Fluvial carbon dioxide emission from the Lena River basin during spring flood

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Key words: CO_2, C, emission, permafrost, river, export, landscape, Siberia

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Abstract

Greenhouse gas (GHG) emission from inland waters of permafrost-affected regions is one of the key factors of circumpolar aquatic ecosystem response to climate warming and permafrost thaw. Riverine systems of central and eastern Siberia contribute a significant part of the water and carbon (C) export to the Arctic Ocean, yet their C exchange with the atmosphere remain poorly known due to lack of *in-situ* GHG concentration and emission estimates. Here we present the results of continuous in-situ pCO_2 measurements over a 2600-km transect of the Lena River main stem and lower reaches of 20 major tributaries (together representing watershed area of 1,661,000 km², 66% of the Lena's basin), conducted at the peak of the spring flood. The pCO_2 in Lena (range 400-1400 µatm) and tributaries (range 400-1600 µatm) remained generally stable (within ca. 20 %) over the night/day period and across the river channels.

The pCO_2 in tributaries increased northward with mean annual temperature decrease and permafrost increase; this change was positively correlated with C stock in soil, the proportion of deciduous needle-leaf forest and the riparian vegetation. Based on gas transfer coefficients obtained from rivers of the Siberian permafrost zone, we calculated CO_2 emission for the main stem and tributaries. Typical fluxes ranged from 1 to 2 g C m^{-2} d^{-1} (>99% CO_2, < 1 % CH_4) which is comparable with CO_2 emission measured in Kolyma, Yukon and Mackenzie and permafrost-affected rivers in western Siberia. The areal C emissions from lotic waters of the Lena watershed were quantified via taking into account the total area of permanent and seasonal water of the Lena basin (28,000 km^2). Assuming 6 months of the year to be open water period with no emission under ice, the annual C emission from the whole Lena basin is estimated as 8.3 ± 2.5 Tg C y^{-1} which is comparable to the DOC and DIC lateral export to the Arctic Ocean.

Introduction

Climate warming in high latitudes is anticipated to result in mobilization, decomposition and atmospheric release of significant amounts of carbon (C) stored in permafrost soils, providing a positive feedback (Schuur et al. 2015). Permafrost thawing is expected to also increase the lateral C export to rivers and lakes (Frey and Smith, 2005). The exported permafrost C is relatively labile and largely degraded to greenhouse gases (GHG) in recipient freshwaters (e.g. Vonk et al., 2015). As a result, assessment of GHG emission in rivers of permafrost affected regions is crucially important for understanding the high latitude C cycle under various climate change scenarios (Chadburn et al., 2017; Vonk et al., 2019). Among six great Arctic rivers, Lena is most emblematic one, situated chiefly within the continuous permafrost zone and exhibiting the highest seasonal variation in discharge. Over the past two decades, there has been an explosive interest to the Lena River hydrology (Yang et al., 2002; Berezovskaya et al., 2005; Smith and Pavelsky, 2008; Ye et al., 2009; Gelfan et al., 2017; Suzuki et al., 2018), organic C (OC) transport (Lara et al., 1998; Raymond et al., 2007; Semiletov et al., 2011; Goncalves-Araujo et al., 2015; Kutscher et al., 2017; Griffin et al., 2018) and general hydrochemistry

(Gordeev and Sidorov, 1993; Cauwet and Sidorov, 1996; Huh et al., 1998a,b; Huh and Edmond, 1999; Wu and Huh, 2007; Kuzmin et al., 2009; Pipko et al., 2010; Georgiadi et al., 2019; Juhls et al., 2020) including novel isotopic approaches for nutrients (Si, Sun et al., 2018) and trace metals such as Li (Murphy et al., 2019) and Fe (Hirst et al., 2020). This interest is naturally linked to the Lena River location within the forested continuous permafrost/taiga zone covered by organic-rich yedoma soil. Under on-going climate warming, the soils of the Lena River watershed are subjected to strong thawing and active (seasonally unfrozen) layer deepening (Zhang et al., 2005) accompanied by overall increase in river water discharge (McClelland et al., 2004; Ahmed et al., 2020), flood intensity and frequency (Gautier et al., 2018). The Lena River exhibits the highest DOC concentration among all great Arctic rivers (i.e., Holmes et al., 2013) which may reflect weak DOC degradation in the water column and massive mobilization of both contemporary and ancient OC to the river from the watershed (Feng et al., 2013; Wild et al., 2019). In contrast to rather limited works on CO_2 and CH_4 emissions from water surfaces of Eastern Siberia (Semiletov, 1999; Denfeld et al., 2013), extensive studies were performed on land, in the polygonal tundra of the Lena River Delta (Wille et al., 2008; Bussman, 2013; Sachs et al., 2008; Kutzbach et al., 2007) and the Indigirka Lowland (van der Molen et al., 2007). Finally, there have been several studies of sediment and particular matter transport by the Lena River to the Laptev Sea (Rachold et al., 1996; Dudarev et al., 2006) together with detailed research of the Lena River Delta (Zubrzycki et al., 2013; Siewert et al., 2016).

Surprisingly, despite such extensive research on C transport, storage, and emission in Eastern Siberian landscapes, C emissions of the Lena River main stem and tributaries remain virtually unknown, compared to a relatively good understanding of those in the Yukon (Striegl et al., 2012; Stackpoole et al., 2017), Mackenzie (Horan et al., 2019), Ob (Karlsson et al., 2021; Pipko et al., 2019) and Kolyma (Denfeld et al., 2013). The only available estimates of C emission from inland waters of the Lena basin are based on few indirect (calculated gas concentration and modelled fluxes) snapshot data with very low spatial and temporal resolution (Raymond et al., 2013). Similar to other regions, this introduces uncertainties and cannot adequately capture total regional C emissions (Abril et al., 2015; Denfeld et al., 2018; Park et al., 2018; Klaus et al., 2019; Klaus and Vachon, 2020; Karlsson et al., 2021). In particular, no detailed studies at the peak of spring flood have been performed and the information on various contrasting tributaries of the Lena River remains very limited. As a result, reliable estimations of magnitude and controlling factors of C emission in the Lena River basin are poorly understood. The present work represents a first assessment of CO_2 and CH_4 concentration and fluxes of the main stem and tributaries during the peak of spring flow, via calculating C emission and relating these data to river hydrochemistry and GIS-based landscape parameters. This should allow identifying environmental factors controlling GHG concentration and emission in the Lena River watershed in order to use this knowledge to foresee future changes in C balance of the largest permafrost-affected Arctic river.

2. Study Site, Materials and Methods

2.1. Lena River and its tributaries

The sampled Lena River main stem and 20 tributaries are located along a 2600 km latitudinal transect SW to NE and include watersheds of distinct sizes, geomorphology, permafrost extent, lithology, climate and vegetation (**Fig. 1, S1 A; Table S1**). The total watershed area of the rivers sampled in this work is approximately 1.66 million km^2, representing 66% of the entire Lena River basin. Permafrost is mostly continuous except some patches of discontinuous and sporadic in the southern part of the Lena basin (Brown et al., 2002). The mean annual air temperatures (MAAT) along the transect ranges from -5 °C in the southern part of the Lena basin to -9 °C in the central part of the basin. The range of MAAT for 20 tributaries is from -4.7 to -15.9 °C. The mean annual precipitation ranges from 350-500 mm y^{-1} in the southern and south-western part of the basin to 200-250 mm y^{-1} in the central and northern parts (Chevychelov and Bosikov, 2010). The lithology of the Siberian platform which is drained by the Lena River is highly diverse and includes Archean and Proterozoic crystalline and metamorphic rocks, Upper Proterozoic, Cambrian and Ordovician dolostones and limestones, volcanic rocks of Permo-Triassic age and essentially terrigenous silicate sedimentary rocks of the Phanerozoic. Further description of the Lena River basin landscapes, vegetation and lithology can be found elsewhere (Rachold et al., 1996; Huh et al., 1999a, b; Pipko et al., 2010; Semiletov et al., 2011; Kutscher et al., 2017; Juhls et al., 2020).

The peak of annual discharge depends on the latitude (**Fig. 1**) and occurs in May in the south (Ust-Kut) and in June in the middle and low reaches of the Lena River (Yakutsk, Kysyr). From May 29 to June 17, 2016, we moved downstream the Lena River by boat with an average speed of 30 km h^{-1} (Gureyev, 2016). As such, we followed the progression of the spring and moved from the southwest to the northeast, thus collecting river water at approximately the same stage of maximal discharge. Note that transect sampling is a common way to assess river water chemistry in extreme environments (Huh and Edmond, 1999; Spence and Telmer, 2005), and generally, a single sampling during high flow season provides the best agreement with time-series estimates (Qin et al., 2006). Regular stops each 80-100 km along the Lena River allowed sampling for major hydrochemical parameters and CH_4 along the main stem. We also moved 500-1500 m upstream of selected tributaries to record CO_2 concentrations for at least 1 h and to sample for river hydrochemistry; see examples of spatial coverage in **Fig. S1 B**. From late afternoon/evening to the next morning, we stopped for sleep but continued to record pCO_2 in the Lena River main stem (15 sites, evenly distributed over the full 2600 km transect) and two tributaries (Aldan and Tuolba).

2.2. CO_2 and CH_4 concentrations

Surface water CO_2 concentration was measured continuously, *in-situ* by deploying a portable infrared gas analyzer (IRGA, GMT222 CARBOCAP® probe, Vaisala®; accuracy ± 1.5%) of two ranges (2 000 and 10 000 ppm). This system was mounted on a small boat in a perforated steel pipe ~0.5 m below water surface. The tube had two necessary opening of different diameter, which allowed free water flow with a constant rate during the moving of the boat. The probe was enclosed within a waterproof and gas-permeable membrane. The key to aqueous deployment of the IRGA sensor is the use of a protective expanded polytetrafluoroethylene (PTFE) tube or sleeve that is highly permeable to CO_2 but impermeable to water (Johnson et al., 2009). The material is available for purchase as a flexible tube that fits over the IRGA sensor (Product number 200-07; International Polymer Engineering, Tempe, Arizona, USA). We also used a copper mesh screen to minimize biofouling effects (i.e., Yoon et al., 2016). However these effects are expected to be low in cold waters of the virtually pristine Lena River and its tributaries. During sampling, the sensor was left to equilibrate in the water for 10 minutes before measurements were recorded.

The probe was enclosed and placed into a tube which was submerged 0.5 m below the water surface. Within this tube, we designed a special chamber that allowed low-turbulent water flow around the probe without gas bubbles. Previous studies (Park et al., 2021; Crawford et al. 2015; Yoon et al., 2016) reported some effects of boat speed on sensor CO_2 measurements due to turbulences. Although the turbulences were minimized in the tube/chamber design used in the present study, on a selected river transect (~10 km) we have also tested the impact of the boat speed (5, 10, 20, 30 and 40 km h^{-1}) on the sensor performance and have not detected any sizable (> 10%, $p < 0.05$, $n = 25$) difference in the CO_2 concentrations recorded by our system.

A Campbell logger was connected to the system allowing continuous recording of the CO_2 concentration (ppm), water temperature (°C) and pressure (mbar) every minute during 5 minutes over 10 minute intervals yielding 4,285 individual pCO_2, water temperature and pressure measurements in total. These data were averaged for 3 consecutive slots of 5 min measurements, which represented the approximate 20-km interval of the main stem route. CO_2 concentrations in the Lena River tributaries were measured over the first 500-2000 m distance upstream of the tributary mouth, and comprised between 5 and 34 measurements for day-time visits and between 305 and 323 individual pCO_2 readings for each tributary for day-time and night-time monitoring.

Sensor preparation was conducted in the lab following the method described by Johnson et al. (2009). The measurement unit (MI70, Vaisala®; accuracy ± 0.2%) was connected to the sensor allowing instantaneous readings of pCO_2. The sensors were calibrated in the lab against standard gas mixtures (0, 800, 3 000, 8 000 ppm; linear regression with $R^2 > 0.99$) before and after the field campaign. The sensors' drift was 0.03-0.06% per day and overall error was 4-8% (relative standard deviation, RSD). Following calibration, post-measurement correction of the sensor output induced by changes in water temperature and barometric pressure was done by applying empirically derived coefficients following Johnson et al. (2009). These corrections never exceeded 5% of the measured values. Furthermore, we tested two different sensors in several sites of the river transect: a main probe used for continuous measurements and another probe used as a control and never employed for continuous measurements. We did not find any sizable (>10%) difference in measured CO_2 concentration between these two probes.

For CH_4 analyses, unfiltered water was sampled in 60-mL Serum bottles and closed without air bubbles using vinyl stoppers and aluminum caps and immediately poisoned by adding 0.2 mL of saturated $HgCl_2$ via a two-way needle system. In the laboratory, a headspace was created by displacing approx. 40% of water with N_2 (99.999%). Two 0.5-mL replicates of the equilibrated headspace were analyzed for their concentrations of CH_4, using a Bruker GC-456 gas chromatograph (GC) equipped with flame ionization and thermal conductivity detectors. After every 10 samples, a calibration of the detectors was performed using Air Liquid gas standards (i.e. 145 ppmv). Duplicate injection of the samples showed that results were reproducible within ±5%. The specific gas solubility for CH_4 (Yamamoto et al., 1976) was used in calculation of total CH_4 content in the vials and then recalculated to μmol L^{-1} of the initial waters.

