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A 240,000-year stable carbon and nitrogen isotope record from a loess-like palaeosol sequence in the Tumara Valley, Northeast Siberia

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Abstract

A 15 m loess-like palaeosol sequence located in the Tumara Valley southwest of the Verkhoyansk Mountains was investigated to reconstruct the Late Quaternary environmental history of Northeast Siberia. Total organic carbon (TOC) and total nitrogen (N) show several distinct and abrupt shifts during the last 240 ka. Both proxies have generally low values (<0.5% and <0.06%, respectively) in brown and weathered horizons, which indicate accelerated soil organic matter (SOM) degradation during periods of favourable climatic conditions. On the contrary, dark horizons in the permafrost profile are characterised by higher TOC and N contents (≥1% and ≥0.12%, respectively). They probably correlate with cold glacial periods, when water logging conditions and preservation of SOM were favoured due to extensive permafrost. The δ^{13}C values of bulk SOM range from approximately −29‰ to −24‰ and show distinct shifts in concert with TOC. Based on the negative correlations of $\delta^{13}C_{org}$ with TOC ($R^2=0.49$; $n=117$) and C_{org}/N ($R^2=0.51$; $n=117$), we suggest that variations of $\delta^{13}C_{org}$ in the Tumara Profile are intensively controlled by SOM degradation. Additionally, also water stress and changes of the atmospheric CO_2 signal should have influenced our stable carbon isotope record. Contrariwise, δ^{15}N – ranging from about +1‰ to +6‰ – showed no significant correlations with our SOM degradation proxies TOC and C_{org}/N. We therefore assume that processes like denitrification, N-fixation, nitrogen losses by frequent fire events and changes in the atmospheric ^{15}N deposition contributed to an opening of the N-cycle and are thus responsible for the observed δ^{15}N signal of the Tumara Profile.

Keywords: Stable carbon and nitrogen isotopes; Palaeosols; SOM degradation; Quaternary; Siberia

1. Introduction

The reconstruction of past climate changes is necessary for a better understanding of the climate system and for the prediction of the future. High-resolution and long records are available from ice-cores and deep-sea sediments (e.g. Lisiecki and Raymo, 2005; McManus et al., 1999; NGRIP members, 2004; Schulz et al., 1998; Siegenthaler et al., 2005). Comparable terrestrial archives are lake sediments (e.g. Karabanov et al., 1998; Last and Smol, 2001; Prokopenko et al., 2001) and loess and loess-like deposits (e.g.: Chlachula, 2003; Liu et al., 1999; Muhs et al., 2003; Rousseau et al., 2002). As only a large number of records allows to investigate regional aspects of past climate and environmental changes, the need and search for further archives especially from hitherto less intensively studied areas is obvious. Here we present results of geochemical (TOC, N), grain size (clay content)

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and stable isotope analyses ($\delta^{13}C_{org}$ and $\delta^{15}N$) from the Tumara Profile. This loess-like palaeosol sequence in the southwest of the Verkhoyansk Mountains probably spans the last 240 ka (Zech et al., submitted for publication).

Within the last decades stable isotope techniques have become an increasingly important tool for current and past ecologic and climatic studies. The natural abundance of ^{13}C in bulk soil organic matter (SOM) is influenced by the input signal of the vegetation and by pedogenetic effects. Specifically, we will discuss our $\delta^{13}C_{org}$ record in terms of (i) the photosynthetic pathway of the surrounding vegetation (C3/C4 metabolic pathway), (ii) water stress, (iii) changes in the atmospheric CO_2 concentration and its isotopic signal, (iv) SOM degradation and (v) methanogenesis. Of course, it is sometimes difficult to disentangle the contribution of all these various factors and therefore especially interpretations of smaller carbon isotopic shifts may be controversial and speculative. Concerning the $\delta^{13}C_{org}$ record of the Tumara Profile, we will show that valuable information can be obtained in combination with TOC and the C_{org}/N ratio.

In contrast to the natural abundance of ^{13}C, which is widely used in palaeoecologic studies, $\delta^{15}N$ analyses have been applied much less so far. Wolfe et al. (1999), for example, studied lake sediments in arctic Russia and found that the natural abundance of ^{15}N reflects the Mid-Holocene transition from forest to tundra vegetation in the catchment. The authors suggest that higher rates of biogeochemical reworking due to warmer climate are responsible for the isotopic enrichment of sediments derived from forest soils, whereas more negative $\delta^{15}N$ values are typical for tundra-soils and lower temperatures. Another example comes from Ethiopia, where Eshetu and Högberg (2000) found isotopic enrichment in soils under relatively young or disturbed forests compared with soils from forests that were established several hundred years ago. The authors explained this difference with a land-use caused opening of the N-cycle, which led to nitrogen losses and fractionating processes.

