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Carbon transport by the Lena River from its headwaters to the Arctic Ocean, with emphasis on fluvial input of terrestrial particulate organic carbon vs. carbon transport by coastal erosion

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Abstract. The Lena River integrates biogeochemical signals from its vast drainage basin, and the integrated signal reaches far out over the Arctic Ocean. Transformation of riverine organic carbon (OC) into mineral carbon, and mineral carbon into the organic form in the Lena River watershed, can be considered to be quasi-steady-state processes. An increase in Lena discharge exerts opposite effects on total organic (TOC) and total inorganic (TCO_2) carbon: TOC concentration increases, while TCO_2 concentration decreases. Significant inter-annual variability in mean values of TCO_2, TOC, and their sum (total carbon, TC) has been found. This variability is determined by changes in land hydrology which cause differences in the Lena River discharge. There is a negative correlation in the Lena River between TC in September and its mean discharge in August; a time shift of about one month is required for water to travel from Yakutsk to the Laptev Sea. Total carbon entering the sea with the Lena discharge is estimated to be almost 10 Tg C yr^{-1}. The annual Lena River discharge of particulate organic carbon (POC) can be as high as 0.38 Tg (moderate to high estimate). If we instead accept Lisytsin's (1994) statement that 85–95 % of total particulate matter (PM) (and POC) precipitates on the marginal "filter", then only about 0.03–0.04 Tg of Lena River POC reaches the Laptev Sea. The Lena's POC export would then be two orders of magnitude less than the annual input of eroded terrestrial carbon onto the shelf of the Laptev and East Siberian seas, which is estimated to be about 4 Tg. Observations support the hypothesis of a dominant role for coastal erosion (Semiletov, 1999a, b) in East Siberian Arctic Shelf (ESAS) sedimentation and the dynamics of the carbon/carbonate system. The Lena River is characterized by relatively high concentrations of the primary greenhouse gases, dissolved carbon dioxide (CO_2) and methane (CH_4). During all seasons the river is supersaturated in CO_2 compared to the atmosphere, by up to 1.5–2 fold in summer, and 4–5 fold in winter. This results in a significant CO_2 supersaturation in the adjacent coastal sea. Localized areas of dissolved CH_4 along the Lena River and in the Lena delta channels may reach 100 nM, but the CH_4 concentration decreases to 5–20 nM towards the sea, which suggests that riverborne export of CH_4 plays but a minor role in determining the ESAS CH_4 budget in coastal waters. Instead, the seabed appears to be the source that provides most of the CH_4 to the Arctic Ocean.

1 Introduction

The Arctic Ocean is surrounded by permafrost which is being degraded at an increasing rate under conditions of warming which are most pronounced in Siberia and Alaska (ACIA, 2004; Richter-Menge et al., 2006). Permafrost thawing accelerates both river discharge (Lyon and Destouni, 2010), and carbon losses from soils (Finlay et al., 2006; Freeman et al., 2001; Guo et al., 2007). Siberian freshwater discharge to the Arctic Ocean is expected to increase with increasing temperatures (Savelieva et al., 2000; Semiletov et

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al., 2000; Peterson et al., 2002), which in turn may result in greater river export of old terrigenous organic carbon (OC) that has recently been documented to be fluvially exported to the ocean (Guo et al., 2004; van Dongen et al., 2008; Vonk et al., 2010a; Gustafsson et al., 2011). Rivers integrate variability in the components of the hydrometeorological regime, including soil condition, permafrost seasonal thaw, and thermokarst development, all the variables that determine the atmospheric and ground water supply for rivers and the chemical weathering in their watersheds. Thus, arctic rivers form an important link between the land and ocean carbon pools. In addition to serving as natural large-scale integrators of carbon-related processes occurring in their extensive drainage basins, the coastal export by the Great Siberian Arctic rivers (GSARs: Yenisey, Lena, Ob, Kolyma, and Indigirka) of carbon and nutrients plays a critical role in the biogeochemical regime of the Arctic Ocean. Evasion of carbon dioxide (CO_2) and methane (CH_4) from river streams can significantly change the regional or even the global carbon cycle (Kling et al., 1991; Richey et al., 2002; Bastviken et al., 2011). These authors cautioned that direct measurements of land-atmosphere CO_2 (and CH_4) exchange that ignore waterborne fluxes might significantly overestimate terrestrial carbon accumulation. A hypothesized climate-change-driven increase in terrestrial OC inputs to the Arctic Ocean through permafrost thawing, increased river runoff, and accelerated coastal erosion could dramatically alter carbon budgets and biogeochemical cycles. While the fate and transport processes of terrestrial OC across the Arctic land/ocean margin and consequent gas exchange are largely unknown, understanding them is critical to our understanding of environmental change on a time scale of human concern.

Documented significant increases in the annual amplitude of GSAR discharges and air temperatures in their watersheds over the past 30 yr (Savelieva et al., 2000; Semiletov et al., 2000) have coincided with an increase in the amplitude of the seasonal atmospheric CO_2 and CH_4 cycles in the north (Conway et al., 1994; IPCC, 2001), indicating a general increase in climatic variability. Therefore, a positive feedback loop could operate between the emission of CO_2 and CH_4 from northern ecosystems and changes in the atmospheric circulation and land hydrology, potentially resulting in greater fluvial export of terrestrial organic matter (terrOM). Changes in the export of terrestrial carbon will certainly affect biogeochemical cycling and carbon balance in the ESAS seas (Alling et al., 2010; Charkin et al., 2011; Romankevich and Vetrov, 2001; Sánchez-García et al., 2011; Semiletov et al., 2005; Stein and Macdonald, 2004; van Dongen et al., 2008). Comprehending the controlling processes and constructing budgets to calculate regional river input and shelf sea processing of biogenic and anthropogenic materials requires comprehensive studies spanning different seasons and years if we aim to avoid being misled by the inevitable variability that is encountered in a single year's expedition.

Among the GSARs the Lena River is considered to be the major source of total OM, which is the sum of dissolved organic carbon (DOC) and particulate organic carbon (POC), to the Arctic Ocean (Gordeev et al., 1996). Cauwet and Sidorov (1996) and Lara et al. (1998) estimated the annual modern Lena River POC discharge as 1.8 Tg (1 Tg = 10^{12} g), higher than that of any other GSAR, and the Lena River DOC discharge as 3.6 Tg, twice as high as that of the Yangtze River – the largest among Asian rivers. Boucsein et al. (2000) argue that a significant change in composition of sediment OM (an increase of freshwater alginite and immature plant material) on the Laptev Sea continental slope at the beginning of the Holocene was due to increased Lena River discharge. Alternatively, Bauch et al. (2001) believe that the increased Holocene deposition of OM in the Laptev Sea was mainly the result of coastal erosion due to sea level transgression. Therefore, it is important to study the role of the Lena River and coastal erosion in different summer months when the major water and solid discharges occur.

Summer runoff from land, dominated by input from organic-rich soils, ice-complexes, and the mires of northern Asia, is an important source of carbon and nutrients to the shelf seas. Recent large-scale studies of terrestrial carbon in the East Siberian Arctic Shelf (ESAS) water column has documented significant degradation of the terrOM (e.g., Semiletov et al., 2007; Anderson et al., 2009, 2011; Alling et al., 2010; Vonk et al., 2010a; Sánchez-García et al., 2011). Further, surface water from the Laptev Sea shelf, rich in dissolved OM, follows Gakkel Ridge towards Fram Strait (Anderson et al., 1998), possibly determining bacterial production over the Amerasian shelf and the Arctic Basin (Wheeler et al., 1996; Opsahl et al., 1999). The Lena River is also considered to be the main sediment source for the Laptev Sea (Alabyan et al., 1995; Rachold et al., 2004), although the amount of suspended sediment delivered to the sea is still under some debate (Charkin et al., 2011). Note that most of the Lena River solid discharge estimates are based on the data obtained near the Kyusur hydrometeorological station which is located upstream from the Lena Delta. Thus, actual sedimentation in the Lena Delta channels and foredelta has yet to be studied.

Hydrometeorological analyses explaining the significant role of the Lena River-Laptev Sea system in the arctic freshwater budget can be found in Nikiforov and Shpaikher (1980), Zakharov (1996), and Semiletov et al. (2000). We view the Lena River as the seminal Siberian river because changes in Lena River hydrology cause significant changes in ice and freshwater input to the Transpolar Current through effects on the freshwater budget of the Laptev Sea. The Lena River also forms a natural boundary between western and eastern Siberian atmospheric circulation patterns, land hydrology, sea-ice conditions, tectonic structure, and biological communities on the Siberian shelf (Semiletov et al., 2000, 2005).

Freshwater discharge from the Lena is highly variable throughout the year; maximum flow occurs during spring breakup (late May–June). It is supposed that a significant portion of the Lena sediment discharge happens at the same time (Stein and Macdonald, 2004). However, sediment transport dynamics during breakup are poorly known and the limited data that exist are controversial. For example, Pivovarov et al. (1999) showed an increase of particulate matter (PM) concentration from the clear winter water in May (0.5–3 $mg\,l^{-1}$) up to 20 $mg\,l^{-1}$ during the flooding peak, with a maximum of 70 $mg\,l^{-1}$ observed at one station located near the main channel (Bol'shaya Trofimovskaya) outlet. A similar range of PM concentrations was observed along the Lena River after breakup time (late June–August), as discussed in this paper. Another "mooring-based" study (Wegner et al., 2005) shows that the main river PM transport onto the mid-shelf occurs during and shortly after river-ice breakup (June–early July), with peak PM concentrations of up to 6.5 $mg\,l^{-1}$. Surprisingly, events with the highest PM concentrations (up to 9.1 $mg\,l^{-1}$) were recorded over the mid-shelf during the ice-free period and at the time of freeze up. Thus, we can consider our current multi-year data to be a product of the soundest basic approach available at present; this approach should be improved in future studies.

This study is based on an extensive observational data set collected from seven field seasons in the Lena River system, extending from the headwaters near Ust'-Kut to the Lena River plume in the recipient Laptev Sea. Studies include one expedition from late June to early August 2003, the time of mid-to-high water levels along the Lena River, from Ust'-Kut to the Lena Delta. Other data obtained in 1995, 1997, 1998, 2002, 2005, and 2008 are also discussed (Fig. 1a, b, c, d). We present results of measurements of dissolved and particulate OC and carbonate system components as a first approach to understanding the inter-annual and seasonal variability of the Lena River and its chemical/carbon signature in the adjacent waters of the Laptev Sea. We briefly discuss connections between the hydrochemical regime of the Laptev Sea, as influenced by river runoff, and atmospheric processes over the Arctic. The emphasis of this work is to summarize information about important biogeochemical fluxes, with a focus on carbon, that link runoff from the Lena River to the Laptev and East Siberian seas, and further to the Arctic Ocean. Note that the annual and intra-seasonal Lena DOC discharge was studied much more thoroughly than was the POC discharge. We examine the fluvial transport of terrestrial POC vs. its export due to coastal erosion, and the interplay between these processes (when possible) across the ESAS which is heavily influenced by Lena River runoff. Data on the partial pressure of CO_2 (pCO_2) and dissolved CH_4, obtained along the Lena River and in the adjacent part of the Laptev Sea, are considered. This information is critical for developing a general understanding of how the Arctic atmosphere-land-shelf system works.