2.3. Chemical analyses of the river water

The dissolved oxygen (CellOx 325; accuracy of ±5%), specific conductivity (TetraCon 325; ±1.5%), and water temperature (±0.2 °C) were measured in-situ at 20 cm depth using a WTW 3320 Multimeter. The pH was measured using portable Hanna instrument via combined Schott glass electrode calibrated with NIST buffer solutions (4.01, 6.86 and 9.18 at 25°C), with an uncertainty of 0.01 pH units. The temperature of buffer solutions was within ± 5°C of that of the river water. The water was sampled in pre-cleaned polypropylene bottle from 20-30 cm depth in the middle of the river and immediately filtered through disposable single-use sterile Sartorius filter units (0.45 μm pore size). The first 50 mL of filtrate was discarded. The DOC and Dissolved Inorganic Carbon (DIC) were determined by a Shimadzu TOC-VSCN Analyzer (Kyoto, Japan) with an uncertainty of 3% and a detection limit of 0.1 mg/L. Blanks of MilliQ water passed through the filters demonstrated negligible release of DOC from the filter material.

2.4. Flux calculation

CO_2 flux (F_{CO_2}) was calculated following Cai and Wang (1998):

$$F_{CO_2} = K_h \, k_{CO_2} \, (C_{water} - C_{air}) \,, \tag{1}$$

where K_h is the Henry's constant corrected for temperature and pressure (mol L^{-1} atm^{-1}), k_{CO_2} is the gas exchange velocity at a given temperature, C_{water} is the water CO_2 concentration, and C_{air} is the CO_2 concentration in the ambient air. We used the average CO_2 concentrations of 402 ppm in May-June 2016 (from 129 stations all over the world, https://community.wmo.int/wmo-greenhouse-gas-bulletins), which is consistent with the value recorded at the nearest Tiksi station in 2016 (404±0.9 ppm, Ivakhov e al., 2019). Temperature-specific solubility coefficients were used to calculate respective CO_2 concentrations in the water following Wanninkhof et al. (1992). To standardize k_{CO_2} to a Schmidt number of 600, we used the following equation (Alin et al., 2011; Vachon et al., 2010):

$$k_{600} = k_{CO_2} \left(\frac{600}{Sc_{CO_2}} \right)^{-n} \tag{2}$$

where Sc_{CO_2} is CO_2 Schmidt number for a given temperature (t, °C) in the freshwater (Wannikhof, 1992):

$$\mathrm{Sc}_{\mathrm{CO_2}} = 1911.1 - 118.11t + 3.4527t^2 - 0.041320t^3 \tag{3}$$

The exponent n (Eqn. 2) is a coefficient that describes water surface (2/3 for a smooth water surface regime while 1/2 for a rippled and a turbulent one), and the Schmidt number for 20°C in freshwater is 600. We used n = 2/3 because all water surfaces of sampled rivers were considered flat and had a laminar flow (Alin et al., 2011; Jähne et al., 1987) with wind speed always below 3.7 m s^{-1} (Guérin et al., 2007).

In this study, we used a k_{CO_2} (a median gas transfer coefficient) value of 4.464 m d^{-1} measured in the 4 largest rivers of Western Siberia Lowalnd (WSL) in June 2015 (Ob', Pur, Pyakupur and Taz rivers, Karlsson et al., 2021). These rivers are similar to Lena and its tributaries in size, but exhibit lower velocity than those of the Lena River. In fact, due to more mountainous relief, the Lena River main stem and tributaries present much higher turbulence than that of the Ob River and tributaries and as such the value k_{CO_2} used in this study can be considered rather conservative. This value is consistent with the k_{CO_2} reported for the Kolyma River and its large tributaries (k = 3.9 ± 2.5 m d^{-1}, Denfeld et al., 2013), tributaries and main stem of the Yukon river basin (k_{600} = 4.9 - 7.6 m d^{-1}, Striegl et al. 2012), large rivers in the Amazon and Mekong basins (k_{600} = 3.5 ± 2.1 m d^{-1}, Alin et al., 2011) and with modelling results of k for large rivers across the world (k = 3 - 4 m d^{-1}, Raymond et al., 2013). Note that decreasing the k to most conservative value of 3 m d^{-1} of Raymond et al. (2013) will decrease specific emissions by ca. 30 %.

Instantaneous diffusive CH_4 fluxes were calculated using an equation similar to 1 with k from western Siberia rivers (Serikova et al., 2018), concentrations of dissolved CH_4 in the water and air–water equilibrium pCH_4 concentration of 1.8 ppm, and mean annual pCH_4 concentration in the air for 2016 (Mauna Loa Observatory fttp://aftp.cmdl.noaa.gov/products/trends/ch4/ch4_annmean_gl.txt) following standard procedures (Serikova et al., 2018, 2019).

2.5. Landscape parameters and water surface area of the Lena basin

The physio-geographical characteristics of the 20 Lena tributaries sampled in this study and the two points of the Lena main stem (upstream and downstream r. Aldan, **Table S1**) were determined by applying available digital elevation model (DEM GMTED2010), soil, vegetation, lithological, and geocryological maps. The landscape parameters were typified using TerraNorte Database of Land Cover of Russia (Bartalev et al., 2020, http://terranorte.iki.rssi.ru). This included various type of forest (evergreen, deciduous, needleleaf/broadleaf), grassland, tundra, wetlands, water bodies and other area. The climate and permafrost parameters of the watershed were obtained from CRU grids data (1950-2016) (Harris et al., 2014) and NCSCD data (doi:10.5879/ecds/00000001, Hugelius et al., 2013), respectively, whereas the biomass and soil OC content were obtained from BIOMASAR2 (Santoro et al., 2010) and NCSCD databases. The lithology layer was taken from GIS version of Geological map of the Russian Federation (scale 1 : 5 000 000, http://www.geolkarta.ru/). To test the effect of carbonate rocks on dissolved C parameters, we distinguished acidic crystalline, terrigenous silicate rocks and dolostones and limestones of upper Proterozoic, Cambrian and Ordovician age. We quantified river water surface area using the global SDG database with 30 m² resolution (Pekel et al., 2016) including both seasonal and permanent water for the open water period of 2016 and for the multiannual average (reference period

2000-2004). We also used a more recent GRWL Mask Database which incorporates first order wetted streams (Allen and Pavelsky, 2018).

The Pearson rank order correlation coefficient (Rs, $p < 0.05$) was used to determine the relationship between CO_2 concentrations and climatic and landscape parameters of the Lena River tributaries. Further statistical treatment of CO_2, DIC and DOC concentration drivers in river waters included a Principal Component Analysis which allowed to test the effect of various hydrochemical and climatic parameters on dissolved C pattern. For the PCA treatment, all variables were normalized as necessary in the standard package of STATISTICA-7 (http://www.statsoft.com) because the units of measurement for various components were different. The factors were identified via the Raw Data method. To run the scree test, we plotted the eigen values in descending order of their magnitude against their factor numbers. There was significant decrease in the PCA values between F1 and F2 suggesting that a maximum of two factors were interpretable.

3. Results

3.1. CO_2, CH_4, DIC and DOC in the main stem and Lena tributaries and C emission fluxes

The main hydrological C parameters of the Lena River and its tributaries (pCO_2, CH_4, pH, DIC, and DOC) are listed in **Tables 1 and 2**. Continuous pCO_2 measurements in the main stem (4285 individual data points) averaged for each 20 km interval over the full distance of the boat route demonstrated a sizable increase (from ca. 380 to 1040 µatm) in pCO_2 northward (**Fig. 2**). There was a positive correlation between the pCO_2 and distance from the head waters of the Lena River ($r = 0.625$, $p < 0.01$**, Fig. 3 A**). The CH_4 concentration was low (0.054 ± 0.023 and 0.061 ± 0.028 µmol L^{-1} in the Lena River and 20 tributaries, respectively) and did not change appreciably along the main stem and among the 20 tributaries (**Fig. 3 B).** The DOC concentration did not demonstrate any systematic variations over the main stem (10.5 ± 2.4 mg L^{-1}, **Fig. 3 C**), however it was higher and more variable in tributaries (15.8 ± 8.6 mg L^{-1}). The DIC concentration decreased about five-fold from the head waters to the middle course of the Lena River (**Fig. 3 D**), and pH decreased by 0.8 units downstream (**Fig. 3 E**).

Generally, the concentrations of DOC measured in the present study during the peak of the spring flood are at the highest range of previous assessments during summer baseflow (around 5 mg L^{-1}; range of 2 to 12 mg L^{-1}, Cauwet and Sidorov, 1996; Lara et al., 1998; Lobbes et al., 2000; Kuzmin et al., 2009; Kutscher et al., 2017). The DIC concentration in the main stem during spring flood was generally lower than that reported during summer baseflow (around 10 mg L^{-1}; range of 5 to 50 mg L^{-1}) but consistent with values reported in Yakutsk during May and June period (7 to 20 mg L^{-1}, Sun et al., 2018). A sizable decrease in DIC concentration between the headwaters (first 500 km of the river) and the Lena River middle course was also consistent with the alkalinity pattern reported in previous works during summer baseflow (Pipko et al., 2010; Semiletov et al., 2011). For the Lena river tributaries, the most comprehensive data set on major ions was acquired in July-August of 1991-1996 by Huh and Edmond's group (Huh and Edmond, 1999; Huh et al., 1998a, b) and by Sun et al. (2018) in July 2012 and at the end of June 2013. For most tributaries, the concentration of DIC was a factor of 2 to 5 lower during spring flood compared to summer baseflow. This result can be explained by the strong dilution of carbonate-rich groundwaters feeding the river in spring high flow compared to summer low flow.

The measured pCO_2 in the river water and published (Karlsson et al., 2021) gas transfer coefficient (4.46 m d^{-1}) allowed for calculation of the CO_2 fluxes over the full length of the studied main stem (2600 km) and the sampled tributaries. Calculated CO_2 fluxes of the main stem and tributaries ranged from zero and slightly negative (uptake) values in the most southern part of the Lena River and certain tributaries (N Katyma), to between 0.5-2.0 g C m^{-2} d^{-1} in the rest of the main stem and tributaries (**Tables 1, 2; Fig. 2 B**). The largest part of the Lena River main stem, 1429 km from Kirenga to Tuolba, exhibited quite stable flux of 1.1±0.2 g C m^{-2} d^{-1}. In the last ~400 km part of the Lena River main stem studied in this work, from Tuolba to Aldan, the calculated fluxes increased to 1.7±0.08 g C m^{-2} d^{-1}.

The river water concentrations of dissolved CH_4 in the tributaries and the main channel (0.059±0.006; IQR range from 0.025 to 0.199 µmol L^{-1}, **Table 1, 2**) did not exhibit any trend with distance from headwaters or landscape parameters of the catchments. These values are consistent with the range of CH_4 concentration in the low reaches of the Lena River main channel (0.03-0.085 µmol L^{-}

^{1}; Bussman, 2013) and are 100-500 times lower than those of CO_2. Consequently, diffuse CH_4 emissions constituted less than 1 % of total C emissions and are not discussed in further detail.

3.2. Diurnal (night/day) pCO_2 variations and spatial variations across the river transect

The diel continuous CO_2 measurements of 3 tributaries (Kirenga, Tuolba and Aldan) and 14 sites of the Lena River main channel showed generally modest variation with diurnal range within 10 % of the average pCO_2 (**Fig. 4** and **Fig. S2**). The observed variations in pCO_2 between day and night were not linked to water temperature ($p > 0.05$), which did not vary more than 1-2 °C between the day and night period.

The spatial variations of hydrochemical parameters were tested in the upper reaches of the Lena main stem and its largest tributary - the Aldan River (**Fig. S3**). In the Lena River, over a lateral distance of 550 m across the river bed, the pCO_2 and CH_4 concentrations were equal to 569±4.6 µatm and 0.0406±0.0074 µmol L^{-1}, respectively, whereas the DIC and DOC concentrations varied < 15% (n = 5). In the Aldan River, over a 2700 m transect across the flow, the pCO_2 and CH_4 concentrations were equal to 1035±95 µatm and 0.078±0.00894 µmol L^{-1}, respectively, whereas DIC and DOC varied within < 20% (n = 4). Overall, these results supported our design of punctual (snap shot) sampling in the middle of the river.

3.3. Impact of catchment characteristics on pCO_2 in tributaries of the Lena River

The CO_2 concentration in the Lena River main stem and tributaries increased from southwest to northeast (**Table 1, 2; Fig. 2**), and this was reflected in a positive (R = 0.66) correlation between CO_2 concentration and continuous permafrost coverage and a negative (R = -0.76) correlation with MAAT (**Table 3)**. Among different landscape factors, C stock in upper 0-30 and 0-100 cm of soil, the proportion of riparian vegetation and bare rocks, the coverage by deciduous needle-leaf forest, and coverage of river watershed by water bodies (mostly lakes) exhibited significant ($p < 0.01$, $n = 19$) positive correlations ($0.54 \leq R \leq 0.86$) with average pCO_2 of the Lena River tributaries (**Fig. 5**). The other potentially important landscape factors of the river watershed (proportion of peatland and bogs, tundra coverage, total aboveground vegetation, type of permafrost, annual precipitation) did not significantly impact the CO_2 concentration in the Lena River tributaries (**Table 3**).