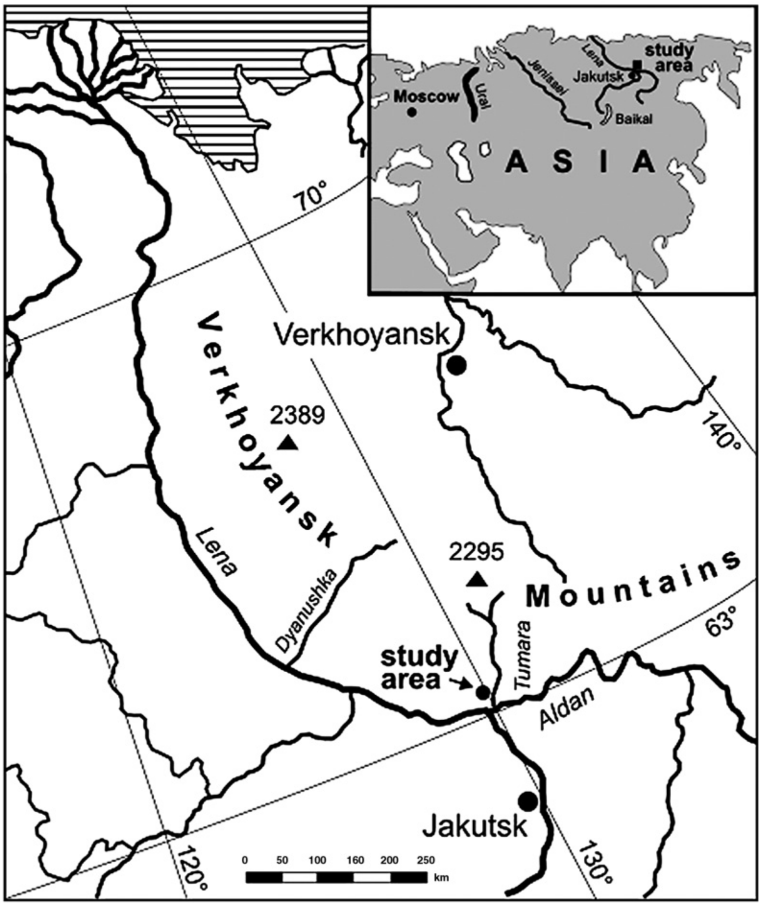

Fig. 1. Location of the study area in Northeast Siberia. The upper 15 m of an undercut slope of the Tumara River is the frozen loess-like palaeosol sequence referred to as 'Tumara Profile'. From Zech et al. (submitted for publication).

According to our knowledge, the here presented study is the first one that applies δ^{15}N analyses at high resolution in a loess-like palaeosol sequence. We will discuss the δ^{15}N variations with respect to (i) SOM degradation, (ii) biological and chemical denitrification, (iii) N-fixation, (iv) N-uptake by plants, (v) nitrogen losses by frequent fire events and (vi) changes in the atmospheric deposition of ^{15}N.

2. Geological setting, stratigraphy and chronology of the Tumara Profile

The investigated loess-like palaeosol sequence, referred to as 'Tumara Profile' (120 m a.s.l., 63°36′ N, 129°58′ E), is located about 300 km north of Jakutsk at the banks of the Tumara River, which drains part of the southern Verkhoyansk Mountains (Fig. 1). The study area is characterised by a boreal climate with a pronounced dry winter season (Jakutsk: 213 mm/a, Verkhoyansk: 155 mm/a) (Müller, 1980). Between the mountains to the north and the debouchure into the Aldan River to the south, the Tumara River cuts a 100 km long transect through Quaternary and Tertiary deposits. Moraines and terraces are typically overlain by several meters of frozen aeolian sediments. Note that in the study area we do not deal with "classical loess" as we find it e.g. in China or Western and Southeast Europe. Similar to aeolian records from Middle Siberia, where the loess is often colluvially reworked or pedogenetically imprinted (Chlachula, 2003; Frechen et al., 2005), our sediments are influenced by pedogenetic and/or cryogenetic processes. Depending on the climatic conditions – glacial or interglacial/interstadial – one can expect that either water saturated pedogenetic conditions prevailed in a thin active permafrost layer with water logging, or weathering processes (e.g. clay mineral formation) and SOM degradation dominated in well aerated and warm surface soils.

Already Kind et al. (1971) reported that the Verkhoyansk Mountains have experienced three major glacial advances during the Upper Pleistocene: the Zyryan during the Early Weichselian, the Zhigansk during the Middle Weichselian and – synchronous with the global Last Glacial Maximum (LGM) – the Sartan Glaciation during Marine Isotope Stage (MIS) 2. More recently, Svendsen et al. (2004) reconstructed four glaciations for northern Eurasia: (1) the Late Saalian (>140 ka), (2) the Early Weichselian (100–80 ka), (3) the Middle Weichselian (60–50 ka) and (4) the Late Weichselian (25–15 ka). This palaeoclimatic history can be expected to be recorded in the loess-like palaeosol sequences covering the oldest moraines and terraces in the study area.

The Tumara Profile, which was sampled during field work in summer 2002, is exposed at a ~60 m high undercut slope of the Tumara River. It is situated several kilometres outside the outermost moraine arc and the surrounding vegetation is taiga. The stratigraphy of the sampled profile is illustrated in Fig. 2B: In the lower part of the brown Unit A (0–1.8 m) several thin organic-rich layers mark the transition to a dark, silty horizon (1.8–4.8 m). This Unit B partly reveals hydromorphic features, is rich in macrofossils and has a grey band at ~3 m depth. The division in three subunits is suggested by Zech et al. (submitted for publication) and based on geochemical and granulometric results. A large ice-wedge protrudes from Subunit B3 (4.0−4.8 m) into the underlying intensively weathered, brown palaeosol (Subunit C1, 4.8–6.7 m). Subunit C2 is less weathered and slightly darker; a thin pebble layer in ~7.7 m depth may indicate a discontinuity. Subunit C3 (7.8–9.7 m) is again generally brown and more clayey than C2; its upper part is rich in red-yellow mottles. Unit D (9.7–12.5 m) is a thick dark horizon, resembling Unit B. The lowermost Unit E (12.5–15 m) is composed of a brown horizon in the upper part and a partly mottled palaeosol complex in the lower part. The basis consists of dark grey fluvioglacial sandy gravels. A more detailed description is given in Zech et al. (submitted for publication).

