



Evidence for past variations in methane availability in a Siberian thermokarst lake based on $\delta^{13}\text{C}$ of chitinous invertebrate remains

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ABSTRACT

Understanding past methane dynamics in arctic wetlands and lakes is crucial for estimating future methane release. Methane fluxes from lake ecosystems have increasingly been studied, yet only few reconstructions of past methane emissions from lakes are available. In this study, we develop an approach to assess changes in methane availability in lakes based on $\delta^{13}\text{C}$ of chitinous invertebrate remains and apply this to a sediment record from a Siberian thermokarst lake. Diffusive methane fluxes from the surface of ten newly sampled Siberian lakes and seven previously studied Swedish lakes were compared to taxon-specific $\delta^{13}\text{C}$ values of invertebrate remains from lake surface sediments to investigate whether these invertebrates assimilated ^{13}C -depleted carbon typical for methane. Remains of chironomid larvae of the tribe Orthocladinae that, in the study lakes, mainly assimilate plant-derived carbon had higher $\delta^{13}\text{C}$ than other invertebrate groups. $\delta^{13}\text{C}$ of other invertebrates such as several chironomid groups (*Chironomus*, Chironomini, Tanytarsini, and Tanytarsini), cladocerans (*Daphnia*), and ostracods were generally lower. $\delta^{13}\text{C}$ of Chironomini and *Daphnia*, and to a lesser extent Tanytarsini was variable in the lakes and lower at sites with higher diffusive methane fluxes. $\delta^{13}\text{C}$ of Chironomini, Tanytarsini, and *Daphnia* were correlated significantly with diffusive methane flux in the combined Siberian and Swedish dataset ($r = -0.72$, $p = 0.001$, $r = -0.53$, $p = 0.03$, and $r = -0.81$, $p < 0.001$, respectively), suggesting that $\delta^{13}\text{C}$ in these invertebrates was affected by methane availability. In a second step, we measured $\delta^{13}\text{C}$ of invertebrate remains from a sediment record of Lake S1, a shallow thermokarst lake in northeast Siberia. In this record, covering the past ca 1000 years, $\delta^{13}\text{C}$ of taxa most sensitive to methane availability (Chironomini, Tanytarsini, and *Daphnia*) was lowest in sediments deposited from ca AD 1250 to ca AD 1500, and after AD 1970, coinciding with warmer climate as indicated by an independent local temperature record. As a consequence the offset in $\delta^{13}\text{C}$ between methane-sensitive taxa and bulk organic matter was higher in these sections than in other parts of the core. In contrast, $\delta^{13}\text{C}$ of other invertebrate taxa did not show this trend. Our results suggest higher methane availability in the study lake during warmer periods and that thermokarst lakes can respond dynamically in their methane output to changing environmental conditions.

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1. Introduction

Methane is an important greenhouse gas in the atmosphere, accounting for approximately 20% of the radiative forcing of all greenhouse gasses (Wuebbles and Hayhoe, 2002; Forster et al., 2007). Predictions of future climate change are highly dependent on accurate predictions of the release and uptake of methane within different compartments of the global carbon cycle under changing environmental conditions. Wetlands, including lakes, are amongst

the most important natural sources of methane to the atmosphere (Zimov et al., 1997; Huttunen et al., 2003; Bastviken et al., 2004; Huber et al., 2006). It has been estimated that natural methane sources emit between 190 and 220 Tg CH₄ y⁻¹ (Denman et al., 2007), and that lakes contribute 71.6 Tg CH₄ y⁻¹ (Bastviken et al., 2011).

Atmospheric methane has been fluctuating over the glacial–interglacial cycles, with concentrations ranging between 350 and 750 ppbv (Brook et al., 1996; Loulergue et al., 2008). It is believed that methane from wetlands and lakes has exerted an important positive feedback to climate change, especially in sensitive regions such as the Arctic (Wuebbles and Hayhoe, 2002; Walter et al., 2006), but very few estimates of past methane emissions from lakes are available (Walter et al., 2007a; van Huissteden et al., 2011). Most reconstructions of methane release from lakes are based on estimates of past changes in the overall surface area comprised of lakes and assume a similar methane release per unit area in the past as measured at present. However, the individual response of lakes in their methane output under changing environments is still poorly studied. It is, therefore, uncertain how methane output per unit area of lake surface has changed in the past and how this process will vary with changing climate.

In lakes, methane is produced by methanogenic bacteria during the degradation of organic matter in anoxic waters or in anoxic sediment (Rudd and Taylor, 1980; Bartlett and Harriss, 1993). This methane is characterized by distinctly depleted stable carbon isotope values relative to the organic matter it is formed from (Whiticar, 1999). Methane can be released from the sediment into the lake water and further into the atmosphere by a range of processes such as diffusion, in-lake currents (e.g. vertical mixing during spring and autumn overturning), transport via vascular plants, and ebullition (bubbles percolating through the sediment and water column) (Kankaala et al., 2007; Walter et al., 2007b; Bastviken et al., 2008). During transport from the sediment a considerable amount of this methane may be oxidized by chemical processes or methane oxidizing bacteria (MOB) (Frenzel et al., 1990; Bastviken et al., 2002; Kankaala et al., 2007).

The distinct carbon isotopic composition of methane leads to very low δ¹³C values in the tissue of aquatic invertebrates feeding on MOB (Bastviken et al., 2003; Kankaala et al., 2006; Jones et al., 2008). δ¹³C of aquatic invertebrates can, therefore, provide insights into the relative importance of MOB in their diet and, indirectly, on whether methane-derived carbon is recycled into a lake's food web. Some aquatic invertebrates produce robust chitinous structures that preserve well in lake sediments and can be used to assess the importance of methane-derived carbon in lake ecosystems in the past (Heiri et al., 2009; van Hardenbroek et al., 2010a). For example, in seven lakes studied in south-central Sweden by van Hardenbroek et al. (2011b) the remains of chironomid larvae of the tribe Chironomini had lower δ¹³C in lakes, and

at sites within lakes, with higher methane production. A significant correlation was observed between δ¹³C of Chironomini and diffusive methane fluxes. It is unclear, however, to what extent this relationship also exists in lakes in other regions.

We examined ten thermokarst lakes in arctic northeast Siberia and compared measured diffusive methane fluxes at the lake surface with δ¹³C of chitinous invertebrate remains from surface sediments of these lakes. In addition, we extended the study of Swedish lakes by van Hardenbroek et al. (2011b) by analysing remains of additional invertebrate taxa. Whereas the previous study focused exclusively on the remains of chironomid larvae, we here also analysed δ¹³C of the remains of the planktonic cladoceran (water flea) genus *Daphnia* and of ostracod (seed shrimp) cuticles in surface sediments, both for the Siberian lakes and the Swedish lakes studied previously by van Hardenbroek et al. (2011b). The aim of these analyses was to assess which invertebrate groups are most sensitive in their δ¹³C to the availability of methane and to examine the relationship between δ¹³C of different invertebrates and diffusive methane flux in a combined dataset consisting of both the Siberian and Swedish lakes. In a second step, we then analysed δ¹³C of chitinous invertebrate remains in a sediment core providing a thousand-year record from a Siberian thermokarst lake named Lake S2. We interpret the changes in δ¹³C of the fossil remains in the context of the modern, taxon-specific relationships between invertebrate δ¹³C and methane flux established based on the studied surface sediments. This allows an assessment of whether invertebrate δ¹³C in Lake S2 changed to values typical of increased or reduced methane availability and methane output during different periods in the past millennium.