Further assessment of landscape factor control on C parameters of the river water was performed via a PCA. This analysis basically confirmed the results of linear regressions and revealed two factors capable explaining only 12.5 and 3.5% of variability (**Fig. S4**). The F1 strongly acted on the sample location at the Lena transect, the content of OC in soils, the watershed coverage by deciduous needle-leaf forest and shrubs, riparian vegetation (a proxy for the width of the riparian zone), but also proportion of tundra, bare rock and soils, water bodies, peatland and bogs (> 0.90 loading). The pCO_2 was significantly linked to F1 (0.72 loading).

3.4. Areal emission from the Lena River basin

The areal emission of CO_2 from the lotic waters of the Lena River watershed were assessed based on total river water coverage of the Lena basin in 2016 (28,197 km², of which 5,022 km² is seasonal water, according to the Global SDG database). This value is consistent with the total river surface area from the GRWL Mask database (22,479 km²). Given that the measurements were performed at the peak of spring flood in 2016, we used the maximal water coverage of the Lena River basin.

Based on past calculated pCO_2 of the Lena River (400 - 1000 µatm, Semiletov, 1999; Semiletov et al., 2011; Pipko et al., 2010) both the seasonality and spatial differences downstream are relatively small. Indeed, for the lower reaches of the Lena River, from Yakutsk to the Lena Delta, Semiletov (1999) and Semiletov et al. (2011) reported, for August-September 1995, the average pCO_2 of 538±96 µatm (range 380-727 µatm). This value is very similar to the one obtained in July 2003 for the low reaches of Lena (559 µatm, Pipko et al., 2010). Over the full length of the Lena River, from Ust-Kut to the Lena mouth, Pipko et al. (2010) reported an average pCO_2 of 450 ± 100 µatm in June-July 2003. At the same time, calculated pCO_2 from previous field campaigns are generally lower than the pCO_2 of the Lena River main stem directly measured in this study: 700-800 µatm for the Ust Kut – Nuya segment (1331 km); 845 – 1050 µatm for the Nuya – Aldan segment (1050 km).

Thus, despite the absolute values of calculated pCO_2 involving uncertainties (our calculated: measured pCO_2 in Lena River main channel and tributaries equaled 0.67±0.15 (n = 47)), this suggests spatial and temporal stability of the pCO_2 in the Lena River waters and allows for extrapolation of the measured pCO_2 in the Lena River from Yakutsk to Aldan to the lower reaches of the river. As for the Lena tributaries, to the best of our knowledge there is no published information on pCO_2 concentration and emission. Overall, the major uncertainty in estimation of the Lena River basin emission stems from a lack of direct pCO_2 measurements in the northern part of the main channel over ca. 1000 km downstream of the Aldan River including the large tributary Vilyi. Further, we noted that the largest northern tributary, the Aldan River providing 70% of the spring time discharge of the Lena River (Pipko et al., 2010), demonstrated sizably higher emissions compared to the Lena River main channel upstream of Aldan (3.2±0.5 and 1.69±0.08 g C m^{-2} d^{-1}, respectively).

For areal emission calculations, we used the range of CO_2 emissions from 1 to 2 g C m^{-2} d^{-1} which covers full variability of both large and small tributaries and the Lena River main channel (**Tables 1-2, Figure 2 B**). This estimation assumes lack of pCO_2 dependence on the size of the watershed in the Lena basin as confirmed by our data (**Fig. S5**). For an alternative areal emission calculations, we explicitly took into account the water area of the main stem (43% relative to the total water area of the Lena catchment) and we introduced the partial weight of emission from the 3 largest tributaries (Aldan, Olekma and Vitim) according to their catchment surface areas (43, 12 and 14% of all sampled territory, respectively). We summed up contribution of the Lena river main stem and the tributaries and we postulated the average emission from the main stem upstream of Aldan (1.25±0.30 g C m^{-2} d^{-1}) as representative of the whole Lena River. This resulted in an updated value of 1.65±0.5 g C m^{-2} d^{-1} which is within the range of 1 to 2 g C m^{-2} d^{-1} assessed previously. Note that this value is most likely underestimated because emissions from the main stem downstream of Aldan are at least 10 % higher (Table 1, Fig. 1 B).

For the two months of maximal water flow (middle of May - middle of July), the C emission from the whole Lena basin equates to 2.8 ± 0.85 Tg C which is 20 to 30% of the DOC and DIC lateral export to the Arctic Ocean. Assuming six months of open water period and no emission during winter, this yields 8.3 ± 2.5 Tg C y^{-1} of annual emission for the whole Lena River basin (2,490, 000 km^2) with a total lotic water area of 28,100 km^2. Considering the only 23,200 km^2 water area in July-October (and maximal water coverage in May-June), these numbers decrease by 12% which is below the uncertainties associated with our evaluation.

4. DISCUSSION

4.1. Possible driving factors of CO_2 pattern in the Lena River basin

Generally, the SW to NE increase in CO_2 concentrations and fluxes of the tributaries was consistent with CO_2 pattern in the main stem (**Fig. 2; Tables 1, 2**), and thus can be considered as a specific feature of CO_2 exchange between lotic waters and atmosphere in the studied part of the Lena Basin. At the same time, there were sizable local variations (peaks) in CO_2 concentration of the main stem along the sampling route (**Fig. 2 A**). Peaks shown on the diagram of the main stem are not necessarily linked to CO_2-rich tributaries, but likely reflect local processes in the main stem, including lateral influx from the shores and shallow subsurface waters, which is typical for permafrost regions of forested Siberian watersheds (i.e., Bagard et al., 2011). Given that the data were averaged over ~20-km distance, we believe that these peaks are not artifacts but reflect local heterogeneity of the pCO_2 pattern in the main stem (turbulences, suprapermafrost water discharge, sediment resuspension and respiration). Note that such a heterogeneity was not observed in the tributaries, at least at the scale of our spatial coverage (see **Fig. S1 B, S3**).

The PCA demonstrated extremely low ability to describe the data variability (12% by F1 and only 3.5% by F2). We believe that the most likely reason of weak PCA capacity is the rather homogeneous distribution of CO_2 and CH_4 among the tributaries, primarily linked to the specific hydrological period, studied in this work - the spring flood. During this high flow period, the local lithological and soil heterogeneities among tributaries or the segments of the main stem virtually disappear and surface flow (via vegetation leaching) becomes the most important driver of riverine chemistry, as is known from adjacent permafrost territories in Central Siberia (i.e., Bagard et al., 2011). Nevertheless, some specific features of the data structure could be established. The first factor, significantly linked to pCO_2 (0.72 loading), strongly acted on the sample location at the Lena transect, the watershed coverage by deciduous needle-leaf forest and shrubs, riparian vegetation, and also the proportion of tundra, bare rock and soils, water bodies, peatland and bogs (> 0.90 loading). This is fully consistent with spatial variation of pCO_2 along the permafrost and climate gradient in the main channel and sampled tributaries. Positive loading of riparian vegetation, peatlands and bogs on F1 (0.927 and 0.989, respectively) could reflect a progressive increase in the feeding of the river basin by mire waters, an increase in the proportion of needle-lead deciduous trees, and in the width of the riparian zone from the SW to the NE direction.

Lack of sizable variation in pCO_2 between the day and night period or across the river bed suggests quite low site-specific and diurnal variability. It may be indicative of a negligible role of primary productivity in the water column given the low water temperatures, shallow photic layer of organic-rich and turbid waters and lack of periphyton activity during high flow of the spring flood. The pCO_2 increased by a factor of 2 to 4 along the permafrost/temperature gradient from the southwest to the northeast, for both the main channel and sampled tributaries. This may reflect progressive increase in the feeding of the river basin by mire waters, increase in the proportion of needle-leaf deciduous forest, and an increase in the width of the riparian zone. Another strong correlation is observed between the stock of OC in soils (both 0-30 and 0-100 cm depth) and the pCO_2 of Lena tributaries. Organic-rich soils are widely distributed in the central and northern part of the basin. The most southern part of the Lena basin is dominated by carbonate rocks and crystalline silicates in generally mountainous terrain, where only thin mineral soils are developed. The northern (downstream of the Olekma River) part of the basin consists of soils developed on sedimentary silicate rocks as well as vast areas of easily eroded yedoma soils. It is likely that both organic matter mineralization in OC-rich permafrost soils and lateral export of CO_2 from these soils, together with particulate and dissolved OC export and mineralization in the water column, are the main sources of CO_2 in the river water. Although some studies have demonstrated high lability of DOM in arctic waters (Cory et al., 2014; Ward et al., 2017; Cory and Kling, 2018), others suggest it is low and does not support the major part of CO_2 supersaturation in water (Shirokova et al., 2019; Payandi-Rolland et al., 2020; Laurion et al., 2021). Note that we have not observed any significant relationship between the DOC and pCO_2 in the Lena River and tributaries (**Fig. S6 A**). Lack of such a correlation and absence of diurnal pCO_2 variations imply that in-stream processing of dissolved terrestrial OC is not the main driver of CO_2 supersaturation in the river waters of the Lena River basin. Furthermore, a lack of lateral (across the river bed) variations in pCO_2 does not support a sizable input of soil waters from the shore, although we admit that much higher spatial coverage along the river shore is needed to confirm this hypothesis.

The role of underground water discharge in regulating pCO_2 pattern of the tributaries is expected to be most pronounced in the SW part of the basin (Lena headwaters), where carbonate rocks of the basement would provide sizable amounts of CO_2 discharge in the river bed. However, there was no relationship between the proportion of carbonate rocks on the watershed and the pCO_2 in the tributaries (**Fig. S6 B**). Furthermore, for the Lena River main stem, the lowest CO_2 concentrations were recorded in the upper reaches (first 0-800 km) where carbonate rocks dominate. Altogether, this makes the impact of CO_2 from underground carbonate reservoirs on river water CO_2 concentrations unlikely. This is further illustrated by a lack of correlation between pCO_2 and DIC or pH (**Fig. S7 A** of the Supplement). The pH did not control the CO_2 concentration in the main stem and only weakly impacted the CO_2 in the tributaries (Fig. **S7 B**). The latter could reflect an increase in pCO_2 in the northern tributaries which exhibited generally lower pH compared to the SW tributaries hosted within the carbonate rocks. Overall, such low correlations of CO_2 with DIC and pH reflected a generally low predictive capacity to calculate pCO_2 from measured pH, temperature and alkalinity (see section 3.4).

Therefore, other sources of riverine CO_2 may include particulate organic carbon processing in the water column (Attermeyer et al., 2018), river sediments (Humborg et al., 2010) and within the riparian zone (Leith et al., 2014, 2015) which require further investigation. In addition, although there was no sizable variation in pCO_2 between the day and night period or across the river bed, the flux could show different spatial and temporal patterns if *k* shows larger variability (cf., Beaulieu et al., 2012). This calls for a need of direct flux measurements in representative rivers and streams of the Lena River basin. Overall, the present study demonstrates highly dynamic and non-equilibrium behavior of CO_2 in the river waters, with possible *hot spots* from various local sources. For these reasons, *in-situ*, high spatial resolution measurements of CO_2 concentration in rivers—such as those reported for the Lena Basin in this study—are crucially important for quantifying the C emission balance in lotic waters of high latitudes.

4.2. Areal emission from the Lena River basin vs lateral export to the Arctic Ocean

The estimated CO_2 emissions from the Lena River main channel over 2600 km distance (0.8 – 1.7 g C m^{-2} d^{-1}) are comparable to values directly measured in rivers and streams of the continuous permafrost zone of western Siberia (0.98 g C m^{-2} d^{-1}, Serikova et al., 2018), the Kolyma River (0.35 g C m^{-2} d^{-1} in the main stem; 2.1 g C m^{-2} d^{-1} for lotic waters of the basin), and the Ob River main channel (1.32±0.14 g C m^{-2} d^{-1} in the permafrost-free zone, Karlsson et al., 2021). At the same time, the Lena River flux (FCO_2) values are lower than typical emissions from running waters in the contiguous Unites States (3.1 g C m^{-2} d^{-1}, Hotchkiss et al., 2015), small mountain streams in Northern Europe (3.3 g C m^{-2} d^{-1}, Rocher-Ros et al., 2019), and small streams of the northern Kolyma River (6 to 7 g C m^{-2} d^{-1}, Denfeld et al., 2013) and Ob River in the permafrost-affected zone (3.8 to 5.4 g C m^{-2} d^{-1}, Karlsson et al., 2021). In contrast to the main stem, the range of FCO_2 in the tributaries is larger (0.2 to 3.2 g C m^{-2} d^{-1}) and presumably reflects a strong variability in environmental conditions across a sizable landscape and climate transect.

Total C emissions from other major Eastern Eurasian permafrost-draining rivers (i.e. sum of Kolyma, Lena and Yenisei rivers) based on indirect estimates (40 Tg C y^{-1}, Raymond et al., 2013) are generally supportive of the estimations of the Lena River in this study (5 to 10 Tg C y^{-1}). At the same time, the C emission from the Lena river basin (28,100 km^2 water area) are lower than those of the lotic waters of western Siberia (30 Tg C y^{-1} for 33,389 km^2 water area, Karlsson et al., 2021). The latter drain through thick, partially frozen peatlands within the discontinuous, sporadic and permafrost-free zones, which can cause high OC input and processing and, thus, enhanced C emissions (Serikova et al., 2018).