According to the chronology discussed in Zech et al. (submitted for publication), the Tumara Profile spans the last 240,000 years (Fig. 2D). The authors developed different palaeopedologic proxies (TOC, clay content, "U-ratio", weathering indices and D/L amino acids) indicating warm interglacial/interstadial climatic conditions or cold glacial ones. Accordingly, the brown palaeosols revealing generally low TOC but increased clay contents characterise warm and favourable periods with enhanced SOM degradation and clay mineral production. On the contrary, SOM accumulation and reduced brunification are characteristic for dark palaeosol, which developed during cold/glacial periods. The palaeopedologic proxies were used to derive a "warm vs. cold stratigraphy", in which the numeric dating results were be embedded and allow a tentative correlation with marine isotope stages (MIS). A short summary of the chronological proposal of Zech et al. (submitted for publication) is given in the following:

Despite of inconsistencies in the radiocarbon ages, the Holocene is supposed to be represented by the brunified Unit A (Fig. 2). Presumably, ^{14}C ages are overestimated for instance by the deposition of "too old" recalcitrant windblown carbon or by mobilisation and vertical translocation of "too old" carbon with ascending water from underlying and formerly permafrosted palaeosols.

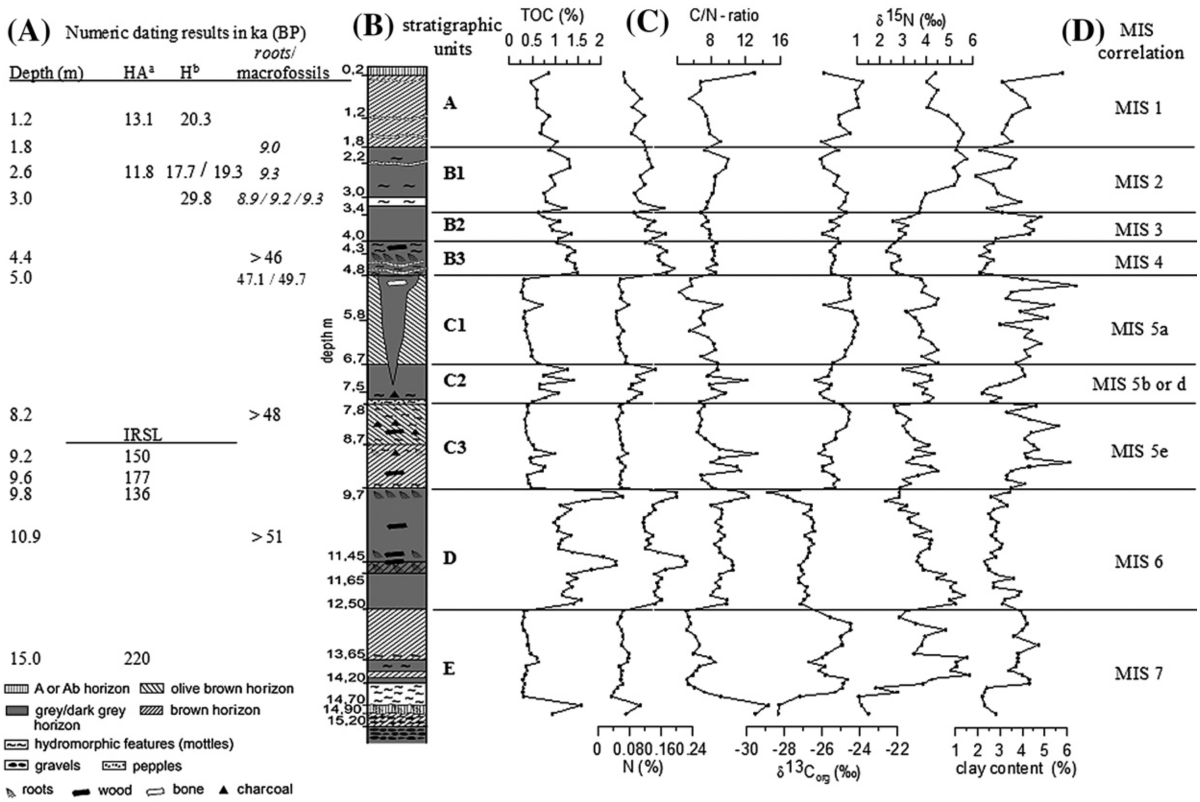

Fig. 2. (A) Numeric dating results in ka BP, (B) stratigraphy, (C) depth-profiles of the analysed parameters (TOC, N, C/N, $\delta^{13}C_{org}$, $\delta^{15}N$ and clay content) and (D) correlation of the stratigraphic units with marine oxygen isotope stages. Modified after Zech et al. (submitted for publication).

Although several radiocarbon ages of approximately 9 ka BP were derived for roots down to 3 m depth (Fig. 2A), Subunit B1 is supposed to have been deposited during MIS 2. Note that this inconsistency among the datable materials does not necessarily belittle the chronological and palaeoenvironmental interpretation, but may enrich it: probably, the warm Late Glacial and Early Holocene summers caused an intensive thawing of the active permafrost layer and allowed roots to penetrate this depth (3 m). This is in agreement with the so-called Boreal thermal optimum reported by Andreev et al. (2002) for Northern Siberia.

