation types of modern plants and respective top-soils in the Khar Nuur catchment, i.e. between grasses/herbs and Betula nana (L.). However, those differences are not reflected in the sediment samples from Lake Khar Nuur and vegetation reconstructions might be biased by aquatic n-alkane contributions.

5.3 Traces of anthropogenic activity reflected in Khar Nuur sediments

In semi-arid Mongolia, anthropogenic activity has strongly influenced soil erosion and landscape changes since the introduction of horse-borne mobile pastoralism ~5 cal. ka BP (Fernández-Giménez et al., 2017; Taylor et al., 2020). Even today, large parts of the country are used by nomadic pastoralism with large herds of sheep, goats, camels, cows/yaks and horses (Upton, 2010). However, over-grazing in semi-arid regions strongly influences landscape stability since physical soil compaction (trampling) and removal of vegetation cover favours both sheet and gully erosion and thus the irreversible loss of fine particles by erosion (Wiesmeier et al., 2015).

The Khar Nuur catchment is settled by many nomadic families and their large herds during summertime. Although the northern steep slopes of the catchment are only covered by very shallow soils and marginal vegetation, animals graze there. In contrast, especially the flatter areas in the southern part of the Khar Nuur catchment are ideal sites where the majority of grazing and family housing takes place (Figures 1d, 2a). As a result of grazing and soil exposure to weathering and erosion, several gullies in the southern part of the catchment developed deeply in the well-developed soils (Figure 2c). This anthropogenic-induced soil erosion can be traced in the surface samples of Lake Khar Nuur.

High TOC and N values are accompanied by slightly 13C enriched 13C<sub>Toc</sub> values and distinctly 15N depleted 15N<sub>TOC</sub> values at the central southern shore, indicating that large proportions of allochthonous-derived organic matter became preferentially eroded from the southern part of the catchment. Although it has to be noted that TOC and N and their isotopic composition are not direct evidence for anthropogenic activity, both show values that are high for sites outside the accumulation zones. Strongly 15N depleted 15N<sub>TOC</sub> values at the southern shore are likewise exceptional compared to the other parts of the lake and might indicate the input of organic matter from allochthonous sources. Consequently, the most likely cause for the observed TOC, N and 15N<sub>TOC</sub> distribution at the southern shore is soil erosion induced by the extensive nearby anthropogenic activity.

Those findings are supported by the conspicuous distribution of the element S, which shows highest concentrations at the central southern shore (Figure 4c). Sulphur is a part of the proteinogenic amino acids which are involved in several enzyme-reactions during the degradation of plants and faunal remains in the digestive tract and partly defecated (Brosnan & Brosnan, 2006; Kertesz & Mirleau, 2004). Consequently, S from faunal and human faeces in the central southern part of the Khar Nuur catchment can be stored in the catchment soils and eroded into the lake and/or directly transported into the lake with the faeces by surficial flow. Thus, high S concentrations at the central southern shore could be a further indicator of high allochthonous input through anthropogenic activity and soil erosion.

Traces of anthropogenic activity and soil erosion in the Khar Nuur sediments are further supported by the significant correlation of TOC, N and S ($r$ between 0.66 and 0.96; $\alpha < 0.05$). Thus, anthropogenic activity in the catchment by grazing and human occupation most likely leads to the characteristic spatial distribution of those elements in the lake. However, trace-elements of anthropogenic activity are difficult to disentangle and more detailed analyses of source-specific biomarkers are necessary to confirm this assumption.

6 CONCLUSIONS

Our grain size and biogeochemical investigations on surface sediment samples from Lake Khar Nuur, a small lake with a small hydrological catchment from the Mongolian Altai, gave the following results:

- The grain size distribution shows higher transport energy levels at the shoreline compared to the two basins of Lake Khar Nuur. The accumulation of finer sediments in those basins is accompanied by high amounts of the inorganic sediment compounds Al and Fe, which are of allochthonous origin. Consequently, the two basins act as the main sediment accumulation zones where sediment focussing is observed.
- Disentangling the autochthonous and allochthonous origin of organic material in the surface sediments is not trivial. While TOC and N accumulate preferentially in the accumulation zones, molar TOC/N ratios indicate high autochthonous production throughout the lake, which is mostly underlined by 15N depleted 15N<sub>TOC</sub> values. The respective 13C depleted 13C<sub>TOC</sub> values could indicate contributions from both allochthonous C3 plants and autochthonous production or at the shorelines changes in available dissolved inorganic carbon (CO₂) for aquatic use. However, those proxies reflect the whole heterogeneous organic matter pool which most likely comprise a mixture of both allochthonous and autochthonous sources. In contrast, the more homogenous and source-specific n-alkanes are constituents of the organic matter pool and indicate that n-alkanes from allochthonous and autochthonous sources are apparent in the surface sediments with higher proportion of allochthonous compounds compared to autochthonous ones.
- Leaf wax-derived long-chain n-alkanes in modern plants and top-soils enables differentiation between the dominating vegetation types in the Khar Nuur catchment, i.e. between grasses/herbs and the shrub Betula nana (L.). Grasses/herbs show a distinct nC29 pre-dominance and Betula nana (L.) a dominance of nC27. However, those differences are not reflected in the surface sediment samples from Lake Khar Nuur, where nC27 is the dominant n-alkane throughout the lake. This indicates that grasses become...
preferentially incorporated into the lake, also at those sites in the catchment where Betula nana (L.) is growing.

- Anthropogenic activity in the southern Khar Nuur catchment is clearly observable in the surface sediments from the central southern shoreline since TOC, N and $\delta^{15}$N indicate increased allochthonous input that is most likely due to increased soil erosion. This is accompanied by increased input of faecal remains as possibly indicated by increased contributions from the element S. However, those are only indirect indicators of anthropogenic activity and more direct analyses such as source-specific biomarkers are necessary to confirm this assumption.

Overall, sediments deposited in the accumulation zones in the two basins of Lake Khar Nuur reflect the recent environmental conditions of the lake and its hydrological catchment and are therefore ideal sediments for paleoenvironmental reconstructions. However, the proxies which possibly will be used in such paleoenvironmental studies have to be carefully evaluated by modern reference studies, although one has to be aware that environmental changes can affect the spatial distribution of sediment compounds through time. Future work on surface sediments should focus on dating the different organic matter compounds for more information about the time of the integrated signal, as well as on compound-specific isotopes of the more homogenous organic matter compounds.

DECLARATION OF COMPETING INTEREST
The authors declare no competing interests.

DATA AVAILABILITY STATEMENT
All data used in this study is available either from the corresponding author on reasonable request or from the public resources as referenced in the text.

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