2. Materials and methods

2.1. Site description

The main study area (70°48' N, 147°26' E) is situated in the Kytalyk wildlife reserve, 30 km northwest of the town of Chokurdakh (Yakutia, Russia; Fig. 1). Surface sediments from ten shallow thermokarst lakes in the arctic tundra were examined in this study (Table 1). Lakes N1 to N8 are located on the floodplain of the River Elon, a tributary of the River Indigirka. With the exception of Lake N2 these lakes are flooded by the river regularly during snowmelt. Lake S1 and Lake S2, the site where the downcore sediment record was obtained from, are located on higher tundra and above river influences. The study area has a very strong continental climate with mean annual, January, and July temperatures of −10.5, −34.2, and 10.4 °C, respectively (van der Molen et al., 2007). The lakes are ice-covered from September until May/June and, therefore, have a very short open-water season. Thermokarst lakes generally develop when ice-rich permafrost soils thaw, causing subsidence and the formation of lakes that increase in size

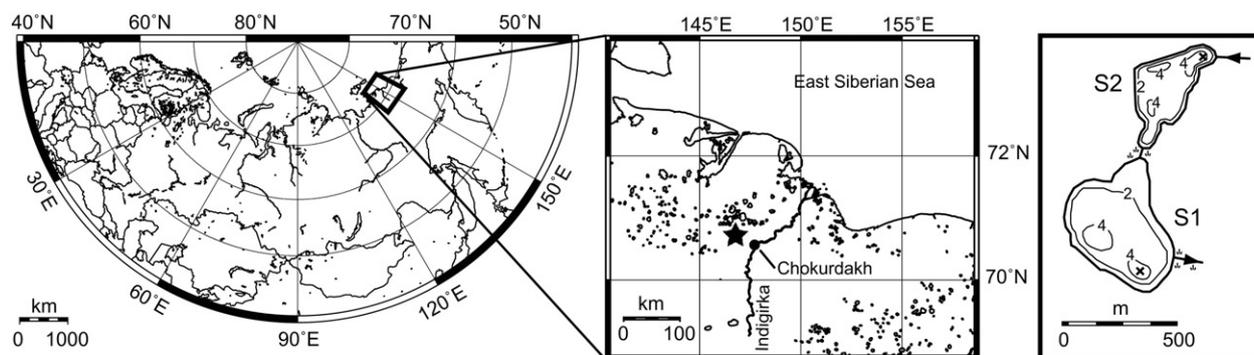


Fig. 1. Study area (star) near the town Chokurdakh in arctic Siberia. Location of lakes S1 and S2 is shown in right panel, including bathymetric maps and coring locations (marked by X).

Table 1
Physical characteristics, water chemistry, and bulk sediment geochemistry of the studied lakes. Diffusive methane fluxes are average values of two campaigns in 2007 and 2009. Water chemistry values are based on average values of two water samples taken at 0.5 m depth and at 1 m from the bottom.

Lake	N1	N2	N3	N4	N5	N6	N7	N8	S1	S2
Coordinates	70°49'29"N 147°27'13"E	70°49'47"N 147°25'40"E	70°49'16"N 147°25'16"E	70°49'33"N 147°30'36"E	70°49'34"N 147°30'54"E	70°49'28"N 147°31'50"E	70°49'21"N 147°32'8"E	70°48'25"N 147°29'50"E	70°44'26"N 147°34'53"E	70°44'47"N 147°35'8"E
Elevation (m a.s.l.)	5	5	5	5	5	5	5	5	7	7
Surface area (ha)	9.2	50	3.7	0.9	1.1	1.5	2.5	3.7	13.7	5.7
Maximum depth (m)	2.3	5.6	2.6	2.9	3.3	5.4	3.1	4.6	5.5	5.2
Secchi depth (m)	1.8	2.8	n.a.	1	2.75	1.8	1.7	1.5	3.3	2.5
CH ₄ flux (mmol m ⁻² d ⁻¹)	1.04 ± 0.44 (n = 4)	0.37 ± 0.09 (n = 5)	0.64 ± 0.15 (n = 5)	1.2 ± 0.70 (n = 4)	1.33 ± 0.77 (n = 5)	3.21 ± 2.00 (n = 6)	3.78 ± 0.55 (n = 4)	1.71 ± 0.51 (n = 6)	1.62 (n = 2)	0.85 ± 0.28 (n = 6)
Conductivity (µS cm ⁻¹)	28.3	222	29.3	41	31.2	46.3	47.2	44.3	72.2	39.5
pH	6.6	7.8	6.3	6.3	6.3	6.4	6.4	6.3	6.8	6.6
Alkalinity (meq L ⁻¹)	0.19	1.88	0.22	0.31	0.22	0.39	0.39	0.38	0.62	0.31
DOC (mg L ⁻¹)	26.5	26.7	36.9	34.4	35.7	35.7	39.2	36.7	35.6	34.3
Color (mg Pt L ⁻¹)	94	11	94	141	76	135	153	129	19	27
TP (µg L ⁻¹)	14	15	12	17	15	13	18	15	15	17
TN (mg L ⁻¹)	0.45	0.40	0.41	0.46	0.38	0.36	0.54	0.40	0.41	0.43
LOI _{bulk} (%)	11.0	8.9	11.5	9.7	25.8	8.4	14.7	15.8	7.0	21.2
δ ¹³ C _{bulk} (‰ VPDB)	-29.3	-28.4	-30.6	-30.1	-31.8	-30.9	-32.5	-31.7	-28.4	-32.3
C:N _{bulk}	8.9	9.3	10.9	10.0	11.0	9.3	9.3	10.7	9.1	9.5

as the underlying thaw bulb expands (Jorgenson and Shur, 2007; West and Plug, 2008). Thermokarst lakes can emit 'old' methane formed during anaerobic decomposition of thawing underlying Pleistocene deposits, sometimes resulting in extremely active ebullition and methane seeps (Walter et al., 2006). Our study lakes are shallow, however, and no exceptionally active sites of ebullition were observed. In addition to surface sediment samples from Siberian lakes, we also present new δ¹³C analyses based on lake sediment samples that originate from south Central Sweden (59° N, 15–17° E). These lakes cover a maximum depth gradient of 4–20 m, include oligotrophic to eutrophic sites, and are discussed in detail in van Hardenbroek et al. (2011b).

2.2. Methane fluxes and water chemistry of Siberian lakes

Diffusive methane emission rates from the Siberian study lakes were measured using floating chambers (6.9 L volume, 0.071 m² area) constructed of dark PVC, adapted from Bastviken et al. (2004). Two to four replicate measurements along a transect from the shore to the deepest point were taken during one day per lake between 14 July and 7 August 2007. Between 16 July and 1 August, 2009 two additional measurements were taken on six of the lakes, whereas four lakes could not be re-examined due to technical and logistical difficulties. Chambers were left floating on the lake surface during 15 min for each measurement. Gas samples were withdrawn at the start and after every 5 min through a rubber septum and transferred to evacuated infusion vials using a syringe. Methane concentrations were analysed with a HP 5890A (Hewlett Packard) gas chromatograph and methane fluxes were calculated by linear interpolation of the measurements and expressed per square metre and day. Flux measurements were rejected if the increase of methane was nonlinear, resulting e.g. from ebullition or sample loss.

Samples for water chemistry analyses were taken from 10 cm below the water surface and from 1 m above sediments between 8 July and 7 August 2007 and stored cold and dark in plastic bottles until processing upon return from the field campaign. Water temperature and conductivity were measured *in situ* with a GMH 3410 conductivity probe (Greisinger Electronic GmbH) and pH with an Eijkelkamp 18.21 multi-meter (Eijkelkamp Agrisearch Equipment). Samples for dissolved organic carbon (DOC) were filtered through 0.2 µm glassfiber-filters and fixed with a few drops of concentrated HCl to keep pH below 2. DOC concentrations were analysed by thermal oxidation on a Shimadzu TOC-5050A analyzer.

Alkalinity was measured by potentiometric titration with HCl. Total phosphorus (TP) and total nitrogen (TN) were measured spectrophotometrically in unfiltered samples. TP was measured using a modified molybdate method (Murphy and Riley, 1962) after oxidation with K₂S₂O₈ + H₂SO₄. TN was determined on nitrite-ions using Griss reagent after oxidation with K₂S₂O₈ + NaOH and reduction of NO₃⁻ to NO₂⁻ in a Cu–Cd-reducer. Colour was determined by comparing the sample against colorimetric titration of a standard Pt-Co-solution in demineralized water.