The Subunits B2 and B3 can tentatively be correlated with MIS 3 and 4 (Fig. 2D). The three radiocarbon ages around 48 ka BP at the basis of Unit B are expected to be minimum ages. Cold glacial conditions during the early MIS 3 – from 50 to 60 ka BP according to Svendsen et al. (2004) – likely provoked the epigenetic ice-wedge to protrude into the underlying brown palaeosol, which is supposed to correspond to MIS 5a. Subunit C2 probably represents the Early Weichselian glaciation of Svendsen et al. (2004) (MIS 5b) and overlies the Eemian soil (MIS 5e: Subunit C3), for which 3 IRSL ages were obtained (136, 150 and 177 ka). Based on the comparison of TOC in the Tumara Profile with marine oxygen isotope records and another IRSL age from Unit E (220 ka), Zech et al. (submitted for publication) correlate Unit D with MIS 6 and Unit E with MIS 7, respectively.

3. Materials and methods

After cleaning the upper 15 m of the Tumara Profile (100 cm width and 30 cm depth), a total number of 117 samples were taken at 10 to 20 cm intervals. The samples were air dried, sieved (<2 mm) and stored in plastic bags. Analyses were performed following standard laboratory procedures at the Institute of Soil Science and Soil Geography, University of Bayreuth. Total organic carbon (TOC) and total nitrogen (N) were determined after removal of carbonate (10% HCl) by dry combustion of a finely ground homogeneous 50 mg sub-sample followed by thermal conductivity detection on a Vario EL elemental analyser (Elementar, Hanau, Germany). The detection limits of our machine were calculated by measuring blanks with increasing net weights of wolfram oxide in tin capsules (~0.0002% for TOC and ~0.007% for N). Precision was determined by measuring acetanilide as standard in quadruplicate. Mean standard errors were better than 0.02% for TOC and 0.05% for N, respectively.

The δ^{13}C and δ^{15}N values of the bulk organic matter (OM) were obtained by dry combustion of decalcified subsamples on a Carlo Erba NC 2500 elemental analyser coupled with a Delta$^{\text{plus}}$ continuous-flow isotope ratio mass spectrometer (Thermo Finnigan MAT, Bremen, Germany) via a Conflow II interface (Thermo Finnigan MAT, Bremen, Germany). Sucrose (ANU, IAEA, Vienna, Austria), $CaCO_3$ (NBS 19, Gaithersburg, USA), and ammoniumsulfate (N1 and N2, both IAEA, Vienna, Austria) were used as calibration standards. Natural abundances of carbon and nitrogen stable isotopes are expressed in the usual δ-scale in parts per thousand according to the equation

$$\delta_{\text{sample}}(‰) = \left(\frac{R_{\text{sample}} - R_{\text{standard}}}{R_{\text{standard}}}\right) \times 1000,$$

where R_{sample} and R_{standard} are the ^{13}C/^{12}C or ^{15}N/^{14}N abundance ratios of a sample or a standard, respectively. Precision was determined by measuring acetanilide as standard in quadruplicate. Mean standard errors were better than 0.15‰ for δ^{13}C and 0.25‰ for δ^{15}N, respectively. Grain-size analyses were performed on a Beckman Coulter particle size analyser after the removal of OM with H_2O_2 (30%) and the removal of carbonate with HCl (10%).

4. Results and discussion

The depth-profiles of TOC, N, C/N, $\delta^{13}C_{\text{org}}$, δ^{15}N and the clay content are illustrated in Fig. 2C. All parameters show distinct shifts for most transitions between brown and dark grey horizons and therefore corroborate the stratigraphic units deduced from field observations.

4.1. Carbon and nitrogen contents

The organic carbon and nitrogen contents display similar variations throughout the profile and range from 0.27% to 2.48% and from 0.04% to 0.22%, respectively (Fig. 2C). Several abrupt shifts occur at the transitions from the brown units (C1, C3 and E) to the dark grey units (B, C2 and D), with TOC and N being generally low in the former and high in the latter ones. High TOC and N values also coincide with higher C/N ratios (Fig. 2C).

On the one hand, the elemental composition of litter may influence the C/N ratio in soils and cannot be completely ruled out. On the other hand, decomposition of SOM lowers the C/N ratio due to the loss of carbon in form of CO_2. However, we assume that the latter effect dominates in our record and hence TOC and C/N are appropriate proxies for SOM degradation. As one may expect, reduced SOM degradation generally coincides with reduced weathering, the latter being inferred from lower clay contents in the Units B1, B3, C2 and D. Vice versa, the brown units are characterised by higher clay contents (increased weathering) and lower TOC and C/N ratios (intensive SOM degradation).

With regard to the chronostratigraphy (Fig. 2D), our findings show that warm climatic conditions during MIS 1, 3, 5 and 7 favoured SOM degradation and weathering (Units A, C and E), whereas during the cold periods MIS 2, 4, 5b/d and 6 weathering and SOM degradation were reduced (Units B1, B3, C2 and D).