Despite the high uncertainty on our regional estimations [due to lack of directly measured gas transfer values and low seasonal resolution] we believe that these estimations are conservative and can be considered as first order values pending further improvements. In order to justify extrapolation of our data to all seasons and the entire area of the Lena basin, we analyzed data for spatial and temporal variations in pCO_2 of the Lena River main stem from available literature. From the literature there were three important findings. First, based on published data, the seasonal and spatial variabilities of pCO_2 across the majority of the Lena River main stem are not high during open water period, although the low reaches of the Lena River may exhibit higher emissions compared to the middle and upper course (see **section 3.4**). Second, although small mountainous headwater streams of the tributaries may exhibit high *k* due to turbulence, this could be counteracted by lower CO_2 supply from low OC in mineral soil, lack of riparian zone and scarce vegetation. Third, although these small streams (watershed area < 100 km²) may represent > 60% of total watershed surfaces of the Lena basin (Ermolaev et al., 2018), their contribution to the total water surface is < 20% (19% from combined analysis of DEM GMTED2010 and 16% from the GRWL or Global SDG database as estimated in this study). Therefore, given that (*i*) within the stream-river continuum, the CO_2 efflux increases only two-fold demonstrating a discharge decrease by a factor of 10,000 (from 100 to 0.01 $m^3 s^{-1}$, Hotchkiss et al., 2015), and (*ii*) the watershed area had no impact on pCO_2 in the river water (**Fig. S5**), this uncertainty is likely less important. As such, instead of integrating indirect literature data, we used the pCO_2 values measured in the present study to calculate the overall CO_2 emission from all lotic waters of the Lena basin.

The C evasion from the Lena basin assessed in the present work is comparable to the total (DOC+DIC) lateral export by the Lena River to the Arctic Ocean (10 Tg C y^{-1} by Semiletov et al. (2011), or 11 Tg C y^{-1} (5.35 Tg DIC y^{-1} + 5.71 Tg DOC y^{-1} by Cooper et al. (2008)). Moreover, the C evasion strongly exceeds sedimentary C input to the Laptev Sea by all Siberian rivers (1.35 Tg C y^{-1}, Rachold et al. (1996) and Dudarev et al. (2006)), the Lena River annual discharge of particulate organic carbon (0.38 Tg y^{-1}, Semiletov et al., 2011), and OC burial on the Kara Sea Shelf (0.37 Tg C y^{-1}, Gebhardt et al., 2005).

Typical concentrations of CH_4 in the Lena tributaries and the main channel are 100 to 500 times lower than those of CO_2. Given that the global warming potential (GWP) of methane on a 100-year scale is only 25 times higher than that of CO_2, the long-term role of diffuse methane emission from the Lena River basin is still 4 to 20 times lower than that of CO_2. However, on a short-term scale (20 years), the

GWP of methane can be as high as 96 (Alvarez et al., 2018) and its role in climate regulation becomes comparable to that of the CO_2. This has to be taken into account for climate modeling of the region.

The follow up studies of this large heterogenous and important system should include CO_2 measurements in 1) the low reaches of the Lena River, downstream of Aldan, notably large organic-rich tributaries such as Vilyi (454,000 km²) and where the huge floodzone (20-30 km wide) with large number of lakes and wetlands is developed, and 2) highly turbulent eastern tributaries of the Lena River downstream of Aldan, which drain the Verkhoyansk Ridge and are likely to exhibit elevated gas transfer coefficients.

5. Conclusions

Continuous pCO_2 measurements over 2600 km of the upper and middle part of the Lena River main channel and 20 tributaries during the peak of spring flood allowed to quantify, for the first time, in-situ pCO_2 variations which ranged from 500 to 1700 µatm and exhibited a 2 to 4-fold increase in CO_2 concentration northward. There was no major variation in pCO_2 between the day and night period or across the river bed which supports the chosen sampling strategy. The northward increase in pCO_2 was correlated with an increased proportion of needle-lead deciduous trees, the width of the riparian zone and the stock of organic C in soils. Among the potential drivers of riverine pCO_2, changes in the vegetation pattern (northward migration of larch tree line in Siberia; Kruse et al., 2019) and soil OC stock are likely to be most pronounced during ongoing climate warming and thus the established link deserves further investigation. The total C emission from the lotic waters of the Lena River basin ranges from 5 to 10 Tg C y^{-1} which is comparable to the annual lateral export (50% DOC, 50 % DIC) by the Lena River to the Arctic Ocean. However, these preliminary estimations of C emission should be improved by direct flux measurements across seasons in different types of riverine systems of the basin, notably in the low reaches of the Lena River.

Acknowledgements.

We acknowledge support from an RSF grant 18-17-00237_P, the Belmont Forum Project VULCAR-FATE, and the Swedish Research Council (no. 2016-05275). We thank the Editor Ji-Hyung Park and 3 anonymous reviewers for their very constructive comments. Chris Benker is thanked for English editing.

Authors contribution.

SV and OP designed the study and wrote the paper; SV, YK and OP performed sampling, analysis and their interpretation; MK performed landscape characterization of the Lena River basin and calculated water surface area; JK provided analyses of literature data, transfer coefficients for FCO_2 calculations and global estimations of areal emission vs export.

Competing interests.

The authors declare that they have no conflict of interest.

References

Abril, G., Bouillon, S., Darchambeau, F., Teodoru, C. R., Marwick, T. R., Tamooh, F., Ochieng Omengo, F., Geeraert, N., Deirmendjian, L., Polsenaere, P., and Borges, A. V.: Technical Note: Large overestimation of pCO2 calculated from pH and alkalinity in acidic, organic-rich freshwaters, Biogeosciences, 12, 67–78, https://doi.org/10.5194/bg-12-67-2015, 2015.

Ahmed, R., Prowse, T., Dibike, Y., Bonsal, B., and O'Neil H.: Recent trends in freshwater influx to the Arctic Ocean from four major Arctic-draining rivers, Water, 12, 1189; doi:10.3390/w12041189, 2020.

Alin, S. R., Rasera, M. F. F. L., Salimon, C. I., Richey, J. E., Holtgrieve, G. W., Krusche, A. V., et al. : Physical controls on carbon dioxide transfer velocity and flux in low-gradient river systems and implications for regional carbon budgets. J. Geophys. Res. Biogeosci. 116, G01009. doi: 10.1029/2010jg001398, 2011.

Allen, G. H. and Pavelsky, T. M.: Global extent of rivers and streams, Science 361, 585-58, doiI:10.1126/science.aat0636, 2018.

Alvarez, R. A., Zavala-Araiza, D., Lyon, D. R., Allen, D. T., Barkley, Z. R., Brandt, A. R., Davis, K. J.,Herndon, S. C., Jacob, D. J., Karion, A., Kort, E. A., Lamb, B. K., Lauvaux, T., Maasakkers, J. D.,Marchese, A. J., Omara, M., Pacala, S. W., Peischl, J., Robinson, A .L., Shepson, P. B.,Sweeney, C., Townsend-Small, A., Wofsy, S.C., Hamburg, S. P.: Assessment of methane emissions from the U.S. oil and gas supply chain. Science 361, 186–188, doi:10.1126/science.aar7204, 2018.

Attermeyer, K., Catalan, N., Einarsdottir, K., Freixa, A., Groeneveld, M., Hawkes, J.A., et al.: Organic carbon processing during transport through boreal inland waters: particles as important sites, J. Geophys. Res. - Biogeosci. 123, 2412–2428, doi:10.1029/2018JG004500, 2018.

Bagard, M. L., Chabaux, F., Pokrovsky, O. S., Viers, J., Prokushkin, A. S., Stille, P., Rihs, S., Schmitt, A. D., Dupre, B.: Seasonal variability of element fluxes in two Central Siberian rivers draining high latitude permafrost dominated areas. Geochim. Cosmochim. Acta 75, 3335-3357, 2011.

Bartalev, S. A., Egorov, V. A., Ershov, D. V., Isaev, A. S., Lupyan, E. A., Plotnikov, D. E., and Uvarov, I. A.: Remote mapping of vegetation land cover of Russia based on data of MODIS spectroradmeter, Modern Problems of Earth Remote Sensing from Space, 8 (No 4), 285-302. http://d33.infospace.ru/d33_conf/2011v8n4/285-302.pdf, 2011.

Beaulieu, J. J., Shuster, W. D., and Rebholz, J. A.: Controls on gas transfer velocities in a large river, J. Geophys. Res., 117, G02007, doi:10.1029/2011JG001794, 2012.

Berezovskaya, S., Yang, D., and Hinzman, L.: Long-term annual water balance analysis of the Lena River, Glob. Planet. Change 48, 84–95, https://doi.org/10.1016/j.gloplacha.2004.12.006, 2005.

Brown, J., Ferrians Jr., O. J., Heginbottom, J. A., and Melnikov, E. S.: Circum-Arctic Map of Permafrost and Ground Ice Conditions. National Snow and Ice Data Center/World Data Center for Glaciology, Boulder, CO, USA, Digital media, 2002.

Bussmann I.: Distribution of methane in the Lena Delta and Buor-Khaya Bay, Russia, Biogeosciences 10, 4641-4652, https://doi.org/10.5194/bg-10-4641-2013, 2013.

Cai, W.-J. and Wang, Y.: The chemistry, fluxes, and sources of carbon dioxide in the estuarine waters of the Satilla and Altamaha Rivers, Georgia, Limnol. Oceanogr. 43, 657–668. https://doi.org/10.4319/lo.1998.43.4.0657, 1998.

Cauwet, G., and Sidorov I.: The biogeochemistry of Lena River: organic carbon and nutrients distribution, Marine Chem. 53, 211-227, https://doi.org/10.1016/0304-4203(95)00090-9, 1996.

Chadburn, S. E., Krinner, G., Porada, P., Bartsch, A., Beer C. et al.: Carbon stocks and fluxes in the high latitudes: using site-level data to evaluate Erath system models, Biogeosciences 14(22), 5143-5169, https://doi.org/10.5194/bg-14-5143-2017, 2017.

Chevychelov, A. P., and Bosikov, N. P.: Natural Conditions, in: The Far North, edited by E.I. Troeva et al., pp. 123, Springer, Netherlands, https://link.springer.com/chapter/10.1007/978-90-481-3774-9_1, 2010.

Cooper, L. W., McClelland, J. W., Holmes, R. M., Raymond, P. A., Gibson, J. J., Guay, C. K., and Peterson, B. J.: Flow-weighted values of runoff tracers (δ^{18}O, DOC, Ba, alkalinity) from the six largest Arctic rivers, Geophys. Res. Lett., 35, L18606, doi:10.1029/2008GL035007, 2008.

Cory, R. M., Ward, C. P., Crump, B. C., and Kling, G. W.: Sunlight controls water column processing of carbon in arctic fresh waters, Science, 345, 925-928, doi: 10.1126/science.1253119, 2014.

Cory, R. M., Kling, G. W.: Interactions between sunlight and microorganisms influence dissolved organic matter degradation along the aquatic continuum, Limnol. Oceanogr. Lett., 3, 102–116, https://doi.org/10.1002/lol2.10060, 2018.

Crawford, J. T., Loken, L. C., Casson, N. J., Smith, C., Stone, A. G, and Winslow, L. A.: High-speed limnology: using advanced sensors to investigate spatial variability in biogeochemistry and hydrology, Environ. Sci. Technol. 2015, 49, 1, 442–450, https://doi.org/10.1021/es504773x, 2015.

Denfeld, B.A., Frey K.E., Sobczak, W.V., Mann P.J., and Holmes, R.M.: Summer CO_2 evasion from streams and rivers in the Kolyma River basin, north-east Siberia, Polar Res., 32, Art No 19704, https://doi.org/10.3402/polar.v32i0.19704, 2013.

Denfeld, B. A., Baulch, H. M., del Giorgio, P. A., Hampton, S. E., and Karlsson, J.: A synthesis of carbon dioxide and methane dynamics during the ice-covered period of northern lakes, Limnology and Oceanography Letters 3, 117-131, doi:10.1002/lol2.10079, 2018.

Dudarev, O. V., Semiletov, I. P., and Charkin, A. N.: Particulate material composition in the Lena River - Laptev Sea system: Scales of heterogeneities, Doklady Earth Sciences, 411A (9), 1445-1451, https://link.springer.com/content/pdf/10.1134/S1028334X0609025X.pdf, 2006.

Ermolaev, O. P., Maltzev K. A., Mukharamova S. S., Khomyakov P. V., and Shynbergenov E. A.: Cartographic model of small rivers of the Lena River basin. Ychenue Zapiski Kazansky Univ., Ser. Natural Sciences, 160(1), 126-144, https://cyberleninka.ru/article/n/kartograficheskaya-model-basseynovyh-geosistem-malyh-rek-vodosbora-reki-leny/viewer, 2018.