2.3. Sediment characteristics and dating of Siberian lake sediments

For sampling surface sediments in the Siberian lakes in the summer of 2007, the deepest part of each lake basin was located using echo-sounding and sediment cores were collected with a gravity corer (UWITEC, Austria). Replicate surficial samples of 0–1 and 1–2 cm sediment depth were taken in each lake. Longer sediment cores were taken in Lake S2 and in the adjacent and hydrologically connected Lake S1 using the same coring system (Fig. 1). Core S2L1, the sediment core from lake S2, had relatively high concentrations of invertebrate remains and was selected for producing a downcore invertebrate δ¹³C record. The 38 cm long core S2L1 and the 29 cm long core S1L3 from lake S1 were extruded and subsampled on site at 0.5-cm resolution. All samples were stored in plastic zip-loc bags and kept cool and dark until freeze-drying upon arrival in the laboratory.

The freeze-dried sediment samples from core S2L1 and S1L3 were analysed for ²¹⁰Pb, ²²⁶Ra, ¹³⁷Cs, and ²⁴¹Am by direct gamma assay. The age–depth relationship and sedimentation rates for the uppermost section of the core were calculated using the AD 1963 peak in atmospheric ¹³⁷Cs deposition (Wright et al., 1999) and the constant rate of supply (CRS) ²¹⁰Pb dating model (Appleby and Oldfield, 1978). Based on distinct changes in the organic matter content and two conspicuous layers with remains of *in situ* grown aquatic moss, core S2L1 was correlated to core S1L3 (Fig. 1). One terrestrial plant remain (*Salix* sp. twig) was found in core S1L3 and AMS ¹⁴C-dated. In addition, samples of bulk sediment and aquatic moss remains from cores S1L3 and S2L1 were AMS ¹⁴C-dated.

A subsample of sediment for geochemical analysis of bulk organic matter was taken from surface sediments from all 10 Siberian lakes examined in this study and from the core S2L1 (at 1-cm intervals in the top 10 cm and at 2-cm intervals deeper in the core). Organic content in the sediment samples was determined using loss-on-ignition at 550 °C (LOI₅₅₀) and expressed as percent weight loss

after combustion at 550 °C for 4 h (Heiri et al., 2001). Bulk sediment samples for stable carbon isotope analysis were soaked in 2.5% HCl for 15 min to remove carbonates, rinsed three times with demineralized water and centrifuged 4 min at 2000 rpm to remove excess water. C:N ratios and $\delta^{13}\text{C}$ of bulk organic matter in sediment samples were analysed on a PDZ Europa ANCA-GSL elemental analyzer interfaced to a PDZ Europa 20-20 IRMS. The reference materials used were secondary standards of known relation to international standards for VPDB ($\delta^{13}\text{C}$). Replicate sample measurements ($n = 18$) on internal standards gave analytical errors of $\pm 0.03\text{‰}$ (2σ).

2.4. $\delta^{13}\text{C}$ of invertebrate remains in sediments from Siberian lakes

Surface sediments analysed for invertebrate remains were taken from the top 0–1 cm of the gravity cores from all Siberian lakes. For downcore analysis, samples from core S2L1 were analysed at 1-cm intervals in the top 10 cm and at 2-cm intervals for the rest of the record. Samples were deflocculated in 10% KOH for 2 h at room temperature and sieved through 200- and 100- μm sieves (van Hardenbroek et al., 2010b). Sieve residues were soaked in 2.5% HCl for 15 min to remove carbonates, rinsed three times in demineralised water, and stored in the dark. Remains were identified under a dissecting microscope at 40–100 \times magnification following Vanderkerkhove et al. (2004) for the resting stages of *Daphnia* and Brooks et al. (2007) for chironomid head capsules that were sorted into five groups: *Chironomus*, Chironomini (not belonging to the genus *Chironomus*), Orthoclaadiinae, Tanytarsini, and Tanypodinae. Furthermore, the chitinous cuticles of ostracods were collected. Remains were separated into different taxonomic groups and transferred directly into pre-weighed ultraclean tin cups with forceps. The tin cups were dried on a hotplate at 50 °C for 24 h after which they were re-weighed and crimped for stable isotope analysis. Control samples ($n = 5$) of water from sieve residues were evaporated in tin cups and no carbon contamination was detected.

Samples for invertebrate $\delta^{13}\text{C}$ were analysed on a Fisons NA 1500 NCS Elemental Analyzer coupled to a Thermo Electron Delta plus IRMS. The reference material used was a secondary standard of known relation to the international standard of VPDB. Replicate sample measurements ($n = 82$) on this internal standard gave an analytical error of $\pm 0.08\text{‰}$ (2σ). For sediment samples with enough material, replicates of individual taxa were analysed. The average $\delta^{13}\text{C}$ for these replicates are reported (Table 2). Pearson's correlation coefficients were calculated using PAST v2.00 (Hammer et al., 2001).

2.5. $\delta^{13}\text{C}$ of invertebrate remains and methane flux data from Swedish lakes

$\delta^{13}\text{C}$ of invertebrate remains in surface lake sediments from Siberia was compared with a similar dataset from lakes in south Central Sweden. These lakes and methane flux measurements in them are described in detail in van Hardenbroek et al. (2011b). $\delta^{13}\text{C}$

of chironomid remains from these samples are described in the same publication, whereas analyses of $\delta^{13}\text{C}$ of *Daphnia* ephippia and ostracod cuticles have not yet been published and were measured using the same laboratory procedures as for the Siberian lakes.

3. Results

3.1. Methane fluxes of the Siberian lakes

Diffusive methane fluxes ranged from 0.41 ± 0.09 to 4.20 ± 0.55 $\text{mmol m}^{-2} \text{d}^{-1}$ in the studied Siberian thermokarst lakes (Table 1). All lakes were mixed and had an oxic water column in summer. According to their water chemistry the lakes were oligo- to mesotrophic (Table 1). No significant correlation was observed between diffusive methane fluxes and any of the bulk sediment or water chemistry parameters listed in Table 1, except colour ($r = 0.63$, $p = 0.049$). The measured diffusive fluxes were considerably higher than the values measured in the Swedish lakes studied by van Hardenbroek et al. (2011b), which ranged from 0.04 ± 0.01 to 0.51 ± 0.23 $\text{mmol m}^{-2} \text{d}^{-1}$ (Fig. 2).

3.2. Invertebrate $\delta^{13}\text{C}$ in surface sediments

The range of $\delta^{13}\text{C}$ in invertebrate remains isolated from surface sediments of the Siberian lakes varied between taxa (Table 2; Fig. 2). Orthoclaadiinae had the highest $\delta^{13}\text{C}$ of all taxa, ranging between -30.8 and -27.1‰ . Tanypodinae and Tanytarsini had lower $\delta^{13}\text{C}$ that ranged from -33.6 to -27.7‰ and from -35.1 to -28.1‰ , respectively. *Chironomus*, ostracods, and *Daphnia* had the lowest $\delta^{13}\text{C}$ in the Siberian lakes, ranging from -35.9 to -29.6‰ , from -36.9 to -30.9‰ , and from -37.9 to -31.9‰ , respectively. Chironomini had the largest range of $\delta^{13}\text{C}$ (-36.8 to -28.8‰). The lowest $\delta^{13}\text{C}$ of Chironomini (-36.8 and -37.9‰ , respectively) were found in Lakes N6 and N7, where the highest methane fluxes were measured.

Several taxa, including *Chironomus*, Chironomini, Tanytarsini, Tanypodinae, *Daphnia*, and ostracods showed negative correlations between $\delta^{13}\text{C}$ of their chitinous remains and diffusive methane fluxes measured in the Siberian lakes (Fig. 2). For Chironomini and *Daphnia*, these correlations were statistically significant ($r = -0.65$, $p = 0.040$ and $r = -0.66$, $p = 0.039$, respectively). If these data are examined together with $\delta^{13}\text{C}$ measured in the seven Swedish lakes studied by van Hardenbroek et al. (2011b), correlations between diffusive methane flux values and invertebrate $\delta^{13}\text{C}$ are also statistically significant for Tanytarsini ($r = -0.53$, $p = 0.029$) and stronger for Chironomini and *Daphnia* ($r = -0.72$, $p = 0.001$ and $r = -0.81$, $p < 0.001$, respectively). No statistically significant relationships were found between $\delta^{13}\text{C}$ of other invertebrate remains and diffusive methane flux estimates. The relationship between methane flux and $\delta^{13}\text{C}$ of bulk organic matter is statistically significant, though only if both the Swedish and the Siberian lakes are examined together (Fig. 2).