The comparison with northern hemispheric loess records reveals that there is a striking difference between the Tumara Profile and European or Chinese loess profiles. In the Tumara Profile high TOC values correlate with glacial periods. In Europe and China, on the contrary, the interglacial and interstadial palaeosols are often enriched in OM indicating higher biomass production, increased moisture availability and low dust accumulation rates (Bronger, 2003; Fink, 1962; Hatté et al., 1999; Zöller et al., 2004). This difference can be explained with permafrost being a crucial factor for pedogenetic processes in Northeast Siberia. Whereas biomass production was probably never a limiting factor for the accumulation of OM due to still relatively warm summers even during the LGM in Siberia (Tarasov et al., 1999), pedogenetic conditions at our study site presumably differed depending on the climate: during cold glacial periods water logging conditions prevailed in the thin active permafrost layer and inhibited SOM degradation in the topsoil. On the contrary, warmer summers during the interstadials and interglacials were characterised by both deeper melting of the permafrost and hence better drainage and increased evaporation. As in these better aerated topsoils SOM mineralisation was intensified, TOC cannot only be interpreted as “warm vs. cold” proxy in our record, but also as proxy for the degree of SOM degradation. This will be of importance when discussing the natural abundance of ^{13}C in the following paragraphs.

4.2. Natural abundance of ^{13}C

According to Fig. 2C, δ^{13}C values obtained for bulk SOM range from −28.9‰ to −23.9‰. They display systematic variations throughout the profile with more negative values coinciding with TOC maxima (Units D, C2 and B). Besides, there is a general tendency towards more positive $\delta^{13}C_{org}$ values in the younger sediments over the whole profile.

Numerous processes can potentially influence the natural abundance of SOM ^{13}C in soils and/or sediments. This makes a palaeoenvironmental reconstruction with bulk $\delta^{13}C_{org}$ values alone sometimes speculative. On the one hand, $\delta^{13}C$ variations of the deposited OM have to be taken into consideration. On the other hand, pedogenetic processes can lead to isotopic fractionation, too. Both aspect will be discussed in the following for the $\delta^{13}C_{org}$ record of the Tumara Profile.

In many studies, varying contributions of *C3 versus C4 plants* were found to be responsible for changing stable carbon isotope compositions in loess-palaeosol sequences or other palaeoenvironmental archives (e.g.: Aucour et al., 1999; Freitas et al., 2001; Liu et al., 2005b; Wang and Follmer, 1998; Wang et al., 2000). These studies are based on the fact that the photosynthetic pathway of C3 plants produces rather depleted $\delta^{13}C$ values of approximately −27‰ (O'Leary, 1988). On the contrary, the C4-metabolism, which is more competitive under drier and/or warmer conditions and lower atmospheric CO_2 concentrations, leads to plant $\delta^{13}C$ values around −14‰ (Collatz et al., 1998). As one may expect, in the Tumara Profile all $\delta^{13}C_{org}$ values are well within the range of SOM derived from C3 vegetation. The absence of C4 plants is typical for temperate and cold environments and has the advantage that other ecological factors affecting $\delta^{13}C_{org}$ in sediments can be investigated more easily.

One of the main environmental factors known to control the $\delta^{13}C$ signature of plants is *water stress*. Plants react on it with stomata narrowing, which results in lower intercellular CO_2 concentrations, reduced isotope fractionation and thus more positive $\delta^{13}C$ values of the synthesized organic compounds (Farquhar et al., 1982; O'Leary, 1995; Schliesser, 1995). Recently, Liu et al. (2005a) and Stevenson et al. (2005) have shown for plants and soils, respectively, that $\delta^{13}C$ decreases (up to 5‰) with increasing rainfall along precipitation gradients in arid regions of China and the USA. Hatté and Guiot (2005) used this relationship to reconstruct the palaeoprecipitation by using the isotopic signal of loess OM in the Nuβloch loess sequence (Rhine Valley, Germany). A key assumption for their modelling is the absence of pedogenesis in typical loess. According to the authors, this implies that the dry glacial environment favoured the degradation of OM without distortion of the isotopic signal.

Reminding our interpretation of TOC and N for the Tumara Profile (reduced SOM degradation in water logged active permafrost layers versus enhanced SOM degradation in well aerated active permafrost layers), the more positive $\delta^{13}C_{org}$ values in the generally OM-depleted Units A, C1, C3 and E could also be interpreted in terms of water stress. In these units, water could have become a limiting factor for plants during the growing season, whereas no water stress should have occurred during the deposition of the water logged Units B, C2, and D. Furthermore, this mechanism could explain the general tendency towards more positive $\delta^{13}C_{org}$ values in the younger sediments. On the one hand, the thus deduced increasing aridity of the study site during the Late Quaternary can be seen in a large-scale context, as e.g. Svendsen et al. (2004) reported that the Northern Siberian ice sheets got progressively smaller during the last four glaciations (covering the last 160,000 years) and this can be explained with decreasing precipitation. On the other hand, one may not forget the local setting being characterised by the Tumara cliff edge, which approached our study site during the Late Quaternary. Accordingly, wind exposure increased and plants hence should have suffered more and more water stress.