2 Data sets and methods

2.1 Field expeditions

Data were collected in different seasons before and after freeze-up over the 1995–2008 period. The study areas visited in different years are presented in Fig. 1a, b, c, d. Shelf water influenced by Lena River runoff was sampled using Niskin bottles and measured onboard the small hydrographic vessel (H/V) *Dunay* in September 1997 (Fig. 1b). Some data obtained in the Laptev Sea onboard motor vessel (M/V) *Auga* (September 2005), small boat *Neptun*, and M/V *Kapitan Danilkin* (September 2006), M/V *Mekhanik Kulibin* (late August–September, 2008), and during the winter expedition (April–May 2002) are also discussed (Fig. 1b). From late August to early September 1995 (Fig. 1c), surface water samples were taken onboard the M/V *Captain Ponomarev* from Yakutsk to the Lena River Delta and Tiksi Bay in the Laptev Sea using a plastic bucket deployed from the bow of the ship during slow cruising. The water samples were transferred immediately to hermetically-sealed bottles (~500 ml) for gas chromatography (GC) measurements of dissolved CH_4 and total CO_2 (TCO_2). This route was repeated onboard the M/V *Mekhanik Kulibin* in late August to early September 1998 (Fig. 1c). During late June to early August, 2003 Niskin bottles were used onboard M/V *Moskovskiy* to sample surface to bottom water from the upper reaches of the Lena River down to the Laptev Sea (Fig. 1d).

2.2 Salinity, nutrients, and dissolved organic carbon

Chlorinity (titrated, Cl), salinity (S), silicates (Si), and total dissolved nitrogen (N) and phosphorus (P) in water samples were determined by traditional oceanographic techniques prescribed by Ivanenkov and Bordovsky (1978). Total OC (TOC) and DOC contents of samples sealed in glass tubes were determined by the Shimadzu TOC-5000 high-temperature catalytic oxidation technique in the laboratories at the University of Alaska, Fairbanks.

2.3 Particulate organic carbon and nitrogen

During flooding (late June to early August, 2003) the PM dynamics and the composition and isotopic signatures of its organic material components (POC and particulate organic nitrogen, PON) were measured from the upper stream of the Lena River down to the Laptev Sea (Fig. 1d). Concentration and isotopic signatures of POC and PON were measured at the University of Alaska Fairbanks using a Finnigan isotope ratio mass spectrometer (IRMS) (Schell et al., 1998; Guo et al., 2004; see website at http://www.uaf.edu/water/ASIF/ASIF.html).

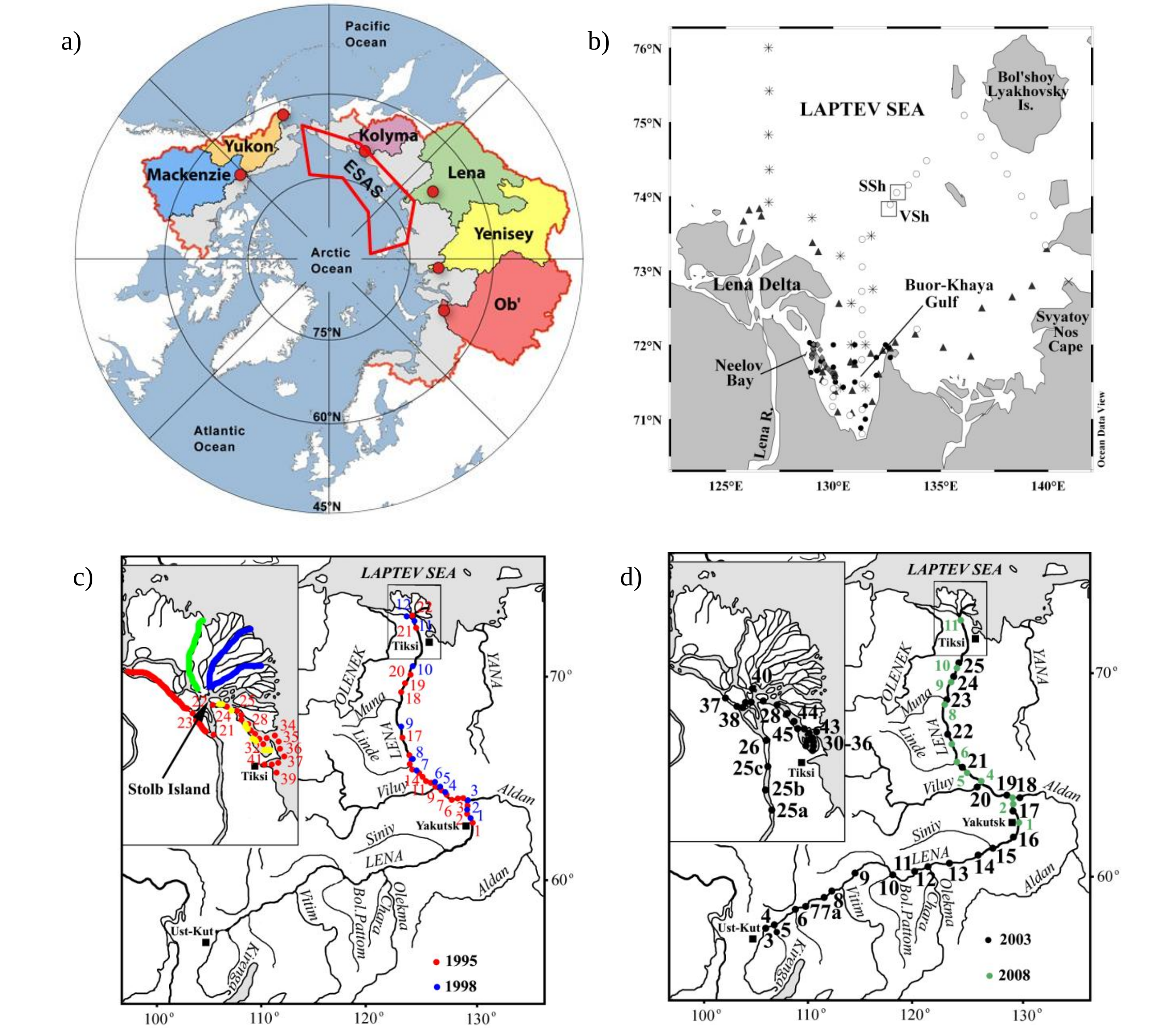

Fig. 1. Map of the study area location: **(a)** watersheds of the six largest arctic rivers and the ESAS position (modified from http://arcticgreatrivers.org/); **(b)** the Laptev Sea stations, *Dunay*, September 1997; ◯ *Auga*, September 2005; ◇ *Neptun* and * *Kapitan Danilkin*, September 2006; • *Winter expedition*, April 2002; SSh – Semenovskaya Shoal; VSh – Vasilievskaya Shoal; **(c)** the Lena River stations, late August–early September 1995 and September 1998; **(d)** the Lena River stations, September 2003 and 2008. Location of major channels is marked by yellow (Bykovskaya Channel), blue (Malya and Bol'shaya Trofimofskaya channels), green (Tumatskaya Channel), and red (Olenekskaya Channel).

2.4 pH

Two different techniques were used for pH measurements. pH measurements were made An open electrochemical cell was used for surface samples and for winter measurements prior to and including 1997 (Semiletov, 1999a). A closed electrochemical cell, which avoided CO_2 leakage into the atmosphere, was used to measure pH in samples taken from sites in all seasons after 1997. NBS (National Bureau of Standards) commercial standard buffers were used to check the Nernst slope of the electrode pairs. Because natural variability in the Arctic river and shelf water samples is large (up to 1 pH unit), we consider the difference of a few 10^{-2} pH units between values measured in open and in closed pH cells to be negligible.

2.5 Total alkalinity and partial pressure of CO_2

Samples for measurements of total alkalinity (TA) were obtained following Dickson and Goyet (1994) and analyzed within 12 to 24 h of collection by direct indicator titration in an open cell using 25 ml of water with 0.02 N hydrochloric acid (HCl) according to Bruevich's method (Ivanenkov and Bordovsky, 1978). Comparison of TA determination by Bruevich's and Edmond's methods (Dickson and Goyet, 1994) showed that the difference in TA did not exceed 0.002 mmol kg^{-1}. This difference is ignored here because the error in calculating pCO_2 is influenced mainly by the uncertainty in the measured pH value (±0.01 pH), which gives an uncertainty of about 10 µatm. The pCO$_2$ was computed from pH and TA, and from pH and TCO_2 according to the methods advocated by UNESCO (1987). We used the equation of Weiss (1974) to represent CO_2 gas solubility as a function of temperature and S, and the equations of Millero (1995) for temperature and S (Cl) dependence of the apparent dissociation constants for boric and carbonic acids in freshwater and seawater, as determined by Lyman and Meerbach et al. in UNESCO (1987). To calculate pCO$_2$ values in freshwater, we applied apparent constants for dissociation of carbonic acid advocated by Millero (1979, 1981), thermodynamic constants from Plummer and Busenburg (1982), and individual ion activity coefficients from Stumm and Morgan (1981). Agreement among results of all applied techniques was good; the pCO$_2$ difference did not exceed 1 %. Detailed descriptions of all measurements and calculations can be found elsewhere (Pipko et al., 2002, 2010, 2011b; Semiletov et al., 2007).

2.6 Total inorganic carbon and methane

To measure TCO_2 we used a TSVET-530 or LKHM-80MD gas chromatograph, with a Poropac T (1.5 m, 80–120 mesh) and a flame-ionization detector (FID) run isothermally at 30 °C with hydrogen carrier gas; a stripping GC technique similar to that described in Weiss (1981) was used. The TCO_2 calibrations were based on 0.73, 1.02, and 1.99 mM standard solutions of sodium carbonate (Na_2CO_3) that were prepared gravimetrically in fresh distilled water. Conversion of CO_2 to CH_4 after the Poropac column was done in a nickel (Ni)-catalyst column (14 cm Chromaton, 80–100 mesh, coated with Ni) at 400 °C (Semiletov, 1993). The measurements are reproducible to within ±1–2 %. The concentration of dissolved CH_4 was also determined by FID. Water for gas analysis was collected in hermetically-sealed glass bottles from just below the sea or river surface in the manner recommended by Dickson and Goyet (1994) for CO_2 system study. Concentration of CH_4 in the equilibrated gas phase (C_G) was determined by a GC head-space technique. The concentration of dissolved CH_4 (C_L) was calculated using the main static head space equation of Vitenberg and Ioffe (1982): $C_L = C_G(V_G/V_L + K)$, where V_G and V_L are volumes in the gas (G) and liquid (L) phases of the closed head-space system with thermostat set to ambient, and K is a gas-partitioning coefficient. Calibration was done with clean CH_4 using a precision multivalve positioned at ambient temperature. The total precision of this technique, which was used until 2003, varied between 3 and 5 %; variation was usually due to instability in shipboard electricity (Semiletov, 1999a; Semiletov et al., 1996a). High-precision measurements of dissolved CH_4 were done in September 2005, 2006, and 2008 using a Micro-Tech-8160 GC equipped with FID. For calibration in 2005, 2006, and 2008 we used a certified CH_4 gas standard in a balance of air (Air Liquide, USA). This technique has been successfully used by Shakhova et al. (2005, 2007a, b, 2010a, b) and Shakhova and Semiletov (2007). Samples stored for more than 4 h before measurements were made of dissolved CH_4 and TCO_2 were treated with mercuric chloride to poison biological activity.