Table 2

Mean (\pm SD) $\delta^{13}\text{C}$ values (‰ VPDB) for invertebrate remains from surface sediment samples from ten study lakes in arctic Siberia.

Lake	<i>Chironomus</i>	Chironomini	Tanytarsini	Orthoclaadiinae	Tanypodinae	<i>Daphnia</i>	Ostracoda
N1		-28.8	-29.4 ($n = 2$)	-27.7	-27.7		
N2	-29.6	-33.3	-29.9	-29.1			-30.9
N3	-32.7 ($n = 2$)	-32.1		-30.1 ($n = 2$)	-31.6	-32.1 \pm 0.9 ($n = 3$)	
N4	-33.9	-34.4	-31.0	-29.9	-30.7	-31.9 ($n = 2$)	-36.9
N5	-35.9 \pm 0.6 ($n = 4$)	-31.6	-32.9 ($n = 2$)	-30.2 \pm 2.3 ($n = 8$)	-33.6	-32.8 \pm 0.4 ($n = 3$)	-32.9
N6	-32.3 ($n = 2$)	-35.0	-35.1 ($n = 2$)	-28.9 ($n = 2$)	-31.5	-37.9 ($n = 2$)	-33.7
N7	-34.3 \pm 0.8 ($n = 4$)	-36.8	-32.3 \pm 1.8 ($n = 9$)	-30.5 \pm 0.8 ($n = 4$)	-32.0 ($n = 2$)	-34.8 ($n = 2$)	-34.8
N8	-33.3 \pm 0.8 ($n = 3$)	-33.9	-34.8 ($n = 2$)	-30.8	-33.2	-34.9 \pm 0.7 ($n = 3$)	-35.0 \pm 0.4 ($n = 4$)
S1	-33.6 \pm 0.8 ($n = 3$)		-28.1	-27.1			-33.7 \pm 0.4 ($n = 5$)
S2	-34.7	-33.6	-32.0	-29.4			-34.1

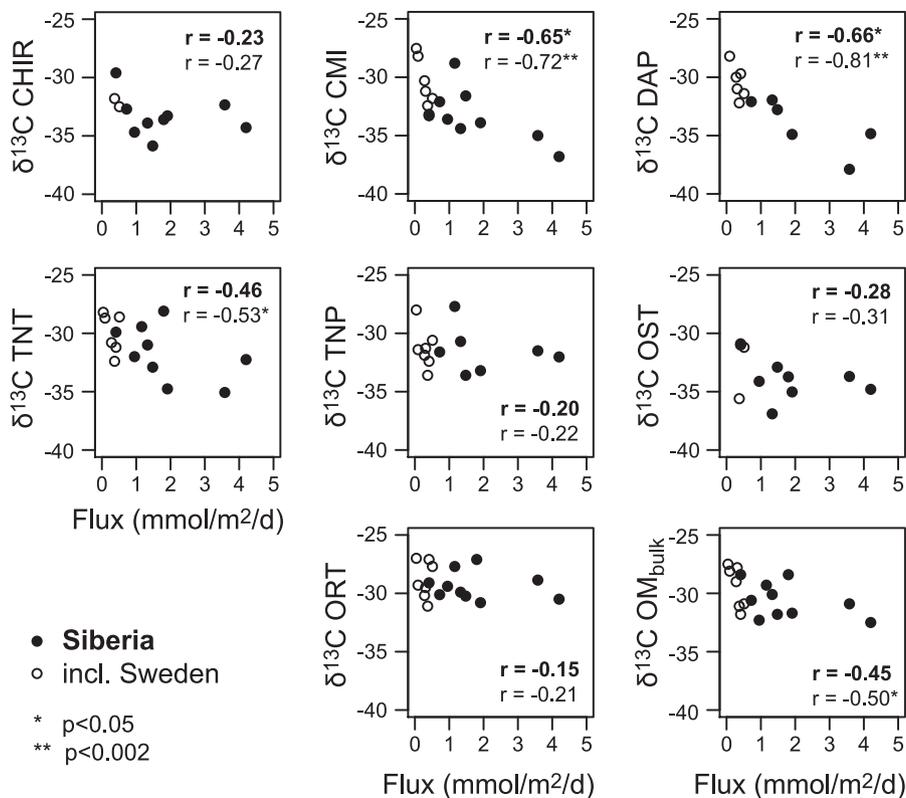


Fig. 2. Scatter plots and correlations between diffusive methane flux from the water surface of Siberian and Swedish lakes and $\delta^{13}\text{C}$ (‰ VPDB) of invertebrate remains and of bulk organic matter in surface sediments. Solid circles represent samples from Siberia, open circles samples from Sweden. CHIR = *Chironomus*, CMI = Chironomini (excluding *Chironomus*), DAP = *Daphnia*, TNT = Tanytarsini, TNP = Tanytopodinae, OST = Ostracoda, ORT = Orthoclaadiinae. r -values calculated for the Siberian lakes are provided in bold, r -values calculated for the combined Siberian and Swedish data are provided in plain text.

If data of all seventeen lakes are combined $\delta^{13}\text{C}$ of Chironomini and *Daphnia* are also significantly correlated with lake depth, LOI_{550} , sediment C:N ratio, DOC concentration, and water colour (Table 3). In addition, $\delta^{13}\text{C}$ of Chironomini and Tanytarsini is significantly correlated to $\delta^{13}\text{C}$ of sediment organic matter. However, most of these correlations are distinctly weaker than the correlation of $\delta^{13}\text{C}$ of Chironomini and *Daphnia* with methane flux (Table 3).

In Siberian lakes with enough invertebrate remains we did replicate measurements of individual taxa to examine the natural variability of $\delta^{13}\text{C}$ that can be expected for chitinous invertebrate remains in lake sediments. For example, replicate measurements of *Chironomus* in lakes N5, N7, N8, and S2 indicated a standard deviation (SD) of 0.6‰ ($n = 4$), 0.8‰ ($n = 4$), 0.8‰ ($n = 3$), and 1.4‰ ($n = 3$), respectively. The average of these SD values was 0.9‰ . Similar average SD estimates in the Siberian dataset have been calculated based on replicate measurements within one to three lakes for Tanytarsini (1.8‰), Orthoclaadiinae (1.6‰), *Daphnia* (2.1‰) and ostracods (0.5‰).

3.3. Sediment record

Core S2L1 obtained from Lake S2 consisted of soft, brown, organic-rich material alternating with grey clay between 16 and 18 cm and below 32.5 cm. The ^{210}Pb CRS age–depth model used for the upper part of the record (Fig. 3) was verified by a peak in ^{137}Cs activity corresponding to the 1963 bomb testing peak. ^{137}Cs and ^{210}Pb dating provides a reliable age control back to ca AD 1890. Due to the absence of suitable remains for ^{14}C -dating, the age of the lower part of the core was more difficult to constrain. ^{14}C -analyses of bulk sediment samples and aquatic mosses in core S2L1 were

inconsistent with the ^{137}Cs - and ^{210}Pb -data and clearly indicated that old carbon from surrounding peatlands and the slopes of the thermokarst lake may have affected the ^{14}C -age of the bulk sediment record. However, sediment core S2L1 could be correlated with sediment core S1L3 from the adjacent and hydrologically connected Lake S1 (Fig. 1) based on two distinctive layers of *in situ* grown aquatic mosses and a clear change in lithology that occurred in both cores (Fig. 3). The upper moss layer in core S1L3 has been ^{14}C -dated to a calibrated age of AD 1490 ± 40 using terrestrial plant remains in the same sample. A second moss layer was observed at the base of both cores. Assuming a constant sedimentation rate in Lake S1 the second moss layer at the base of the core would have an age of ca AD 1060. Furthermore, a distinct change in lithology from brown silty clay to dark clay was observed at 24 and 32.5 cm depth in the cores S1L3 and S2L1, respectively (Fig. 3), which is assumed to represent the same sedimentation event. Assuming again constant sedimentation rates within the section of core S1L3 not constrained by ^{210}Pb and ^{137}Cs , this change in lithology would be dated to ca AD 1250. The age–depth model for core S2L1 outside the ^{210}Pb -dating range was constructed by linear interpolation between the lowermost horizon dated by ^{210}Pb and the three horizons (the two moss layers and the lithological change) that could be correlated to core S1L3. In the age–depth model of core S1L3 the age of sediments older than AD 1490 is estimated based on extrapolation of sedimentation rates of core S1L3 beyond the oldest radiocarbon-dated sample. Age estimates for this section should therefore be treated with caution.