Another important environmental factor, which is supposed to influence $\delta^{13}C$ of plants, is the *CO_2 concentration* in the atmosphere. Lower CO_2 concentrations during glacials (~100 ppm, e.g. Petit et al., 1999) lead to reduced isotopic fractionation, resulting in $\delta^{13}C$ values that are 0.02‰ more positive per 1 ppm CO_2 (Feng and Epstein, 1995). In the Tumara Profile, however, the dark glacial deposits (Units B, C2 and D) would have to be enriched. As this is not the case, we conclude that the changing atmospheric CO_2 concentration does not explain but rather dampens the observed $\delta^{13}C_{org}$ signal. Concerning the *carbon isotope signature* of the atmospheric CO_2, which can be measured on air trapped in ice-cores, only minor changes (0.3±0.2‰) have been reported for glacial–interglacial transitions (Leuenberger et al., 1992). They can therefore be neglected.

Whereas according to Hatté and Guiot (2005) *SOM degradation* does not influence the $\delta^{13}C_{org}$ signal in typical loess records, such fractionation can certainly not be ruled out for our loess-like palaeosol sequence, which is characterised by the alteration of different pedogenetic conditions. Many soil studies reported increasing ^{13}C abundance with soil depth being correlated with a decrease of TOC and with age of SOM despite the absence of C3–C4 shifts (Andreux et al., 1990; Balesdent et al., 1993; Bol et al., 1999; Chen et al., 2002; Krull et al., 2002; Nadelhoffer and

Fry, 1988; Stevenson, 1997). Although the exact mechanisms are not yet well understood, this pattern is mainly attributed to SOM decomposition. Note that there are also studies of aquatic sediments, peat bogs and litter decay revealing no ^{13}C enrichment or even ^{13}C depletion in more degraded OM in contrast to well preserved OM (Balesdent et al., 1993; Benner et al., 1987; Meyers and Ishiwatari, 1993; Pancost et al., 2003; Spiker and Hatcher, 1984; Van Kaam-Peters et al., 1998). According to Balesdent and Mariotti (1998), these apparently divergent results can however be reconciled in an OM decomposition model considering both a ^{13}C enrichment due to OM degradation (e.g. by microbial respiration) and a slower because inhibited decay of ^{13}C-depleted compounds (e.g. lignin) during certain steps of OM decomposition. Reviewing the literature, we conclude that OM degradation in soils – and hence also in our loess-like palaeosol sequence – can account for a ^{13}C enrichment of up to 2–3‰ in temperate and boreal environments.

We tried to assess the SOM degradation effect on $\delta^{13}C_{org}$ in the Tumara Profile by applying cross-plot-analyses. As outlined above, TOC and C/N can be used as proxies for SOM degradation in our study. A significant positive correlation between TOC and C/N ($R^2_{total}=0.46$, $n=117$) corroborates our previous TOC interpretation (low TOC contents are caused by SOM degradation as indicated by coinciding low C/N ratios) and also holds true for most individual stratigraphic units (Fig. 3A). Correlation coefficients range from $R^2=0.06$ ($n=8$, Subunit B3) to $R^2=0.95$ ($n=16$, subunit C3). Trendlines with slopes of ~2 are characteristic for the dark organic-rich units, whereas the slopes are much steeper for the intensively weathered brown units (~10). Both TOC and C/N are negatively correlated with $\delta^{13}C_{org}$ ($R^2_{total}=0.49$ and $R^2_{total}=0.51$, respectively, see Fig. 3B and C). The correlation coefficients for the individual stratigraphic units range from $R^2=0.09$ (C/N vs. $\delta^{13}C_{org}$ in Subunit B3, $n=8$) to $R^2=0.95$ (C/N vs. $\delta^{13}C_{org}$ in Subunit B3, $n=8$). We conclude that most likely SOM degradation exerted a dominant control on TOC, C/N and likewise on $\delta^{13}C_{org}$ of the Tumara Profile.

Our interpretation is in agreement with findings from other permafrost soils in Siberia: Pfeiffer and Jansen (1993) and Gundelwein (1998) found more negative δ^{13}C values in hydromorphic, humic-rich soils in comparison to well drained soils that were enriched in ^{13}C. The authors explained their findings with differences in the SOM decomposition and with anaerobic SOM transformation processes, although climatically induced variations of the plant communities were not completely ruled out (Gundelwein, 1998). The data of Schirrmeister et al. (2002), who studied Ice Complex deposits in the Laptev Sea region, Northern Siberia, reveal the same negative correlation between TOC and C/N versus $\delta^{13}C_{org}$ as our results do. Their 60 ka record, however, shows two distinctive differences compared to our record: firstly, lower TOC contents and more positive $\delta^{13}C_{org}$ values for MIS 2 sediments and TOC maxima and $\delta^{13}C_{org}$ minima for Holocene and MIS 3 layers (i.e. that is the direct opposite signal in the Tumara Profile); secondly, the TOC contents in the peaty Ice Complex deposits are by factor 10 higher than in our loess-like profile. This likely reflects the different environmental conditions at the two study sites, although similar mechanisms could be responsible for the $\delta^{13}C_{org}$ signals.

Another relevant process that might be important when explaining fluctuations of $\delta^{13}C_{org}$ values is *methanogenesis*. Methane emitted from wetlands is strongly depleted in ^{13}C (Bréas et al., 2001). This readily leads to ^{13}C enrichment of soils. Several studies determined methane production rates for various environments (Whiting and Chanton, 1993). Generally, methanogenic bacteria are suppressed by oxygen and high CH_4 emissions only occur when the groundwater table and the soil temperature are high (>10 °C) (Dunfield et al., 1993; Svensson, 1984). According to our interpretation, these two conditions rather exclude each other at our study site. Either the soils were well drained during warm periods, or water logged during cold periods. We therefore assume that methanogenesis is less important than water stress and SOM degradation for the observed $\delta^{13}C_{org}$ variations in the Tumara Profile.