3 General description of the Lena River basin

The Lena River originates near Lake Baikal from a large ($\sim 2.5 \times 10^6$ km^2) drainage basin that lies within 53–71° N and 105–141° E; this basin is located in one of the most remote areas of the Eurasian continent, and contains a large variety of geological features. Phytogeographically, the Lena River basin is an area of transition among various Siberian landscapes: Boreal and northern taiga, forest tundra, shrub tundra, open-woodland tundra, and semi-desert tundra. The Lena watershed area exceeds the entire area of the ESAS which, with a size of $\sim 2.1 \times 10^6$ km^2 (including the Laptev, East Siberian, and Russian part of the Chukchi seas), is the broadest and shallowest shelf in the World Ocean. The ESAS alone constitutes almost 20 % of the total area of the Arctic Ocean, although its mean depth is only 50 m. The changing fresh water and biogeochemical signals from the Lena's vast drainage area in the ESAS can be considered to be an indicator of ongoing changes in east Siberia (Savelieva et al., 2000; Semiletov et al., 2000; Gustafsson et al., 2011).

The Lena River Delta occupies 28.5×10^3 km^2; it is the second largest delta in the world (after the Mississippi River Delta) and the largest delta on the Eurasian continent. About 30 000 lakes are located in the delta, 90 % with a typical size of ~0.25 km^2 and only 100 lakes with surface area >10 km^2. According to official terminology, the delta begins downstream of Stolb Island (Fig. 1a). Following Antonov (1964) for 1936–1950 data and the Tiksi Hydromet data for 1953–1990 (Sidorov, 1992), the Bykovskaya Channel of the mouth receives about 24 % and 28 % of the total discharge during winter low water and summer high water, respectively, while the Trofimovskaya Channel receives about 70 % and 54 %, and the Tumatskaya and Olenekskaya channels together receive about 6 % in winter and 18 % in summer. According to Antonov (1964) and Ivanov (1970), the Bykovskaya Channel hydrological regime reflects all important features of the other Lena River Delta channels. Because the Bykovskaya Channel has been the most accessible and investigated, we collected our main set of samples in this channel in different seasons (Fig. 1c, d).

Thaw and release of OC from Arctic permafrost is postulated to be one of the most powerful and vulnerable to a warming climate, yet least understood of these mechanisms (ACIA, 2004; Canadell and Raupach, 2009; Gruber et al., 2004; Semiletov et al., 2011). Thus, a brief description of major vulnerable to a warming climate permafrost-driven carbon pools is required to better understand the linkage among degrading terrestrial and subsea permafrost, the transport and fate of OC to and within the ESAS, and the processes determining fluxes from the ESAS to the atmosphere. The Lena basin is generally located in the continuous permafrost zone where the frozen layer depth varies from 50 to 1500 m. In the delta area the permafrost extends down to 600 m, though large lakes and rivers can be underlain by taliks extending through permafrost (Grigoriev and Kunitsky, 2000). It is assumed that under the Laptev Sea the depth of the offshore layer of relict permafrost varies between 50 and 200 m. Note that estimation of the subsea permafrost distribution is mostly based on modeling, but some geophysical and borehole data show that the subsea permafrost is the most fragile component of the modern cryosphere because the mean temperature of the upper 100 m subsea sediment layer is roughly $-1°$C, compared to $\sim -12°$ C onshore (see discussion in Shakhova et al., 2010a, b; Nicolsky and Shakhova, 2010; Rachold et al., 2007). A vast amount of buried OM containing an average 3–5 % C by weight is presently reentering biological cycling due to thawing and erosion of onshore and offshore permafrost. The upper 100 m of permafrost in Siberia (tundra and northern taiga, 9.4×10^6 km^2) contains no less than 9400 Gt (1 Gt = 10^{15} g) of OC (assuming a conservative mean OC concentration of 0.5 % w/w) that could be available for biogeochemical cycling through permafrost degradation processes, including coastal erosion (Semiletov, 1999a, b). Taking into account that $>$90 % of the ESAS was land that was flooded by the sea during transgression, we may assume that the upper 100 m shelf sediment layer contains at least 1800 Gt of OC. This reservoir also consists of ESAS hydrate deposits estimated to hold $\sim$540–750 Gt of CH_4 and an additional pool of hydrates 2/3 as large ($\sim$360–500 Gt) trapped below as free gas (Shakhova et al., 2010a). Thus, the Lena River serves as a pathway by which terrestrial OC can relocate between the land and shelf carbon megapools. Additionally, bottom erosion and subsea talik formation (Romanovskii and Hubberten, 2001; Nicolsky and Shakhova, 2010; Shakhova and Semiletov, 2007, 2009; Dudarev et al., 2008) can provide a significant amount of old carbon (predominantly in the form of CH_4 and CO_2) to the water column.

The water balance of the Siberian rivers situated in the permafrost region differs from that of rivers in other regions because the upper surface of the permafrost (permafrost table) beneath the seasonally-thawed layer is a water-impermeable sheet that influences land hydrology and hydrochemistry. The permafrost table depth varies from 10 to 10^2 cm north of the Arctic Circle, but in southern mountainous areas discontinuous permafrost (permafrost islands) may be from 10^3 to 10^4 cm deep. Therefore, the regime of groundwater flow is significantly different among rivers situated in different permafrost environments, and is also different between rivers in any permafrost environment and rivers in non-permafrost environments (van Dongen et al., 2008; Guo et al., 2004; Semiletov et al., 2000).

4 Results and discussion

4.1 Fluvial transport and sedimentation in the Lena River Delta and adjacent seas: river-derived particulate matter and particulate organic carbon

Our observations of PM distribution along the Lena River from the upper reaches (Ust'-Kut) in late June (21) to mid August (11) of 2003, to midway (Yakutsk) in late August to early September of 1995 and 1998, and down to the Laptev Sea (Fig. 1) demonstrate that almost all riverine PM settles in the delta, even during spring/summer high river discharge as observed in June–July 2003 (Fig. 2a, b, c). PM concentration increased from low (10–15 mg l^{-1}) in the lower river to as much as 30 mg l^{-1} in the Bykovskaya Channel of the delta, but decreased to 5–10 mg l^{-1} in the channel outlets (Fig. 2). Note that two stations (encircled area in Fig. 2) were accomplished in the Bykovskaya Channel in early August at the beginning of the "second flooding wave" caused by rains (Antonov, 1964). Both August stations recorded a doubling of the PM concentrations compared with the survey accomplished one week earlier. The observed PM increase in the delta channels may also be related to coastal erosion, which is common in this area (Grigoriev, 1993). The mean PM content we measured in the river was 19.4 mg l^{-1}, about half of the mean value (38.5 mg l^{-1}) reported by Romankevich and Vetrov (2001) or the mean (34 mg l^{-1}) from Gordeev et al. (1996). Both used the data obtained by the Soviet Union Hydrometeorological Service. The difference may be related to the use of filters with non-standard, most likely smaller, pores in the latter studies compared to the now-canonical Whatman GF/F borosilicate filters.

The δ^{13}C isotope ratios in river POC ranged between -30 ‰ and -25 ‰ with a mean value near -27 ‰ (Fig. 2d). A similar range of POC δ^{13}C values was reported by Rachold and Hubberten (1999) for the Lena, Khatanga, and Yana basins. The δ^{13}C values of the organic surface bottom sediment fraction, measured in the delta and the southeastern part of the Laptev Sea during September 1997, indicate a terrestrial origin for the OM (Semiletov, 1999a, b). These results agree with Rachold and Hubberten (1999) who found nearly constant δ^{13}C values for particulate OM throughout

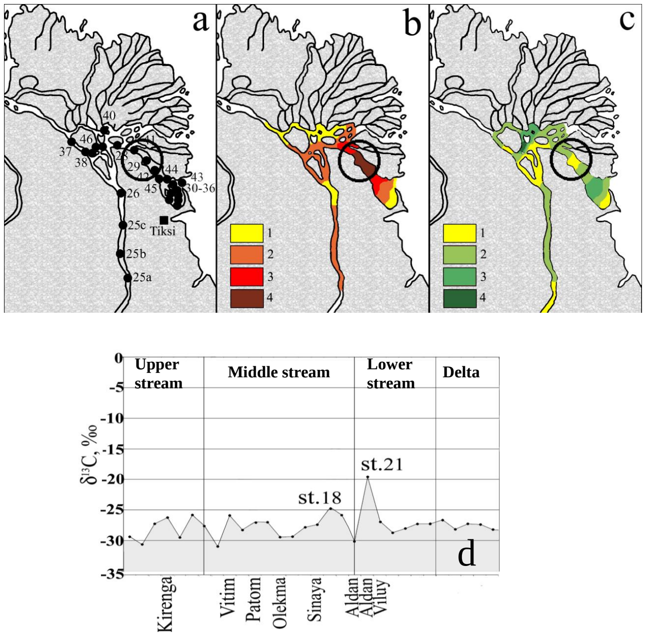

Fig. 2. **(a)** Distribution of sampling sites in the lower Lena River-Lena delta channels in late July–early August 2003; **(b)** PM distribution in the surface layer, mg l^{-1}: (1) <10, (2) 11–20, (3) 21–30, (4) >30; **(c)** POC distribution in the surface layer, mg l^{-1}: (1) <1, (2) 1–1.3, (3) 1.4–1.6, (4) >1.6; **(d)** distribution of particulate organic carbon isotope ratios (δ^{13}C) in the surface layer of the Lena River: from the upper stream to the delta.

the river, with an average of -27.0 ± 0.8 ‰, reflecting the dominant contribution of C_3 plants. Similar values of δ^{13}C were found in the Yenisey estuary (Kodina et al., 1999). Reasons for the variability in POC δ^{13}C are discussed briefly by Rachold and Hubberten (1999), though clarification of this issue will require additional study. Note that the δ^{13}C value (−27.70 ‰) is constant in Neelov Bay of the delta and close to the values (−27.16 to −27.54 ‰) measured in the sediments of the Laptev Sea between Bol'shoy Lyakhovsky Island and Cape Svyatoy Nos where coastal erosion dominates (Semiletov, 1999b).