Bulk organic matter $\delta^{13}\text{C}$ remained relatively constant (SD 1.5‰) throughout core S2L1 (Fig. 4) with values ranging between -32.5 and -27.1‰ . In contrast, isotopic analyses of invertebrate remains in core S2L1 revealed a differential behaviour of different

Table 3Correlations between $\delta^{13}\text{C}$ of invertebrate taxa and physical characteristics, water chemistry, and bulk sediment geochemistry of the combined dataset of Siberian and Swedish lakes.

	<i>Chironomus</i>	Chironomini	Tanytarsini	Orthoclaadiinae	Tanypodinae	<i>Daphnia</i>	Ostracoda
Surface area (ha)	0.40	-0.04	0.14	0.33	-0.17	0.33	0.49*
Maximum depth (m)	-0.02	-0.54*	-0.25	0.08	-0.14	-0.59*	0.15
CH ₄ flux (mmol m ⁻² d ⁻¹)	-0.27	-0.72**	-0.53*	-0.21	-0.22	-0.81***	-0.31
Conductivity ($\mu\text{S cm}^{-1}$)	0.47	-0.05	0.2	0.11	0.07	0.1	0.49*
pH	0.25	-0.37	0.01	0.18	-0.08	-0.38	0.19
Alkalinity (meq L ⁻¹)	0.46	-0.07	0.19	0.11	0.07	0.09	0.48
DOC (mg L ⁻¹)	-0.34	-0.66**	-0.50*	-0.36	-0.42	-0.52*	-0.31
Colour (mg Pt L ⁻¹)	-0.17	-0.60*	-0.52*	-0.38	-0.18	-0.71**	-0.51*
TP ($\mu\text{g L}^{-1}$)	0.03	-0.21	-0.07	0.13	-0.32	0.23	0.21
TN (mg L ⁻¹)	0.14	-0.03	0.01	-0.23	-0.31	0.2	-0.11
LOI _{bulk} (%)	-0.13	0.62**	0.26	0.05	0.27	0.61**	0.13
C:N _{bulk}	-0.1	0.60*	0.23	-0.11	0.29	0.50*	-0.03
$\delta^{13}\text{C}_{\text{bulk}}$ (‰ VPDB)	0.43	0.69**	0.66**	0.38	0.56*	0.41	0.12

* $p < 0.05$; ** $p < 0.01$; *** $p < 0.001$.

ecosystem components in their $\delta^{13}\text{C}$. As in the surface sediment dataset, $\delta^{13}\text{C}$ of Orthoclaadiinae and ostracod cuticles remained relatively constant (SD of 1.3‰ for both groups). $\delta^{13}\text{C}$ of other remains revealed distinct changes, with the strongest variations observed in Chironomini, Tanytarsini, and *Daphnia* (SD of 1.9, 1.9, and 2.6‰, respectively). $\delta^{13}\text{C}$ of bulk organic matter is characterized by relatively low values between ca AD 1300 and 1550 and again in the youngest sediments from AD 1970 to present. $\delta^{13}\text{C}$ of

invertebrate groups such as ostracods and Orthoclaadiinae track these changes in $\delta^{13}\text{C}$ with a relatively constant offset, the Orthoclaadiinae being characterized by very similar values as $\delta^{13}\text{C}$ of organic matter and the ostracods by clearly lower values. In contrast, distinct changes are apparent in the offset between Chironomini, Tanytarsini, and *Daphnia* $\delta^{13}\text{C}$ and bulk organic matter $\delta^{13}\text{C}$, although all three indicators usually have lower $\delta^{13}\text{C}$ than bulk organic matter. The offset appears largest in the period ca

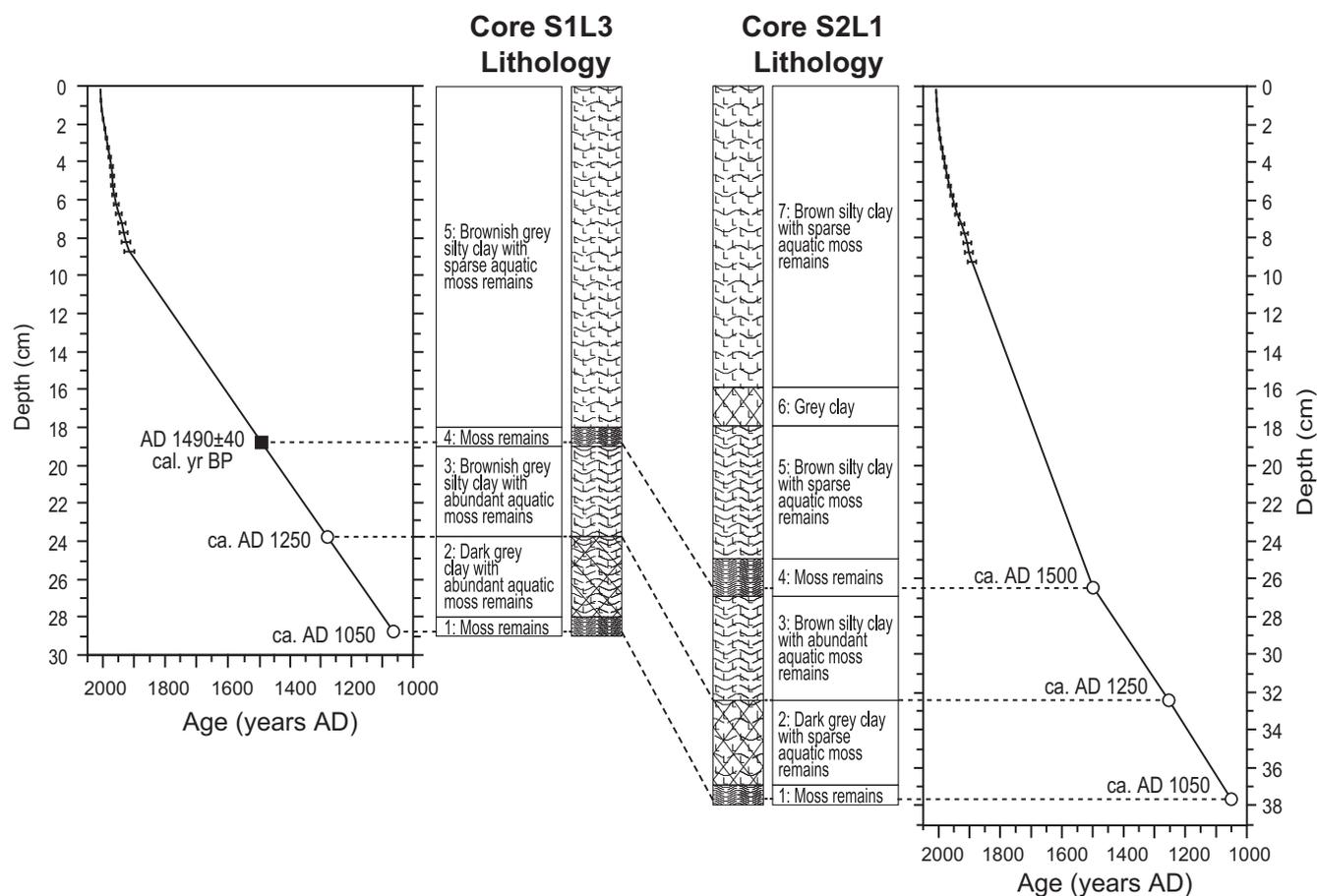


Fig. 3. Age–depth relationship and lithology for the sediment records S1L3 from Lake S1 (left) and S2L1 from Lake S2 (right). Dashed lines indicate the aquatic moss layers and the lithographic contact zones that allowed a correlation of the two records. The age–depth relationship for the top ~10 cm of these sediments was obtained via ^{210}Pb dating using the CRS model. A terrestrial plant macrofossil (*Salix* sp. twig) in the upper aquatic moss layer (18–19 cm depth) in S1L3 was AMS ^{14}C -dated to AD 1490 ± 40. The age of deeper lithographic contact zones in S1L3 was based on extrapolation of this age–depth relationship. The age–depth relationship in S2L1 not constrained by the ^{210}Pb chronology was obtained by linear interpolation of the sedimentation rates calculated between the ^{210}Pb dated layers and the aquatic moss layer dated to AD 1490.