Feng, X. J., Vonk, J. E., van Dongen, B. E., Gustafsson, O., Semiletov, I. P., Dudarev, O. V., Wang, Z. H., Montlucon, D. B., Wacker, L., and Eglinton, T.I.: Differential mobilization of terrestrial carbon pools in Eurasian Arctic river basins, P. Natl. Acad. Sci. USA, 110, 14168–14173, https://doi.org/10.1073/pnas.1307031110, 2013.

Frey, K. E., and Smith, L.C.: Amplified carbon release from vast West Siberian peatlands by 2100, Geophys. Res. Lett., 32, L09401, doi:10.1029/2004GL022025, 2005.

Gautier, E., Depret, T., Costard, F., Virmoux, C., Fedorov, A. et al. : Going with the flow : Hydrologic response of middle Lena River (Siberia) to the climate variability and change, J. Hydrol. 557, 475

- 488, https://doi.org/10.1016/j.jhydrol.2017.12.034, 2018.
Gebhardt, A. C., Gaye-Haake, B., Unger, D., Lahajnar, N., and Ittekkot, V.: A contemporary sediment and organic carbon budget for the Kara Sea shelf (Siberia), Marine Geology 220(1-4), 83-100, https://doi.org/10.1016/j.margeo.2005.06.035, 2005.
Gelfan, A., Gustafsson, D., Motovilov, Y., Arheimer, B., Kalugin, A., Krylenko, I., and Lavrenov A.: Climate change impact on the water regime of two great Arctic rivers: modelling and uncertainty issues, Climate Change 414(3), 499-515, DOI: 10.1007/s10584-016-1710-5m, 2017.
Georgiadi, A. G., Tananaev, N. I., and Dukhova L.A.: Hydrochemical conditions at the Lena River in August 2018, Oceanology 59(5), 797-800, https://doi.org/10.1134/S0001437019050072, 2019.
Goncalves-Araujo, R., Stedmon, C. A., Heim, B., Dubinenkov, I., Kraberg, A., Moiseev, D., and Brachler A.: From fresh to marine waters: Characterization and fate of dissolved organic matter in the Lena River delta region, Siberia, Front. Marine Sci., 2, Art No 108, https://doi.org/10.3389/fmars.2015.00108, 2015.
Gordeev, V. V. and Sidorov, I. S.: Concentrations of major elements and their outflow into the Laptev Sea by the Lena River, Mar. Chem., 43, 33–46, 1993.
Griffin, C. G., McClelland, J. W., Frey, K. E., Fiske, G., and Holmes, R. M.: Quantifying CDOM and DOC in major Arctic rivers during ice-free conditions using Landsat TM and ETM+ data. Remote Sens. Environ. 209, 395-409, doi: 10.1016/j.rse.2018.02.060, 2018.
Guérin, F., Abril, G., Serça, D., Delon, C., Richard, S., Delmas, R., Tremblay, A., and Varfalvy, L.: Gas transfer velocities of CO_2 and CH_4 in a tropical reservoir and its river downstream, J. Mar. Syst., 66, 161–172. https://doi.org/10.1016/j.jmarsys.2006.03.019, 2007.
Gureyev, D.: Tomsk State University: The expedition on the Lena River from the headwaters to the Aldan River, 2016. https://www.youtube.com/watch?v=7IEiO4bgxc8, 2016.
Harris, I., Jones, P. D., Osborn, T. J., and Lister, D. H.: Updated high-resolution grids of monthly climatic observations – the CRU TS3.10 Dataset, Int. J. Climatol., 34: 623–642, doi: 10.1002/joc.3711, 2014.
Hirst, K., Andersson, P., Kooijman, E., Kutscher, L., Maximov, T., Moth, C.-M., Porcelli, D.: Iron isotopes reveal the sources of Fe-bearing particles and colloids in the Lena River basin, Geochim. Cosmochim. Acta, 269, 678-692, doi: 10.1016/j.gca.2019.11.004, 2020.
Holmes, R.M., Coe, M.T., Fiske, G.J., Gurtovaya, T., McClelland, J.W., Shiklomanov, A.I., Spencer, R.G.M., Tank, S.E., and Zhulidov, A.V.: Climate change impacts on the hydrology and biogeochemistry of Arctic Rivers, In: Climatic Changes and Global warming of Inland Waters: Impacts and Mitigation for Ecosystems and Societies, Eds. C.R. Goldman, M. Kumagi, and R.D. Robarts, John Wiley and Sons, p. 1-26, 2013.
Horan, K.; Hilton, R. G., Dellinger, M., Tipper, E., Galy, V., Calmels, D., Selby, D., Gaillardet, J., Ottley, C.J., Parsons, D.R., and Burton, K.W.: Carbon dioxide emissions by rock organic carbon oxidation and the net geochemical carbon budget of the Mackenzie River Basin, American J. Sci. 319 (6) 473-499, DOI: https://doi.org/10.2475/06.2019.02, 2019.
Hotchkiss, E., Hall Jr, R., Sponseller, R. et al.: Sources of and processes controlling CO_2 emissions change with the size of streams and rivers, Nature Geoscience, 8, 696–699, https://doi.org/10.1038/ngeo2507, 2015.
Hugelius, G., Tarnocai, C., Broll, G., Canadell, J. G., Kuhry, P., and Swanson, D. K.: The Northern Circumpolar Soil Carbon Database: spatially distributed datasets of soil coverage and soil carbon storage in the northern permafrost regions, Earth Syst. Sci. Data, 5, 3–13, https://doi.org/10.5194/essd-5-3-2013, 2013.
Huh, Y., Tsoi, M. Y., Zaitsev, A., and Edmond, J. M.: The fluvial geochemistry of the rivers of eastern Siberia: I. Tributaries of the Lena River draining the sedimentary platform of the Siberian Craton, Geochim. Cosmochim. Acta, 62, 1657-1676, doi: 10.1016/S0016-7037(98)00107-0, 1998a.
Huh, Y., Panteleyev, G., Babich, O., Zaitsev, A., and Edmond, J. M.: The fluvial geochemistry of the rivers of Eastern Siberia: II. Tributaries of the Lena, Omoloy, Yana, Indigirka, Kolyma, and Anadyr draining the collisional/accretionary zone of the Verkhoyansk and Cherskiy ranges, Geochim. Cosmochim. Acta, 62, 5063-5075, 1998b.

Huh, Y., and Edmond, J. M.: The fluvial geochemistry of the rivers of Eastern Siberia: III. Tributaries of the Lena and Anabar draining the basement terrain of the Siberian Craton and the Trans-Baikal Highlands, Geochim. Cosmochim. Acta, 63, 967–987, doi:10.1016/S0016-7037(99)00045-9, 1999.

Humborg, C., Morth, C.-M., Sundbom, M., Borg, H., Blenckner, T., Giesler, R., and Ittekkot, V.: CO_2 supersaturation along the aquatic conduit in Swedish watersheds as constrained by terrestrial respiration, aquatic respiration and weathering, Glob. Change Biol., 16, 1966–1978, doi:10.1111/j.1365-2486.2009.02092.x, 2010.

Ivakhov, V. M., Paramonova, N. N., Privalov, V. I., Zinchenko, A. V., Loskutova, M. A., Makshtas, A. P., Kustov, V. Y., Laurila, T., Aurela, M., and Asmi, E.: Atmospheric Concentration of Carbon Dioxide at Tiksi and Cape Baranov Stations in 2010–2017, Russian Meteorol. Hydrol., 44(4), 291–299, DOI: 10.3103/S1068373919040095, 2019.

Jähne, B., Heinz, G., and Dietrich, W.: Measurement of the diffusion coefficients of sparingly soluble gases in water, J. Geophys. Res. Oceans 92, 10767–10776, https://doi.org/10.1029/JC092iC10p10767, 1987.

Johnson, M. S., Billett, M. F., Dinsmore, K. J., Wallin, M., Dyson, K. E., and Jassal, R. S.: Direct and continuous measurement of dissolved carbon dioxide in freshwater aquatic systems-method and applications, Ecohydrology 3(1), 68-78, doi:10.1002/eco, 2009.

Juhls, B., Stedmon, C. A., Morgenstern, A., Meyer, H., Holemann, J., Heim, B., Povazhnyi, V., and Overduin P. P.: Identifying drivers of seasonality in Lena River biogeochemistry and dissolved organic matter fluxes, Front. Environ. Sci., 8, Art No 53, https://doi.org/10.3389/fenvs.2020.00053, 2020.

Karlsson, J., Serikova, S., Rocher-Ros, G., Denfeld, B., Vorobyev, S. N., and Pokrovsky, O. S.: Carbon emission from Western Siberian inland waters, Nature Communication 12, 825, https://doi.org/10.1038/s41467-021-21054-1, 2021.

Klaus, M. and Vachon, D.: Challenges of predicting gas transfer velocity from wind measurements over global lakes, Aquatic Sciences 82, Art No 53, doi:10.1007/s00027-020-00729-9, 2020.

Klaus, M., Seekell, D. A., Lidberg, W., and Karlsson, J.: Evaluations of climate and land management effects on lake carbon cycling need to account temporal variability in CO_2 concentration, Global Biogeochemical Cycles, 33, 243-265, doi:10.1029/2018gb005979, 2019.

Kruse, S., Gerdes, A., Kath, N. J., Epp, L. S., Stoof-Leichsenring, K. R., Pestryakova, L. A., and Herzschuh, U.: Dispersal distances and migration rates at the arctic treeline in Siberia – a genetic and simulation-based study, Biogeosciences, 16, 1211–1224, https://doi.org/10.5194/bg-16-1211-2019, 2019.

Kutscher, L., Mörth, C.-M., Porcelli, D., Hirst, C., Maximov, T. C., Petrov, R. E., and Andersson, P. S.: Spatial variation in concentration and sources of organic carbon in the Lena River, Siberia, J. Geophys. Res. Biogeosciences, 122, 1999-2014, https://doi.org/10.1002/2017JG003858, 2017.

Kutzbach, L., Wille, C., and Pfeiffer, E.-M.: The exchange of carbon dioxide between wet arctic tundra and the atmosphere at the Lena River Delta, Northern Siberia, Biogeosciences 4(5), 869-890, https://doi.org/10.5194/bg-4-869-2007, 2007.

Kuzmin, M. I., Tarasova, E. N., Bychinskii, V. A., Karabanov, E. B., Mamontov, A. A., and Mamontova, E. A.: Hydrochemical regime components of Lena water, Water Resources 36(4), 418-430, https://doi.org/10.1134/S0097807809040058, 2009.

Lara, R. J., Rachold, V., Kattner, G., Hubberten, H. W., Guggenberger, G., Annelie, S., and Thomas, D. N.: Dissolved organic matter and nutrients in the Lena River, Siberian Arctic: Characteristics and distribution, Marine Chemistry 59, 301-309, doi: 10.1016/S0304-4203(97)00076-5, 1998.

Laurion, I., Massicotte, P., Mazoyer, F., Negandhi, K., and Mladenov, N.: Weak mineralization despite strong processing of dissolved organic matter in Eastern Arctic tundra ponds, Limnol. Oceanogr., 66, (S1), S47-S63, doi: 10.1002/lno.11634, 2021.

Leith, F. I., Garnett, M. H., Dinsmore, K. J., Billett, M. F., and Heal, K. V.: Source and age of dissolved and gaseous carbon in a peatland-riparian-stream continuum: a dual isotope (^{14}C and $\delta^{13}C$) analysis, Biogeochemistry, 119, 415–433, doi:10.1007/s10533-014-9977-y, 2014.

Leith, F. I., Dinsmore, K. J., Wallin, M. B., Billett, M; F., Heal, K. V., Laudon, H., Öquist, M. G., and Bishop, K.: Carbon dioxide transport across the hillslope–riparian–stream continuum in a boreal headwater catchment, Biogeosciences, 12, 1–12, doi:10.5194/bg-12-1-2015, 2015.

Lobbes, J. M., Friznar, H. P., and Kattner, G.: Biogeochemical characteristics of the dissolved and particulate organic matter in Russian rivers entering the Arctic Ocean, Geochim. Cosmochim. Acta, 64(17), 2973–2983, 2000.

McClelland, J. W., Holmes, R. M., Peterson, B. J., and Striegltz, M.: Increasing river discharge in the Eurasian Arctic: Consideration of dams, permafrost thaw, and fires as potential agents of change, J. Geophys. Res. Atmospheres, 109 (D18), Art No D18102, doi:10.1029/2004JD004583, 2004.

McClelland, J. W., Déry, S. J., Peterson, B. J., Holmes, R. M., and Wood, E. F.: A pan-Arctic evaluation of changes in river discharge during the latter half of the 20^{th} century, Geophys. Res. Lett., 33, L06715, https://doi.org/10.1029/2006GL025753, 2006.

Murphy, M., Porcelli, D., Pogge von Strandmann, P., Hirst, K., Kutscher, L., Katchinoff, J., Morth, C.-M., Maximov, T., and Andresson, P.: Tracing silicate weathering processes in the permafrost-dominated Lena River watershed using lithium isotopes, Geochim. Cosmochim. Acta, 245, 154-171, doi:10.1016/j.gca.2018.10.024, 2018.

Park, J. H., Jin, H., Yoon, T. K., Begum, M. S., Eliyan, C., Lee, E.-J., Lee, S.-C., and Oh, N.-H.: Wastewater-boosted biodegradation amplifying seasonal variations of pCO2 in the Mekong–Tonle Sap river system, Biogeochemistry, https://doi.org/10.1007/s10533-021-00823-6, 2021.