Using the mean PM concentration of 20 mg l^{-1} and an annual river discharge of 525 km^3 we calculated the mean solid discharge delivered through the delta channels to be 10.5 Tg. Annual discharge of POC may be equal to 0.38 Tg if we use a mean POC value of 0.75 mg l^{-1}, which was obtained in 2003 along the Lena River during late flooding (a moderate-to-high estimate). If we accept Lisytsin's (1994) statement that 85–95 % of total PM (and POC) precipitates on the marginal "filter", then only about 1 Tg of PM, and 0.03–0.04 Tg of POC reaches the Laptev Sea from the Lena River. The Lena's POC export would then be two orders of magnitude less than the annual input of eroded terrestrial carbon onto the shelf of the Laptev and East Siberian seas, which is estimated to be about 4 Tg (Rachold et al., 2004). Observations support the hypothesis of a dominant role for coastal erosion (Semiletov, 1999a, b) in ESAS sedimentation and the dynamics of the carbon/carbonate system. The marginal filter hypothesis is supported by the fact that the Lena Delta is growing (Grigoriev, 1993). Similarly, in summer and fall the solid discharge of other major East Siberian Sea rivers, the Indigirka and Kolyma, is also limited by the near-mouth areas (Ivanov and Piskun, 1999). Note that OM exported by the eastern GSARs (Lena, Indigirka, Kolyma) and originating from local coastal erosion, despite high bulk radiocarbon ages, is less degraded than OM carried by their western Siberian counterparts (Ob and Yenisey). This is consistent with increasing permafrost and a shorter annual thaw period in eastern Siberia compared with western Siberia (Guo et al., 2004; Gustafsson et al., 2011; van Dongen et al., 2008).

A second Lena River "flooding wave" in summer 2008 brought an unusually high amount of PM and POC with a light δ^{13}C signature to the Laptev Sea; this material was dispersed further north of the Lena Delta (Sánchez-García et al., 2011). At that time the contribution from coastal erosion was small, because low winds and small waves did not move the eroded material from the foreshore zone to the sea (Charkin et al., 2011). Thus, the partitioning between fluvial and coastally-eroded POC input to the sea can vary significantly from year to year depending on hydrometeorological conditions over land and ocean.

The spatial trends of $\delta^{13}C_{org}$ in the bottom sediment can be used to quantitatively estimate the contribution of terrestrial organic matter (CTOM) to the ESAS sediment. Following Walsh et al. (1989), the amount of OC derived from terrestrial end-member $\delta^{13}C_{ter}$ (terrestrial C) can be calculated from the data as

$$\text{CTOM } (\%) = (\delta^{13}C_o - \delta^{13}C_{mar}) \cdot 100 / (\delta^{13}C_{ter} - \delta^{13}C_{mar}),$$

where "o", "mar", and "ter" refer to the δ^{13}C values of observed, marine, and terrestrial sediment, respectively. If we take the two end members to be a $\delta^{13}C_{ter}$ of −27 ‰, typical of higher plants, and a $\delta^{13}C_{mar}$ of −21 ‰, typical of phytoplankton (Walsh et al., 1989; Naidu et al., 2000), then the CTOM values for the East Siberian Sea western geochemical area and eastern area (bounded by CTOM = 70 %) become 86 % and 52 % of terrestrial OC, respectively (Fig. 3). Note that the $\delta^{13}C_{mar}$ value given for phytoplankton is for pelagic organisms; the $\delta^{13}C_{mar}$ of ice algae is significantly different. However, the ice algae and melt water do not play a role in September (Nikiforov and Shpaikher, 1980; Anderson at al., 2009, 2011), which is the end of hydrological summer, and is also when most of our offshore data were obtained.

Calculations made for the entire ESAS indicate that there is a significant amount of terrestrial OC stored within sediments, especially in the near-shore zone most strongly influenced by coastal erosion; between the Dmitry Laptev Strait and the Kolyma mouth, the OC is almost all of terrestrial origin (from 81 to 100 %, mean CTOM = 93 %). In contrast, the sediments underneath transformed Pacific-origin water (the

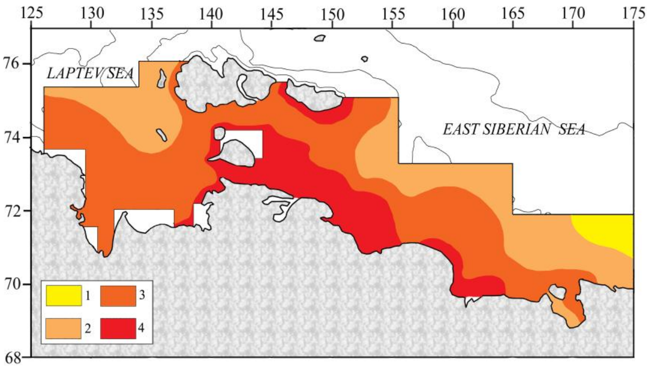

Fig. 3. CTOM (%) in the ESAS surface sediment: (1) <40 %, (2) 40–69 %, (3) 70–98 %, (4) 99–100 %.

eastern area) are almost half of marine origin (Fig. 3). That result agrees well with n-alkane measurements in the surface sediment OC (Vetrov et al., 2008) and a three end-member isotopic mixing model for POC applied along the Kolyma paleo-river transect (Vonk et al., 2010a) which revealed that the proportion of OC derived from coastal erosion was substantial (51–60 %).

The PM data obtained in the Dmitry Laptev Strait (strongly impacted by the Lena River freshwater plume: Semiletov et al., 2000, 2005) where the river PM signal is negligible (Dudarev et al., 2003) show that "new production" is formed from the old terrigenous carbon with a typical terrestrial signal of $\delta^{13}C_{ter} < -26.5$ ‰. Thus, the river OM discharge probably has no direct influence on marine productivity, while coastal erosion and consequent degradation of "fresh" old terrestrial organics builds the pCO$_2$ value up to 1500–2000 µatm in summer and 5000 µatm in winter, and plays a significant role in biogeochemical processes especially in the southeastern area of the Laptev Sea, and western area of the East Siberian Sea where coastal retreat is highest (Pipko et al., 2005, 2008, 2011b; Semiletov, 1999a, b; Semiletov et al., 2004b, 2007; Anderson et al., 2009, 2011; Alling et al., 2010). A detailed discussion of Lena River-Laptev Sea carbonate system dynamics can be found below (Sect. 4.3).

PM in the western part of the ESAS ranged between 4.7 mg l^{-1} and 79.7 mg l^{-1} in the surface water, and between 5.2 mg l^{-1} and 106.4 mg l^{-1} in the near-bottom water (Semiletov et al., 2005), vs. up to 20 mg l^{-1} during the flooding peak, with a maximum of 70 mg l^{-1} observed at one station located near the main channel (Bol'shaya Trofimovskaya) outlet (Pivovarov et al., 1999). During August-September, when the river discharge is low but the rates of coastal erosion are highest, the mean PM concentration (24.2 mg l^{-1} in the surface layer and 25.4 mg l^{-1} in the near bottom layer, Semiletov et al., 2005) obtained over the western part of the ESAS inner shelf (>40 m in depth) is several times higher than that reported for the Lena River-Laptev Sea shelf system during the "surge freshet" (Pivovarov et al., 1999). This controversy between the paradigm of a "dominant" role for the surge freshet in Lena River discharge of solids vs. the importance of the Lena's summer discharge can be resolved because rates of river bank erosion are maximum during the warmest months of August and September; this erosion has the potential to bring more PM/POC per Lena River water unit than in flooding time (May–early June) when the upper soil body in the low Lena stream is still frozen. Note that the highest PM concentrations (>100 mg l^{-1}) were observed in the Dmitry Laptev Strait which is outside the area of direct river sediment input (Dudarev et al., 2003). The finding of anomalously high concentrations of benthic organisms (up to 100–200 g C m^{-2}) in the coastal bottom depressions of the Laptev Sea (Gukov et al., 1999) could be associated with near-shore accumulation of eroded old OC.

Several studies have shown that the amount of coastally-eroded material transported into the Laptev and East Siberian seas ranges between 40–70 Tg (Are, 1999; Grigoriev and Kunitsky, 2000; Rachold et al., 2004) and the mean OC concentration of this material is 3 %. Annual transport of eroded terrestrial carbon onto the shelf may therefore range from 1.2 to 2.1 Tg. A higher annual estimation (∼4 Tg) was made by Rachold et al. (2004); this value is about two orders of magnitude higher than the Lena River POC discharge into the sea (0.03–0.04 Tg).

4.2 Processes controlling the distribution and loss of fluvial POC across the ESAS

The ESAS is strongly impacted by the Lena River plume during summer time (Anderson et al., 2011; Pipko et al., 2008, 2011b; Semiletov et al., 2000; Shakhova and Semiletov, 2007; Alling et al., 2010). However, the fate of the Lena POC plume across the ESAS and its contribution to sedimentation have not been studied. To illustrate anomalous export of terrOM into the ESAS we show the integrated plume of PM and CTOM in POC for the period 2003–2007 (Fig. 4) using data obtained east of the Lena Delta. These observations support the hypothesis of a dominant role for coastal erosion (Semiletov, 1999a, b) in ESAS sedimentation and in the dynamics of the carbon/carbonate system; this hypothesis is also supported by the CTOM distribution in the surface sediment shown in Fig. 3. It can be seen that the PM and CTOM/POC plume from the Lena is limited to the narrow nearshore zone where most of the Lena River burden settles. The pattern also shows a negligible direct influence of the Lena transport of PM into the East Siberian Sea.

The Quartz/Feldspar (Q/FS) ratios in the western and eastern East Siberian Sea are the same (Q/FS = 0.26), while ratios typical of the Lena River are 10 times higher (between 2 and 2.3) (Serova and Gorbunova, 1997). This evidence also indicates an insignificant direct influence of Lena River transport of PM into the East Siberian Sea. Note that in summer-fall

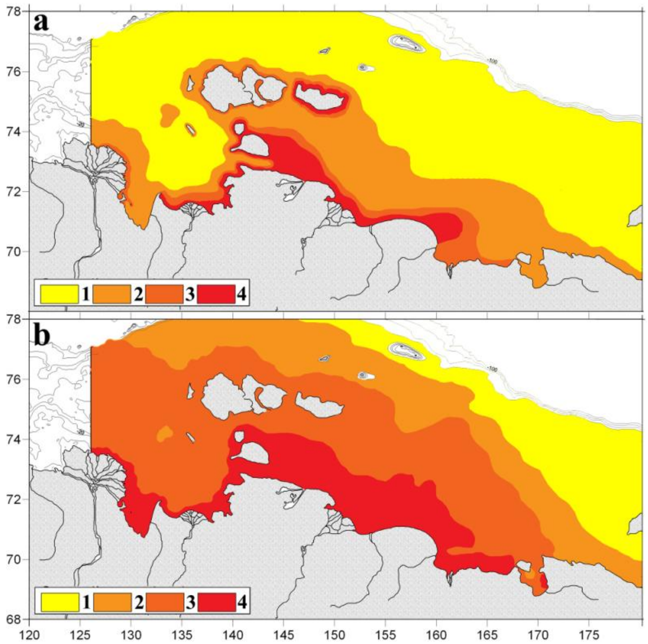

Fig. 4. The secondary role of the Lena River and the dominating role of coastal erosion in ESAS sedimentation is illustrated by the surface distribution of: **(a)** suspended particular materials (PM, mg l^{-1}): (1) <2, (2) 2.1–13, (3) 13.1–24, (4) >24; **(b)** CTOM of POC (%) (1) <25, (2) 25–50, (3) 51–75, (4) >75.

the solid discharge of the major east Siberian rivers, Indigirka and Kolyma, is also limited by the near-mouth areas (Ivanov and Piskun, 1999). We can argue that transport of the eroded material (Semiletov, 1999a, b) plays a major role in the accumulation of carbon in this part of the Arctic Ocean. Note that summer/fall 2008 was the only time that we measured a significant contribution of Lena POC burden to the adjacent part of the Laptev Sea and western East Siberian Sea (Sánchez-García et al., 2011); this occurred because the abrupt second summertime flooding wave combined with low wind conditions prevented removal of eroded carbon from the foreshore to the sea (Charkin et al., 2011).