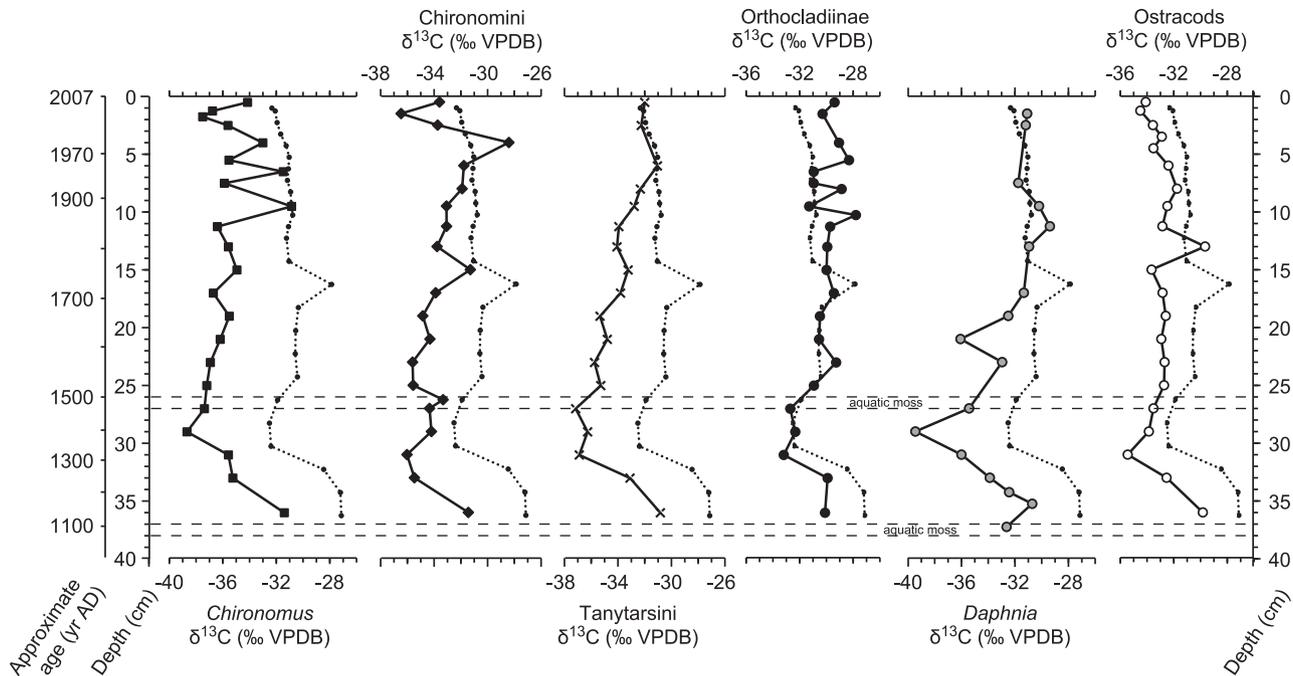


Fig. 4. $\delta^{13}\text{C}$ (‰ VPDB) of invertebrate remains in the sediment record from Lake S2. Dotted lines represent bulk organic matter $\delta^{13}\text{C}$, dashed horizontal lines indicate layers with aquatic mosses.

AD 1300–1700 and for the Chironomini in the youngest sediments. *Chironomus* is also characterized by variable $\delta^{13}\text{C}$ and values more negative than in organic matter. However, since $\delta^{13}\text{C}$ of this taxon shows a high degree of variability in the sediments deposited in the uppermost sediment layers of the lake the changes in the offset between *Chironomus* $\delta^{13}\text{C}$ and bulk organic matter $\delta^{13}\text{C}$ are more difficult to determine than for the other taxa. Concentrations of Tanytarsini head capsules in the sediment core were too low to provide enough material for $\delta^{13}\text{C}$ analysis.

4. Discussion

4.1. Invertebrate $\delta^{13}\text{C}$ in surface sediments

$\delta^{13}\text{C}$ of chitinous remains obtained from lake surface sediments suggest that the different invertebrate groups had access to different dietary carbon sources. Typically, $\delta^{13}\text{C}$ of material from terrestrial and littoral sources in lakes range between -33 and -8 ‰ (Meyers and Teranes, 2001). Kling et al. (1992) report $\delta^{13}\text{C}$ of -28 and -27 ‰ for peat and *Carex*, respectively for lakes in arctic Alaska, which is probably representative for allochthonous input in the Siberian lakes in our study. $\delta^{13}\text{C}$ of phytoplankton varies mostly between -35 and -25 ‰ (Yoshioka et al., 1994; Grey and Jones, 1999; Bade et al., 2006; Vuorio et al., 2006), but can be higher in periods of increased productivity (Hollander et al., 1993) and lower in oligotrophic lakes (Jones et al., 1999; Kankaala et al., 2010). In our Siberian lakes, nutrient concentrations indicated mesotrophic conditions (TP 12–18 $\mu\text{g L}^{-1}$, Table 1) and, therefore, we would not expect $\delta^{13}\text{C}$ of phytoplankton biomass below -35 ‰. Concentrations were more variable in the Swedish lakes analysed by van Hardenbroek et al. (2011b) (TP 9–41 $\mu\text{g L}^{-1}$).

Orthoclaadiinae identified in preliminary screening of the Siberian surface sediments were predominantly littoral taxa typically feeding on algae and associated microorganisms growing on hard substrates such as rocks and macrophytes. This diet is reflected in the $\delta^{13}\text{C}$ of Orthoclaadiinae remains that are similar to $\delta^{13}\text{C}$ of bulk sediment organic matter and highest of all invertebrate groups in all ten