Park, J.-H., Nayna, O.K., Begum, M.S., Chea, E., Hartmann, J., Keil, R.G., Kumar, S., Lu, X., Ran, L., Richey, J.E., Sarma, V.V.S.S, Tareq, S.M., Xuan, D. T., and Yu, R.: Reviews and syntheses: Anthropogenic perturbations to carbon fluxes in Asian river systems—concepts, emerging trends, and research challenges, Biogeosciences 15, 3049–3069. https://doi.org/10.5194/bg-15-3049-2018, 2018.

Payandi-Rolland, D., Shirokova, L. S., Nakhle, P., Tesfa, M., Abdou, A., Causserand, C., Lartiges, B., Rols, J.L., Guérin, F., Bénézeth, P., and Pokrovsky, O.S.: Aerobic release and biodegradation of dissolved organic matter from frozen peat: Effects of temperature and heterotrophic bacteria, Chem. Geol. 536, Art No 119448, https://doi.org/10.1016/j.chemgeo.2019.119448, 2020.

Pekel, J. F., Cottam, A., Gorelick, N. et al.: High-resolution mapping of global surface water and its long-term changes, Nature, 540, 418–422, https://doi.org/10.1038/nature20584, 2016.

Pipko, I. I., Pugach, S. P., Savichev, O. G., Repina, I. A., Shakhova, N. E., Moiseeva, Yu. A., Barskov, K. V., Sergienko, V. I., and Semiletov, I. P.: Dynamics of dissolved inorganic carbon and CO2 fluxes between the water and the atmosphere in the main channel of the Ob River, Doklady Chemistry 484(2), 52-57, doi:10.1134/S0012500819020101, 2019.

Pipko; I. I., Pugach, S. P., Dudarev, O. V., Charkin, A. N., and Semiletov, I. P.: Carbonate parameters of the Lena River: Characteristics and distribution, Geochem. Internat., 48(11), 1131-1137, https://doi.org/10.1134/S0016702910110078, 2010.

Qin, J., Huh, Y., Edmond, J. M., Du, G., and Ran, J.: Chemical and physical weathering in the Min Jiang, a headwater tributary of the Yangtze River, Chem. Geol., 227, 53–69, doi:10.1016/j.chemgeo.2005.09.011, 2006.

Rachold, V., Alabyan, A., Hubberten, H.-W., Korotaev, V. N., and Zaitsev, A. A.: Sediment transport to the Laptev Sea - hydrology and geochemistry of the Lena River, Polar Research, 15(2), 183-196, doi: https://doi.org/10.3402/polar.v15i2.6646, 1996.

Raymond, P. A., McClelland, J. W., Holmes, R. M., Zhulidov, A. V., Mull, K. et al.: Flux and age of dissolved organic carbon exported to the Arctic Ocean: A carbon isotopic study of the five largest arctic rivers, Global Biogeochemical Cycles, 121, Art No GB4011, https://doi.org/10.1029/2007GB002934, 2007.

Raymond, P. A., Hartmann, J., Lauerwald, R., Sobek, S., McDonald, C., Hoover, M., Butman, D., Striegl, R., Mayorga, E., Humborg, C., Kortelainen, P., Dürr, H., Meybeck, M., Ciais, P., and Guth, P.: Global carbon dioxide emissions from inland waters, Nature, 503, 355–359, doi:10.1038/nature12760, 2013.

Rocher-Ros, G., Sponseller, R. A., Lidberg, W., Mörth, C-M., and Giesler, R.: Landscape process domains drive patterns of CO_2 evasion from river networks, Limnol. Oceanogr. Lett., 4, 87-95, https://doi.org/10.1002/lol2.10108, 2019.

Sachs, T., Wille, C., Boike, J., and Kutzbach, L.: Environmental controls on ecosystem-scale CH_4 emission from polygonal tundra in the Lena River Delta, Siberia, J. Geophys. Research Biogeosciences, 113, Art No G00A03, https://doi.org/10.1029/2007JG000505, 2008.

Santoro, M., Beer, C., Cartus, O., Schmullius, C., Shvidenko, A., McCallum, I., Wegmueller, U., and Wiesmann, A.: The BIOMASAR algorithm: An approach for retrieval of forest growing stock volume using stacks of multi-temporal SAR data, In: Proceedings of ESA Living Planet Symposium, 28 June-2 July 2010 (https://www.researchgate.net/publication/230662433, http://pure.iiasa.ac.at/id/eprint/9430/), 2010.

Schuur, E. A. G., McGuire, A. D., Schädel, C., Grosse, G., Harden, J. W., Hayes, D. J., Hugelius, G., Koven, C. D., Kuhry, P., Lawrence, D. M., Natali, S. M., Olefeldt, C., Romanovsky, V. E., Schaefer, K., Turetsky, M. R., Treat, C. C., and Vonk. J. E.: Climate change and the permafrost carbon feedback, Nature 520, 171–179, http://dx.doi.org/10.1038/nature14338, 2015.

Semiletov, I. P.: Aquatic sources and sinks of CO_2 and CH_4 in the polar regions, J. Atmospheric Sci., 56, 286-306, https://doi.org/10.1175/1520-0469(1999)056<0286:ASASOC>2.0.CO;2, 1999.

Semiletov, I. P., Pipko, I. I., Shakhova, N. E., Dudarev, O. V., Pugach, S. P., Charkin, A. N., McRoy, C. P., Kosmach, D., Gustafsson Ö.: 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, Biogeosciences, 8, 2407–2426, https://doi.org/10.5194/bg-8-2407-2011, 2011.

Serikova, S., Pokrovsky, O. S., Ala-aho, P., Kazantsev, V., Kirpotin, S. N. Kopysov, S. G., Krickov, I. V., Laudon, H., Manasypov, R. M., Shirokova, L. S., Sousby, C., Tetzlaff, D., and Karlsson, J.: High riverine CO_2 emissions at the permafrost boundary of Western Siberia, Nat. Geosci., 11, 825–829, https://doi.org/10.1038/s41561-018-0218-1, 2018.

Serikova S., Pokrovsky O. S., Laudon, H., Krickov, I. V., Lim, A. G., Manasypov, R. M., and Karlsson, J.: C emissions from lakes across permafrost gradient of Western Siberia, Nat. Commun., 10, 1552, https://doi.org/10.1038/s41467-019-09592-1, 2019.

Siewert, M. B., Hugelius, G., Heim, B., and Faucherre, S.: Landscape controls and vertical variability of soil organic carbon storage in permafrost-affected soils of the Lena River Delta, Catena, 147, 725-741, doi:10.1016/j.catena.2016.07.048, 2016.

Smith, L. C., Pavelksky, T. M.: Estimation of river discharge, propagation speed,a nd hydraulic geometry from space: Lena River, Siberia, Water Resources Res., 44(3), W03427, https://doi.org/10.1029/2007WR006133, 2008.

Spence, J. and Telmer, K.: The role of sulfur in chemical weathering and atmospheric CO_2 fluxes: evidence from major ions, $\delta^{13}C_{DIC}$, and $\delta^{34}S_{SO4}$ in rivers of the Canadian Cordillera, Geochim. Cosmochim. Acta, 69, 5441–5458, doi:10.1016/j.gca.2005.07.011, 2005.

Stackpoole, S. M., Butman, D. E., Clow, D. W., Verdin, K. L., Gaglioti, B. V., Genet, H., and Striegl, R. G.: Inland waters and their role in the carbon cycle of Alaska, Ecological Applications 27(5), 1403-1420, doi:10.1002/eap.1552/full, 2017.

Striegl, R. G., Dornblaser, M. M., McDonald, C. P., Rover, J. R., and Stets E. G.: Carbon dioxide and methane emissions from the Yukon River system, Global Biogeochem. Cycles, 26, GB0E05, doi:10.1029/2012GB004306, 2012.

Sun X., Mörth C.-M., Porcelli D., Kutscher L., Hirst C., Murphy M. J., Maximov T., Petrov R. E., Humborg C., Schmitt M. and Andersson P. S.: Stable silicon isotopic compositions of the Lena River and its tributaries: Implications for silicon delivery to the Arctic Ocean, Geochim. Cosmochim. Acta 241, 120–133, doi: 10.1016/j.gca.2018.08.044, 2018.

Suzuki, K., Matsuo, K., Yamazaki, D., Ichii, K., Iijima, Y., Papa, F., Yanagi, Y., and Hiyama, T.: Hydrological variability and changes in the Arctic circumpolar tundra and the three largest Pan-Arctic river basins from 2002 to 2016, Remote Sensing 10, Art No 402, doi:10.3390/rs10030402, 2018.

Vachon, D., Prairie, Y. T., and Cole, J. J.: The relationship between near-surface turbulence and gas transfer velocity in freshwater systems and its implications for floating chamber measurements of gas exchange, Limnology and Oceanography, 55(4), 1723–173, doi:10.4319/lo.2010.55.4.1723, 2010.

Van der Molen, M. K., van Huissteden J., Parmentier F. J. W., Petrescu, A. M. R., Dolman, A. J., Maximov, T. C. et al.: The growing season greenhouse gas balance of a continental tundra site in the Indigirka lowlands, NE Siberia, Biogeosciences 4(6), 985-1003, https://doi.org/10.5194/bg-4-985-2007, 2007.

Vonk, J. E., Tank, S. E., Mann, P. J., Spencer, R. G. M., Treat, C. C., Striegl, R. G., Abbott, B. W., and Wickland, K. P.: Biodegradability of dissolved organic carbon in permafrost soils and aquatic systems: a meta-analysis, Biogeosciences, 12, 6915–6930, https://doi.org/10.5194/bg-12-6915-2015, 2015.

Vonk, J. E., Tank, S. E., and Walvoord, M. A.: Integrating hydrology and biogeochemistry across frozen landscapes, Nat. Commun. 10, 1–4. https://doi.org/10.1038/s41467-019-13361-5, 2019.

Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean, J. Geophys. Res. 97, 7373–7382, https://doi.org/10.4319/lom.2014.12.351, 1992.

Ward, C. P., Nalven, S. G., Crump, B. C., Kling, G. W.,and Cory, R. M.: Photochemical alteration of organic carbon draining permafrost soils shifts microbial metabolic pathways and stimulates respiration, Nat. Commun., 8, 772, https://doi.org/10.1038/s41467-017-00759-2, 2017.

Wild, B., Andersson, A., Bröder, L., Vonk, J., Hugelius, G., McClelland, J. W., Song, W., Raymond P. A., and Gustafsson, Ö.: Rivers across the Siberian Arctic unearth the patterns of carbon release from the thawing permafrost, PNAS 116(21), 10280-10285, https://doi.org/10.1073/pnas.1811797116, 2019.

Wille, C., Kutzbach, L., Sachs, T., Wagner, D., and Pfeiffer, E.M.: Methane emission from Siberian arctic polygonal tundra: eddy covariance measurements and modeling, Global Change Biology 14(6), 1395-1408, https://doi.org/10.1111/j.1365-2486.2008.01586.x, 2008.

Wu, L. and Huh, Y.: Dissolved reactive phosphorus in large rivers of East Asia, Biogeochemistry 85, 263-288, doi:10.1007/s10533-007-9133-z, 2007.

Yang, D. Q., Kane, D. L., Hinzman, L. D., Zhang, X. B., Zhing, T. J., and Ye, H. C.: Siberian Lena River hydrological regime and recent change, J. Geophys. Res. Atmopsheres 107, D23, Art No 4694, https://doi.org/10.1029/2002JD002542, 2002.

Yamamoto, S., Alcauskas, J. B., and Crozier, T.E.: Solubility of methane in distilled water and seawater, J. Chem. Eng. Data, 21(1), 78– 80, doi:10.1021/je60068a029, 1976.

Ye, B., Yang, D., Zhang, Z., and Kane, D. L.: Variation of hydrological regime with permafrost coverage over Lena basin in Siberia, J. Geophys. Res., 114, D07102, doi:10.1029/2008JD010537, 2009.

Yoon, T. K., Jin, H., Oh, N.-H., and Park, J.-H.: Technical note: Assessing gas equilibration systems for continuous pCO_2 measurements in inland waters, Biogeosciences, 13, 3915–3930, https://doi.org/10.5194/bg-13-3915-2016, 2016.

Zhang, T., Frauenfeld, O. W., Serreze, M. C., Etringer, A., Oelke, C. et al.: Spatial and temporal variability in active layer thickness over the Russian Arctic drainage basin, J. Geophys. Res., 110, D16101, doi: 10.1029/2004JD005642, 2005.

Zubrzycki, S., Kutzbach, L., Grosse, G., Desyatkin, A., and Pfeiffer, E. M.: Organic carbon and total nitrogen stocks in soils of the Lena River Delta, Biogeosciences 10, 3507-3524, doi:10.5194/bg-10-3507-2013, 2013.

Table 1. Measured water temperature, pCO_2, calculated CO_2 flux, CH_4, DOC, and DIC concentrations and pH in the Lena River main stem (average ± s.d.; (n) is number of measurements).