All the geochemical data obtained in the 1999–2007 summertime expeditions (in contrast to 2008 when an anomalously high Lena River solid discharge was measured, see Sánchez-García et al., 2011; Charkin et al., 2011) clearly show that the depositional environment in the Lena River prodelta and the entire ESAS is dominated by coastal erosion rather than by riverine input. This conclusion agrees well with available CN stable isotope data, C/N ratios, and the distribution of biomarkers in the Buor-Khaya Gulf of the Laptev Sea (Charkin et al., 2011; Karlsson et al., 2011). Thus, we argue that in the 1995–2007 period, the Lena River solid discharge played a minor role, compared with offshore transport of eroded material, in accumulation of carbon in this part of the Arctic Ocean; this conclusion also supports the conclusions of Bauch et al. (2001) and Vonk et al. (2010a).

Our studies to date show that terrOM entering sub-Arctic Baltic and Eurasian Arctic seas follows continent-scale trends in molecular and isotopic composition. *Sphagnum* is a key contributor to the pre-aged (by 1000 s of ^{14}C yr) terrOM in these coastal waters; the greatest *Sphagnum* contribution but the youngest terrOM occurs toward the west (Vonk and Gustafsson, 2009; Vonk et al., 2010a). There was also rapid degradation during settling and in the surface sediment (Sánchez-García et al., 2011; Vonk et al., 2010a) that is supported by numerous $p\mathrm{CO_2}$ observations (Semiletov et al., 2007; Anderson et al., 2009, 2011; Pipko et al., 2011b). A simple box model was parameterized with measurements of advective river input, settling fluxes, and advective export, and was solved for degradation (Gustafsson et al., 2000; van Dongen et al., 2008). The model was run for both bulk POC and bulk DOC individual biomarkers. Rapid terrPOC degradation was constrained (65 % during the 5-day box transit). This translated into a degradation rate for terrPOC nearly 20 times more rapid than for terrDOC, which makes both pools equally important for total terrOM degradation. Similar trends for POC biomarkers and degradation indexes between the Kalix-Baltic and GSAR systems suggest that the early processing may be similar on the Eurasian Arctic shelf (Vonk et al., 2008, 2010b; Gustafsson et al., 2011). Taken together, all recent benchmark studies of the current composition of terrestrially-exported POM suggest that continent–scale trends in the molecular composition of the OM across the west-east Eurasian shelf seas, which reflect differences in both vegetation and climate (Guo et al., 2004; Gustafsson et al., 2011; van Dongen et al., 2008; Vonk et al., 2010a, b), can be distinguished. If the climate in the ESAS region becomes more like the current west Siberian regional climate, these results would predict a greater degree (compared with western Siberia) of decomposition of the old terrOM released by coastal erosion and by the Lena and other eastern GSARs, and thus greater remineralization and release as CO_2.

4.3 Dynamics of the carbonate system in the Lena River-Laptev Sea system

4.3.1 The carbonate system in the river and near-shore zone

In summer 1995, we began a study of the carbonate system and dissolved CH_4 in the Lena River Delta and adjacent shelf waters (Fig. 1). In general we sampled the surface layer, though in 1996–1998 and in 2006 vertical sampling sections were also conducted. We consider the summer surface chemical data from the river to be representative of the whole water column because measurements of temperature, conductivity, dissolved oxygen, colored dissolved OM (CDOM), TOC, pH, and TCO_2 were the same at all depths except close to the bottom. The total carbon (TC) transported by the Lena River is a significant contribution to the sea, especially during flood periods. The fate of this carbon pool, whether consumed more or less rapidly in the adjacent sea or accumulated in the coastal zone, is one of the important questions posed by this system.

Summer

To understand the carbon cycle in the Lena River we carried out a shipboard investigation along a 1700 km section from Yakutsk to Tiksi on the Laptev Sea coast. During a 5-day voyage, in late August to early September 1995, 41 sites were surveyed (Fig. 1c). At that time, the surface waters of the Laptev Sea near the delta were strongly freshened by river discharge. The major components of the carbon cycle (TCO_2, TOC, pH) and Si (an indicator of continental crust weathering) were sampled (Table 1). Because the river includes water entering from a vast basin that exceeds the entire area of the ESAS, the signature from the major tributaries, the Aldan and Viljuy rivers, can be identified.

TCO_2 varies inversely to TOC concentration down the Lena River (Figs. 5, 6). In the coastal waters of Tiksi Bay (sites 40 and 41) TCO_2 and TOC concentrations increase drastically. It is true that primary production should decrease TCO_2 if no additional sources of CO_2 are available, but here an additional source exists in the form of oxidized terrOC. Thus, increased TOC, TCO_2, and TC values are consistent with the maximum primary production (432 mg C m^{-3} d^{-1}) and mesozooplankton biomass (890 mg m^{-3}) measured in the Lena River-Laptev Sea system in the same month (September 1991) (Sorokin and Sorokin, 1996). Therefore Tiksi Bay is a summer biogeochemical oasis, where the estuarine environment replaces the winter sea environment when river discharge is low.

The highest TOC concentrations (Table 1) were found in the Lena, mid-stream between the inflow of the Aldan River and the Viljuy River (Figs. 1, 5a). The TC value increases by 0.3 mM (∼0.2 mM as TCO_2, ∼0.1 mM as TOC) due to inflow of the Viljuy River in comparison to a TC increase by 0.2 mM (∼0.0 as TCO_2, ∼0.2 mM as TOC) contributed by the Aldan River. The plot of TCO_2 and TOC shows high variability: (1) TCO_2 ranges from 0.63–1.07 mM (mean = 0.85 mM) from Yakutsk to Stolb Island, and between 0.84–1.38 mM (mean = 1.06 mM) in the delta and Tiksi Bay; (2) TOC decreases from 0.39–0.93 mM (mean = 0.59 mM) from Yakutsk to Stolb Island, and from 0.38–0.53 mM (mean = 0.47 mM, except at site 40) from Stolb Island to Tiksi (Fig. 6). In general, the distributions of TCO_2 and TOC in river waters, excluding Tiksi Bay, are negatively correlated ($r = -0.60$).

Comparing our results obtained along the Lena River and in the nearshore zone near the Lena Delta at the same time (September) in 1995 and 1998 with the data obtained by Cauwet and Sidorov (1996) in 1991 within the framework of the Russian-French SPASIBA-2 expedition reveals significant inter-annual variability in mean values of total TCO_2, TOC, and their sum (TC), Table 2. We suggest that this variability is determined by changes in land hydrology which cause differences in the Lena discharge (Pipko et al., 2010; Semiletov et al., 2000). About one month is required for water to travel from Yakutsk to the Laptev Sea. For example, the mean Lena discharge (18 800 m^3 s^{-1}) in August 1995 was

Table 1. Major components of carbon cycling and silicates sampled between Yakutsk, the Lena Delta, and Tiksi Bay (late August–early September, 1995).

N_{CT}	pH_{25}	TCO_2, mM	TOC, mM	Si, µM	pCO_2, µatm	TCO_2+TOC, mM
Yakutsk – Stolb Is.						
1	7.94	0.92	0.46	91.0	479	1.38
2	7.90	0.75	0.59	89.9	428	1.34
3	7.79	0.78	0.45	85.9	569	1.23
4	7.78	0.64	0.57	90.3	486	1.21
5	7.67	0.66	0.75	88.2	577	1.41
6	7.57	0.66	0.89	88.4	727	1.55
7	7.66	0.66	0.93	83.4	613	1.59
8	7.68	0.65	0.69	82.4	575	1.34
9	7.67	0.63	0.63	81.7	568	1.26
10	7.78	0.82	0.75	85.7	597	1.57
11	7.81	0.75	0.63	81.6	499	1.38
12	7.84	0.84	0.58	80.8	532	1.42
13	7.90	1.00	0.47	78.8	558	1.47
14	7.94	1.07	0.67	80.8	558	1.74
15	7.96	0.99	0.52	77.0	474	1.51
16	7.95	1.01	0.48	77.0	494	1.49
17	7.95	1.02	0.43	85.8	498	1.45
18	8.01	0.95	0.56	74.6	380	1.51
19	7.95	1.06	0.58	77.0	487	1.64
20	7.86	0.92	0.43		522	1.35
21	7.80	0.93	0.52	72.4	611	1.45
22	7.81	0.93	0.39	72.3	593	1.32
Mean	7.83	0.85	0.59	82.1	538	1.44
The Lena Delta – Tiksi Bay						
23	7.83	0.90	0.40	72.1	540	1.30
24	7.86	0.84	0.53	71.4	470	1.37
25	7.85	1.03		71.1	589	
26	7.86	0.99	0.38	63.9	544	1.37
27	7.87	0.99	0.38		520	1.37
28	7.75	1.01	0.38	52.5	717	1.39
29	7.85	0.95	0.39	52.5	534	1.34
30	7.82	0.93	0.39	52.5	551	1.32
31	7.87	0.98	0.40		526	1.38
32	7.78	0.94	0.40	52.5	609	1.34
33	7.82	1.04	0.42	63.9	608	1.46
34	7.76	1.14	0.45	41.3	715	1.59
35	7.83	0.96	0.39	38.7	523	1.35
36	7.81	0.96	0.47	43.8	513	1.43
37	7.81	1.16	0.38	52.5	593	1.54
38	7.78	1.36	0.43	47.2	688	1.79
39	7.78	1.25	0.47	37.4	585	1.72
40	7.75	1.38	1.30	31.5	682	2.68
41	7.79	1.32		35.0	736	
Mean	7.81	1.06	0.47	51.8	592	1.51