Siberian lakes and six of the Swedish lakes. $\delta^{13}\text{C}$ of Orthoclaadiinae is not significantly correlated with diffusive methane flux in the Siberian, the Swedish, or the combined set of lakes. $\delta^{13}\text{C}$ of Chironomini and *Daphnia* remains in Siberian surface sediments were lower in most lakes than Orthoclaadiinae and negatively correlated with diffusive methane fluxes. Larvae of the Chironomini are typically deposit feeders burrowing in lake sediments, although some taxa inhabit aquatic macrophytes as well. Chironomini have previously been reported to incorporate methanogenic carbon by feeding on MOB. For example, markedly ^{13}C -depleted larvae have been found in several mud-dwelling genera including *Chironomus*, *Sergentia*, and *Stictochironomus* (Kiyashko et al., 2004; Jones et al., 2008). At least some tube-dwelling Chironomini larvae seem to actively promote the growth of methanotrophic bacteria (Deines et al., 2007). van Hardenbroek et al. (2011b) have shown that $\delta^{13}\text{C}$ of Chironomini remains is negatively correlated with diffusive methane flux in Swedish lakes as well. Interestingly, $\delta^{13}\text{C}$ of remains of the genus *Chironomus* show a weaker correlation with methane flux measurements than the remains of other Chironomini even though the $\delta^{13}\text{C}$ are similarly low in both taxa. This is unexpected since larvae of *Chironomus* have been widely reported as being exceptionally susceptible for incorporating methanogenic carbon into their biomass (Jones et al., 2008) and one would therefore have expected $\delta^{13}\text{C}$ of this taxon to be strongly related with methane availability. *Daphnia* are planktonic filter feeders that feed on fine particulate organic matter. A number of studies have indicated that $\delta^{13}\text{C}$ of *Daphnia* can be depleted in lake ecosystems with a high abundance of MOB indicating that *Daphnia* can ingest MOB, or organisms feeding on these microorganisms, and incorporate methanogenic carbon into its biomass (Bastviken et al., 2003; Taipale et al., 2008). In contrast to the remains of Chironomini and *Daphnia*, $\delta^{13}\text{C}$ of Tanytarsini and Tanytarsini are more weakly related to methane flux and do not reach similarly low $\delta^{13}\text{C}$ in the Siberian surface sediment samples. Similarly, $\delta^{13}\text{C}$ of the chitinous cuticles of ostracods are only weakly correlated with methane availability in both the Siberian and Swedish study lakes, although it is noteworthy that ostracod $\delta^{13}\text{C}$ is consistently lower than $\delta^{13}\text{C}$ of other invertebrate remains in most of the lakes.

The relationships between invertebrate $\delta^{13}\text{C}$ and methane flux measurements found for the Siberian dataset are similar to the relationships observed for the Swedish lakes sampled by van Hardenbroek et al. (2011b). In these seven Swedish lakes $\delta^{13}\text{C}$ of Chironomini remains is also negatively correlated with diffusive methane flux measured on the lake surface ($r = -0.90$, $p = 0.006$), whereas $\delta^{13}\text{C}$ of other remains does not reveal a statistically significant relationship with methane flux ($r = -0.57$ to 0.17 , $p = 0.18$ – 0.92). If the two datasets are combined the following relationships are apparent (Fig. 2): $\delta^{13}\text{C}$ of Chironomini and *Daphnia* shows a significant and negative correlation with diffusive methane flux, whereas $\delta^{13}\text{C}$ of most other groups is not significantly related with the methane flux data ($r = -0.31$ to -0.21 , $p = 0.23$ – 0.41). The exception here are the Tanytarsini, for which a relatively weak but statistically significant relationship is detected ($r = -0.53$, $p = 0.03$). The Tanytarsini are also predominantly deposit feeders although their larvae are smaller than Chironomini and probably more restricted in their burrowing behaviour to shallower sediment layers. To our knowledge, larvae of Tanytarsini have not previously been identified as potential grazers on MOB.

No significant correlation was observed between nutrient concentrations and methane fluxes, $\delta^{13}\text{C}$ of sedimentary organic matter, or $\delta^{13}\text{C}$ of invertebrate remains in the Siberian lakes, or in the combined dataset of seventeen lakes in Sweden and Siberia. Therefore, we assume that differences in productivity between the study lakes did not have strong effects on $\delta^{13}\text{C}$ of phytoplankton, bulk sediment, or invertebrate remains. However, significant correlations between invertebrate $\delta^{13}\text{C}$ and some other physical and chemical parameters were observed (Table 3), although all of these were weaker than the correlations between Chironomini and *Daphnia* $\delta^{13}\text{C}$ and methane flux. Some of these correlations would be expected if methanogenic carbon enters the water column and food web of a part of the studied lakes. For example, DOC concentrations and colour of the lakes influence the extent of the photic zone and the oxygenation of lakes. Lakes with a high DOC content are therefore expected to be more susceptible to anoxia in the bottom waters, which would allow for relatively high methane concentrations in the lake water (Houser et al., 2003). Similarly, if methane is incorporated into benthic and planktonic biomass one would also expect the organic matter $\delta^{13}\text{C}$ to be affected. A number of other processes, however, may have a strong influence in invertebrate $\delta^{13}\text{C}$, including the incorporation of respired, non-methanogenic carbon into lake foodwebs and differences in the relative contribution of allochthonous and autochthonous sources of organic matter to the lakes (see van Hardenbroek et al. (2011b) for a detailed discussion of some of these processes). Since the strongest correlations apparent in our dataset are with methane flux measurements it seems likely that these other processes are of secondary importance in explaining $\delta^{13}\text{C}$ of invertebrate remains in the examined lake surface sediments.

$\delta^{13}\text{C}$ of Chironomini, Tanytarsini and *Daphnia* remains are not as negative as reported in the literature for some living specimens that assimilate methane-derived carbon. ^{13}C -depletion of invertebrate biomass due to ingestion of methanogenic carbon seems to be most pronounced at the oxic–anoxic interface in lake basins and during autumn overturn when surface sediments are re-oxygenated (Grey et al., 2004; Jones and Grey, 2011). It must be kept in mind that invertebrate remains in surface sediment samples provide an assemblage that is integrated over space and time (Frey, 1988; van Hardenbroek et al., 2011a) and at present it is not clear whether $\delta^{13}\text{C}$ of fossil assemblages can attain $\delta^{13}\text{C}$ below -40‰ even in lakes in which their living counterparts attain exceptionally low values. Nevertheless, the negative correlations between diffusive methane flux measured at the lake surface and $\delta^{13}\text{C}$ of Chironomini and *Daphnia* seem to be a robust feature, both present within the two

regional surface sediment datasets from Sweden and Siberia, whereas the relationship between $\delta^{13}\text{C}$ of Tanytarsini and methane flux is more uncertain and will have to be confirmed by future studies.

4.2. Sediment record

In the sediment record from Siberian thermokarst Lake S2 lower $\delta^{13}\text{C}$ were observed for Chironomini, Tanytarsini, and *Daphnia* between approximately AD 1250 and 1500 (32.0–25.5 cm depth) (Fig. 4). Furthermore, a general increasing trend in $\delta^{13}\text{C}$ of these groups was observed between ca AD 1600 and 1900. Considering the correlation of $\delta^{13}\text{C}$ of these taxa with diffusive methane flux and the amplitude of these changes, which amounts to 8.1, 6.3, and 10.1‰ for Chironomini, Tanytarsini, and *Daphnia*, respectively, this suggests major changes in the carbon cycling of Lake S2 during the past 1000 years. In contrast, some decadal-scale variations in $\delta^{13}\text{C}$ are apparent in all indicator groups as well as in bulk organic matter, including a relatively prominent but moderate decrease in $\delta^{13}\text{C}$ centred on ca AD 1350 (Fig. 4), suggesting that the baseline $\delta^{13}\text{C}$ of inorganic carbon in Lake S2 may have changed during this period.

$\delta^{13}\text{C}$ of *Chironomus* showed variable values but overall a similar trend as the Chironomini, Tanytarsini and *Daphnia*. In contrast, $\delta^{13}\text{C}$ of Orthoclaadiinae and ostracods was less variable and generally followed the pattern of bulk organic matter $\delta^{13}\text{C}$. In the uppermost sediment layers the $\delta^{13}\text{C}$ trajectories of the different organism groups diverge, with Chironomini remains again characterized by very low $\delta^{13}\text{C}$ whereas the values of *Daphnia* and Tanytarsini did not seem to respond in the youngest part of the record. A pronounced difference in $\delta^{13}\text{C}$ between Orthoclaadiinae and the groups Chironomini and *Daphnia* is characteristic for lakes with a high methane flux in lakes in both Sweden and Siberia (Fig. 2). Taxa with the strongest changes in $\delta^{13}\text{C}$ in core S2L1 (Chironomini and *Daphnia*) are also the ones that show a significant correlation with methane availability in the modern datasets. Furthermore, both *Daphnia* and the larvae of the Chironomini are known to ingest methanogenic carbon either directly by ingesting on MOB or indirectly by feeding on higher trophic levels that assimilate carbon originating from MOB (Jones et al., 2008; Taipale et al., 2008). It seems likely, therefore, that changes in methane production and availability were responsible for the distinct shifts in the difference between $\delta^{13}\text{C}$ of different invertebrate groups in Lake S2, with higher methane availability within the lake during phases when $\delta^{13}\text{C}$ of Chironomini, Tanytarsini and *Daphnia* was distinctly more negative than $\delta^{13}\text{C}$ of Orthoclaadiinae and bulk sediment organic matter. As briefly mentioned above, alternative explanations for relatively low $\delta^{13}\text{C}$ in aquatic invertebrate remains include, e.g., the incorporation of respired carbon into lake foodwebs or variations in the primary productivity which may lead to moderate variations in algal $\delta^{13}\text{C}$ (see van Hardenbroek et al. (2011b) for a more detailed discussion). Changes in $\delta^{13}\text{C}$ of algal biomass would be expected to lead to similar shifts in $\delta^{13}\text{C}$ in the remains of deposit feeders, algivores, and predators in core S2L1, though. However, only taxa expected to be sensitive in their $\delta^{13}\text{C}$ to methane availability based on the surface sediment survey are characterized by clearly more pronounced variations in $\delta^{13}\text{C}$ than the other invertebrate groups in the record.