4.3. Natural abundance of ^{15}N

The δ^{15}N values of the Tumara Profile vary from +1‰ and +6‰ and are well within the range typically found in soils (Nadelhoffer and Fry, 1988). The Units A and C generally reveal smaller δ^{15}N fluctuations around +4‰, whereas distinct long-term trends occur in the organic-rich Units B and D. The largest variations occur in the lowermost Unit E (Fig. 2C).

Typically, δ^{15}N is higher in soils compared to the surrounding vegetation and increases with soil depth (Nadelhoffer and Fry, 1988). This points to an isotopic enrichment during *SOM degradation*. We can test this effect on our δ^{15}N record in a similar way as previously performed for $\delta^{13}C_{org}$. Interestingly, no significant correlation can be found between δ^{15}N and any of the other organic parameters (TOC, C/N and $\delta^{13}C_{org}$),

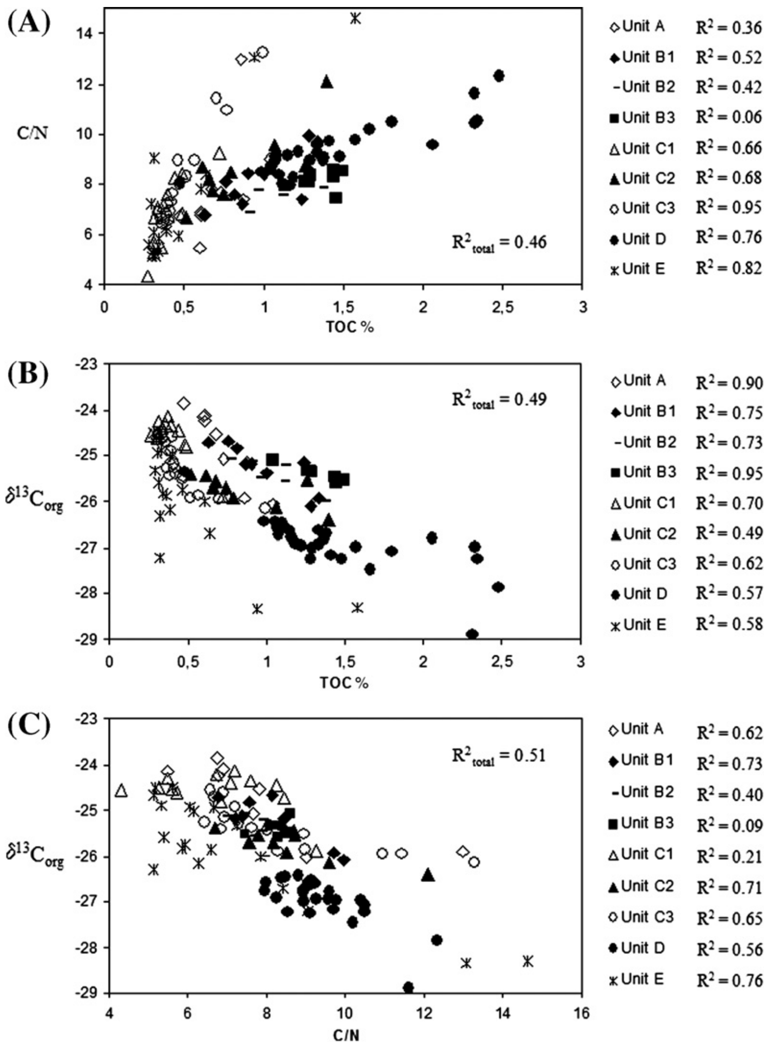

Fig. 3. Correlation between TOC, C/N and $\delta^{13}C_{org}$ ($n=117$): (A) TOC vs. C/N ($R^2_{total}=0.46$), (B) TOC vs. $\delta^{13}C_{org}$ ($R^2_{total}=0.49$) and (C) C/N vs. $\delta^{13}C_{org}$ ($R^2_{total}=0.51$). Correlation coefficients for individual stratigraphic units are given in the legend. The highly significant correlations indicate that $\delta^{13}C_{org}$ in the Tumara Profile is intensively influenced by SOM degradation.

which are controlled by SOM degradation. Other processes of the N-cycle apparently exert a dominant control on the nitrogen isotopic composition in the Tumara Profile.

It is well-known that *denitrification* strongly discriminates against the heavier ^{15}N-isotope and causes a preferential emission of ^{14}N in form of N_2 and N_2O. This results in more positive $\delta^{15}N$ values in the remaining SOM. Dörsch et al. (1993), for instance, found significant N_2O emissions especially during melting after frost periods — probably due to short-term but strongly increased microbial activity. Papen and Butterbach-Bahl (1999) confirmed the importance of freeze/thaw cycles and emphasized that also the forest type greatly influences the N_2O release. Their beech control site released 4–5 times more N_2O than their spruce control site. Chemical denitrification, as opposed to biological denitrification, might play a role at temperatures just below 0 °C. Christianson and Cho (1983) measured N_2 emissions and showed that the chemical denitrification generally decreases with temperature, however, at −3.5 °C it suddenly increases again and reaches values as they are typical for ~20 °C. They explained this temperature anomaly with higher solute concentration of NO_2^- in the unfrozen interfacial water, which exists at temperatures as low as −40 °C.