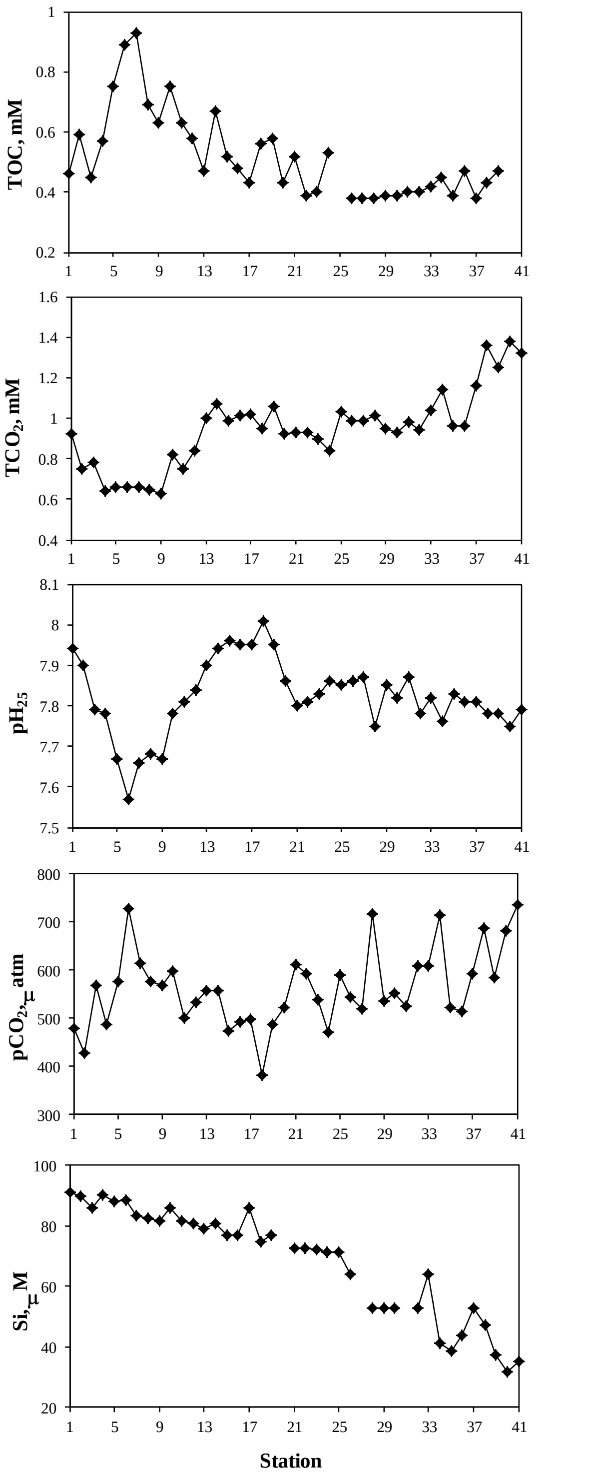

Fig. 5. Distribution of measured TOC, TCO_2, pH_{25}, Si, and $p$$CO_2$ along the Lena River from Yakutsk to Tiksi: stations 1–22 (from Yakutsk downstream to the delta, Stolb Island); stations 23–38 (from Stolb Island to the Laptev Sea), late August–early September 1995.

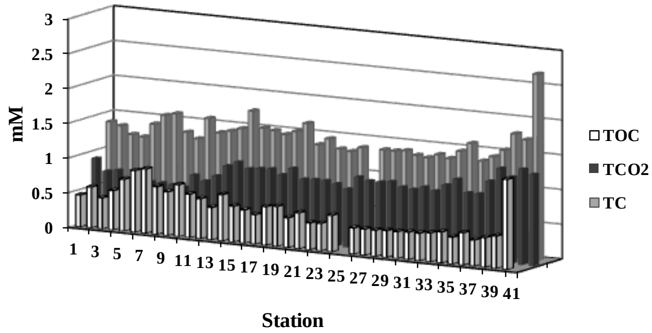

Fig. 6. Distribution of measured TOC (mM), TCO_2 (mM), and TC (mM) along the Lena River from Yakutsk to Tiksi (late August–early September 1995).

Table 2. Mean concentrations of TOC, TCO_2, and TC obtained along the Lena River (from Yakutsk to the Laptev Sea), and Lena River discharge (W) in August of different years.

Time	TCO_2, mM	TOC, mM	TC, mM	W, km^3
September 1995	0.95	0.53	1.42	50.35
September 1998	0.65	0.58	1.24	56.51
September 1991 (SPASIBA-2)	0.60	0.60	1.20	62.94

about 89 % of the discharge in August 1998 (21 100 $m^3\,s^{-1}$), while the mean TC value in 1998 was 87 % of the TC value in 1995. These changes appear to reflect a dilution effect. Cauwet and Sidorov (1996) also found a positive correlation between TOC values and river discharge. Increasing Lena discharge causes opposite effects on dissolved organic and inorganic carbon: TOC concentration increases, while TCO_2 concentration decreases (Pipko et al., 2010). This phenomenon was also observed in the Kalrix River in sub-Arctic Scandinavia (Ingri et al., 2005).

The mean TCO_2 in the river from Yakutsk to Stolb Island (where the major Lena stream separates into delta channels) was 0.65 mM in 1998, about 76 % of the mean TCO_2 obtained in 1995. We found a high negative correlation (-0.91) between TCO_2 values and river discharge for the 40 sites represented in Table 1. In the river system the TC distribution was quasi-homogeneous with a mean of 1.48 ± 0.03 mM (except for site 40 which exhibits an extremely high TOC), while TOC and TCO_2 changed in an opposite manner (Fig. 6). This indicates that transformation of OC into mineral carbon in the aquatic environment, and mineral carbon into the organic form in the Lena River watershed, can be considered quasi-equilibrated processes. However, the geochemical processes responsible for that transformation require additional study. The mean pH_{25} was 7.83 and 7.81 in the delta and coastal waters of the Laptev Sea,

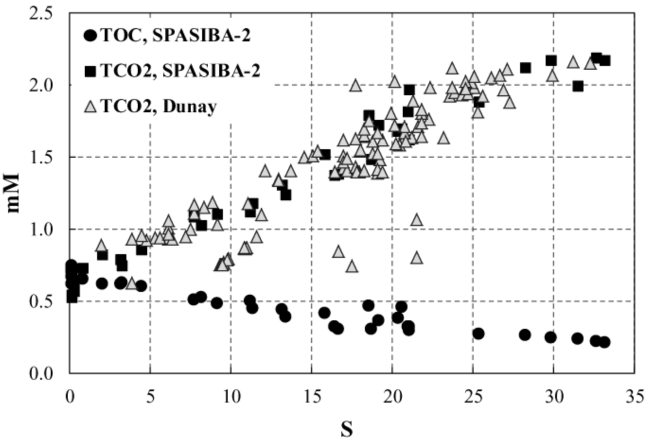

Fig. 7. The mean values of TOC and TCO_2 vs. S measured in September 1991 (adopted by Cauwet and Sidorov, 1996) and in September 1997 onboard H/V *Dunay*.

respectively; the value is similar in the middle and downstream waters of the Lena. Relatively high pH_{25} values, up to 8.01 (site 18) in the lower river, are similar to the pH_{25} values measured near outlets of small streams draining from lakes in the taiga-tundra zone of the Kolyma lowland (Semiletov et al., 1996a).

The interannual difference in the hydrological regime is also reflected in the temperature gradient of the river. In early September 1998 a 12 °C gradient was measured, from 18.6 °C near Yakutsk to 6.6 °C in Tiksi Bay, compared to a 7 °C change in 1995. The larger temperature gradient in 1998 resulted from greater cooling in Tiksi Bay, down to 6.6 °C in 1998 vs. 10.4 °C in 1995. An increase in river discharge is in agreement with an increase in the horizontal gradient of the river's surface temperature between Yakutsk and Tiksi. This parameter can be used as an additional indicator of interannual and long-term thermal changes across the watershed of the Lena (and of other rivers). A decrease of TCO_2 in September 1998 relative to September 1995 could be the result of an abundant supply of snow and glacial melt water from mountain areas in the Lena River watershed.

To explore the fate of organic-rich Lena River water in the Laptev Sea, when our measurements from September 1997 or data collected by Cauwet and Sidorov (1996) in 1991 are plotted against S, results indicate a similarity between mean values of TCO_2 and TOC (0.58 mM and 0.60 mM, respectively) near zero S (Fig. 7). These values reflect the contributions from all rivers that discharge into the Laptev Sea (Lena, Khatanga, Yana, Anabar). TCO_2 values obtained in September of 2003 and 2004 in the East Siberian Sea and extrapolated to zero S were 0.52 mM and 0.50 mM, respectively (Semiletov et al., 2005). Thus, assuming the melt water signal is negligible over the shallow East Siberian Sea shelf at the end of hydrological summer, we can conclude that an integrative TCO_2 signal from the Lena and other rivers flowing into the Laptev Sea and further east is slightly higher than the signal from rivers that discharge into the East Siberian Sea.

The mean pCO_2 value increased from 538 µatm in the middle and lower portions of the river to 592 µatm in the delta and nearshore sea. This increase is caused by a corresponding increase of TCO_2 that dominates the effect of the temperature shift on the carbonate system. These results generally agree with those measured in the mouths of the Ob and Yenisei rivers (Makkaveev, 1994), and in arctic Alaskan rivers (Kling et al., 1991).

A simple calculation shows that each year the Lena River alone brings to the sea a significant amount of dissolved inorganic and organic carbon; if the mean $TCO_2 = 1$ mM, TOC = 0.5 mM, TC = 1.5 mM ($\sim$18 mg l^{-1}), and W = 525 km^3 yr^{-1}, then TC entering the sea would be almost 10 Tg C yr^{-1}. Understanding the proportions of "primary river carbon" that are redistributed to the main carbon pools is an important problem that is starting to be addressed (e.g., Stein and Macdonald, 2004; Alling et al., 2010).

Winter

In winter 1995 pCO_2 was 1267–1579 µatm in the Lena Delta channels, 1045–1500 µatm in Neelov Bay, and 2140–5800 µatm in Tiksi Bay due to an increase in TCO_2 and a decrease in pH (Semiletov et al., 1996b). Maximum pCO_2 values were found in May when winter OM is oxidized and CO_2 accumulates beneath the ice cover (Semiletov et al., 1999b). May is a period of spring phytoplankton blooms when dissolved CO_2 is removed from ocean surface waters by photosynthesis, but here the winter accumulation primarily determines the spring concentration of CO_2. Data obtained in April–May 2002 in the Buor-Khaya Gulf of the Laptev Sea, adjacent to the eastern part of the river, show dramatic pCO_2 changes throughout the water column from summer (Fig. 8a) to winter (Fig. 8b), when fast-ice thickness exceeds 2.0 m. In winter, when OM decomposition dominates, pCO_2 was greater than 4000 µatm near the bottom. Data show that supersaturation beneath the sea ice is a principal feature of the carbon cycle in the Arctic Siberian seas (Semiletov, 1999a; Semiletov et al., 1996a, b, 2004a, b) and in the central basin of the Arctic Ocean (Semiletov et al., 2007). Wintertime CO_2 and CH_4 accumulation beneath the ice was also found in northern Siberian lakes (Semiletov, 1999a).

4.3.2 The influence of the Lena River on the carbonate system dynamics in the East Siberian Arctic Shelf seas

Here we present CO_2 data that were obtained in the Laptev Sea, which is influenced strongly by inflow of the Lena River (Figs. 9, 10).