If the offset between $\delta^{13}\text{C}$ of bulk organic matter and invertebrate remains is examined (Fig. 5), it becomes apparent that variations in this offset for invertebrate groups correlated with methane flux in the modern environment are similar to trends in temperature reconstructed using tree ring data from the Chokurdakh region (Sidorova and Naurzbaev, 2005; Sidorova et al., 2008). The most negative $\delta^{13}\text{C}$ of Chironomini, Tanytarsini and *Daphnia* relative to bulk organic matter were registered for the

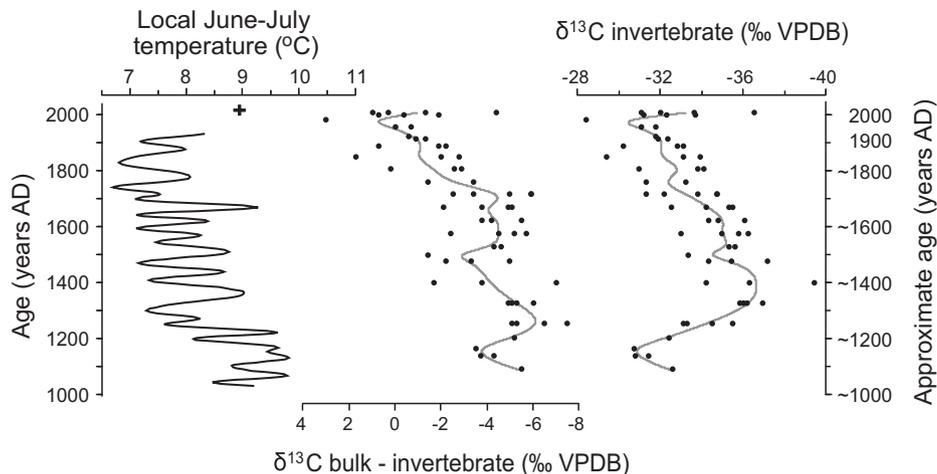


Fig. 5. June–July temperature reconstruction (smoothed using a 50-year low-pass filter) based on local tree-ring series from the region around the town of Chokurdakh, northern Yakutia (Sidorova and Naurzbaev, 2005; Sidorova et al., 2008). 1997–2006 average June–July temperature indicated by (+) was measured at the nearest meteorological station in Chokurdakh (30 km from study site). The temperature record is compared with offsets between bulk organic matter $\delta^{13}\text{C}$ and $\delta^{13}\text{C}$ of Chironomini, Tanytarsini, and *Daphnia* (in center) and absolute $\delta^{13}\text{C}$ of Chironomini, Tanytarsini, and *Daphnia* (on right). A loess smoother (bandwidth 0.15) is fitted through the invertebrate data. Note inverted x-axis for $\delta^{13}\text{C}$.

period preceding ca AD 1350, when temperatures were warm. After ca AD 1350 the offsets decreased and the smallest values in this offset are recorded for the 18th, 19th and 20th century, the coldest part of the temperature reconstruction. This pattern suggests a correspondence between past changes in carbon cycling of Lake S2 and variations in temperature. Changes in the offset between $\delta^{13}\text{C}$ of bulk organic matter and invertebrate groups that could reflect methane availability seem to lag behind reconstructed temperature changes (Fig. 5). However, the observed lag could also be an artefact of uncertainties in the age model.

In recent decades inferred temperatures increase again and $\delta^{13}\text{C}$ of Chironomini track this recent temperature rise by a decrease in $\delta^{13}\text{C}$. However, $\delta^{13}\text{C}$ of Tanytarsini and *Daphnia* does not respond to the recent warming. There are a number of possible reasons for this differential response of the $\delta^{13}\text{C}$ of different methane-sensitive invertebrate groups to climate change. Responses in methane availability to climatic variations may differ within different ecosystem components and this may have affected the studied invertebrate groups, which includes deep-burrowing zoobenthos (most Chironomini), surface feeding zoobenthos (most Tanytarsini) and zooplankton (*Daphnia*), to varying extents. Changes in the thermal stratification of Lake S2 could have affected $\delta^{13}\text{C}$ for *Daphnia* differently than for chironomids, as *Daphnia* are only able to incorporate MOB at the oxycline in the water column, whereas Chironomini can burrow down to the oxic/anoxic interface in the sediment. Alternatively, methane availability in the lake may have varied seasonally which may have led to a differential response in the various invertebrate groups. For example, in many lakes *Daphnia* produce ephippia late in the year and $\delta^{13}\text{C}$ of *Daphnia* remains may therefore reflect lacustrine conditions during autumn, whereas chironomid larvae typically grow and produce cuticles during a more extended period. Processes possibly leading to higher methane availability in lake S2 during warmer periods in the past millennium may have included higher autochthonous primary productivity in the lake associated with higher temperatures or enhanced methane formation as the talik of unfrozen ground expanded beneath the lake in response to warmer temperature (Burn and Smith, 1990). The latter process could also be expected to lag behind air temperature changes and would be a potential explanation for the different timing of changes in reconstructed summer temperatures based on tree rings and variations in methane availability as assessed by $\delta^{13}\text{C}$ of aquatic invertebrates in Lake S2.

5. Conclusions

Methane emissions from lakes have been considered marginal in models that simulate methane emissions from high-latitude wetlands (Zhuang et al., 2004; van Huissteden, 2004). However, recent studies have indicated that lakes can contribute substantially to atmospheric methane (Bastviken et al., 2011), especially thermokarst lakes such as the ones examined here (Walter et al., 2006). Our results, indicating distinct variations in the carbon cycle within Lake S2 during the past millennium, suggest variable methane availability in the lake and methane emissions to the atmosphere. The finding that methane availability seemed to have been highest during warmer climatic phases agrees with model-based reconstructions that suggest that arctic and boreal wetlands produced more methane during warmer interstadials than the cooler stadials of the last glaciation (Velichko et al., 1998; Huber et al., 2006; Fischer et al., 2008). If other high-latitude lakes responded similarly to past climatic changes as our results suggest Lake S2 did, this would suggest that thermokarst lakes react dynamically in their methane output to environmental change even during the climatologically relatively stable Holocene period. This potential feedback of lake methane emissions needs to be taken into account when predicting the response of terrestrial methane sources to future climate change. Clearly, additional analyses will be necessary to corroborate our results, confirm the relationship between $\delta^{13}\text{C}$ of selected invertebrate taxa and methane flux in lakes, and to examine whether methane availability in other arctic lakes has shown similar variations in the past as our results suggest for Lake S2. However, our analyses of $\delta^{13}\text{C}$ of invertebrate remains in Siberian lake sediments confirm the strong relationship between $\delta^{13}\text{C}$ of selected invertebrate taxa and methane availability that has previously been described from boreal lakes in Sweden (van Hardenbroek et al., 2011b) and demonstrate that downcore analyses of $\delta^{13}\text{C}$ of chitinous invertebrate remains can provide insights into past changes in the carbon cycle and methane production of high-latitude thermokarst lakes.

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