River transect	T_{water}, °C	pCO_2, µatm	FCO_2, g C m^{-2} d^{-1} k = 4.464
Lena upstream of Kirenga (0-578 km)	12.65±0.22 (99)	714±22 (99)	0.849±0.061 (99)
Lena Kirenga – Vitim (579-1132 km)	9.17±0.15 (87)	806±8.8 (87)	1.19±0.024 (87)
Lena Vitim -Nuya (1132-1331 km)	8.10±0.115 (27)	797±22 (27)	1.22±0.072 (27)
Lena Nuya – Tuolba (1331-2008 km)	9.61±0.09 (95)	846±12 (95)	1.29±0.034 (95)
Lena Tuolba – Aldan (2008-2381 km)	10.6±0.21 (52)	1003±28 (52)	1.69±0.081 (5)

	CH_4, µmol L^{-1}	DOC, mg L^{-1}	DIC, mg L^{-1}	pH
Lena upstream of Kirenga (0-578 km)	0.068±0.003 (6)	13.9±1.4 (6)	20.0±1.2 (6)	8.12±0.203 (7)
Lena Kirenga – Vitim (579-1132 km)	0.040±0.002 (12)	7.55±0.246 (14)	6.30±0.485 (14)	7.77±0.040 (14)
Lena Vitim -Nuya (1132-1331 km)	0.038±0.003 (5)	9.02±0.29 (3)	4.55±0.70 (3)	7.69±0.063 (3)
Lena Nuya – Tuolba (1331-2008 km)	0.037±0.002 (6)	10.4±0.78 (2)	5.09±1.157 (2)	7.62±0.052 (2)
Lena Tuolba – Aldan (2008-2381 km)	0.088±0.034 (5)	11.6±0.27 (5)	5.24±0.102 (5)	7.49±0.044 (5)

Table 2. Measured water temperature, pCO_2, calculated CO_2 flux, CH_4, DOC, DIC concentration and pH in the tributaries (average ± s.d.; (n) is number of measurements).

Tributary	T_{water}, °C	pCO_2, µatm	FCO_2, g C m^{-2} d^{-1}
№4 Orlinga (208 km)	8.0±0.0 (13)	515±2.9 (13)	0.347±0.01 (13)
№5 Nijnaya Kitima (228 km)	6.8±0.0 (11)	462±9.4 (11)	0.193±0.03 (11)
№8 Taiur (416 km)	8.5±0.0 (10)	575±31 (10)	0.523±0.095 (10)
№10 Bol. Tira (529 km)	11.9±0.0 (15)	788±12 (15)	1.04±0.03 (15)
№12 Kirenga (579 km)	10.2±0.0 (323)	448±4 (323)	0.131±0.01 (323)
№25 Thcayka (1025 km)	8.6±0.01 (8)	856±13 (8)	1.37±0.04 (8)
№28 Tchuya (1110 km)	5.9±0.0 (5)	751±5.7 (5)	1.16±0.019 (5)
№29 Vitim (1132 km)	6.8±0.0 (10)	654±10 (10)	0.812±0.03 (10)
№32 Ykte (1265 km)	4.9±0.0 (11)	676±4.8 (11)	0.943±0.02 (11)
№34 Kenek (1312 km)	7.60±0.0 (11)	710±2.6 (11)	0.964±0.01 (11)
№36 Nuya (1331 km)	11.8±0.0 (10)	752±6.0 (10)	0.947±0.02 (10)
№38 Bol. Patom (1670 km)	6.9±0.0 (5)	730±12 (5)	1.05±0.04 (5)
№39 Biriuk (1712 km)	14.2±0.0 (5)	929±19 (5)	1.32±0.05 (5)
№40 Olekma (1750 km)	6.4±0.0 (11)	802±14 (11)	1.30±0.05 (11)
№43 Markha (1948 km)	17.5±0.0 (15)	844±15 (15)	0.998±0.03 (15)
№44 Tuolba (2008 km)	12.3±0.0 (305)	1181±6 (305)	2.08±0.02 (305)
№46 Siniaya (2118 km)	18.5±0.0 (24)	894±19 (24)	1.08±0.04 (24)
№48 Buotama (2170 km)	18.5±0.0 (24)	1160±25 (24)	1.66±0.06 (24)
№52-54 Aldan (2381 km)	14.8±0.02 (316)	1715±12 (316)	3.23±0.03 (316)

Table 2, continued.

	CH_4, μmol L^{-1}	DOC, mg L^{-1}	DIC, mg L^{-1}	pH
№4 Orlinga (208 km)	0.064	13.4	27.9	8.64
№5 Nijnaya Kitima (228 km)	0.033	16.7	13.1	8.48
№8 Taiur (416 km)	0.079	10.0	11.2	8.36
№10 Bol. Tira (529 km)	0.084	22.7	14.9	8.13
№12 Kirenga (579 km)	0.036	5.13	6.86	7.97
№25 Thcayka (1025 km)	0.066	16.7	22.5	8.30
№28 Tchuya (1110 km)	0.037	7.08	3.44	7.57
№29 Vitim (1132 km)	0.057	10.1	2.18	7.70
№32 Ykte (1265 km)	0.037	5.49	15.3	7.86
№34 Kenek (1312 km)	0.053	21.1	16.0	8.12
№36 Nuya (1331 km)	0.048	26.6	11.7	7.80
№38 Bol. Patom (1670 km)	0.026	6.99	4.56	7.76
№39 Biriuk (1712 km)	0.047	29.2	11.3	7.87
№40 Olekma (1750 km)	0.046	13.3	3.3	7.53
№43 Markha (1948 km)	0.088	27.4	10.9	8.00
№44 Tuolba (2008 km)	0.035	14.5	14.7	7.98
№46 Siniaya (2118 km)	0.113	33.2	7.73	7.97
№48 Buotama (2170 km)	0.124	12.2	31.6	8.45
№52-54 Aldan (2381 km)	0.088 (4)	9.07±0.75 (4)	6.67±0.13 (4)	7.59±0.02 (4)

Footnote : in all tributaries except Aldan, there was only one measurement of CH_4, DOC, DIC and pH

Table 3. Pearson correlations between pCO_2 and landscape parameters of the Lena tributaries. Significant correlations ($p < 0.05$) are marked by asterisk. Methane concentration did not exhibit any significant correlation with all tested parameters.

% coverage of the watershed and climate	**$R_{Pearson}$**
Broadleaf Forest	0.04
Humid Grassland	-0.52*
Shrub Tundra	-0.05
Riparian Vegetation	0.87*
Croplands	-0.31
Bare Soil and Rock	0.54*
Evergreen Needle-leaf Forest	-0.59*
Deciduous Broadleaf Forest	-0.14
Mixed Forest	-0.34
Deciduous Needle-leaf Forest	0.56*
Bogs and marches	0.44
Palsa bogs	0.29
Recent burns	-0.25
Water bodies	0.63*
Aboveground biomass	-0.55*
Soil C stock, 0-30 cm	0.54*
Soil C stock, 0-100 cm	0.65*
Carbonate rocks	0.20
Continuous permafrost	0.66*
Discontinuous permafrost	-0.27
Sporadic permafrost	-0.43
Isolated permafrost	-0.19
Mean annual air temperature	-0.76*
Mean annual precipitation, mm	0.10

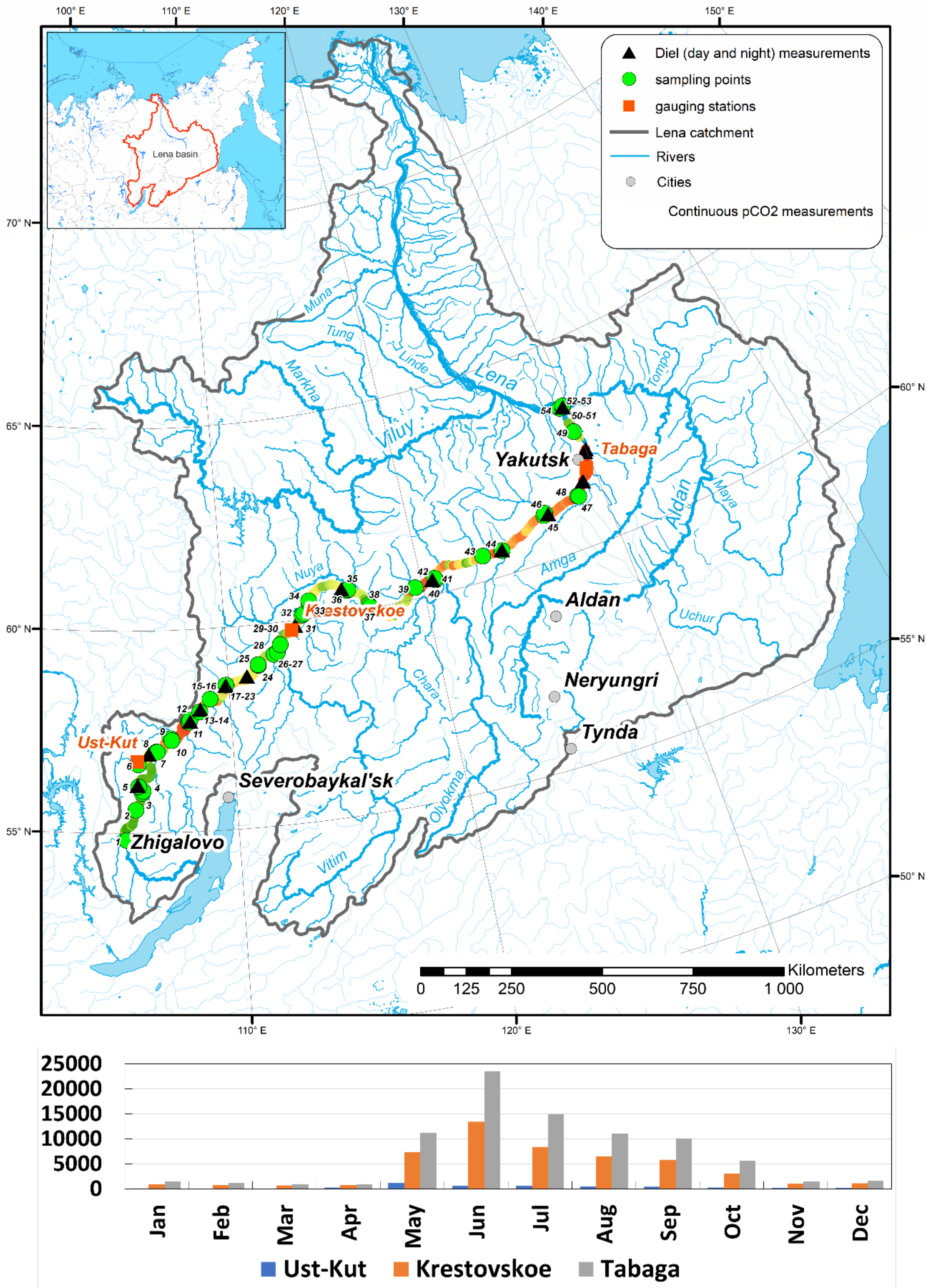

Fig. 1. Map of the studied Lena River watershed with continuous pCO_2 measurements in the main stem. Mean multi-annual hydrographs of Ust-Kut, Krestovskoe and Tabaga station (labelled in red on the map) are provided in the insert.