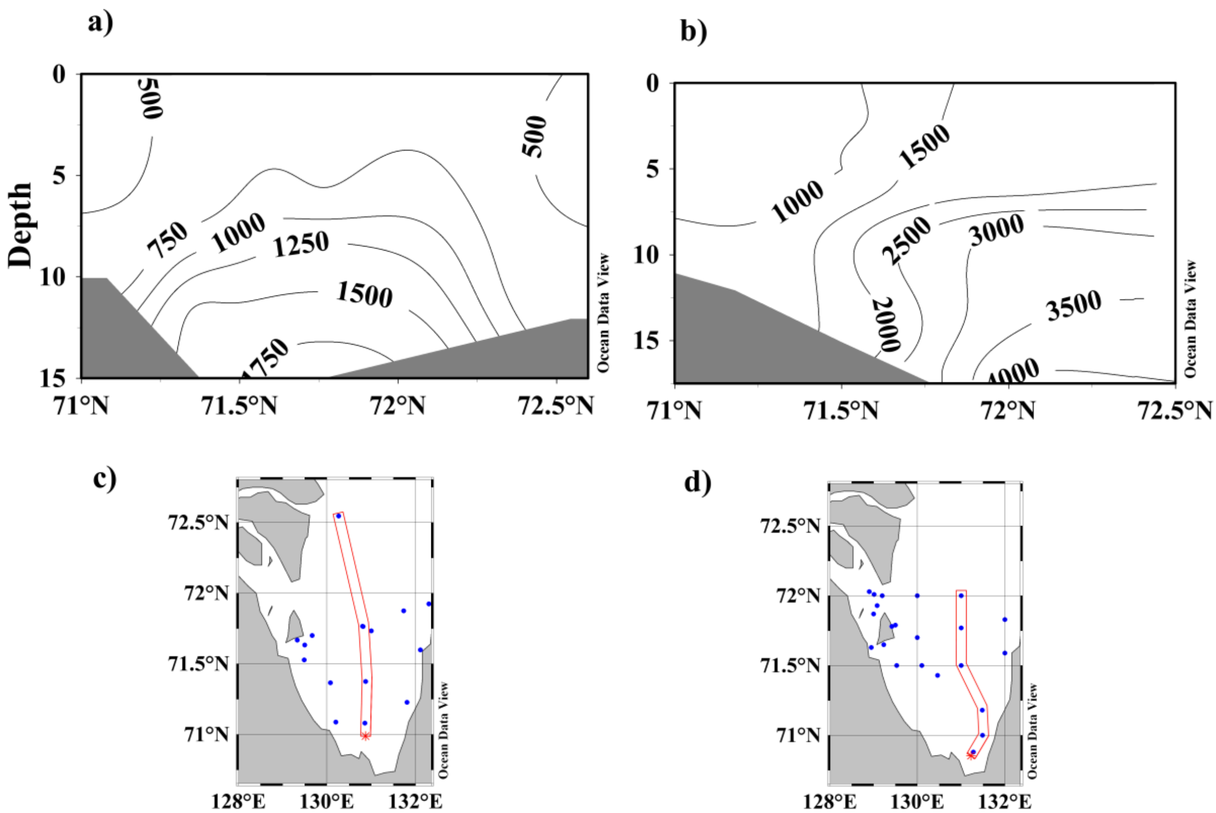

Fig. 8. Meridian vertical pCO_2 (μatm) profile across the Bur-Khaya Gulf of the Laptev Sea: **(a)** summertime distribution (September 1997); **(b)** wintertime distribution (April 2002); transect positions in **(c)** 1997, **(d)** 2002.

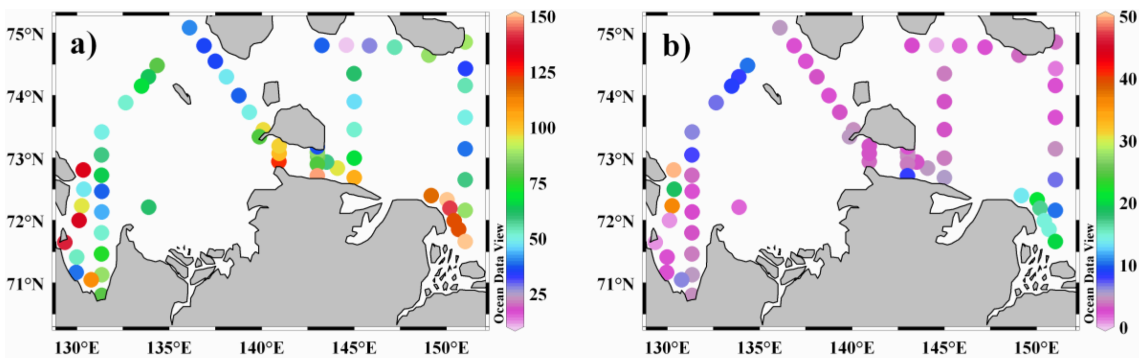

Fig. 9. Distribution of ΔpCO_2 (μatm) and CO_2 fluxes (FCO_2, mmol m^{-2} d^{-1}) between the surface water and atmosphere in the Laptev Sea, September 2005.

East of the delta

Year-round study of the CO_2 system in the Lena River-Laptev Sea system shows that river waters are a source of CO_2 into the atmosphere during all seasons. Likewise, long-term hydrochemical investigations by the Russian Arctic and Antarctic Research Institute (Rusanov and Shpaikher, 1979) and a continuing study by the Tiksi Hydrometeorological Service show that dissolved oxygen concentrations are inversely proportional to the pCO_2 concentrations obtained in our data. The measured concentrations of dissolved CO_2 and oxygen are a result of OM oxidation and interaction between the physical and biological processes of cooling or warming, photosynthesis, and respiration, as well as mixing of different waters.

The ΔpCO_2 ($\Delta pCO_2 = pCO_2^{sw} - pCO_2^{air}$) data show that the surface layer of the eastern Laptev Sea is supersaturated in CO_2 relative to the atmosphere (Fig. 9); the

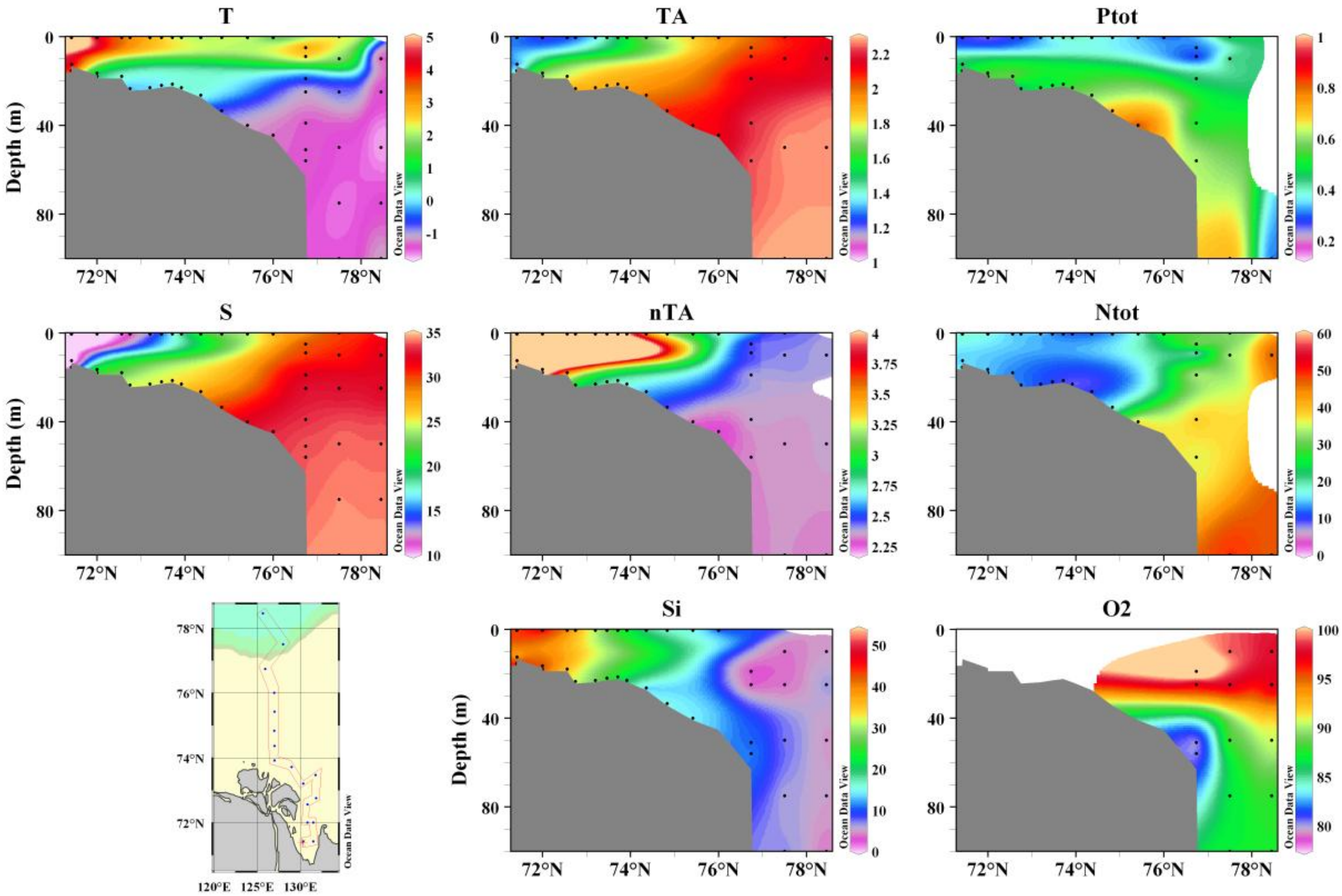

Fig. 10. Vertical distribution of temperature (T, °C), S, TA (mmol kg^{-1}), normalized TA (nTA, mmol kg^{-1}), Si (µM), total dissolved phosphorus (P_{tot}, µM), total dissolved nitrogen (N_{tot}, µM), and oxygen saturation (O_2, %) within the south-north transect across the Laptev Sea, September 2006.

highest supersaturation was found near the delta and over the Vasilievskaya-Semenovskaya shoals which are characterized by high rates of bottom erosion and elevated values of POC (Dudarev et al., 2008), perhaps as a result of the upwelling of bottom water enriched in CO_2. Because the uptake of CO_2 by photosynthesis is relatively high only in the surface waters of the Siberian river deltas (in the Lena River up to 0.1–0.3 g C m^{-2} d^{-1}) and decreases 10–100 times toward the shelf edge (Sorokin and Sorokin, 1996), we assume that the level of CO_2 saturation is not strongly affected by photosynthesis especially in low-transparency water that is strongly impacted by the Lena River (Pipko et al., 2011b; Semiletov et al., 2007). Available data show that the eastern Siberian coastal zone, influenced by river run-off and coastal erosion, is a source of atmospheric CO_2 during summer and winter, whereas areas of the Laptev Sea remote from the coastline tend to be sinks in summer (Semiletov, 1999a, b; Semiletov et al., 2007; Pipko et al., 2008, 2011a, b; Anderson et al., 2009, 2011). Therefore, the CO_2 exchange between air and sea exhibits a spatial-temporal mosaic pattern across the different Arctic seas.

The summer data show that the sea surface layer that is strongly influenced by the Lena River is 10–50 % (Figs. 8a, 9a) higher than saturation in CO_2 while it is 200–500 % higher in areas strongly affected by coastal erosion (Semiletov et al., 1996a, b, 2007). These data show that the coastal zone pCO_2 is higher than the pCO_2 of the rivers. The combined effect of CO_2export by rivers and aerobic oxidation of coastline-eroded carbon are responsible for that increase (Semiletov, 1999a, b).