We tentatively conclude that the very positive $\delta^{15}N$ values in the lower parts of the Units A and D as well as in Unit B1 and partly in Unit E may be caused by extraordinarily frequent freeze/thaw cycles having occurred in these sediments (Fig. 2C). At least for the Units A and B1 the numeric dating result support this idea. Several ^{14}C ages around 9 ka BP – obtained for roots down to 3 m depth – indicate a regular and deep melting of the active permafrost layer during the warm summers of the Late Glacial and Early Holocene climatic optimum. However, the $\delta^{15}N$ pattern for the whole profile is not consistent with the alternating brown and dark grey palaeosols. Therefore denitrification is unlikely to be the only process responsible for the observed nitrogen isotopic composition.

As mentioned before, vegetation changes may affect the biological denitrification. Additionally, vegetation changes could influence the nitrogen isotopic composition of soils especially when atmospheric nitrogen is fixed. *N-fixation* is well-known to cause more negative $\delta^{15}N$ values of the biomass and hence also of the SOM. Benson and Silvester (1993) reported that in cold environments N-fixation by actinomycetes (*Francia*), living in symbiosis with trees and shrubs, especially alnus, may be underestimated. Apart from N-fixation, further differences in the *N-uptake* have to be considered when interpreting $\delta^{15}N$ variations in plants. For example the arctic sedge *Eriophorum vaginatum*, gets up to 60% of their nitrogen in form of free amino acids instead of inorganic NO_3^- and NH_4^+ (Chapin III et al., 1993). This confirms that different soil nitrogen sources contribute to the wide range of $\delta^{15}N$ values found in various plant species collected from tundra ecosystems — e.g. very low values (−8 to −6‰) in *Picea* and ericaceous species and even positive values in sedges and grasses (Nadelhoffer et al., 1996; Schulze et al., 1994). Accordingly, apart from SOM degradation N-uptake by plants is often accounted when interpreting ^{15}N enrichment with soil depth. However, on the one hand, we have no stable surface in our accumulating loess-like sediments — making considerations concerning this isotopic depth gradient difficult. On the other hand, we argue that the N-uptake by plants (N-fixation is not included here) should not have the potential to alter the $\delta^{15}N$ signal if the N-cycle remains closed and all plant material converts again into SOM by decomposition.

Therefore, other mechanisms leading to gains/losses of depleted/enriched N-pools should be further investigated. For instance, frequent *fire events* could have contributed significantly to an open N-cycle and thus to a preferential loss of isotopically lighter plant OM. Apart from nitrogen losses, also changes in the nitrogen input by dry and wet *atmospheric deposition* have to be considered. Rain contains significant amounts of dissolved nitrogen (as NO_3^-, NO_2^- and NH_4^+) with relatively low $\delta^{15}N$ values (Heaton, 1987; Paerl and Fogel, 1994), whereas aeolian dust containing SOM is relatively enriched in ^{15}N. The distinctive shift from low $\delta^{15}N$ values in Unit B2 and 3 to very positive values in Unit B1 could thus indicate decreasing wet and increasing dry nitrogen deposition. This would confirm the trend towards more arid conditions in Siberia in the course of the last glacial cycle (Hubberten et al., 2004; Svendsen et al., 2004). Whether changing atmospheric ^{15}N deposition contributed to the observed trend in Unit D or the large variations in Unit E is however even more speculative.

5. Conclusions

In this study we presented the carbon and nitrogen contents and the respective isotopic compositions of a probably ~240,000 year old loess-like palaeosol sequence in the Tumara Valley, Northeast Siberia. In contrast to European and Chinese loess records, glacial and stadial soils in the Tumara Profile are characterised by higher TOC and N contents than the interglacial and interstadial ones. Water logging conditions likely favoured the preservation of SOM during cold periods with extensive permafrost. The highly significant correlations between TOC, C/N, $\delta^{13}C_{org}$ and the clay content suggest that all these parameters reflect the alternation of accelerated and reduced SOM degradation/weathering. Although probably intensively controlled by SOM degradation, the $\delta^{13}C_{org}$ record should have also been influenced by water stress and the atmospheric CO_2 signal.

The interpretation of $\delta^{15}N$ is more speculative. SOM degradation probably affects the nitrogen isotopic composition, but the lack of a significant correlation with TOC, C/N or $\delta^{13}C_{org}$ indicates that additional processes of the N-cycle played a crucial role. For instance, denitrification, depending on temperature and on the frequency of freeze/thaw cycles, causes ^{15}N enrichment of soils and in all likelihood influenced the $\delta^{15}N$ record of the Tumara Profile. Vegetation changes should not have contributed significantly to the observed $\delta^{15}N$ variations, if they were not accompanied by an opening of the N-cycle. On the contrary, N-fixation and nitrogen losses by fire events had the potential to alter the $\delta^{15}N$ pattern. Eventually, dry and wet atmospheric N-deposition should be further investigated because of their different isotopic signal.

Both $\delta^{13}C_{org}$ and $\delta^{15}N$ are subjected to pedogenetic processes, which have to be considered carefully when deriving palaeoclimatic information from SOM stable isotopic variations. The application of compound-specific isotope analyses on degradation-resistant biomarkers could be a promising methodological approach in order to disentangle the influence of SOM degradation on $\delta^{13}C_{org}$. Palynological work for the Tumara Profile is in progress.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemgeo.2007.04.002.

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