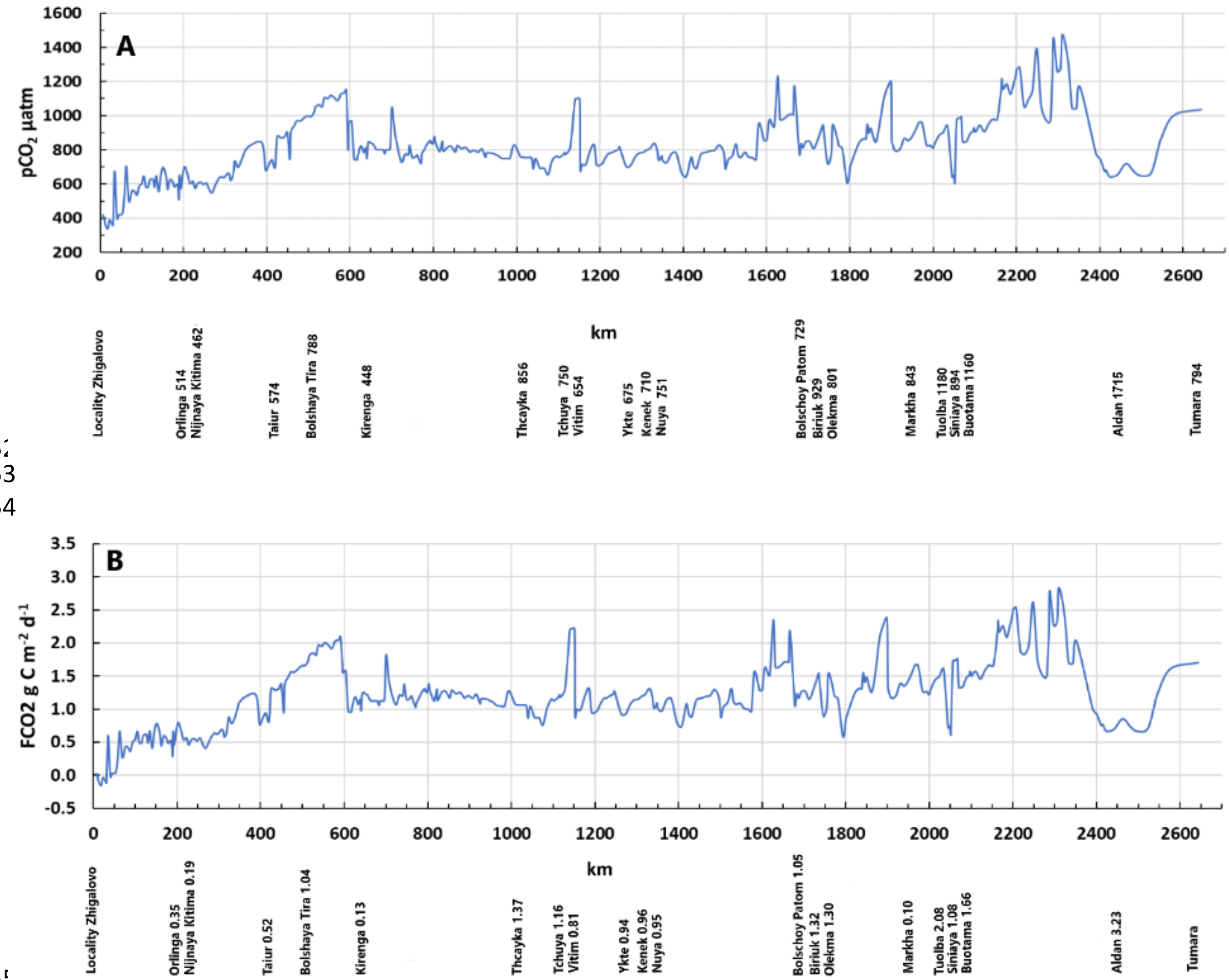

Figure 2. A 20-km averaged pCO_2 profile (**A**) and calculated CO_2 fluxes (**B**) of the Lena River main stem of over 2600 km distance, from Zhigalovo to the Tumara River. The average pCO_2 (µatm) and fluxes (g C m^{-2} d^{-1}) of the main sampled tributaries are provided as numbers below X axes. Note that peaks of CO_2 concentration at the main stem are not linked to conflux with tributaries.

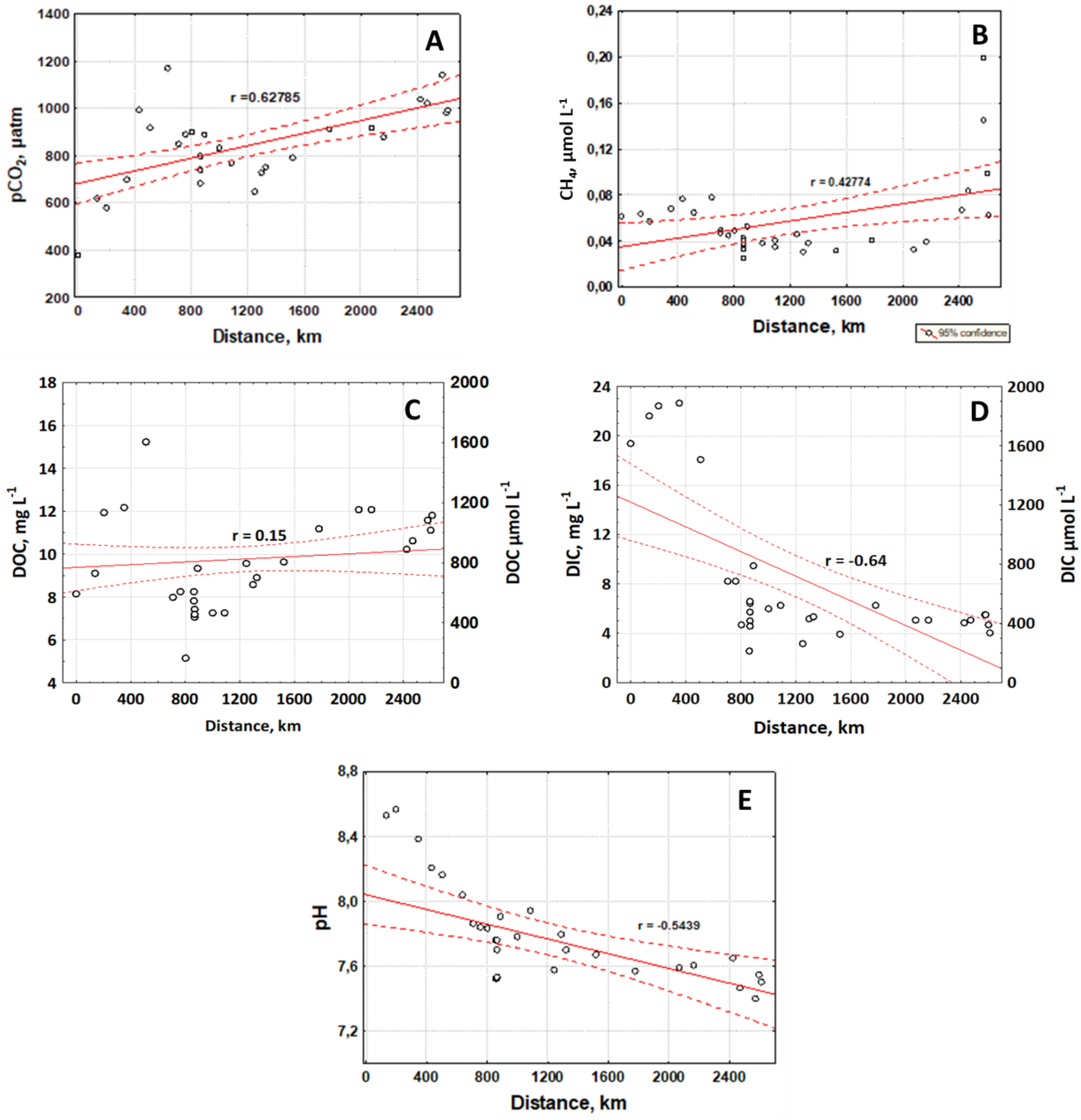

Figure 3. Averaged (over 20-km distance) CO_2 (A), CH_4 (B), DOC (C), DIC (D) and pH (E) concentration over the distance of the boat route at the Lena River, from the south-west to north-east.

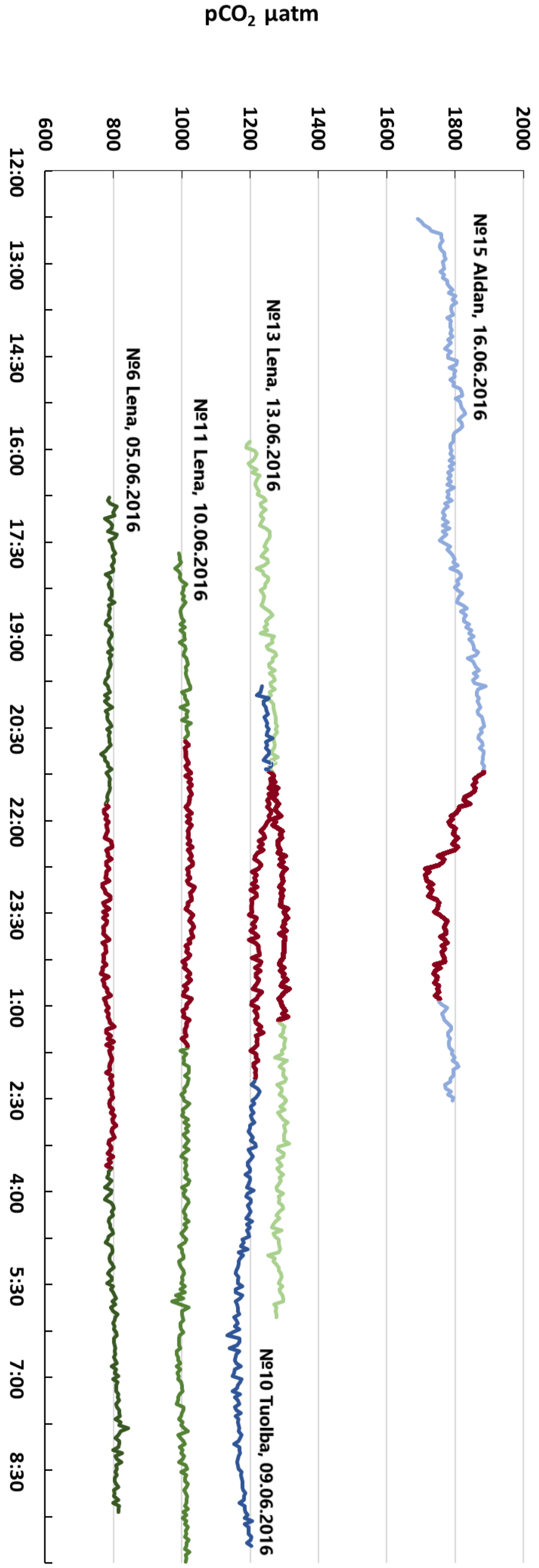

Figure 4. Continuous pCO_2 concentration in the Lena River and two tributaries from late afternoon to morning next day. Red part of the line represents night time. Variations of water temperature did not exceed 2 °C.

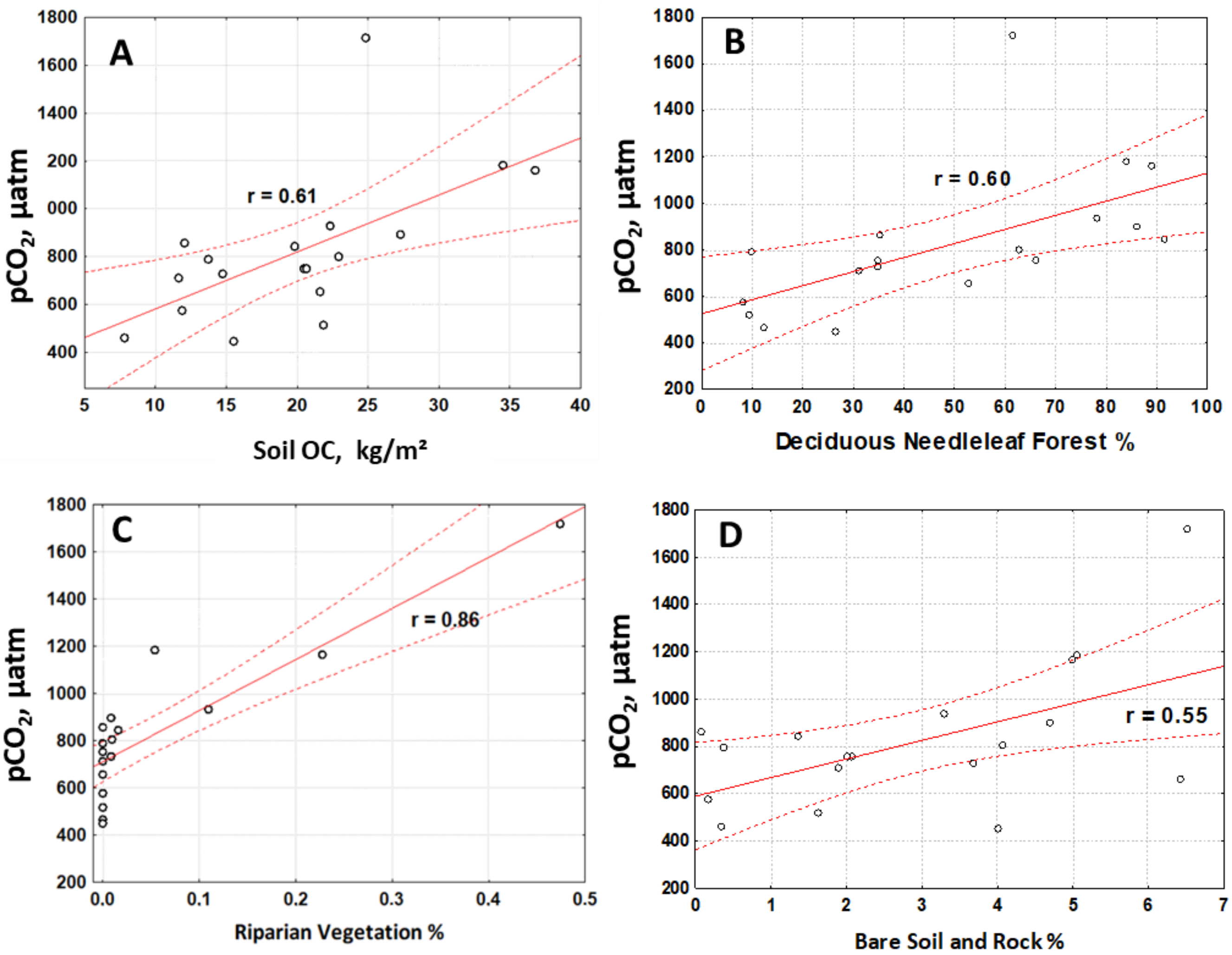

Figure 5. Significant ($p < 0.05$) positive control of landscape parameters – OC stock in 0-100 cm of soil (A), and proportion of deciduous needle-leaf forest (B), riparian vegetation (C) and bare soil and rock (D) in the watershed on pCO_2 in the Lena River tributaries.