4.4 North of the delta

The September 2006 vertical transect from the Lena Delta to the north illustrates how far the Lena River signal is propagated (Fig. 10). It has been shown that the relatively fresh and warm (<2 °C) riverine plume is characterized by high Si and normalized TA (nTA) relative to the typical Atlantic water layer in the Arctic Ocean, where S = 35. Total dissolved phosphorus (P_{tot}) and total dissolved nitrogen (N_{tot}) are depleted in the Lena plume at the end of biological summer, but their concentrations (0.3 and 10–15 µM, respectively) are still significant because primary production is low (Sorokin and Sorokin, 1996). Vertical fluxes in the inner ESAS yield a strong efflux of CO_2 into the atmosphere ranging from 0.4 to 49.9 $mM\,m^{-2}\,d^{-1}$ (Fig. 9b), while the northern periphery of the Lena River plume (up to 80° N) is a lesser source of CO_2 to the atmosphere (Semiletov et al., 2007). Measurements of CO_2 turbulent flux taken in September 2005 above open water over the outer Laptev Sea shelf and slope ranged between the *positive* (evasion) and *negative* (invasion) values of +1.7 $mmol\,m^{-2}\,d^{-1}$ and −1.2 $mmol\,m^{-2}\,d^{-1}$ (Semiletov et al., 2007). Comparing distribution of CO_2 fluxes with surface temperature and S shows that warmer and fresher water, which is probably the Lena River plume, acts as a source of atmospheric CO_2, while relatively colder and saltier water near the ice edge is a sink (Semiletov et al., 2007).

Ice-tethered observations made using an autonomous SAMI CO_2 sensor in the central Arctic Ocean by the Russian North Pole-33 drifting station in summer 2005 show that $p\mathrm{CO_2}$ values ranged between 425 µatm and 475 µatm with a drop to 375 µatm in mid-August; these values are higher than the mean summer atmospheric value in the Arctic (∼340–345 µatm). The photosynthetically active radiation (PAR) was near zero until mid-June when the snow and sea ice began to melt; it reached its maximum value in July, but no correlation between PAR and values of $p\mathrm{CO_2}$ has been found (Semiletov et al., 2007). The source of those high values may be high rates of bacterial respiration (Rich et al., 1997), perhaps indicating a rapid turnover of sub-ice dissolved OM which is closely connected with high sea-ice algal production (e.g., Gosselin et al., 1997). Another explanation for such a phenomenon involves the Lena River plume. Surface water from the Laptev Sea shelf, rich in runoff, dissolved CO_2, and DOM, follows the Gakkel Ridge towards Fram Strait (Anderson et al., 1998). This could explain the high concentration of DOM measured over the Amerasian shelf and Arctic Basin (Wheeler et al., 1996; Opsahl et al., 1999). Surface water concentrations of humic substances were ten times higher in the 87–89° N latitudes than in the 70–85° N latitudes; the former area is associated with S = 31 instead of 34, and presumably this pattern reflects a clear signal of terrestrial carbon near the North Pole which originated from the Lena plume and traveled as part of the Transpolar Drift (Sobek and Gustafsson, 2004). Hence, the influence of the Lena River propagates throughout the interior Arctic basins.

The carbonate system parameters from all seasons show that the river is supersaturated in CO_2 compared to the atmosphere, up to 1.5–2 fold in summer (Figs. 5, 8, 9), and 4-5 fold in winter. This results in CO_2 supersaturation in the shallow-mid shelf sea, although near-ice-edge waters are generally undersaturated (Semiletov et al., 2007; Anderson et al., 2009, 2011; Pipko et al., 2011a, b). Macro-synoptical conditions determine the distribution of $p\mathrm{CO_2}$ in the southeastern Laptev Sea; either the anti-cyclonic pattern dominates over the Arctic Basin (Proshutinsky and Johnson, 1997; Semiletov et al., 2000) and the transport of river runoff is northward, causing a northward extension of the surface layer supersaturated in CO_2 as has been shown for September of 2008 by Pipko et al. (2011b), or the cyclonic pattern dominates and the northward transport of river water decreases compared to the anticyclonic situation, resulting in a decrease of the supersaturated surface layer north off the Lena Delta and eastward propagation of the Siberian Coastal Current enriched by CO_2. Note that the highest regional CO_2 supersaturation (by up to 15 times) induced by oxidation of eroded carbon was found in winter in the nearshore zone adjacent to the Lena Delta (Semiletov, 1999a; Semiletov et al., 1996b, 2004a, 2007).

5 Dissolved methane in the Lena River-Laptev Sea system

The first dissolved CH_4 measurements were taken in the Lena River Delta channels in fall of 1994 (Semiletov et al., 1996a, b). In the Lena River Delta the concentration of dissolved CH_4 was usually below detection limit (which was ∼15 nM at that time) during summertime, while the highest concentrations (50–70 nM) were found in the Neelov Bay in the southeastern part of the delta. However, during the ice-covered season the concentration of CH_4 varied from trace to 100 nM in the Bykovskaya Channel and Neelov Bay and in the Olenekskaya Channel (Semiletov, 1999a). This indicates that high wintertime concentrations of CH_4 may be somehow related to degradation of permafrost in the Lena basin, especially in the fault zones, and drainage of thaw lakes enriched in dissolved CH_4 (Semiletov et al., 1996a, 2004b; Shakhova et al., 2005, 2010a, b). Thaw lakes with dissolved CH_4 concentrations ranging over two orders of magnitude, from $\sim 10^{-2}$ to 10^0 µM in the high Arctic (Lena Delta/Tiksi area) to ∼10 to 10^1 µM in the subarctic (Kolyma Lowland/Cherskiy area), which are connected to the Siberian permafrost rivers by numerous channels and through taliks, would be an additional source of riverine CH_4, dissolved CO_2, and DOC (Shakhova et al., 2005; Semiletov et al., 1996a, b, 2004b). This hypothesis is supported by the observed increase in dissolved CH_4 concentrations in the Lena River compared with the Ob; the latter is located mostly in a non-permafrost zone (Shakhova et al., 2007a). In an early phase of these investigations, based on the data obtained in the Laptev and East Siberian seas during cruises in 2003 and 2004, Shakhova and Semiletov (2007) found a significant correlation between integrated CH_4 storage and either integrated S values ($r = 0.61$) or integrated values of TCO_2 ($r = 0.62$); both these features characterize marine water as compared to the fresh water flowing onto the shelf from rivers and streams. This suggests that riverine dissolved CH_4 export contributed only a minor amount to the dissolved CH_4 content of shallow shelf water.

In contrast, anomalously high concentrations of dissolved CH_4 (up to 5 µM) and an episodically (non-gradually)

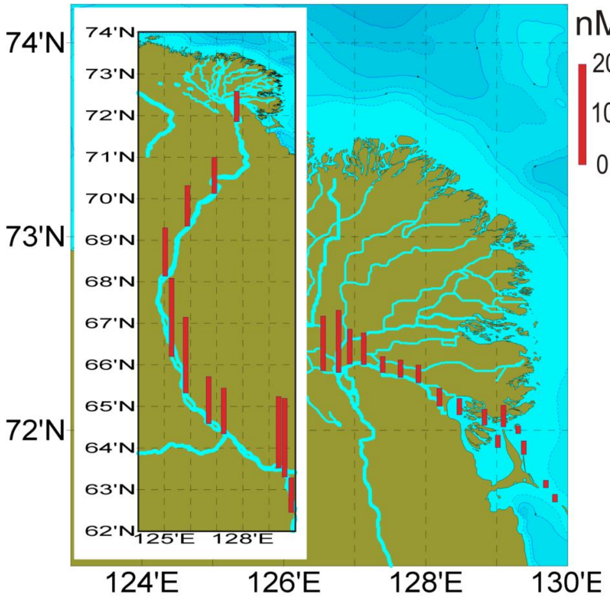

Fig. 11. Distribution of dissolved CH_4 (nM) in the Lena River water, September 2008.

increasing atmospheric mixing ratio (up to 8.2 ppm) were measured in some areas of the ESAS (Semiletov, 1999a; Shakhova et al., 2007b, 2010a, b). Results of studies made in the Lena Delta channels in early September 2006 (Shakhova et al., 2010a, b) and along the Lena River from Yakutsk to the Laptev Sea in late August–September 2008 are shown in Fig. 11. A localized area of high dissolved CH_4 (up to 125 nM) was only detected at the beginning of the Olenekskaya Channel (Fig. 3), where ice-complex bank erosion provides a huge amount of eroded OM, forming an anaerobic bottom environment which is conducive to early diagenetic CH_4 production. The Olenekskaya Channel is also underlain by a fault zone which may allow the formation of through taliks, disturbing the permafrost-associated hydrates (Collett and Dallimore, 2000; Shakhova and Semiletov, 2007, 2009; Shakhova et al., 2010a, b). However, in the Bykovskaya Channel (the deepest of the Lena Delta channels) we detected a drastic decrease in dissolved CH_4 concentration downstream. This picture is further consistent with the low CH_4 atmospheric emissions ($\sim$10 mg m^{-2} d^{-1}) found recently in the Lena Delta (Ulrich et al., 2009) and our high-precision (Shakhova et al., 2010a, b) air CH_4 measurements made in 2006 along the Lena River channels which demonstrated relatively low values ($\sim$1.90 ppm) vs. the moderate (1.95–2.10 ppm) and high (>2.10 ppm) values measured over the ESAS. Taken together, riverine export of CH_4 plays only a minor role in the CH_4 budget of ESAS coastal waters.

6 Conclusions

The Lena River integrates biogeochemical signals from its vast drainage basin, and its signal reaches far out over the Arctic Ocean. The annual Lena River discharge of POC may be equal to 0.38 Tg (a moderate-to-high estimate). If we instead accept Lisytsin's (1994) statement that 85–95 % of total PM (and POC) precipitates on the marginal "filter", then only about 0.03–0.04 Tg of POC reaches the Laptev Sea from the Lena River. The Lena's POC export would then be two orders of magnitude less than the annual input of eroded terrestrial carbon onto the shelf, which ranges from 1.2–2.1 to 4.0 Tg.

Transformation of riverine OC into mineral carbon, and mineral carbon into the organic form in the Lena River watershed, can be considered quasi-equilibrated processes. Significant inter-annual variability in mean values of TCO_2, TOC, and their sum (TC) has been found. This variability is determined by changes in land hydrology which cause differences in the Lena discharge. A negative correlation is between TC in September and mean discharge in August; a time shift of about one month is required for water to travel from Yakutsk to the Laptev Sea. Increasing Lena River discharge causes opposite effects on dissolved organic and inorganic carbon; TOC concentration increases, while TCO_2 concentration decreases. TC entering the sea with the Lena discharge is estimated to be almost 10 Tg C yr^{-1}

The Lena River is characterized by relatively high concentrations of the primary greenhouse gases dissolved CO_2 and CH_4. During all seasons the river is supersaturated in CO_2 compared to the atmosphere, by up to 1.5–2 fold in summer, and 4–5 fold in winter. This results in a narrow zone of significant CO_2 supersaturation in the adjacent coastal sea. Localized areas of dissolved CH_4 in the Lena Delta channels may reach 100 nM, but the CH_4 concentration decreases to 5–20 nM towards the sea, which suggests that riverborne export of CH_4 makes only a minor contribution to the ESAS CH_4 budget in coastal waters. Instead, the seabed appears to be the source that provides most of the CH_4 to the Arctic Ocean.

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